On detecting a trend in the residual circulation from observations of column HCl

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[1] Ground-based measurements show that the HCl column at middle latitudes varies seasonally with a late spring maximum and a minimum in late summer. HALOE measurements of HCl profiles show that this seasonal cycle in the column is controlled by the seasonal variations of the mass and composition of the lowermost stratosphere. The seasonal maxima are produced by wintertime downward transport of stratospheric air. We hypothesize that the amplitude of the seasonal cycle is quantitatively related to the strength of the stratospheric residual circulation and stratosphere-troposphere exchange (STE). We investigate this relationship using a multi-decadal simulation from the GSFC Chemistry and Transport Model driven by output from a general circulation model in which an STE trend exists. We also examine the ground-based record of HCl measurements at Jungfraujoch for evidence of a trend in the HCl seasonal amplitude that would be a signature of a long-term change in the stratospheric circulation. **Citation:** Olsen, M. A., A. R. Douglass, R. S. Stolarski, and M. R. Schoeberl (2006), On detecting a trend in the residual circulation from observations of column HCl, *Geophys. Res. Lett.*, *33*, L14815, doi:10.1029/2006GL026214.

1. Introduction

[2] General circulation models have been shown to exhibit a change in the residual circulation as a result of changes in atmospheric composition [e.g., *Rind et al.*, 1998; *Sigmond et al.*, 2004; *Eichelberger and Hartmann*, 2005]. If such a trend is real, it would contribute to observed trends in stratospheric trace gases including ozone. *Butchart and Scaife* [2001] have demonstrated that increasing the rate of mass exchange between the stratosphere and troposphere accelerates the removal of chlorofluorocarbons (CFCs) from the stratosphere and would alter ozone recovery. The present study introduces a novel method of detecting a change in the residual circulation.

[3] Both the change in the mass of the extratropical lowermost stratosphere (the region between the tropopause and the 380K surface) and the change in the HCl mixing ratio through transport contribute to the seasonal variation observed in the column. The mass of the lowermost stratosphere is a maximum in the winter when the tropopause is lowest and a minimum during summer when the tropopause is highest [*Schoeberl*, 2004]. The composition of the middle latitude lowermost stratosphere during winter is dominated by downward transport of stratospheric air from the region above the 380K surface which typically lies between 100–150 hPa. In the summer, the composition of this region becomes tropospheric in character due to two-way, quasi-isentropic exchange of air with the upper tropical troposphere [e.g., *Ray et al.*, 1999; *Pan et al.*, 2004]. Middle latitude ground based measurements of the HCl column exhibit a maximum in late winter and a minimum in late summer. The stratospheric column above 100 hPa computed from profiles measured by the Halogen Occultation Experiment (HALOE) on the Upper Atmosphere Research Satellite is nearly constant throughout the year, thus the seasonal variability of the lowermost stratospheric mass and HCl are manifest in the column measurements.

[4] Based on the discussion above it seems likely that changes in the amplitude of the seasonal cycle of mid-latitude column HCl are related to changes in the residual circulation. The annual maximum in the HCl column is sensitive to a change in the stratospheric residual circulation through the winter and spring downward flux of HCl into the lowermost stratosphere. Stronger (weaker) transport during these seasons will produce a larger (smaller) late winter maximum in the HCl column. The seasonal minimum of the column will remain relatively unchanged since the late summer/fall composition of the mid-latitude lowermost stratosphere is tropospheric (low HCl) in character, as noted above. Thus, since the HCl mixing ratio increases with height, more vigorous winter/spring transport across the 380K surface will lead to a greater seasonal amplitude of the HCl column. Here we use a multi-decadal simulation with an increasing trend in the mass stratosphere-troposphere exchange (STE) to quantify the relationship between STE and the mid-latitude column HCl annual amplitude. We argue that such an increase in transport is related to a strengthening the residual circulation. We also examine the existing ground-based observation record for a signature of such an increase.

2. Simulation and Data

2.1. Chemistry and Transport Model and Simulation

[5] We performed a simulation for 1973–2023 using the GSFC Chemistry and Transport Model (CTM) [*Stolarski et al.*, 2006]. The model horizontal resolution is 2° latitude by 2.5° longitude and fields are saved at half of this resolution. The vertical domain extends from the surface to 0.43 hPa with 28 levels spaced by about 1 km in the upper troposphere and lower stratosphere, with spacing increasing to 4 km at the upper boundary. The vertical transport is calculated to satisfy continuity with the horizontal divergence. Boundary conditions of source gases, including observed and projected CFCs and other sources of chlorine, are taken

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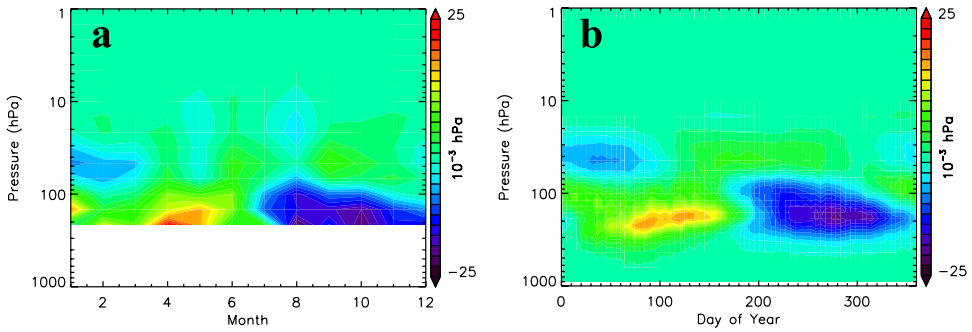

Figure 1. Seasonal cycle of partial pressure of HCl from (a) HALOE at 45°–55°N and (b) CTM at 46°–54°N. The annual mean at each pressure surface has been removed to emphasize the seasonal cycle. Both panels show six-year means (1992–1997) and the HALOE data are monthly mean values.

from scenario A2 [*World Meteorological Organization*, 2003]; *Stolarski et al.* [2006] provide details of the simulation. Meteorological fields from the GEOS-4 general circulation model (GCM) [*Bloom et al.*, 2005] are used to drive the CTM. The GCM uses Hadley Centre monthly mean sea surface temperatures for the years 1949 to 1998. *Douglass et al.* [2003] use comparisons of observations of long-lived tracers with fields from a CTM simulation driven by an earlier version of GEOS-4 to show that the large-scale stratospheric transport of the GCM is realistic. *Schoeberl et al.* [2003] show that the age-of-air and residual circulation produced by these GCM fields are reasonable. *Olsen et al.* [2004] show that the flux of mass and ozone from the stratosphere to the troposphere compares well with estimates based on observations.

2.2. HALOE and FTIR Observations of HCl

[6] The Halogen Occultation Experiment (HALOE) on the Upper Atmosphere Research Satellite (UARS) measured the composition of the stratosphere and mesosphere by solar occultation limb sounding from 1991–2005 [*Russell et al.*, 1993]. Thirty vertical profiles of seven chemical species, including HCl, were made daily until problems with the solar array forced power sharing among instruments and reduced sampling of the seasonal cycle at middle latitudes beginning in 1995. In 1997, power sharing necessitated by battery problems further reduced HALOE sampling.

[7] Regular ground based measurements of column HCl have been made by Fourier Transform Infrared (FTIR) spectrometers of the Network for the Detection of Atmospheric Composition Change (NDACC, formerly the Network for the Detection of Stratospheric Change). Jungfraujoch measurements are the longest-on going record and extend back to 1989. The random and systematic errors of each measurement are ±3% and ±5%, respectively [*Rinsland et al.*, 2003].

3. Results

[8] The simulated difference of the partial pressure of HCl from the annual mean is similar to that calculated from HALOE observations (Figure 1). The seasonal cycle in the mid- to upper-stratosphere is relatively flat and almost all of the variability in the column of HCl comes from the lowermost stratosphere. The simulation is biased high with respect to the observations but is within the calibration error estimates of HALOE [*Russell et al.*, 1996]. Any mean bias of the simulated HCl compared with observations is irrelevant to this study and is removed from the comparison in Figure 1. The key point of this comparison is that the seasonal cycle in the lowermost stratosphere is reproduced well by the simulation and the seasonal amplitudes are similar.

[9] Figure 2 compares the monthly mean Fourier Transform Infrared Spectrometer (FTIR) HCl column measured at Jungfraujoch (46.55°N) with the simulated zonal mean column at 50°N. We choose this comparison with a latitudinal offset so that the mean magnitudes are similar. This offset does not alter our conclusions presented in this work. Overall, the interannual trend and seasonal amplitude of the simulated HCl column compare well with the available FTIR data. The interannual trend in the simulation reflects the specified boundary conditions of chlorine species. The CTM simulation includes the aerosol loading of two major volcanic eruptions, El Chichon in 1982 and Pinatubo in 1991. The simulated eruption of Pinatubo in 1991 results in a substantial decrease of the seasonal maximum in the following year. Significantly low values of column HCl are observed over Jungfraujoch during winter 1991/1992. This results from the interaction of HCl and chlorine nitrate on aerosol surfaces followed by the initial reformation into chlorine nitrate.

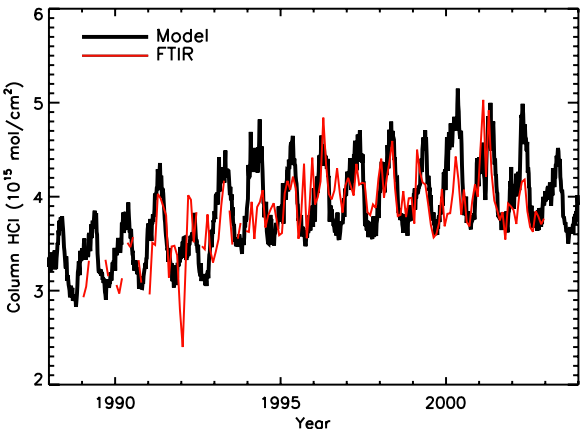

Figure 2. Time series of HCl column. Bold line is the model zonal mean at 50°N. Thin red line is the monthly mean FTIR data from Jungfraujoch (46.55°N).

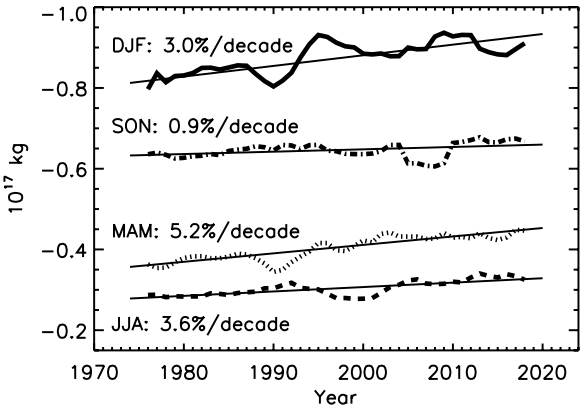

Figure 3. Seasonal mass transport across the extratropical 380 K surface for the Northern Hemisphere from a fifty-year integration of the GEOS-4 GCM. Solid line is total STE for December–February. Dotted line is March–May. Dashed line is June–August. Dash-dot line is September–November. Negative values indicate net "downward" transport. Values are smoothed using a five-year running mean.

[10] We have calculated the extratropical STE of mass in the GCM simulation using the mass balance diagnostic of *Appenzeller et al.* [1996]. The annual STE exhibits an increasing trend of 3% per decade in the Northern Hemisphere and 2% per decade in the Southern Hemisphere (not shown). These trends are similar to those found with previous studies [e.g., *Butchart and Scaife*, 2001; *Eichelberger and Hartmann*, 2005]. A change in the net annual extratropical STE requires a change in the residual circulation if the annual mass and location of the lowermost stratosphere remains constant. In our simulation, increasing wave driving in the polar stratosphere produces a strengthening residual circulation and forces the trend in STE. We limit the focus of the present discussion to the relationship of the seasonal amplitude of the HCl column to the residual circulation and STE.

[11] The net annual extratropical STE is ultimately determined by the net transport across the extratropical 380K surface [*Appenzeller et al.*, 1996]. Figure 3 shows time series of the mass transport across the 380K surface for each season. The mass transport increases during all four seasons. The greatest seasonal trend is found during MAM, the season of the maximum HCl column (Figure 1). The trend is smallest during SON when the HCl column is at a seasonal minimum. Although the SON 380K flux increases, the trend is comparatively small and is unlikely to significantly increase the column HCl seasonal minimum since the composition of the mid-latitude lowermost stratosphere is predominantly forced by the influx of HCl-poor tropospheric air.

[12] We determine the zonal mean column HCl seasonal cycle amplitude by fitting a linear function of an annual sine and cosine. The amplitude is divided by the annual mean and expressed as a percent to remove the trend of atmospheric chlorine. This normalization also removes the impact of the volcanic aerosol loading.

[13] The time series of the zonal mean column HCl seasonal amplitude at 50°N from the simulation is shown in Figure 4a. Over the entire time series, the HCl amplitude at this latitude increases at 9.7% per decade. The zonal mean trend tends to decrease both poleward and equatorward. Figure 4b shows that the simulated annual HCl amplitude and STE are significantly correlated at greater than 99% confidence. The residual circulation is forced by wave activity as expressed through the eddy heat flux. As previously noted, the STE trend in the simulation is driven by increased wave forcing in the polar stratosphere. The winter and spring eddy heat flux increases throughout the stratosphere poleward of 60°N. The winter and spring eddy heat flux in this entire region is correlated with the mid-latitude zonal mean column HCl amplitude at greater than 99% significance (not shown). This is consistent with greater HCl seasonal maximums caused by an increasing residual circulation.

[14] To separate the change in residual circulation from the changes in chlorine loading, a second CTM simulation for 1979–2001 was run with source gas boundary conditions fixed to 1979 levels. All other boundary conditions and driving meteorological fields were identical to the longer simulation. The column HCl seasonal amplitude in the fixed chlorine simulation increases at a rate of about 13% per decade and is also significantly correlated with STE (not shown). Over the same time period, the varying chlorine simulation exhibits a 20% per decade trend. Thus, the strength of the residual circulation is a primary factor in determining the amplitude of the column seasonal cycle. However, our normalization by the annual mean does not completely remove the trend in the HCl column due to the trend in atmospheric chlorine. It remains advantageous to normalize using the annual mean rather than normalize using atmospheric chlorine since aerosol influences are eliminated and this method can be easily applied to ground-based data records. Atmospheric chlorine is projected to continue to slowly decrease at present day rates over the next few decades, so the significant correlation of the HCl amplitude and STE will persist.

[15] The HCl seasonal amplitude has considerable interannual variability. We can estimate the number of years of observations that are needed to isolate a trend in the residual circulation given the same trend and standard deviation of the HCl amplitude as found in the simulation. Our results from the time series of the zonal mean HCl amplitude at 50°N (Figure 4a) indicate that 11 and 19 years are needed to isolate the signal with 95% and 99% confidence, respectively. If the actual trend of the amplitude is less or if the

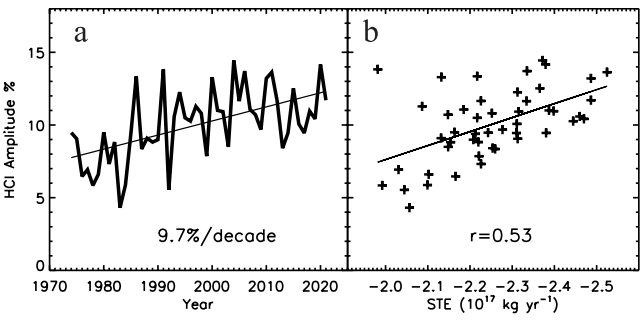

Figure 4. (a) Time series of zonal mean HCl seasonal amplitude from CTM at 50°N. (b) Correlation of seasonal amplitude with annual extratropical STE.

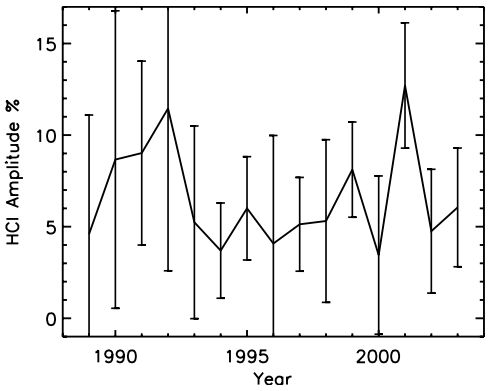

Figure 5. Time series of the seasonal amplitude of column HCl measured by the FTIR at Jungfraujoch.

variance is greater, then the number of years needed will increase, and vice versa. The results using time series at different latitudes within 38°N to 50°N do not vary greatly. At these latitudes the calculated number of years needed range from 10 to 13 years at the 95% confidence level.

[16] Because power issues on UARS led to undersampling the HCl seasonal cycle at middle latitudes, ground-based measurements are most likely to provide a long enough, consistent record of column HCl. Figure 5 shows the seasonal amplitudes calculated from the FTIR measurements at Jungfraujoch. We use a Monte Carlo bootstrap method following *Hood et al.* [1993] to estimate the error bars for the fit of the annual sine and cosine. There are many significant gaps in the early years of the data record as can be seen in Figure 2. Fewer than seventy days have measurements in each of the years 1989–1994 and many months lack any measurements. These years are significantly undersampled, resulting in very large estimated errors in the seasonal amplitude. Beginning in 1997, at least 100 measurements per year are recorded and all months are represented. A positive trend exists after the mid 1990's, but it is not significantly different from zero given the variance and length of the record. Thus, no evidence of an increasing residual circulation can be determined from the present FTIR data.

[17] In the simulation, we used zonal mean HCl columns at a single latitude to compute the seasonal amplitude. The variability of the column HCl is much greater at a single location than the zonal mean. However, the number of sites of ground-based measurements of column HCl from the NDACC is few. We have found that the mean measurement from several middle latitude sites reduces the single station geographical and meteorological variability. We have simulated NDACC measurements by sampling the model at grid points nearest to Northern Hemisphere middle latitude NDACC sites that have any multi-year record of FTIR measurements. The variability of the average of five locations between 38° and 47°N (Jungfraujoch at 8°E, Zugspitze at 11°E, Moshiri at 142°E, Rikubetsu at 144°E, and Mt. Barcroft at 118°W) is similar to that of the simulated zonal mean at 50°N. Using the mean of the values sampled at these five sites we find that 11 years are needed to detect a signal at 95% confidence, the same length of time estimated for the zonal mean. This suggests that coordination of simultaneous mid-latitude NDACC FTIR measurements to reduce longitudinal variability could identify a trend in a reasonable time frame.

4. Discussion and Conclusions

[18] We have investigated other trace gases, including ozone, methane and CFC's. The seasonal cycle of ozone in the lowermost stratosphere is a convolution of transport driven changes and the seasonal variation in ozone above 380 K [e.g., *Olsen et al.*, 2004]. The seasonal cycle of gases such as methane and fluorocarbons are not as sensitive to the residual circulation since their vertical gradient in the lowermost stratosphere is relatively small. HCl is most sensitive to the residual circulation and STE since it has a strong gradient in the lowermost stratosphere and little seasonal variation above 380 K.

[19] In summary, we have identified a method of detecting a change in the residual circulation from observations of HCl. The seasonal amplitude of the middle latitude HCl column provides a marker of the magnitude of the residual circulation once the interannual trend of atmospheric chlorine is removed. Currently, ground-based observations do not adequately define the seasonal cycle for a long enough period to isolate a signal of increased circulation. Our results indicate that such a signal could be detected in a reasonable time with coordination by mid-latitude NDACC sites.

[20] **Acknowledgments.** The Jungfraujoch FTIR data used in this publication was obtained as part of the Network for the Detection of Atmospheric Composition Change (NDACC) and is publicly available (see http://www.ndacc.org). This work was supported by NASA's EOS IDS and ACMAP programs.

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