Evidence for slowdown in stratospheric ozone loss: First stage of ozone recovery

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[1] Global ozone trends derived from the Stratospheric Aerosol and Gas Experiment I and II (SAGE I/II) combined with the more recent Halogen Occultation Experiment (HALOE) observations provide evidence of a slowdown in stratospheric ozone losses since 1997. This evidence is quantified by the cumulative sum of residual differences from the predicted linear trend. The cumulative residuals indicate that the rate of ozone loss at 35–45 km altitudes globally has diminished. These changes in loss rates are consistent with the slowdown of total stratospheric chlorine increases characterized by HALOE HCl measurements. These changes in the ozone loss rates in the upper stratosphere are significant and constitute the first stage of a recovery of the ozone layer. *INDEX TERMS:* 0340 Atmospheric Composition and Structure: Middle atmosphere—composition and chemistry; 0341 Atmospheric Composition and Structure: Middle atmosphere—constituent transport and chemistry (3334); 1610 Global Change: Atmosphere (0315, 0325); *KEYWORDS:* stratospheric ozone trends, CFCs, Montreal Protocol

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1. Introduction

[2] Stratospheric ozone depletion has been a significant international concern ever since it was first recognized as a consequence of anthropogenic Ozone Depleting Substances (ODS) [*Molina and Rowland*, 1974]. Although the Montreal Protocol and its Amendments have been effective in reducing the production and emission of ODS [*Montzka et al.*, 1999], we have seen, as expected, continued, relatively steady declines in upper stratospheric ozone levels [*Newchurch et al.*, 2000; *Randel et al.*, 1999; *Staehelin et al.*, 2001; *World Meteorological Organization* (*WMO*), 1999]. We expect to see signs of ozone recovery first in the 40-km region [*Jucks et al.*, 1996]; however, because of interactions with increasing greenhouse gases, decreasing temperature, and circulation changes, these climate changes can mask an ozone recovery from chlorine-catalyzed loss [*Shindell et al.*, 1998]. Furthermore, detecting signs of ozone recovery first in the 40-km region is extremely important for confirming our understanding of ozone chemistry in the upper stratosphere, a research area with significant heritage in measurements, modeling, and laboratory investigations [*Chen et al.*, 1997; *Crutzen et al.*, 1995; *Dessler et al.*, 1998; *Eluszkiewicz and Allen*, 1993; *Froidevaux et al.*, 1985; *Grooß et al.*, 1999; *Jackman et al.*, 1996; *Jucks et al.*, 1996; *Kegley-Owen et al.*, 1999; *Khosravi et al.*, 1998; *Lipson et al.*, 1999; *Michelsen et al.*, 1994; *Minschwaner and Siskind*, 1993; *Natarajan and Callis*, 1991; *Randel et al.*, 1999; *Reinsel et al.*, 1999; *Russell et al.*, 1996b; *Siskind et al.*, 1995; *Siskind and Summers*, 1998; *Stolarski et al.*, 1992; *Stolarski and Douglass*, 1986; *Summers et al.*, 1997; *Viggiano et al.*, 1995; *Waters et al.*, 1996; *Wennberg et al.*, 1994] and in the effects on ground-level ultraviolet radiation [*McKenzie et al.*, 1999; *Rozema et al.*, 2002]. Most recently, for example, *Shindell and Grewe* [2002] show that the 40-km region is not only the optimum location to identify recovery but also is the ideal location to ascribe attribution due to CFC reductions and complicating greenhouse gas effects [*Shindell et al.*, 1999; *Shindell*, 2001]. However, because this high-altitude region contains only a few percent of the total ozone column, changes in this region should not be over interpreted as indications of a recovery of the entire ozone layer [*WMO*, 1999].

[3] Decreases in tropospheric chlorine loading after 1993 have been convincingly reported from ground-based measurements [*Montzka et al.*, 1996; *Prinn et al.*, 2000]. In the upper stratosphere, however, models predict a change in the halogen trends to begin between 1998 and 2001. *Considine et al.* [1999] showed statistically significant slowdowns in increasing HCl and HF concentrations by using the version-18 HALOE measurements at 54–56 km by 1997. *Waugh et al.* [2001] indicated the increase of total chlorine abundance during 1992–1997 and confirmed its subsequent decrease as early as 1997 at 55 km. They

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focused on the change in trends of these reservoir species for halogen atoms, not on the associated change in ozone depletion. Models also predict an inflection in the 40-km ozone trend to occur in the late 1990s, with a sharper inflection expected when time-dependent CO_2 trends are considered [*WMO*, 1999, Figure 12–36]. The purpose of this study is to show direct evidence of a slowdown in upper-stratospheric ozone depletion by using the SAGE I/II, HALOE, and Dobson Umkehr ozone measurements for 1979–2003.

2. Measurements

[4] The Stratospheric Aerosol and Gas Experiment I and II (SAGE I/II) instruments comprise the longest (1979 to 2000) source of global upper-stratospheric ozone measurements. For complete version-6.1 data set and details of the retrieval algorithm, see http://www-sage2.larc.nasa.gov. As a result of extensive analyses with respect to ground-based Dobson Umkehr, microwave, and lidar measurements and to other satellite measurements (Atmospheric Trace Molecule Spectroscopy Experiment, ATMOS; Halogen Occultation Experiment, HALOE; Microwave Limb Sounder, MLS; and Solar Backscatter UltraViolet, SBUV) these SAGE observations are widely regarded as the most accurate measurements available [*Cunnold et al.*, 2000a; *Harris et al.*, 1998; *WMO*, 1999]. HALOE retrieves vertical HCl and HF profiles as well as ozone profiles [*Russell et al.*, 1996a; *Russell et al.*, 1993; *Russell et al.*, 1996b]. Because of the robust geophysical relationship between these halogen reservoir species and inorganic halogen emissions [*Zander et al.*, 1996], stratospheric HCl provides a good indication of trends in total chlorine loading, Cl_y. The version-19 HALOE data for 1991–2003 are available at http://haloedata.larc.nasa.gov.

[5] We analyze SAGE I/II version-6.1 and HALOE version-19 ozone measurements over an altitude range between 20 and 50 km and in a latitude range between 60°S and 60°N, within 10° × 1 km latitudinal and vertical boxes. Although the SAGE I and SAGE II time series are not contiguous, the veracity of the continuity of these records was carefully inspected by comparison to independent satellite and ground-based observations and determined to represent the actual changes in stratospheric ozone [see also *Cunnold et al.*, 2000a]. Owing to the occasional contamination of ozone measurements by aerosols, the lower stratospheric ozone data during the Pinatubo period are excluded from the SAGE II analysis [*Harris et al.*, 1998; *Cunnold et al.*, 2000b]. In order to remove volcanic aerosol and cloud effects, SAGE II ozone data are discarded (1) if aerosol extinction at 0.525 μm are greater than 6×10^{-3} km^{-1} and (2) if aerosol extinction at 0.525 μm are less than 6×10^{-3} km^{-1} but greater than 1×10^{-3} km^{-1} and if the aerosol extinction ratios (0.525/1.02 μm) are less than 1.4. In addition, the whole profile is omitted if the 1-σ measurement error exceeds 10% between 30 and 50 km altitude [*Wang et al.*, 2002].

[6] Dobson Umkehr measurements of layer-8 (~40 km) ozone are selected from three stations, Arosa (47°N), Boulder (40°N), and Tateno (36°N) (data at http://www.msc-smc.ec.gc.ca/woudc/, the World Ozone and Ultraviolet Radiation Data Centre). A level shift intervention term is included and corrected for station Tateno due to change of instrument from January 1994 [see *Reinsel*, 2002]. In order to avoid the periods of most extreme volcanic aerosol contamination, Umkehr layer-8 ozone data are omitted from November 1982 to June 1983 and from November 1991 to January 1993 [*Reinsel*, 2002].

3. Analysis

[7] The SAGE I/II and HALOE trend calculations employ a linear regression model similar to the underlying model typically used in ozone trend analysis [*Harris et al.*, 1998; *Newchurch et al.*, 2000; *WMO*, 1999]. Ozone variability caused by the Quasi Biennial Oscillation (QBO) which appears as one of several exogenous effects included in the underlying model, has little influence on ozone trend estimates because of its relatively short periodicity (about 2.5 years compared to about 20 year trend calculations). The otherwise small QBO effect (~4–8%) [*WMO*, 1999], however, becomes an important contributor to systematic ozone variations in the six-year testing period (January 1997–February 2003) introduced here for diagnosing and quantifying changes in ozone trends. The underlying standard regression model, unfortunately, does not properly account for the seasonally modulated QBO signals in the extratropics (i.e., 30-, 20-, and 8-months components), resulting in underestimation of the magnitude and a discrepancy in phase [*Randel and Wu*, 1996; *Tung and Yang*, 1994a; *Tung and Yang*, 1994b]. In the present study, the QBO signals are identified at the 95% confidence level by a stepwise regression method [*Draper and Smith*, 1981] which examines periodicities between 3 to 30 months. This stepwise process is similar to the spectral analysis of *Kane et al.* [1998] who identified periodicities of the QBO with sine waves and calculated their amplitudes based on a multiple regression analysis.

[8] Our trend model can be described as

$$[O_3]_t = \mu + \omega t + [\text{Seasonal terms}] + [\text{QBO periodic terms}] + \gamma[F10.7]_t + N_t,$$

where μ is the mean level, ω is a linear trend coefficient, and the seasonal terms represent the 12-, 6-, 4-, and/or 3-months cosine terms each with a time lag to obtain the best estimate of its coefficient. The QBO periodic terms consist of cosines with time lags to represent QBO signal with periods between 3 and 30 months excluding 12-, 6-, 4-, and/or 3-months terms, which are included in the seasonal coefficients. The traditional approach of using Singapore winds with a fitted lag produces similar results, but with less precise trend estimates and more fluctuations in the residuals. The details of the exogenous variable treatment and the resulting residual ozone time series appear in Appendix A. $[F10.7]_t$ is the F10.7-cm radio flux density which is used to provide a solar variation proxy, and γ is a solar signal regression coefficient. An aerosol term is not included in the regression because SAGE ozone data above 30 km are essentially free of aerosol interference [*Cunnold et al.*, 1996] and, in the lower stratosphere where aerosol interference occurs, we have omitted data during the Pinatubo period [*Cunnold et al.*,

2000b]. N_t is the autocorrelated error term, for which a first order autoregressive process is assumed ($N_t = a_1 N_{t-1} + \varepsilon_t$). The ε_t residuals, after removing the autoregressive component, $a_1 N_{t-1}$, are the residuals that are used to compute the cumulative sums of residuals described in Appendix B.

4. Results

[9] Compared to the previous SAGE trends for 1979–1996 [*Randel et al.*, 1999] or 1979–1998 [*Newchurch et al.*, 2000] the current SAGE results for 1979–2000 (Figure 1) show somewhat less negative ozone trends. For example, near 40 km at 45°N, the SAGE trend estimates presented here are about −7%/decade for 1979–2000 compared to −8 to −9%/decade previously reported for shorter periods [see also *Li et al.*, 2002]. The detection of ozone trend changes is possible by comparing the linear trends in two different periods. However, if there is a real change in ozone trend for the test period (1997–2003), the trend is not necessarily linear because, since 1997, all data are close to the turnaround point. The failure of the linear trend assumption since 1997 results in biased trend and error estimates. Therefore, we quantify the change in ozone trends after December 1996 in a self-consistent manner using cumulative sums (CUSUM) which does not require a linear assumption on trend after 1997.0 [e.g., *Reinsel*, 2002].

[10] For better detection of ozone trend changes, ozone records should be extended after June 2000 (end of the available SAGE II data). We determine from calculation that the HALOE and SAGE ozone measurements at 35–45 km show excellent agreement except a slight difference in mean level (∼3%). No significant trend difference is detected between the SAGE II and HALOE ozone series for 1991–2000. The left panels in Figure 2 show the SAGE (black line) and HALOE (red line) residuals, which are independently obtained by using the trend model described in the previous section. The HALOE residuals remarkably resemble the SAGE residuals for 1991–2000. The trend estimates of the SAGE (HALOE) ozone residuals for 1991–2000 are −3.24 ± 2.55 (−3.39 ± 1.45), −4.08 ± 2.20 (−4.25 ± 0.96), and −4.92 ± 2.44 (−3.41 ± 1.56) in %/decade ±2σ at 30–50°N, 30°S–30°N, and 30–50°S, respectively. *WMO* [1999] also indicates that the differences in SAGE and HALOE ozone trends are less than 2%/decade at 25–50 km, 60°S–60°N and they are not statistically significant. Note that the variance of the HALOE residuals is slightly less than that of the SAGE residuals. In this study we use the SAGE I/II data for 1979–2000.5 and HALOE data afterwards.

[11] The ozone residuals plus the linear trends (left panels in Figure 2) result from the difference between the monthly mean of the observed ozone and the ozone seasonal, QBO, solar, and AR(1) terms for the total periods, 1979–2000 (SAGE) and 1991–2003 (HALOE). The blue line is the linear trend line and the trend value ± two standard error are indicated numerically for the regression-model period (1979–1996). In all three of the latitude regions in Figure 2 more negative ozone trends are shown during the model period than during the test period. If the model-period trend line is extended after December 1996 (i.e., linear trend forecast), the difference between the residual ozone and that trend line will indicate how the ozone depletion during the later test period compares with that in the earlier period. If the ozone depletion rate is less, the black/red traces during the test period would systematically be above the blue trend line. A cumulative sum (CUSUM) of ozone residuals can assess the systematic departure of ozone from the trend line. The right panels in Figure 2 show the CUSUM over the three latitude regions for the model and test periods. The magnitude of the residual CUSUM is a function of the sampling periodicities (monthly in this analysis). The positive CUSUM implies a smaller ozone loss rate during the test period than during the earlier period. The 95% confidence limits due to unresolved fluctuations and empirical model uncertainty appear as the blue traces in Figure 2 implying that the three latitude regions experienced ozone deviations from the pre-1997 trend which are statistically significant. The 95% confidence envelopes are adjusted to the change in variance from the SAGE residuals to the HALOE residuals after 2000.5.

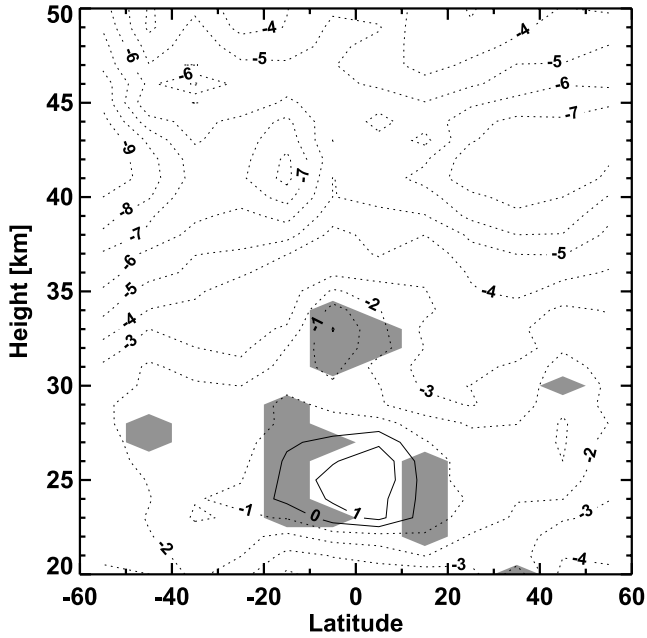


Figure 1. SAGE I/II ozone linear trends in percent per decade for 1979–2000. Shaded regions are not statistically different from zero at 95% confidence level.

[12] If the ozone record comprised two linear trends with slopes that changed abruptly in 1997, the CUSUM would be an integral of a linear quantity and one would expect its functional form to be quadratic in time. This quadratic, corresponding to a trend of zero after December 1996, is indicated by the green traces in Figure 2 (right panels) over the 1997–2003 period. In the upper-stratospheric traces in Figure 2, one sees the CUSUM traces equal or exceed the indications of a quadratic form over the entire period after 1996. These functional forms are consistent with the residual ozone time series in the left panels of Figure 2. The choice of the break point between the regression period and the projected period does not

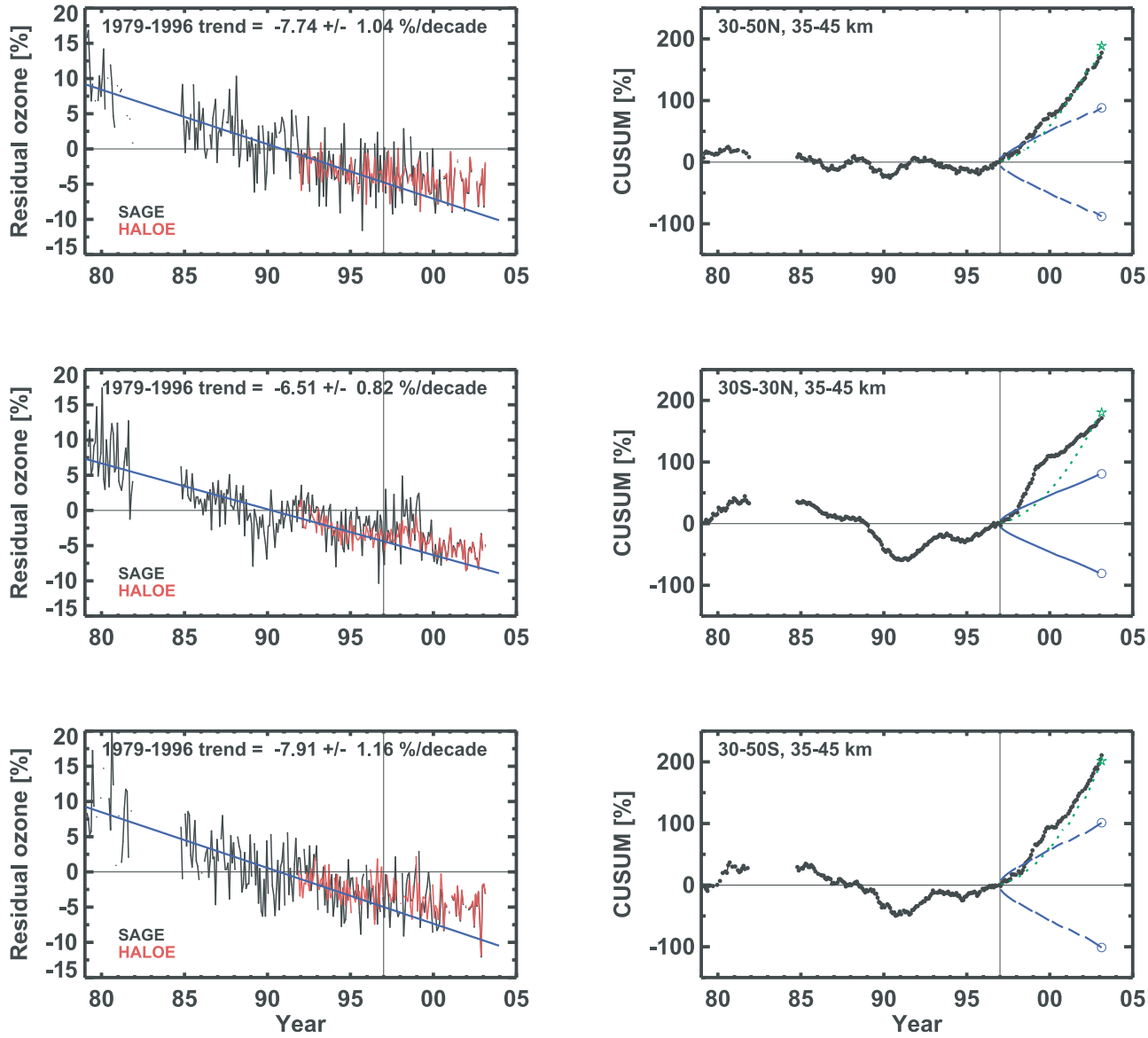


Figure 2. SAGE I/II and HALOE monthly average ozone residuals plus linear trend (left) and cumulative sum (CUSUM) of residuals (right) in percent at 35–45 km: 30–50°N (top panels) 30°S–30°N (center panels), and 30–50°S (bottom panels). The SAGE (black traces) and HALOE (red traces) residuals are independently obtained by subtracting the seasonal, solar, QBO, and AR(1) terms from ozone series. The blue line indicates the ozone trend calculated from observations for 1979–1996 and forecasted linearly afterward. Linear trends and 95% confidence intervals for 1979–1996 are listed. The last value in the cumulative residual time series represents the cumulative difference of all monthly residuals after 1997.0 with respect to the predicted trend line. The green line represents the hypothetical case where the ozone values remained at their 1997.0 values until 2003. The blue lines indicate the 95% confidence envelopes of departure from natural variability and model uncertainty.

significantly change these conclusions for choices of 1996, 1997, or 1998 (see Appendix C). Those years are all reasonable choices and produce similar results. Furthermore, calculation of the ozone CUSUM magnitude over the SAGE II period only (omitting SAGE I period) does not significantly alter these conclusions. The CUSUM of ozone residuals in Figure 2 graphically shows relatively large deviations from the zero line and a scarcity of zero crossings compared to random fluctuations, especially in the early 1980s (SAGE I) and during the Pinatubo period. The low frequency dominance of the CUSUM is expected due to the cumulative characteristics of random fluctuations and it might be noted that seasonal and QBO fluctuations have already been removed from the ozone time series. However, we cannot entirely rule out the possible contamination of the ozone values by unknown terms, especially, direct or indirect aerosol effects following the Pinatubo volcanic eruption.

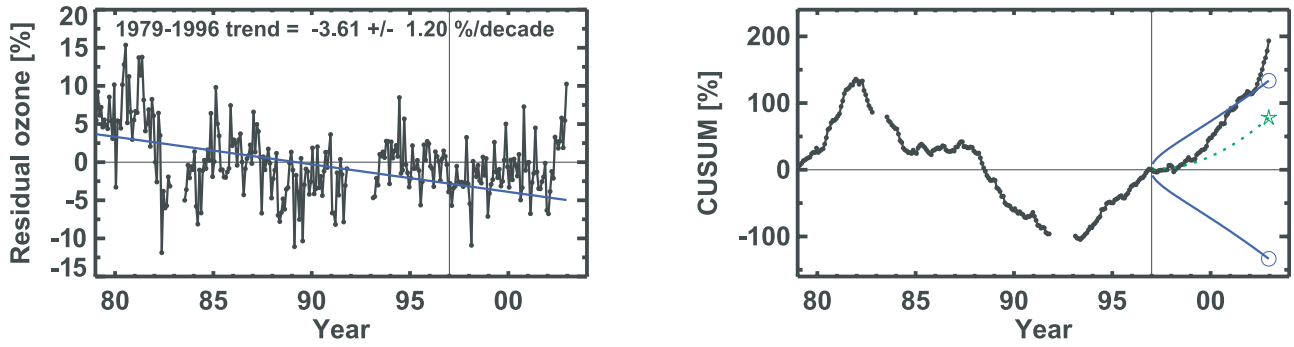


Figure 3. Results as in Figure 2 but derived from Arosa, Boulder, and Tateno Dobson Umkehr observations in layer 8 (~40 km).

[13] Analysis of Dobson Umkehr measurements of the ozone profile at Arosa, Boulder, and Tateno shown in Figure 3 corroborate the findings from the SAGE observations by exceeding the quadratic form in the test period. However, because of the much less frequent sampling, the time series residual fluctuations are higher and the commensurate test-period significance is smaller (i.e., the confidence envelope is larger, blue traces in Figure 3). *Reinsel* [2002] reaches the same conclusion from his analysis of Dobson Umkehr measurements.

5. Attribution

[14] HCl constitutes the majority of inorganic chlorine in the upper stratosphere [*Zander et al.*, 1996]. Consequently, HCl measurements have been used as indirect evidence for a slowdown in ozone trends [*Considine et al.*, 1997; *Considine et al.*, 1999; *Waugh et al.*, 2001]. Analyzing HALOE version-19 HCl data for 10° × 3 km latitude-altitude boxes in the same altitude-latitude ranges as for SAGE I/II ozone (20–50 km altitude, 60°S–60°N latitude) for 1993–2003 results in the zonal mean traces shown in Figure 4. The red quadratic traces in that figure represent the hypothetical case in which HCl remained at its 1997.0 values. The HALOE residual HCl at 35–45 km, after dominant seasonal, QBO, and solar effects are removed, show relatively monotonic increases until 1997 and then slower rates of increase for 2 or 3 years. These rate changes in HCl are significant as indicated by the departure of the HCl CUSUM well outside the 95% confidence envelope (blue traces in Figure 4). Correspondingly, SAGE combined with HALOE ozone time series indicate a lower ozone depletion rate after December 1996 (Figure 2).

[15] Owing to the coupling of CH_4 and H_2O with ozone in the upper stratosphere, the ozone trend could reflect changes in CH_4 and H_2O [*WMO*, 1999]. Consequently, the change in the ozone trend could be affected by the changes in CH_4 and H_2O trends. Between 1993 and 2003, HALOE observations at 35–45 km indicate that the fluctuations of monthly mean CH_4 are out of phase with the H_2O variations; they show larger percent variations in CH_4 consistent with approximate conservation of $2CH_4 + H_2O$ molecules. In order to investigate the sensitivity of the change in ozone trends to CH_4, H_2O, temperature (T), and HCl ($CH_4/H_2O/T/HCl$) variations, $CH_4/H_2O/T/HCl$ series should be included in the regression. However, both ozone and $CH_4/H_2O/T/HCl$ time series have signals caused by trend, season, QBO, and solar UV flux. Those signals in ozone may be in phase or out of phase with those in $CH_4/H_2O/T/HCl$, resulting in overestimation or underestimation of regression coefficients [*Pankratz*, 1991]. Therefore we remove trend, seasonal, QBO, and solar terms from the original ozone and $CH_4/H_2O/T/HCl$ series, then regress the ozone residuals on the residual $CH_4/H_2O/T/HCl$ series to obtain regression coefficients.

[16] The first panel in Figure 5 shows the residual $CH_4/H_2O/T/HCl$ variations at 35–45 km, 60°S–60°N, where the trend component is included. That figure indicates that it is more reasonable to assume two different linear trends for $CH_4/H_2O/T/HCl$ data before and after 1997.0 than to fit a linear or quadratic trend for the entire datasets. For regression purpose, two different trends, one before and one after 1997.0, are assumed and removed from the original series. Thus the change in ozone trend caused by $CH_4/H_2O/T/HCl$ variations is estimated from the regression coefficient determined from higher frequency variations multiplied by $CH_4/H_2O/T/HCl$ trend plus residuals. The responses of $CH_4/H_2O/T/HCl$ on ozone are 0.20 ± 0.05, 0.59 ± 0.20, −1.71 ± 0.50, and −0.41 ± 0.20 in % ±2σ for CH_4, H_2O, T, and HCl, respectively. However, because of the strong negative correlation (−0.76) between CH_4 and H_2O variations, it is better to use the orthogonal components (CH_4 in % − $4H_2O$ in %) and (CH_4 in % + $4H_2O$ in %). The latter corresponds approximately to ($2CH_4 + H_2O$) molecules variations. The regression coefficients then approximately agree with model calculations. For example, *Li et al.* [2002] calculated the sensitivity as 0.1 for CH_4, −0.1 for H_2O, and −1.0 for T at 1.8 mb (~43 km). Our positive correlation between O_3 and (CH_4 in % + $4H_2O$ in %) is consistent with the positive correlation between O_3 and H_2O found by *Siskind et al.* [1998, 2002] at 42–65 km altitude.

[17] The second panel in Figure 5 shows the combined effect of CH_4 and H_2O on ozone (blue line) whose individual long-term effect roughly cancels out each other except for 1993–1994 (right after the Pinatubo volcanic eruption). More ozone recovery would have been expected if the effects of CH_4 and H_2O had been excluded because the

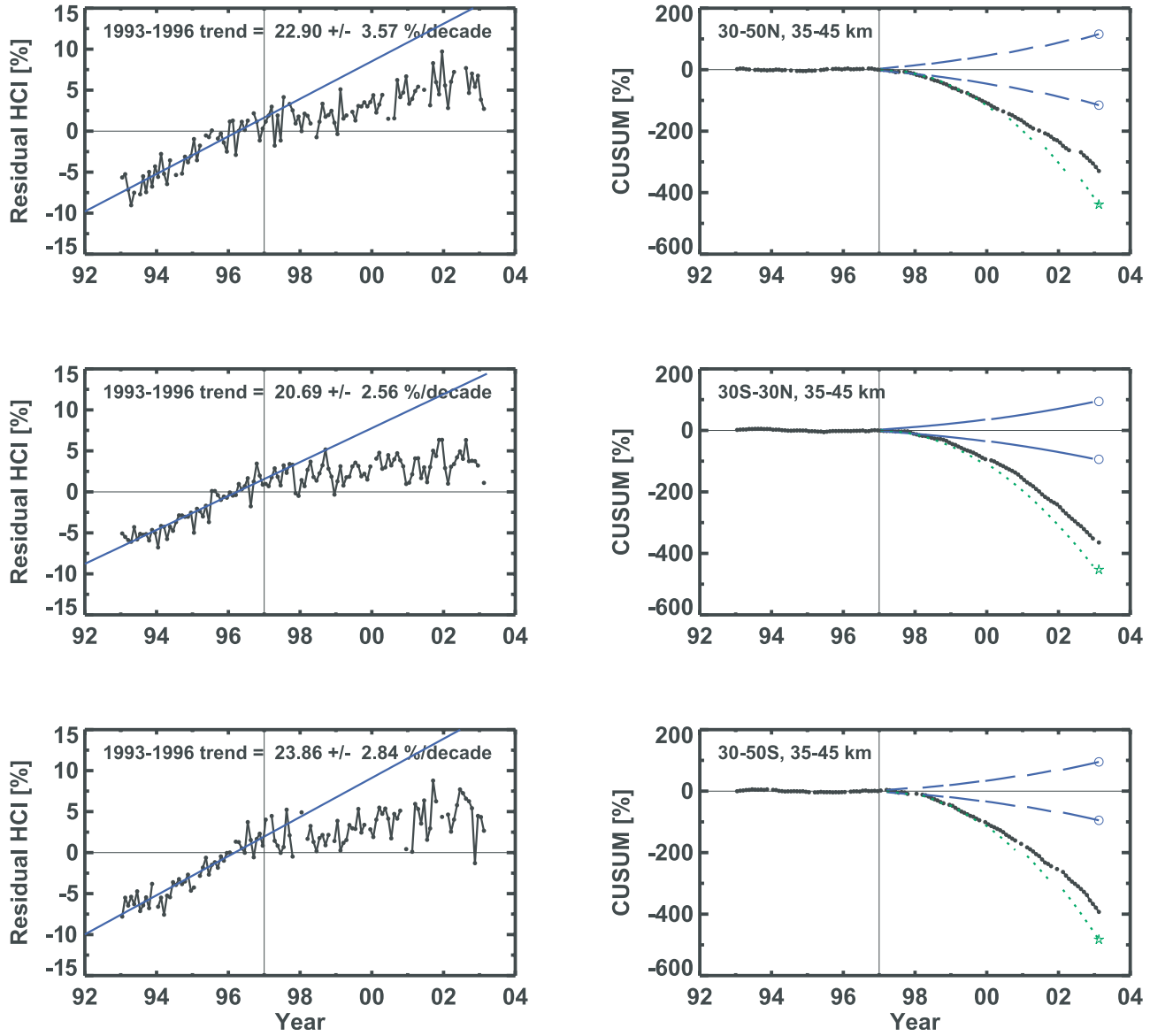
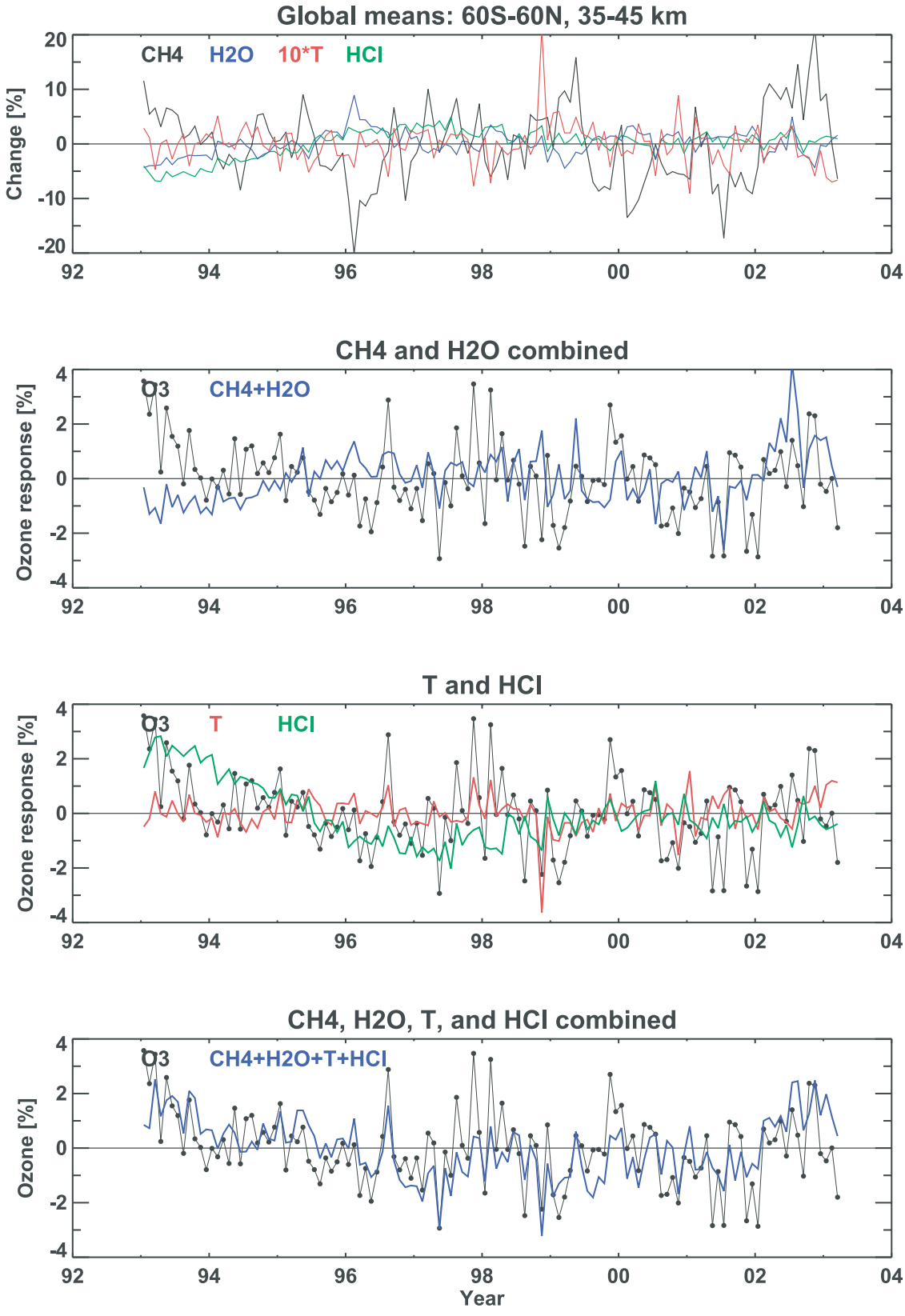


Figure 4. HALOE HCl time series, linear trends, residuals, and cumulative sums (CUSUMs) between 35 and 45 km analogous to the CUSUM in Figure 2.

net ozone loss, not caused by CH_4 and H_2O coupling with ozone, should be higher for 1993–1994 (before 1997.0). Therefore the slowdown of ozone loss rate after 1997.0 is unlikely to be due to the coupling of CH_4 and H_2O with ozone. The temperature and HCl impacts on the ozone series are shown in the third panel of Figure 5 (red and green lines). The temperature is negatively correlated with ozone, but there is little influence of temperature on the ozone trend due to the negligible temperature trend. The HCl series, included as a ODS proxy, on the other hand well reproduces the change in ozone trend (although temporal fluctuations of ozone are better represented by the $CH_4/H_2O/T$ variations). The bottom panel in Figure 5 shows that the observed ozone trend plus residuals is in good agreement with the estimated ozone trend plus residuals estimated from the $CH_4/H_2O/T/HCl$ sensitivities. Therefore the slowdown

Figure 5. (opposite) Coupling of CH_4, H_2O, and T with ozone at 35–45 km, 60°S–60°N. First panel: HALOE monthly averages for CH_4, H_2O, T, and HCl residuals plus trend in percent. The seasonal, solar, and QBO effects are subtracted from the original CH_4, H_2O, T, and HCl observations, respectively. Note that the vertical scale for T is 10 times larger. Second panel: combined CH_4 and H_2O impacts on ozone (blue line), which are empirically estimated from the regression analysis. For comparison, ozone trend plus residuals are shown with a black line. Third panel: estimated influence of T on ozone (red line) and ozone change corresponding to HCl variations (green line). HCl is selected as a proxy for stratospheric CFC. Fourth panel: observed ozone residuals plus trend (black line) compared to estimated ozone residuals plus trend by $CH_4/H_2O/T/HCl$ impacts on ozone (blue line).


Global means: 60S-60N, 35-45 km
CH4
H2O
10*T
HCl
Change [%]
20
10
0
-10
-20
92
94
96
98
00
02
04
CH4 and H2O combined
O3
CH4+H2O
Ozone response [%]
4
2
0
-2
-4
T and HCl
O3
T
HCl
CH4, H2O, T, and HCl combined
O3
CH4+H2O+T+HCl
Ozone [%]
Year

of ozone loss rate at 35–45 km is most likely due to the slowdown of ODS increase.

6. Conclusions

[18] SAGE I/II, ozone combined with HALOE ozone observations and Dobson Umkehr ozone measurements provide convincing evidence for a slowdown in the stratospheric ozone loss rates at 35–45 km. After seasonal, solar, and QBO effects are removed from the ozone series, the residual ozone shows a global slowdown of stratospheric ozone depletion after 1997.0. This slowdown is quantified by the cumulative residual ozone differences from the predicted linear trend. The systematic positive values of the cumulative residuals for 1997–2003 are significant at confidence levels much greater than 95%, showing less stratospheric ozone depletion after 1997.0. This result corresponds to a slowdown in stratospheric HCl increases after 1997.0 measured by HALOE. These changes in the ozone loss rates in the upper stratosphere are significant and constitute the beginning of a recovery of the ozone layer.

Appendix A

[19] In this work the QBO signals are obtained directly from ozone observations. With a few cosine terms that easily reproduce the modulated ozone QBO (8- and 20-month periods) at midlatitudes. For example, at 40 km, 45°N, a stepwise regression scheme selects 8 cosine terms as temporary QBO signals: 28, 30 (close to the 30-month period), 17, 19, 20, 21 (close to the 20-month period), 9 (close to the 8-month period), and 13 (perhaps the 2nd harmonic of the 30-month period or a modulated solar signal. This component, however, does not change the overall QBO signal). There is no overfitting problem because the stepwise regression scheme allows only statistically significant periodicities, which are again confirmed by regression coefficients and their uncertainties. On the other hand, when using the Singapore winds alone, the modulated ozone QBO signal with 8- and 20-month periods in the extratropics is not developed as observed.

[20] When using the Singapore winds as the QBO proxy, we assume time lags as functions of latitude and altitude. Because the Singapore winds have a dominant 30-month periodicity, the multiple regression model may sometimes show unreasonable time lags and signs. For example, a 1-month time lag in the tropics can be identified as a 16-month time lag with an opposite sign, which seems to be physically unreasonable. Even in the extratropics, time lags of ozone QBO at the same latitude range from a few months to near 30 months. Using the harmonic approach avoids these problems.

[21] The harmonic approach explains the QBO signal in ozone at any latitude and altitude and results compare favorably to other results where applicable (e.g., *Randel and Wu* [1996] using SAGE II; Figure 4–7 of *WMO* [1999] updated from *Zerefos et al.* [1994] using TOMS total ozone; Figure A1 using Umkehr layer-5 QBO signals at 30–52°N). Importantly, the harmonic approach produces the QBO signals similar to the results by the traditional approach of using Singapore winds with a lag but with more precise trend estimates and less fluctuation in the residuals.

[22] Figure A1 shows the vertical structure of the QBO signal in the tropics (top) and extratropics (middle). It is apparent that the phase propagates downward in the tropics and remains nearly vertically constant in the extratropics. Independently, Dobson Umkehr layer-5 ozone (~25 km) exhibits very small solar and aerosol effects but still a significant QBO effect [*Reinsel et al.*, 1999]. Therefore the deseasonalized Umkehr layer-5 ozone itself provides an approximate QBO signal without any complicated statistics. The bottom panel of Figure A1 shows the deseasonalized layer-5 ozone averaged from the following 7 Umkehr stations between 30° and 52°N: Belsk (52°N), Arosa (47°N), Haute Provence (44°N), Sapporo (43°N), Boulder (40°N), Tateno (36°N), and Cairo (30°N). The deseasonalized layer-5 Umkehr ozone (bottom, black trace) well reproduces the magnitude and phase of the SAGE II QBO signal at 25 km obtained from the sum of nonseasonal harmonic terms (bottom, dotted line).

[23] The solar term is included in the linear regression model and filtered out from the ozone series. This trend model is well tested and documented (e.g., 4-4 of *WMO* [1999]). The peak-to-peak magnitudes of the solar signals at 35–45 km over the period 1991–2000 are 4.2% (2.1%), 4.4% (4.3%), and 3.0% (3.4%) for SAGE (HALOE) at 30–50°N, 30°S–30°N, and 30–50°S, respectively. The magnitude is in good agreement with previous results: 4.5% at 40–45 km from Umkehr and SBUV(2) [*Miller et al.*, 1996]; 2–4% at 40 km from SBUV(2) [*McCormack and Hood*, 1996]. The 2-D model calculations produce a maximum of solar signals, approximately 2–2.5% at 35–40 km [*Brasseur*, 1993]. The current results are in good agreement with previous estimates from measurements and are unlikely to be too small.

[24] The timing of the solar maximum is important to empirically see if the solar component has been successfully removed from the data. The solar activity increases from 1986 and attains its maximum in 1989–1991. It again increases from 1997 and reaches its maximum in 2000/2001 (same shape as the upper panel of Figure A2). To demonstrate that the solar effect is not responsible for the CUSUM increase after 1997.0, we calculate the features of the expected CUSUM under the assumption that the solar effect has not been completely removed from the ozone residuals. If the solar cycle effect were higher than our model estimate by 2% per 100 units of F10.7 as an ozone response to F10.7 solar flux, the resulting estimates of the variations of ozone and CUSUM values expected from that assumption appear in the (lower panel of Figure A2), which shows that the CUSUM values are negative right after 1997.0 and then become positive in 1999.0, followed by a rapid increase of CUSUM afterward. This behavior is quite different from the CUSUM pattern in Figures 2 and 3 which show a rapid increase of the CUSUM right after 1997.0 and then slow increase near 2000. These CUSUM values at the end of data in Figure A2 derived from the exaggerated solar effect are still only about 80%, a value much smaller than our best estimate of all effects shows in Figures 2 and 3 (~200%). Therefore the solar effect alone cannot explain the positive CUSUM values in Figures 2 and 3.

[25] For one particular case at 35–45 km, 30–50°N, we show the deseasonalized SAGE data series in which the solar, QBO, and trend components are still present. By

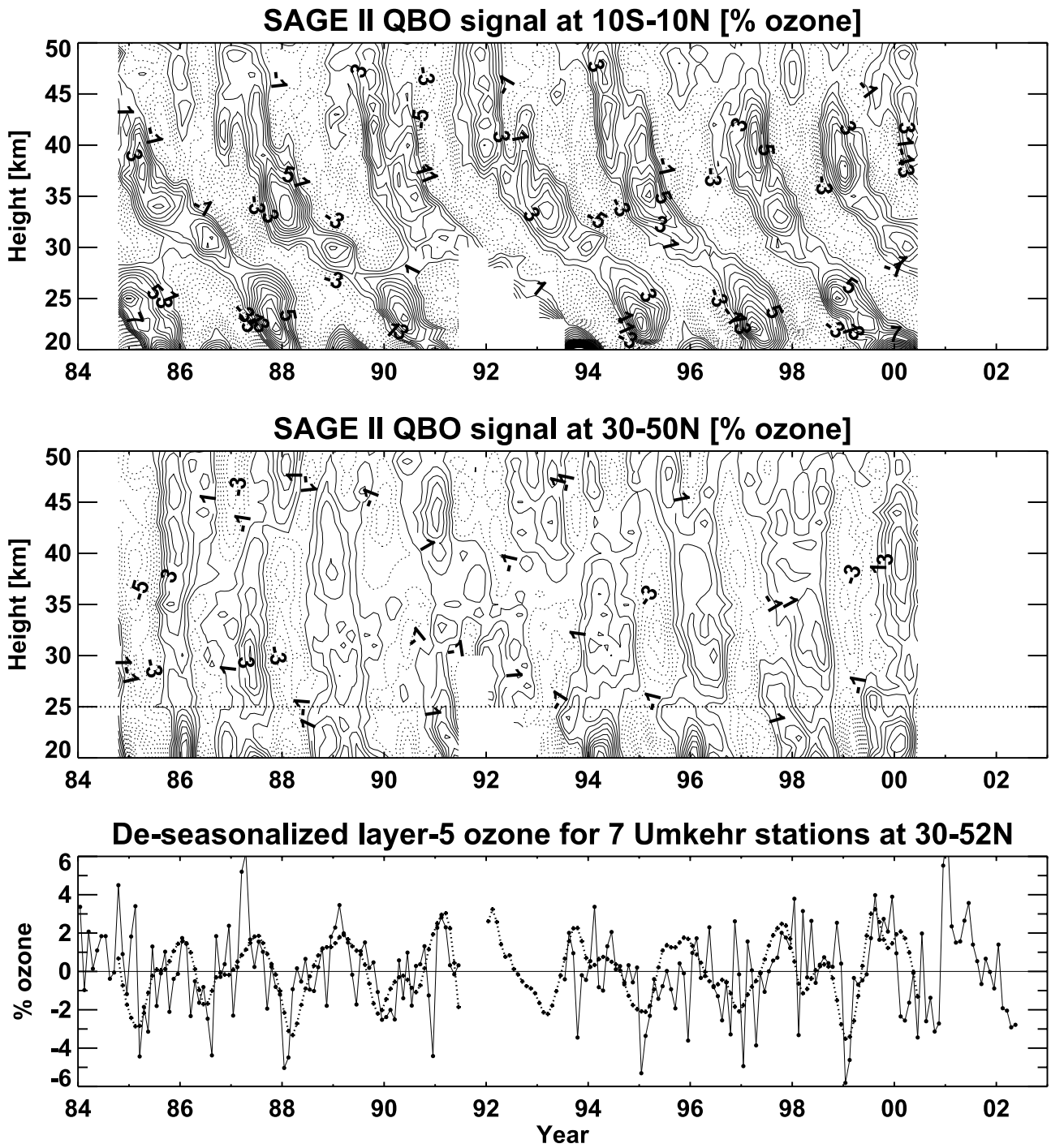


Figure A1. The vertical structure of the QBO signal at 10°S–10°N (top) and 30–50°N (middle) derived from the SAGE II ozone observations. The bottom panel shows the deseasonalized layer-5 ozone averaged from the following 7 Umkehr stations between 30° and 52°N: Belsk (52°N), Arosa (47°N), Haute Provence (44°N), Sapporo (43°N), Boulder (40°N), Tateno (36°N), and Cairo (30°N). The SAGE II QBO signal at 25 km obtained from the sum of non-seasonal harmonic terms (bottom, dotted line) well reproduces the magnitude and phase of the deseasonalized layer-5 Umkehr ozone (bottom, solid line).

adding or removing the fitted solar, QBO, and trend terms, we can depict what the statistical model is actually capturing and what it is missing. Figure A3 shows the partitioning of the deseasonalized ozone series into its QBO, solar, and trend components. The deseasonalized ozone was independently obtained by using 1 constant and 8 harmonic terms (constant and cosine/sine 12-, 6-, 4-, and 3-months terms). The top panel in Figure A3 shows this deseasonalized ozone residual time series that still contains the QBO, solar, and trend components. The QBO signal alone also appears in

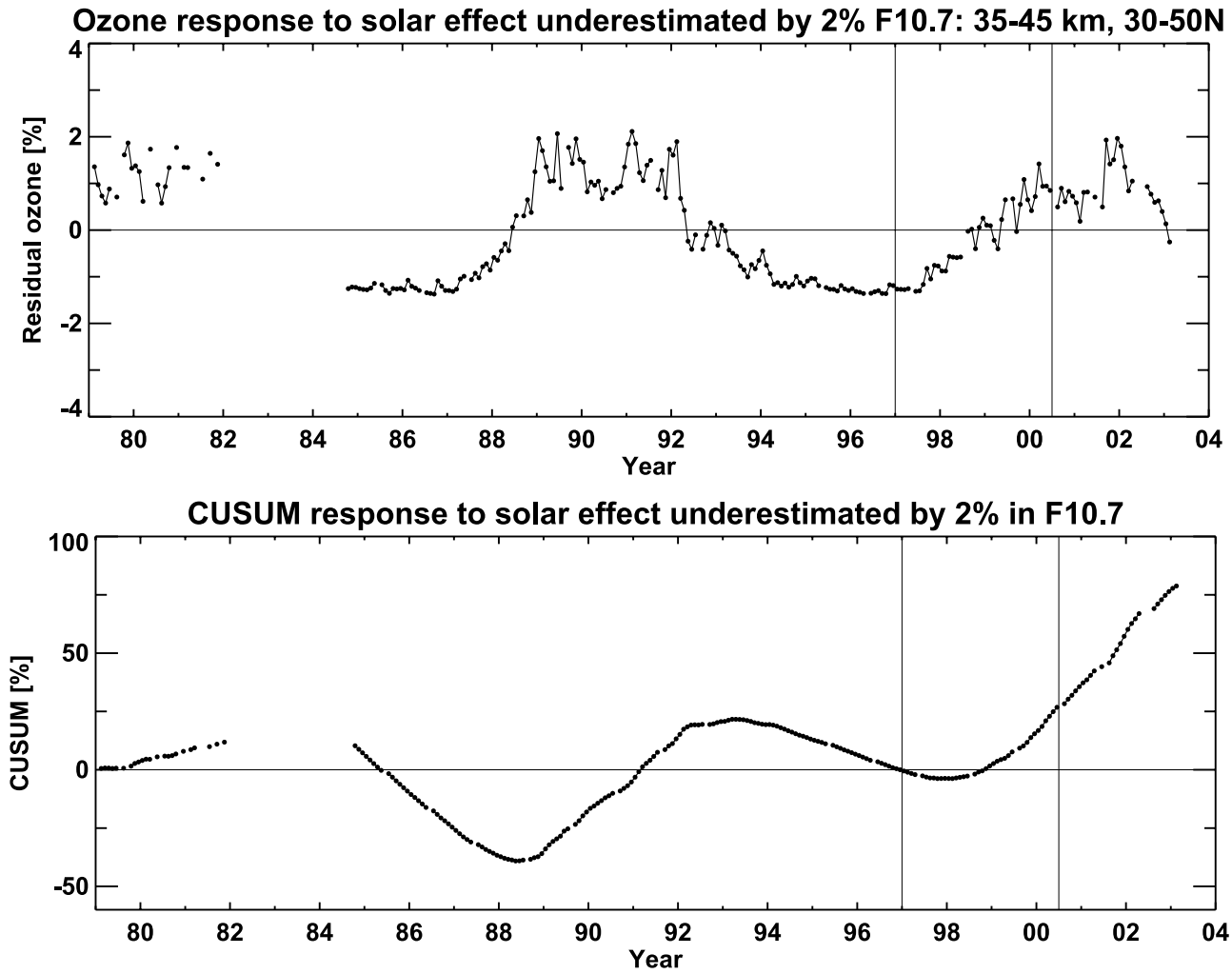


Figure A2. Expected ozone (upper) and CUSUM (lower) variations at 35–45 km, 30–50°N for the hypothetical case that the solar effect was greater than our model estimates by 2% per 100 units of F10.7. The vertical lines indicate 1997.0 (turnaround point) and 2000.5 (end of SAGE II data).

separate traces for SAGE and HALOE observations. The middle panel of Figure A3 shows the deseasonalized ozone series with the QBO signal removed (solar and trend terms remain) along with the separate SAGE and HALOE solar terms. The bottom panel shows the deseasonalized time series with both the QBO and solar signals removed leaving only the trend component and the residuals as in Figure 2. The trend term fitted to the SAGE data (-7.74 ± 1.04 in %/decade $\pm 2\sigma$) also appears in the bottom panel. Note that the trend is fitted over 1979–1996 and projected to 2003. This partitioning demonstrates the veracity of the model and plainly portrays the positive deviation of the residuals above the projected trend line in the bottom panel.

Appendix B

[26] As introduced by *Reinsel* [2002], the cumulative sum (CUSUM) of residuals visually depicts a relatively small change in pattern such as a trend change in this study. The change in ozone trends could be obtained simply from the difference of two linear trends for observations before and after 1997.0 [e.g., *Weatherhead et al.*, 1998, 2000]. The standard error of the trend estimate for 1997–2003 is then larger than that for 1979–1996 because there are relatively fewer ozone data for 1997–2003. Consequently, the significance of the difference depends more on uncertainty of the trend estimate for 1997–2003. In addition, if ozone loss rate is changing at and after the turnaround point, a linear-trend-assumption may not be valid after 1997, resulting in a biased trend estimate for 1997–2003. Instead of directly comparing two linear trend estimates from observations over two different periods before and after 1997.0, we use consecutive sums of residual ozone after removing seasonal, QBO, solar, and first order autoregressive AR(1) terms from original ozone series for the entire period. The CUSUM should follow a random walk processes after removing the trend from the residuals.

[27] Starting with the hypothesis that the ozone trends for observations before and after 1997.0 are the same, we calculate an ozone trend for observations before 1997.0 and use it to forecast thereafter. When the trend for observations before 1997.0 is calculated and subtracted from the residuals after 1997.0, the residuals after 1997.0 should be random and independent of each other if there is

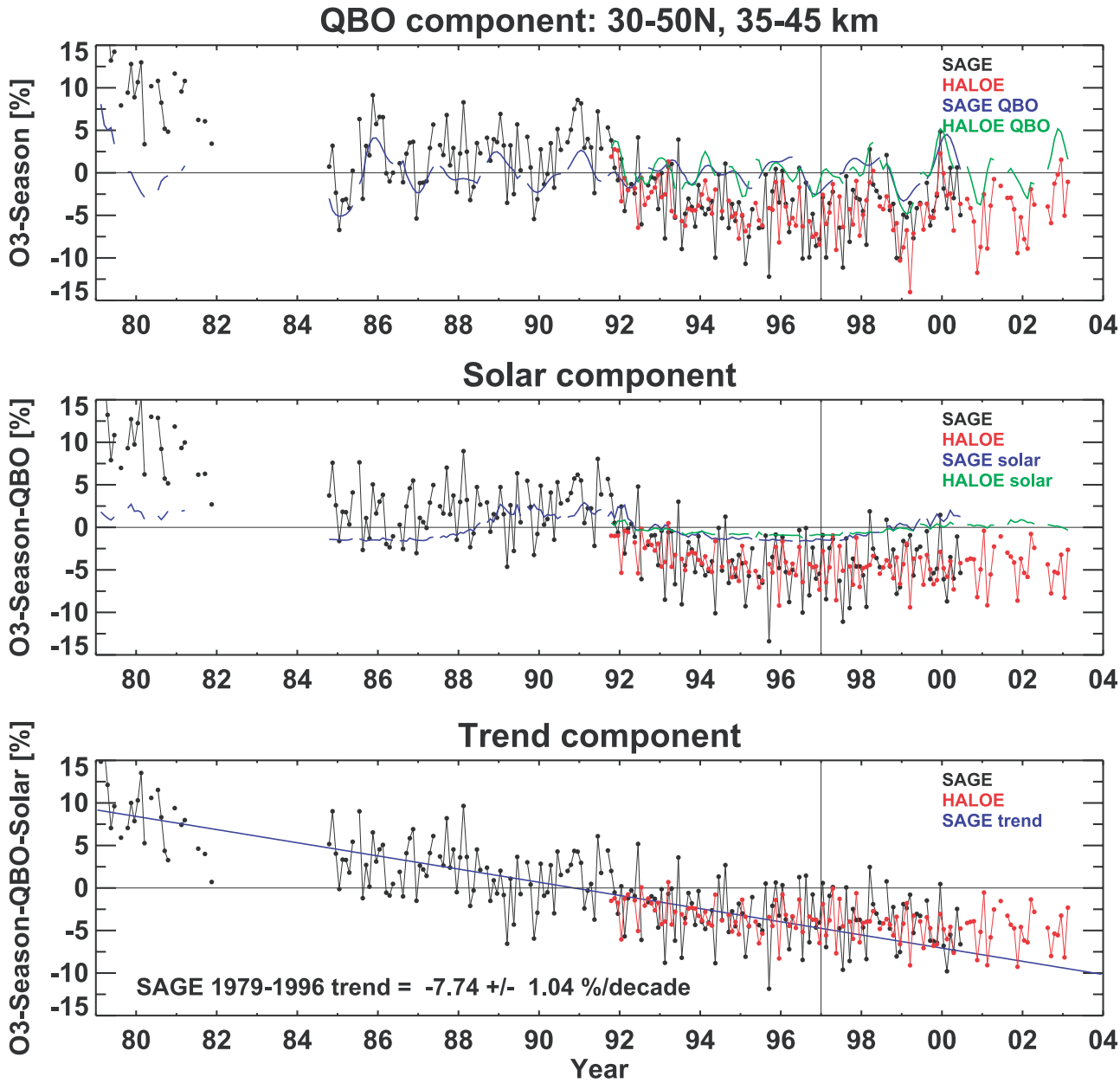


Figure A3. Fitted QBO, solar, and trend components for the SAGE I/II (black dots) and HALOE ozone series (red dots) at 35–45 km, 30–50°N. Top panel: time series of deseasonalized SAGE and HALOE ozone containing QBO, solar, trend, and residual terms (solid lines between symbols) and the fitted harmonic QBO signals (blue line for SAGE QBO and green line for HALOE QBO). Center panel: deseasonalized ozone with the QBO signal removed (solid line between symbols) and the fitted F10.7 cm flux solar signal (blue line for SAGE and green line for HALOE). Bottom panel: deseasonalized ozone with both the QBO and solar signals removed (i.e., only trend and residual terms remain; solid line between symbols) and the fitted linear trend (blue line for SAGE 1979–1996 observations).

no error in the trend and mean level estimates. The CUSUM of those residuals, then, would follow a normal distribution with mean zero and variance proportional to number of time steps. A systematic positive or negative CUSUM indicates failure of that hypothesis, implying systematic difference between ozone trends before and after 1997.0. If there is less (more) ozone depletion after 1997, the ozone residuals will not represent a white noise process but will show systematically positive (negative) values. The significance of the departure from the same-trend-hypothesis can be determined from the CUSUM value and the number of time steps, that is, n, the number of months if using monthly means. When uncertainties in the regression model are included, the variance of CUSUM becomes,

$$\mathrm{VAR}\{\mathrm{CUSUM}\} = \sigma^2\left\{n_2 + n_2^2/n_1 + [\Sigma_2(t - t_o)]^2/\Sigma_1(t - t_o)^2\right\},$$

where

σ : standard error of the residuals for 1979 – 1996,

n_1 : number of data for 1979 – 1996,

n_2 : number of data after 1997.0,

t_o : mean value of t for 1979 – 1996,

$\Sigma_1(t - t_o)^2 = (t_1 - t_o)^2 + (t_2 - t_o)^2 + \ldots + (t_{n1} - t_o)^2$, and

$[\Sigma_2(t - t_o)]^2 = [(t_{n1+1} - t_o) + (t_{n1+2} - t_o) + \ldots + (t_{n1+n2} - t_o)]^2$.

[28] The first term of the above equation is due to the random fluctuations whose variance dominates the early stage of the test period, a few years after 1997.0. The second and third terms result from the uncertainties in mean level and trend estimates of the regression model. Note that the second and third terms increase with time ($n_2^2 \sim t^2$ and $[\Sigma_2 (t - t_o)]^2 \sim t^4$). As time increases, the third term dominates the total variance of CUSUM. At the end of the SAGE data, the variance at 30–50°N due to random walk, uncertainty in the mean level, and uncertainty in the trend contribute to the total variance by 47%, 12%, and 41%, respectively. When the HALOE data are concatenated, the contributions become 24%, 14%, and 63%, respectively.

Appendix C

[29] The selection of 1997 does not change much with altitude or data sources: ~1997 for SAGE and HALOE ozone and ~1997/1998 for HALOE HF and HCl. The selection of the turnaround point is rather flexible because ozone trends do not change abruptly at a certain time. For example, for 1996–2003, 1997–2003, and 1998–2003, the CUSUMs and their 95% confidence bounds differ due to a different number of months and slightly different trend values for each regression time period. However, the statistical significance of the CUSUM is very similar, regardless of our choice of a specific projection time within a year of 1997.

[30] The selection of the turnaround point (1997.0) also comes from our interest in the detection of stratospheric ozone changes caused by the Montreal Protocol and its Amendments. Since the Montreal Protocol, ozone depletion substances (ODS) in the troposphere have shown definite declines [*Montzka et al.*, 1999; *Prinn et al.*, 2000]. The turnaround point for the ODS production is the early 1990s. Considering the transport time of tropospheric gases to stratosphere is up to 6 years [*WMO*, 1999], 1997.0 is a reasonable choice for detecting stratospheric ozone changes caused by a reduction in ODSs.

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