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Global stratospheric chlorine inventories for 2004–2009 from Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) measurements

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Abstract

We present chlorine budgets calculated between 2004 and 2009 for four latitude bands (70° N–30° N, 30° N–0° N, 0° N–30° S, and 30° S–70° S). The budgets were calculated using ACE-FTS version 3.0 retrievals of the volume mixing ratios (VMRs) of 9 chlorine-containing species: CCl_4, CFC-12 (CCl_2F_2), CFC-11 (CCl_3F), $COCl_2$, COClF, HCFC-22 (CHF_2Cl), CH_3Cl, HCl and $ClONO_2$. These data were supplemented with calculated VMRs from the SLIMCAT 3-D chemical transport model (CFC-113, CFC-114, CFC-115, H-1211, H-1301, HCFC-141b, HCFC-142b, ClO and HOCl). The total chlorine profiles are dominated by chlorofluorocarbons (CFCs) and halons up to 24 km in the tropics and 19 km in the extra-tropics. In this altitude range CFCs and halons account for 58 % of the total chlorine VMR. Above this altitude HCl increasingly dominates the total chlorine profile, reaching a maximum of 95 % of total chlorine at 54 km. All total chlorine profiles exhibit a positive slope with altitude, suggesting that the total chlorine VMR is now decreasing with time. This conclusion is supported by the time series of the mean stratospheric total chlorine budgets which show mean decreases in total stratospheric chlorine of 0.38 ± 0.03 % per year in the Northern Hemisphere extra-tropics, 0.35 ± 0.07 % per year in the Northern Hemisphere tropical stratosphere, 0.54 ± 0.16 % per year in the Southern Hemisphere tropics and 0.53 ± 0.12 % per year in the Southern Hemisphere extra-tropical stratosphere for 2004–2009. Globally stratospheric chlorine is decreasing by 0.46 ± 0.02 % per year. Both global warming potential-weighted chlorine and ozone depletion potential-weighted chlorine are decreasing at all latitudes. These results show that the Montreal Protocol has had a significant effect in reducing emissions of both ozone-depleting substances and greenhouse gases.

1 Introduction

In the stratosphere stable chlorine-containing molecules undergo photolysis or react with $O(^1D)$ to produce chlorine species which can catalyse the destruction of ozone.

In the "natural" atmosphere this ozone loss process is not significant because non-anthropogenic chlorine-containing molecules such as CH_3Cl have relatively small volume mixing ratios (VMRs) in the stratosphere. Anthropogenic emissions of chlorine-containing species grew from the early 20th century until the mid-1990s. From 1974 there was a growing body of research, starting with the work of Molina and Rowland, which showed that emissions of anthropogenic halogen-containing gases would ultimately lead to ozone loss. After the publication of the paper by Farman, Gardiner and Shanklin in 1985, which documented the discovery of the "ozone hole" above Antarctica, the global community was galvanized into action leading to the Montreal Protocol on Substances that Deplete the Ozone Layer. The Montreal Protocol prohibits the use of a number of halogen-containing species, and came into force in 1987 (UNEP, 2009).

Thanks to the effectiveness of the Montreal Protocol in reducing the emissions of ozone depleting substances (Montzka et al., 2011), and despite the ever growing concentration of greenhouse gases which decrease the temperature of the stratosphere, stratospheric ozone is predicted to return to the same levels as in 1980 by 2050 in both the tropics and the poles (Montzka et al., 2011). Rigorous assessments of ozone depletion require measurements of the stratospheric total chlorine and its change over time; thus a number of chlorine budgets have been calculated previously. Measurements made by the Atmospheric Trace MOlecule Spectroscopy (ATMOS) instrument were used to calculate a stratospheric chlorine budget at 30° N for 1985 using VMRs of HCl, CH_3Cl, $ClONO_2$, CCl_4, CFC-12, CFC-11 and HCFC-22. A value of 2.58 ± 0.1 ppb was calculated at that time (Zander et al., 1992). A second chlorine budget was produced using ATMOS in 1994, which used ClO, CH_3CCl_3, CFC-112, HOCl and COClF in addition to the species listed previously. This calculation produced a value of 3.53 ± 0.1 ppb, representing a growth rate of 3.3 % per year in the total chlorine VMR for the 1985-1994 period (Zander et al., 1996). Measurements of HCl from HALogen Occultation Experiment (HALOE) between 1991 and 1995 showed concentrations rising at a rate of 0.102 ± 0.006 ppb per year (Russell et al., 1996). Measurements made by the MK-IV balloon borne instruments in 1997 showed a total chlorine VMR of

3.7 ± 0.2 ppb suggesting further increases in atmospheric chlorine at that time (Sen et al., 1999). Data from the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) was used to calculate a stratospheric chlorine budget using measurements made in 2004. This work calculated a mean stratospheric total chlorine VMR of 3.65 ± 0.13 ppb (Nassar et al., 2006a) and provided evidence of the beginning of a slow decline in total stratospheric chlorine. Measurements made by the Microwave Limb Sounder (MLS) between August 2004 and February 2006 showed a reduction in stratospheric total chlorine at a rate of 0.78 ± 0.08 % per year (Froidevaux et al., 2006b). A long-term stratospheric chlorine trend was produced from measurements made by 7 space-borne instruments (ACE-FTS, ATMOS, Aura MLS v1, Cryogenic Limb Array Etalon Spectrometer (CLAES), Cryogenic Infrared Spectrometer & Telescope for the Atmosphere (CRISTA), HALOE and Upper Atmosphere Research Satellite (UARS) MLS v5), for the years between 1991 and 2006. This series showed a general increase in the concentration of stratospheric chlorine until the late 1990s, followed by a slow decrease (Lary et al., 2007).

The chlorine budgets presented in this paper were calculated using ACE-FTS measurements supplemented with data from the SLIMCAT 3D Chemical Transport Model (CTM). Eighteen chlorine-containing species, which are listed in Sect. 2, were used to calculate vertical profiles of total chlorine in 4 latitude bands between 70° N and 70° S. These budgets represent the most comprehensive stratospheric chlorine budget to date. The ACE-FTS instrument has been active since late February 2004 and we are therefore able to chart the long-term changes in VMRs of chlorine-containing molecules for 2004 to 2009. Since atmospheric chlorine can catalyse ozone destruction in the stratosphere the long-term changes in atmospheric chlorine are important for forecasting the recovery of the ozone layer. The large number of chlorine-containing species used in our study allows the calculation of the contribution of each species on the total chlorine VMR. The Global Warming Potential (GWP) and Ozone Depletion Potential (ODP) weighted trends in total chlorine are also presented and illustrate how changes in atmospheric chlorine have affected both climate forcing and ozone depletion. Coupled with the corresponding fluorine budget (Brown et al., 2013), produced from ACE-FTS data this work allows the full implications of the Montreal Protocol on fluorine- and chlorine-containing molecules to be quantified.

Section 2 provides a brief description of ACE-FTS and a discussion on the validation of chlorine-containing species retrieved from ACE-FTS spectra. A very brief description of the SLIMCAT3-D Chemical Transport Model (CTM) is given in Sect. 3 (for a full description of the model readers are directed to Chipperfield, 2006). Section 4 describes our method for calculating the chlorine budget before the results of this work are presented and discussed in Sect. 5.

2 The atmospheric chemistry experiment

The atmospheric chemistry experiment (ACE) Fourier transform spectrometer (FTS) was launched by NASA on-board the Canadian satellite SCISAT-1 in August 2003. ACE-FTS is now in its tenth year of operation with routine data being available from February 2004. SCISAT-1 is in a low circular orbit (650 km altitude) with an inclination angle of 74°to the equator (Bernath et al., 2005), which gives almost global coverage from the Antarctic to the Arctic, and allows ACE-FTS to study “the chemical and dynamical processes that control the distribution of ozone in the stratosphere and upper troposphere” (Bernath, 2006). ACE’s primary instrument is a high-resolution (0.02 cm^{-1}) FTS which operates between 750 and 4400 cm^{-1}. ACE-FTS is a limb viewing solar-occultation spectrometer, i.e. the sun acts as a source of infrared radiation allowing ACE-FTS to record transmission spectra of the limb of the atmosphere at a series of tangent heights during sunrise and sunset. The current version 3.0 of the ACE retrieval includes profiles of over 30 molecules (http://www.ace.uwaterloo.ca), retrieved using the methods outlined by Boone et al. (2005).

The ACE-FTS currently measures 9 chlorine-containing species, CCl_4, CCl_2F_2 (CFC-12), CCl_3F (CFC-11), $COCl_2$, COClF, CHF_2Cl (HCFC-22), CH_3Cl, HCl and $ClONO_2$ with version 3.0 retrievals and 10 years of data from 2004-2013; version 3.0

Title Page
Abstract Introduction
Conclusions References
Tables Figures
◀| |▶
◀ ▶
Back Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

has some problems with the temperature/pressure retrievals after Sept. 2010 (Boone et al., 2013). However, the work presented in this paper was carried out when only data between 2004 and 2009 had been fully processed. The retrieval altitude limits for these species can be seen in Table 1. Version 3.0 also has retrievals for CFC-113, HCFC-142b, and HCFC-141b (Brown et al., 2011), but the retrieved concentrations have substantial biases so we decided to use SLIMCAT 3D Chemical Transport Model data instead. This study used the following SLIMCAT species to supplement ACE-FTS data: CFC-113, CFC-114, CFC-115, H-1211, H-1301, HCFC-141b, HCFC-142b, ClO and HOCl.

2.1 Chlorine-containing species retrieved by the atmospheric chemistry experiment

2.1.1 Carbon tetrachloride – CCl_4

Carbon tetrachloride (CCl_4) was first retrieved from ACE observations for the 2004 global stratospheric chlorine budget (Nassar et al., 2006a). They found that 3 % of the total stratospheric chlorine budget was made up of CCl_4 in 2004. A study of the global atmospheric distribution of CCl_4 from ACE-FTS data was published in Allen et al. (2009). The production of CCl_4 is banned by the Montreal Protocol, and a previous study of measurements of this species from ACE-FTS showed decreasing atmospheric concentrations between 2004 and 2010 (Brown et al., 2011).

2.1.2 Methyl chloride – CH_3Cl

CH_3Cl is the main source of natural chlorine in the atmosphere and was included in the 2004 global stratospheric chlorine budget (Nassar et al., 2006a). Measurements made by ACE-FTS of CH_3Cl biomass plumes have been studied by Rinsland et al. (2007).

2.1.3 CFC-11 – CCl_3F

CFC-11 is the second most abundant CFC in the atmosphere and is banned under the Montreal Protocol. ACE-FTS retrievals of CFC-11 have been validated using measurements from the balloon-borne FIRS-2 instrument, showing an agreement to within 10 % below 16 km (Mahieu et al., 2008). Comparisons between retrievals from ACE-FTS and the balloon-borne FTS, Mk-IV show agreement to 10 % above 12 km and 20 % below 12 km (Mahieu et al., 2008). Non-coincident Mk-IV balloon profiles were also used for validation, producing differences of less than ± 20 % between 17 km and 24 km (Velazco et al., 2011).

2.1.4 CFC-12 – CCl_2F_2

CFC-12 is the most abundant CFC in the atmosphere and is banned under the Montreal Protocol. When compared to FIRS-2 measurements ACE-FTS retrievals showed an agreement to 10 % above 12 km and 20 % below 12 km (Mahieu et al., 2008). Non-coincident retrievals from ACE-FTS were compared by Velazco et al. (2011) to measurements from the Mk-IV instrument who found differences of around ± 10 %.

2.1.5 HCFC-22 – $CHClF_2$

HCFC-22 is the most abundant HCFC in the atmosphere having been widely used since the 1950s. CFCs have been temporarily replaced by HCFCs under the Montreal Protocol. The HCFC-22 retrieval used in this paper is new; a paper is currently in preparation which discusses this new retrieval.

2.1.6 Carbonyl chlorofluoride – COClF

COClF is produced from the decomposition of molecules which contain a single fluorine atom, such as CFC-11. The first global observations of atmospheric COClF from ACE-FTS were made using data from between 2004 and 2007 (Fu et al., 2009). The ACE-

FTS retrieval of COClF was used in both the stratospheric chlorine and fluorine budgets (Nassar et al., 2006a, b).

2.1.7 Phosgene – $COCl_2$

The first study of atmospheric phosgene from ACE-FTS spectra was carried out by Fu et al. (2007). They showed lower concentrations of $COCl_2$ than observations made in the 1980s and 1990s. Comparisons of observations of $COCl_2$ made between 2004 and 2010 by ACE-FTS showed a small, and statistically insignificant, decrease in the stratospheric concentration of $COCl_2$ during this time (Brown et al., 2011).

2.1.8 Chlorine nitrate – $ClONO_2$

Chlorine nitrate ($ClONO_2$) is a reservoir species for atmospheric chlorine, produced by the reaction of ClO with NO_2. During the day $ClONO_2$ is photolysed and so its VMR decreases. In the absence of light $ClONO_2$ increases as it reforms. This diurnal cycle must be accounted for when analysing data because ACE-FTS measures only at sunrise and sunset. The mean difference between ACE-FTS and MIPAS measurements of $ClONO_2$ showed differences of 0.03 ppb at the peak of the profile (Höpfner et al., 2007). Further comparisons have been made between ACE partial column and ground based FTS measurements. The largest difference between these measurements was 21 %, within the uncertainty of ground-based instruments and ACE (Mahieu et al., 2005). $ClONO_2$ was included in the 2004 stratospheric chlorine budget (Nassar et al., 2006a).

2.1.9 Hydrogen chloride – HCl

Hydrogen chloride (HCl) is the main chlorine reservoir in the stratosphere. The concentration of stratospheric HCl from ACE-FTS data has been studied in both the Arctic and Antarctic (Dufour et al., 2006; Mahieu et al., 2005; Santee et al., 2008). The Microwave Limb Sounder (MLS) retrieval of HCl was validated using ACE-FTS measurements (Froidevaux et al., 2006a). MLS HCl profiles were within 5 % of the ACE profiles. ACE-

FTS measurements of HCl have also been compared to balloon-borne Mk-IV instrument and the difference was 7 %. Rinsland et al. (2005) sought to build on the legacy of the ATMOS instrument by calculating the trend in stratospheric HCl between 1985 and 2004 (Rinsland et al., 2005). This work found that between 1994 and 2004 there was a marked decrease in the mixing ratio of HCl, which helped illustrate the success of the Montreal Protocol in reducing the concentration of stratospheric chlorine.

3 SLIMCAT 3-D chemical transport model

ACE observations have been supplemented with output from the SLIMCAT off-line three-dimensional (3-D) chemical transport model (CTM). SLIMCAT contains a detailed treatment of stratospheric chemistry including the major species in the O_x, NO_y, HO_x, Cl_y and Br_y chemical families (Chipperfield, 1999;Feng et al., 2007). Winds from meteorological analyses are used to specify horizontal transport, vertical motion in the stratosphere is calculated from diagnosed heating rates. This approach gives a realistic stratospheric circulation (Chipperfield, 2006; Monge-Sanz et al., 2007).

For this study SLIMCAT was integrated from 1977 to the present day at a horizontal resolution of 5.6° × 5.6° and 32 levels from the surface to about 60 km (run 540). The model used a σ-θ vertical coordinate (Chipperfield, 2006) and was forced by European Centre for Medium Range Weather Forecasts (ECMWF) reanalyses (ERA-Interim from 1989 onwards). The volume mixing ratio of source gases at the surface level were specified using data files compiled for WMO (2007) (Chipperfield et al., 2007). These global mean surface values define the long-term tropospheric source gas trends in the model. The model zonal mean monthly output was averaged to create annual means for 4 latitude bins (70° N–30° N, 30° N–0° N, 0° N–30° S and 30° S–70° S) on a 1 km altitude grid. Two species, ClO and HOCl, were sampled at the local space and time coordinates of the ACE-FTS occultations. This was carried out so that the profiles of ClO and HOCl from SLIMCAT would correspond to the profiles of $ClONO_2$ from ACE-FTS. These species exhibit strong diurnal cycles and so the evening and morning profiles of these species should not be mixed in the overall total chlorine calculations. Chlorine containing species retrieved by ACE-FTS and simulated in the SLIMCAT model have been compared with ACE-FTS in a previous paper (Brown et al., 2013). Generally, there is good agreement between ACE-FTS and SLIMCAT, for a full discussion of the comparisons the reader is directed to Brown et al. (2013).

4 Chlorine budget method

In the latitudes between 70° and the equator, vertical transport in the stratosphere can be broadly divided into two bands (Plumb, 2002). Outside of the tropics (between 70° and 30°) the stratosphere is well mixed by breaking planetary scale waves. In the tropics (between 30° and 0°) powerful convective transport penetrates the tropopause mixing the lower layers of the stratosphere in this area (Jacob, 1999). For this reason in this study the globe has been divided into 4 latitude bands: 70° N–30° N, 30° N–0° N, 0° N–30° S and 30° S–70° S. Here we have used 16,186 ACE occultations; the distribution of these in terms of year of occultation and latitude band are shown in Table 2. Dividing data by year and latitude produced 24 batches of occultations. Each of these groups was filtered individually for outlying data using the median absolute deviation (MAD) of the data. The MAD of the data was used as a filter because it is more robust to outlying data than the standard deviation. Values which were greater than 2.5 times the MAD from the median of the raw data were discarded. Using 2.5 times the MAD allowed between 80 and 90 % of the data to be included (depending on the altitude). Profiles within the polar vortex feature as outliers and so were removed by the MAD filtering.

Once the outliers had been removed an average profile of each species was produced by calculating the average mixing ratio at each altitude. Profiles were extended by using the SLIMCAT profiles of the corresponding species scaled to match the ACE-FTS data at its highest and lowest retrieved altitude point. $COCl_2$ did not have corresponding SLIMCAT data and so were extended vertically upwards using a scaled

COClF SLIMCAT profile from the final retrieved altitude. SLIMCAT runs for HOCl and ClO were sampled for local ACE-FTS occultation time and location. This ensured that the total chlorine concentrations would be calculated correctly for these photosensitive molecules. Some of the profiles of these species (HOCl and ClO) from 2004 appeared to peak at a higher altitude, to a significantly higher maximum VMR than the profiles from the other years. In this case the value for the mixing ratio in 2005 was used instead. Data from 2004 contained only a small number of occultations and so small atmospheric anomalies had a large effect on these profiles. All profiles were extended up to 54.5 km corresponding to the maximum altitude of the corresponding fluorine budget produced from ACE-FTS data (Brown et al., 2013).

The local time of an occultation was calculated using the universal time and the longitude (λ) of the occultation and Eq. (1). This equation produces a local time which is relative to the date of the universal time of the occultation. A negative local time represents an occultation whose local time is a day before the universal time, in this case 24 h should be added to the calculated time for a true local time. A positive local time represents an occultation whose local time is a day ahead of the universal time, in this case 24 h should be subtracted from the calculated time for the true local time. Occultations with local times greater than 12 were considered to be evening occultations. Occultations with local times less than 12 were considered to be morning occultations. Both morning and evening stratospheric chlorine budgets have been constructed as part of this work.

$$\text{Local Time} = \text{Universal Time} + \left(\frac{24}{360}\right)\lambda \tag{1}$$

The total chlorine volume mixing ratio was calculated at 54 levels between 0.5 km and 53.5 km. This spacing reflected the ACE retrieval altitude grid. The equation used for this calculation can be seen below, where square brackets indicate volume mixing ratio (VMR). The total chlorine volume mixing ratio can be expressed as the sum of the total inorganic and organic chlorine, and is defined below.

$$[\text{Total Chlorine}] = [\text{Total Inorganic Chlorine}] + [\text{Total Organic Chlorine}]$$

$$[\text{Total Inorganic Chlorine}] = [\text{ClONO}_2] + [\text{COClF}] + 2[\text{COCl}_2] + [\text{HCl}] + [\text{ClO}] + [\text{HOCl}]$$

$$[\text{Total Organic Chlorine}] = 4[\text{CCl}_4] + 2[\text{CFC-12}] + 3[\text{CFC-11}] + [\text{CH}_3\text{Cl}] + [\text{HCFC-22}] +$$

$$3[\text{CFC-113}] + 2[\text{CFC-114}] + [\text{CFC-115}] + [\text{HCFC-142b}] + 2[\text{HCFC-141b}] + [\text{H-1211}]$$

5 Results and discussion

This section is divided into 6 subsections. In Sect. 5.1 the vertical profiles of the total chlorine calculations are presented along with a focus on the slope of the line and its implications for the changes in total chlorine VMR in the atmosphere. The slope of the correlation plot between inorganic chlorine species and organic chlorine species is presented and discussed in Sect. 5.2. The impact of the individual species included in the total chlorine calculations on the total chlorine VMR are presented in Sect. 5.3. The subsequent sections present the changes in total chlorine between 2004 and 2009. Section 5.4 presents the changes in total stratospheric chlorine during this time. Sections 5.5 and 5.6 discuss the implications of these changes on climate and ozone loss using weighted total chlorine budgets.

5.1 Total chlorine profiles

The results of the total chlorine calculations are shown in Figs. 1 to 4, the error bars shown on the plots are the errors calculated from the standard deviation of each species which was included in the budget. The SLIMCAT profiles have been given a flat 5 % error in their VMR. This value was chosen as it is an overestimation of the error for the ground-based measurements, on which SLIMCAT is based, and accounted for slight errors in transport. The percentage of the total chlorine which comes from ACE-FTS measurements varies between 80 % and 96 %. At lower altitudes ACE-FTS measurements account for around 90 % of the total chlorine VMR. This percentage contribution rises until it peaks at between 23 and 28 km which corresponds to a peak in the percentage contribution of $ClONO_2$. Above this altitude the percentage contribution from ACE-FTS decreases slightly to around 80 %; as the percentage contributions of HOCl and ClO (SLIMCAT) increases. As the VMRs of HOCl and ClO decrease, the percentage contribution to the total chlorine from ACE-FTS increases to around 96 % at 53 km. The total chlorine profiles follow a straight line with slight deviations around 40 km. The mean morning profiles of the inorganic chlorine species between 25 and 55 km between 30° N and the equator can be seen in Fig. 5. It appears that this decrease in the VMR of HCl (that can be seen in Fig. 5) is not compensated for by an increase in the VMR of ClO. It is possible that these dips may be due to a peak in stratospheric OH at this altitude. Stratospheric OH reacts with HCl to form chlorine atoms and water.

The mean total stratospheric chlorine and the slope of the total chlorine profile can be seen in Tables 3 and 4. There is no significant latitudinal difference between the mean total chlorine in the different latitude bands. All total chlorine profiles exhibit a positive slope, showing the VMR of total chlorine generally increases with altitude. Since age-of-air also increases with altitude (Waugh and Hall, 2002) this slope suggests that the concentration of chlorine has been decreasing over time. There does not appear to be a systematic difference between the slopes of the total chlorine in the tropical and sub-tropical latitudes.

5.2 Inorganic chlorine and organic chlorine correlations

Inorganic chlorine species are produced by the breakdown of the organic chlorine species. Therefore, correlating the inorganic and organic species gives an indication as to whether the major chlorine containing species have been included in the budget; a slope close to one would show that all of the major chlorine containing species were included in the budget. The results of these plots can be seen in Table 5. All calculated slopes are greater than 1.01 and smaller than 1.11. These values suggest that the most important chlorine containing species have been included in this budget.

5.3 The contribution of species to the total chlorine budget

Total chlorine profiles are dominated by CFCs in the lower altitudes, with CFCs and halons contributing around 58 % of the total chlorine. The majority of this contribution comes from CFC-11 and CFC-12 which contribute around 20 % and 30 %, respectively. In contrast HCFCs, one of the class of replacement gases for CFCs under the Montreal Protocol, account for 7 % of the total chlorine in this region. As expected above 24 km in the tropics and 19 km in the extra-tropics HCl rapidly dominates the chlorine budget. At its peak, at around 53 km, HCl accounts for around 95 % of the total stratospheric chlorine. The second largest contribution comes from the chlorine reservoir species $ClONO_2$ which peaks at between 20 and 25 % of stratospheric chlorine between 26 and 28 km (dependent on the latitude). It is interesting to note that the contribution of $ClONO_2$ is slightly higher in the extra-tropics than the tropics. The contribution of each species to the total chlorine VMR can be found in the Appendix.

5.4 The change in stratospheric chlorine between 2004 and 2009

The mean of the total stratospheric chlorine was calculated for the years between 2004 and 2009 to form a time series; these plots can be seen in Fig. 6. There does not seem to be significant latitudinal dependence to these time series. The difference between the morning and evening means is not statistically significant. The VMR of the mean total chlorine is decreasing in every latitude band for both the evening and morning measurements. This rate is similar in the Northern Hemisphere and the southern tropics. In the Northern Hemisphere morning occultations show total chlorine decreasing at a rate of 0.40 ± 0.05 % per year and evening show a decrease at a rate of 0.30 ± 0.22 % per year. The errors quoted here are the 1-σ fitting error of a linear least squared fit to the data. This difference in the rate of change in stratospheric chlorine between morning and evening occultations is exhibited in both the northern and southern tropics (30° N–0° N and 0° N–30° S respectively) and the southern sub-tropics (30° S–70° S). The calculated rate of change is larger for morning measurements than for evening measurements. Both southern and northern tropics morning occultations show a similar means with overlapping error ranges. The southern tropics morning occultations show total chlorine decreasing at a rate of −0.43 ± 0.31 % per year and evening occultations show total chlorine decreasing at a rate of −0.65 ± 0.14 % per year. Measurements from the northern extra-tropical stratosphere exhibit a trend very similar to the tropical trends. Morning occultations show mean stratospheric VMR decreasing at a rate of 0.40 ± 0.17 % per year, with evening occultations exhibiting a similar 0.36 ± 0.21 % per year. The rate of decrease appears to be slightly higher in the southern mid-latitude stratosphere. This is exhibited by both the evening and morning results, 0.69 ± 0.17 % per year and 0.84 ± 0.17 % per year respectively. This increased rate of change is due to a slightly higher mean total chlorine value for 2004 (yellow and purple points in Fig. 6). If this value is removed then the rate of change becomes −0.62 ± 0.18 % per year for morning occultations and −0.44 ± 0.13 % per year for evening occultations. These values show stratospheric chlorine is decreasing at a very similar rate independent of latitude, and have been used in the calculation of the global trends.

The global trend for morning occultations is −0.47 ± 0.10 % per year, the evening data produces a trend of −0.44 ± 0.12 % per year. The mean trend (calculated using both morning and evening data) from the Northern Hemisphere is a decrease of −0.37 ± 0.05 % per year. The Southern Hemisphere data exhibits a mean decrease of −0.70 ± 0.08 % per year. The difference between these two rates is likely due to the difference in emissions of chlorine containing molecules from the different hemispheres. The temporary replacement HCFCs contain chlorine which can enter the stratosphere in the tropics before being transported throughout the stratosphere. Whilst these species are rapidly being phased out in the European Union and the United States, their atmospheric concentrations continue to increase (O'Doherty et al., 2004; Montzka et al., 2009; Brown et al., 2011). The emission of these additional chlorine-containing molecules in the Northern Hemisphere would slow the loss rate of chlorine in the Northern Hemisphere producing the different rates seen in this work. The global mean value is a decreasing VMR of total chlorine of 0.46 ± 0.02 % per year. These results are presented in Table 6. Changes in the mean VMR in the total chlorine between 48.5km and 53.5 km between 2004 and 2009 have also been calculated. These results can be been in Table 7. These calculations show no statistically significant hemispheric dependence on the rate of decrease of chlorine. Globally the total chlorine is decreasing at a rate of −0.70 ± 0.12 % per year between these altitudes. This value is very close to the rate calculated by Froidevaux et al. (2006b) of 0.78 ± 0.08 % per year. This suggest that there has been no statistically significant change in the rate at which chlorine is decreasing at higher altitudes during this time.

5.5 Global Warming Potential weighted chlorine budget trends

Many of the species used in this work are potent greenhouse gases; any changes in the VMR of these species will therefore have an effect on climate. Different species have different GWPs leading to different effects on global warming. A useful measure of the effects of changes in the VMR of chlorine-containing species can be gained by calculating the GWP-weighted trend of total chlorine during this time. Since the GWP is a measure of the global warming potential of 1 kg of substance compared to 1 kg of CO_2, the GWP total chlorine cannot be calculated by simply weighting the chlorine source species by their GWP. Instead, the total chlorine must be weighted by the mass of the species and the GWP. The method used in this work to calculate this value was to weight each source species by its GWP and relative molecular mass. The VMR of these species was also weighted by the pressure, which is a measure of the atmospheric mass, at the corresponding altitude. Since the effects of these species are concentrated in the troposphere, data above the tropopause was not included in the calculations of the GWP-weighted total chlorine. The GWP of a species on a 20-yr timeframe was used to weight the individual species. The time series of these means can be seen in Fig. 7.

The results of this analysis can be seen in Table 8. This work shows GWP-weighted total chlorine decreasing most rapidly in the extra-tropical region. The rate of decrease is largest in the extra-tropical Northern Hemisphere, where GWP-weighted chlorine is decreasing at a rate of 0.37 ± 0.09 % per year. In the Southern Hemisphere extra-tropical latitudes the GWP weighted total chlorine is decreasing at a rate of 0.27 ± 0.13 % per year. The tropical latitudes show smaller changes in GWP total chlorine with trends of −0.19 ± 0.11 % per year (Northern Hemisphere) and −0.19 ± 0.13 % per year (Southern Hemisphere). It seems likely that the difference between the rates of decrease of the extra-tropical and tropical latitudes is due to the increase in HCFCs during this time.

If HCFCs are not included in the calculations the rate of decrease becomes significantly higher as can be seen in Table 8. As has been previously noted there has been an increase in the atmospheric VMRs of HCFCs in recent years (Brown et al., 2011; Montzka et al., 2011). HCFCs reach the stratosphere through strong upwelling in the tropics leading to higher VMRs of HCFCs in the upper troposphere lower stratosphere region in tropical latitudes than in extra-tropical latitudes. This would have the effect of reducing the rate of decrease of GWP weighted chlorine in these areas.

5.6 Ozone Depletion Potential-weighted chlorine budget trends

The primary aim of the Montreal Protocol is to reduce the emission of ozone-destroying substances. The calculation of the trend in total atmospheric chlorine is valuable to see how effective the Montreal Protocol has been. The calculation of a trend, weighted by the ODP of the individual species, offers a more specific view of the impact of this reduction on ozone destruction. Like the GWP, ODP is a comparative value: the ozone depleting potential of 1 kg of substance as compared to 1 kg of CFC-11. The ODP-weighted total chlorine is calculated in a similar manner to the GWP weighted total fluorine. Each source species was weighted by its ODP, relative molecular mass and pressure. The results of this analysis can be seen in Table 9. As expected the ODP-weighted total chlorine is decreasing in all latitude bands. The weighted total chlorine is decreasing at the fastest rate in the extra-tropics, where ODP-weighted total chlorine is decreasing at a rate of 1.00 ± 0.19 % per year in the Southern Hemisphere and 0.82 ± 0.09 % per year in the Northern Hemisphere. The tropics exhibit a strong, but slower, rate of decrease of 0.66 ± 0.08 (Northern Hemisphere) and 0.49 ± 0.08 (Southern Hemisphere) % per year. ODP weighted total chlorine is decreasing at equal rate in the Northern Hemisphere (0.74 ± 0.05 % per year) and in the Southern Hemisphere (0.73 ± 0.13 % per year). These results show that the Montreal Protocol has had great success in reducing the emissions of ozone-depleting substances.

6 Conclusions

We have presented chlorine budgets calculated between 2004 and 2009 for the latitude bands of 70° N–30° N, 30° N–0° N, 0° N–30° S, and 30° S–70° S, which represent the extra-tropical and tropical latitudes in the stratosphere. The chlorine budgets were calculated using ACE-FTS version 3.0 retrievals of 9 chlorine-containing species: CCl_4, CCl_2F_2 (CFC-12), CCl_3F (CFC-11), $COCl_2$, COClF, CHF_2Cl (HCFC-22), CH_3Cl, HCl and $ClONO_2$, This data was supplemented with data for the following species from the SLIMCAT 3D chemical transport model: CFC-113, CFC-114, CFC-115, H-1211, H-1301, HCFC-141b, HCFC-142b, ClO and HOCl. Since HOCl, ClO and $ClONO_2$ exhibit diurnal variation, it is necessary to separate the data into morning and evening occultations. This allows both morning and evening total chlorine budgets to be calculated.

The total chlorine profiles are dominated by CFCs at lower altitudes, where CFCs and halons account for 58 % of the total chlorine. This remains the case until around 24 km in the tropics and 19 km in the extra-tropics when HCl rapidly dominates the chlorine budget. At its peak, at around 53 km, HCl accounts for around 95 % of the total stratospheric chlorine. All total chlorine profiles exhibit a positive slope with altitude. A positive slope to the total chlorine profile suggests that the atmospheric volume mixing ratio (VMR) of chlorine is decreasing with time. This is supported by the time series of the mean stratospheric total chlorine budgets. This data shows mean decreases in total stratospheric chlorine of 0.38 ± 0.03 % per year in the Northern Hemisphere extra-tropics, 0.35 ± 0.07 % per year in the Northern Hemisphere tropical stratosphere, 0.54 ± 0.16 % per year in the Southern Hemisphere tropics and 0.53 ± 0.12 % per year in the Southern Hemisphere extra-tropical stratosphere. As expected, these values suggest a very similar decrease in total stratospheric chlorine globally. Globally stratospheric chlorine is decreasing by 0.46 ± 0.02 % per year.

Trends in weighted total chlorine have also been calculated using the ODP and GWP of individual species for weighting. These values allow the impact of the Montreal Protocol on global warming and ozone depletion to be further analysed. GWP-weighted chlorine is decreasing in all latitude bands. However, decreases in the tropics are not significant and appear to be suppressed by the increase in HCFCs during this time. The fastest rate of increase occurs in the extra-tropical Northern Hemisphere where GWP-weighted chlorine is decreasing at a rate of 0.35 ± 0.01 % per year. ODP-weighted chlorine is also decreasing in all bands with the fastest rate in the Southern Hemisphere extra-tropical latitude band. This work suggests that the Montreal Protocol has been successful in reducing the VMR of chlorine-containing gases in the atmosphere. It has also been inadvertently successful in reducing the VMR of GWP-weighted chlorine during this time.

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References

Allen, N. D. C., Bernath, P. F., Boone, C. D., Chipperfield, M. P., Fu, D., Manney, G. L., Oram, D. E., Toon, G. C., and Weisenstein, D. K.: Global carbon tetrachloride distributions obtained from the Atmospheric Chemistry Experiment (ACE), Atmos. Chem. Phys., 9, 7449–7459, doi:10.5194/acp-9-7449-2009, 2009.

Bernath, P. F.: Atmospheric chemistry experiment (ACE): Analytical chemistry from orbit, Trends Anal. Chem., 25, 647–654, doi:10.1016/j.trac.2006.05.001, 2006.

Bernath, P. F., McElroy, C. T., Abrams, M. C., Boone, C. D., Butler, M., Camy-Peyret, C., Carleer, M., Clerbaux, C., Coheur, P. F., Colin, R., DeCola, P., DeMazière, M., Drummond, J. R., Dufour, D., Evans, W. F. J., Fast, H., Fussen, D., Gilbert, K., Jennings, D. E., Llewellyn, E. J., Lowe, R. P., Mahieu, E., McConnell, J. C., McHugh, M., McLeod, S. D., Michaud, R., Midwinter, C., Nassar, R., Nichitiu, F., Nowlan, C., Rinsland, C. P., Rochon, Y. J., Rowlands, N., Semeniuk, K., Simon, P., Skelton, R., Sloan, J. J., Soucy, M. A., Strong, K., Tremblay, P., Turnbull, D., Walker, K. A., Walkty, I., Wardle, D. A., Wehrle, V., Zander, R., and Zou, J.: Atmospheric Chemistry Experiment (ACE): Mission overview, Geophys. Res. Lett., 32, L15S01, doi:10.1029/2005gl022386, 2005.

Boone, C. and Walker, K., Version 3 Retrievals for the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS), in: The Atmospheric Chemistry Experiment ACE at 10: A Solar Occultation Anthology, edited by: Bernath, P., A. Deepak Publishing, Hampton, VA, in press, 2013.

Boone, C. D., Nassar, R., Walker, K. A., Rochon, Y., McLeod, S. D., Rinsland, C. P., and Bernath, P. F.: Retrievals for the atmospheric chemistry experiment Fourier-transform spectrometer, Appl. Opt., 44, 7218–7231, 2005.

Brown, A. T., Chipperfield, M. P., Boone, C., Wilson, C., Walker, K. A., and Bernath, P. F.: Trends in atmospheric halogen containing gases since 2004, J. Quant. Spectrosc. Radiat. Transf., 112, 2552–2566, doi:10.1016/j.jqsrt.2011.07.005, 2011.

Chipperfield, M. P.: Multiannual simulations with a three-dimensional chemical transport model, J. Geophys. Res., 104, 1781–1805, doi:10.1029/98jd02597, 1999.

Chipperfield, M. P.: New Version of the TOMCAT/SLIMCAT Off-Line Chemical Transport Model: Intercomparison of Stratospheric Tracer Experiments, Q. J. R. Meteorol. Soc., 132, 1179–1203, 2006.

Chipperfield, M. P. and Fioletov, V. E., (Lead Authors), Bergman, B., Burrows, J., Connor, B. J., Haigh, J. D., Harris, N. R. P., Hauchecome, A., Hood, L. L., Kawa, S. R., Kryscin, J. W., Logan, J. A., Muthama, N. J., Polyani, L., Randel, W. J., Sasaki, T., Stahelin, J., Stolarski, R. S., Thomason, L. W., and Zawodny, J. M.: Global Ozone: Past and Present, Chapter 3 in Scientific Assessment of Ozone Depletion: 2006, Global Ozone Research and Monitoring Project-Report No. 50, 572 pp., World Meteorological Organization, Geneva, Switzerland, 2007.

Dufour, G., Nassar, R., Boone, C. D., Skelton, R., Walker, K. A., Bernath, P. F., Rinsland, C. P., Semeniuk, K., Jin, J. J., McConnell, J. C., and Manney, G. L.: Partitioning between the inorganic chlorine reservoirs HCl and ClONO2 during the Arctic winter 2005 from the ACE-FTS, Atmos. Chem. Phys., 6, 2355–2366, doi:10.5194/acp-6-2355-2006, 2006.

Feng, W., Chipperfield, M. P., Dorf, M., Pfeilsticker, K., and Ricaud, P.: Mid-latitude ozone changes: studies with a 3-D CTM forced by ERA-40 analyses, Atmos. Chem. Phys., 7, 2357–2369, doi:10.5194/acp-7-2357-2007, 2007.

Froidevaux, L., Livesey, N. J., Read, W. G., Jiang, Y. B., Jimenez, C., Filipiak, M. J., Schwartz, M. J., Santee, M. L., Pumphrey, H. C., Jiang, J. H., Wu, D. L., Manney, G. L., Drouin, B. J., Waters, J. W., Fetzer, E. J., Bernath, P. F., Boone, C. D., Walker, K. A., Jucks, K. W., Toon, G. C., Margitan, J. J., Sen, B., Webster, C. R., Christensen, L. E., Elkins, J. W., Atlas, E., Lueb, R. A., and Hendershot, R.: Early validation analyses of atmospheric profiles from EOS MLS on the aura Satellite, IEEE Transact. Geosci. Remote Sens., 44, 1106–1121, doi:10.1109/tgrs.2006.864366, 2006a.

Froidevaux, L., Livesey, N. J., Read, W. G., Salawitch, R. J., Waters, J. W., Drouin, B., MacKenzie, I. A., Pumphrey, H. C., Bernath, P., Boone, C., Nassar, R., Montzka, S., Elkins, J., Cunnold, D., and Waugh, D.: Temporal decrease in upper atmospheric chlorine, Geophys. Res. Lett., 33, L23812, doi:10.1029/2006gl027600, 2006b.

Fu, D., Boone, C. D., Bernath, P. F., Walker, K. A., Nassar, R., Manney, G. L., Global phosgene observations from the atmospheric chemistry experiment (ACE) mission, Geophys Res Lett., 34, 2007.

Fu, D., Boone, C. D., Bernath, P. F., Weisenstein, D. K., Rinsland, C. P., Manney, G. L., and Walker, K. A.: First global observations of atmospheric COClF from the Atmospheric Chemistry Experiment mission, J. Quant. Spectrosc. Radiat. Transf., 110, 974–985, doi:10.1016/j.jqsrt.2009.02.018, 2009.

Höpfner, M., von Clarmann, T., Fischer, H., Funke, B., Glatthor, N., Grabowski, U., Kellmann, S., Kiefer, M., Linden, A., Milz, M., Steck, T., Stiller, G. P., Bernath, P., Blom, C. E., Blumenstock, T., Boone, C., Chance, K., Coffey, M. T., Friedl-Vallon, F., Griffith, D., Hannigan, J. W., Hase, F., Jones, N., Jucks, K. W., Keim, C., Kleinert, A., Kouker, W., Liu, G. Y., Mahieu, E., Mellqvist, J., Mikuteit, S., Notholt, J., Oelhaf, H., Piesch, C., Reddmann, T., Ruhnke, R., Schneider, M., Strandberg, A., Toon, G., Walker, K. A., Warneke, T., Wetzel, G., Wood, S., and Zander, R.: Validation of MIPAS $ClONO_2$ measurements, Atmos. Chem. Phys., 7, 257–281, doi:10.5194/acp-7-257-2007, 2007.

Jacob, D. J.: Introduction to Atmospheric Chemistry, Princeton University Press, New Jersey, p. 79, 1999.

Lary, D. J., Waugh, D. W., Douglass, A. R., Stolarski, R. S., Newman, P. A., and Mussa, H.: Variations in stratospheric inorganic chlorine between 1991 and 2006, Geophys. Res. Lett., 34, L21811, doi:10.1029/2007gl030053, 2007.

Mahieu, E., Zander, R., Duchatelet, P., Hannigan, J. W., Coffey, M. T., Mikuteit, S., Hase, F., Blumenstock, T., Wiacek, A., Strong, K., Taylor, J. R., Mittermeier, R. L., Fast, H., Boone, C. D., McLeod, S. D., Walker, K. A., Bernath, P. F., and Rinsland, C. P.: Comparisons between ACE-FTS and ground-based measurements of stratospheric HCl and ClONO2 loadings at northern latitudes, Geophys. Res. Lett., 32, L15S08, doi:10.1029/2005gl022396, 2005.

Mahieu, E., Duchatelet, P., Demoulin, P., Walker, K. A., Dupuy, E., Froidevaux, L., Randall, C., Catoire, V., Strong, K., Boone, C. D., Bernath, P. F., Blavier, J. F., Blumenstock, T., Coffey, M., De Mazière, M., Griffith, D., Hannigan, J., Hase, F., Jones, N., Jucks, K. W., Kagawa, A., Kasai, Y., Mebarki, Y., Mikuteit, S., Nassar, R., Notholt, J., Rinsland, C. P., Robert, C., Schrems, O., Senten, C., Smale, D., Taylor, J., Tétard, C., Toon, G. C., Warneke, T., Wood, S. W., Zander, R., and Servais, C.: Validation of ACE-FTS v2.2 measurements of HCl, HF, CCl3F and CCl2F2 using space-, balloon- and ground-based instrument observations, Atmos. Chem. Phys., 8, 6199-6221, doi:10.5194/acp-8-6199-2008, 2008.

Monge-Sanz, B. M., Chipperfield, M. P., Simmons, A. J., and Uppala, S. M.: Mean age of air and transport in a CTM: Comparison of different ECMWF analyses, Geophys. Res. Lett., 34, L04801, doi:10.1029/2006gl028515, 2007.

Montzka, S. A., Hall, B. D., and Elkins, J. W.: Accelerated increases observed for hydrochlorofluorocarbons since 2004 in the global atmosphere, Geophys. Res. Lett., 36, L03804, doi:10.1029/2008gl036475, 2009.

Montzka, S. A., Reimann, S., (Coordinating Lead Authors), Engel , A., Krüger, K., O'Doherty, S., Sturges, W. T., Blake, D., Dorf, M., Fraser, P., Froidevaux, L., Jucks, K., Kreher, K., Kurylo, M. J., Mellouki, A., Miller, J., Nielsen, O.-J., Orkin, V. L., Prinn, R. G., Rhew, R., Santee, M. L., Stohl, A., and Verdonik, D.: Ozone-Depleting Substances (ODSs) and Related Chemicals, Chapter 1 in Scientific Assessment of Ozone Depletion: 2010, Global Ozone Research and Monitoring Project–Report No. 52, World Meteorological Organization, Geneva, Switzerland, 2011.

Nassar, R., Bernath, P. F., Boone, C. D., Clerbaux, C., Coheur, P. F., Dufour, G., Froidevaux, L., Mahieu, E., McConnell, J. C., McLeod, S. D., Murtagh, D. P., Rinsland, C. P., Semeniuk, K., Skelton, R., Walker, K. A., and Zander, R.: A global inventory of stratospheric chlorine in 2004, J. Geophys. Res., 111, D22312, doi:10.1029/2006JD007073, 2006a.

Nassar, R., Bernath, P. F., Boone, C. D., McLeod, S. D., Skelton, R., Walker, K. A., Rinsland, C. P., and Duchatelet, P.: A global inventory of stratospheric fluorine in 2004 based on Atmospheric Chemistry Experiment Fourier transform spectrometer (ACE-FTS) measurements, J. Geophys. Res., 111, D22313, doi:10.1029/2006jd007395, 2006b.

O'Doherty, S., Cunnold, D. M., Manning, A., Miller, B. R., Wang, R. H. J., Krummel, P. B., Fraser, P. J., Simmonds, P. G., McCulloch, A., Weiss, R. F., Salameh, P., Porter, L. W., Prinn, R. G., Huang, J., Sturrock, G., Ryall, D., Derwent, R. G., and Montzka, S. A.: Rapid growth of hydrofluorocarbon 134a and hydrochlorofluorocarbons 141b, 142b, and 22 from Advanced Global Atmospheric Gases Experiment (AGAGE) observations at Cape Grim, Tasmania, and Mace Head, Ireland, J. Geophys. Res., 109, D06310, doi:10.1029/2003jd004277, 2004.

Plumb, R. A.: Stratospheric Transport, Journal of Meteorological Society of Japan, 80, 793–809, 2002.

Rinsland, C. P., Boone, C., Nassar, R., Walker, K., Bernath, P., Mahieu, E., Zander, R., McConnell, J. C., and Chiou, L.: Trends of HF, HCl, CCl2F2, CCl3F, CHClF2 (HCFC-22), and SF6 in the lower stratosphere from Atmospheric Chemistry Experiment (ACE) and Atmospheric Trace Molecule Spectroscopy (ATMOS) measurements near 30° N latitude, Geophys. Res. Lett., 32, L16S03, doi:10.1029/2005gl022415, 2005.

Rinsland, C. P., Dufour, G., Boone, C. D., Bernath, P. F., Chiou, L., Coheur, P.-F., Turquety, S., and Clerbaux, C.: Satellite boreal measurements over Alaska and Canada during June-July

2004: Simultaneous measurements of upper tropospheric CO, C_2H_6, HCN, CH_3Cl, CH_4, C_2H_2, CH_3OH, HCOOH, OCS, and SF_6 mixing ratios, Global Biogeochem. Cy., 21, GB3008, doi:10.1029/2006GB002795, 2007.

Russell, J. M., Luo, M., Cicerone, R. J., and Deaver, L. E.: Satellite confirmation of the dominance of chlorofluorocarbons in the global stratospheric chlorine budget, Nature, 379, 526–529, 1996.

Santee, M. L., MacKenzie, I. A., Manney, G. L., Chipperfield, M. P., Bernath, P. F., Walker, K. A., Boone, C. D., Froidevaux, L., Livesey, N. J., and Waters, J. W.: A study of stratospheric chlorine partitioning based on new satellite measurements and modeling, J. Geophys. Res., 113, D12307, doi:10.1029/2007jd009057, 2008.

Sen, B., Osterman, G. B., Salawitch, R. J., Toon, G. C., Margitan, J. J., Blavier, J. F., Chang, A. Y., May, R. D., Webster, C. R., Stimpfle, R. M., Bonne, G. P., Voss, P. B., Perkins, K. K., Anderson, J. G., Cohen, R. C., Elkins, J. W., Dutton, G. S., Hurst, D. F., Romashkin, P. A., Atlas, E. L., Schauffler, S. M., and Loewenstein, M.: The budget and partitioning of stratospheric chlorine during the 1997 Arctic summer, J. Geophys. Res., 104, 26653–26665, doi:10.1029/1999jd900245, 1999.

UNEP: Handbook for the Montreal Protocol on Substances that Deplete the Ozone Layer 8ed., Secretariat for The Vienna Convention for the Protection of the Ozone Layer & The Montreal Protocol on Substances that Deplete the Ozone Layer, Kenya, 2009.

Velazco, V. A., Toon, G. C., Blavier, J.-F. L., Kleinböhl, A., Manney, G. L., Daffer, W. H., Bernath, P. F., Walker, K. A., and Boone, C.: Validation of the Atmospheric Chemistry Experiment by noncoincident MkIV balloon profiles, J. Geophys. Res., 116, D06306, doi:10.1029/2010jd014928, 2011.

Waugh, D. W. and Hall, T. M.: Age of stratospheric air: Theory, observations, and models, Rev. Geophys., 40, 1010, doi:10.1029/2000RG000101, 2002.

Zander, R., Gunson, M. R., Farmer, C. B., Rinsland, C. P., Irion, F. W., and Mahieu, E.: The 1985 chlorine and fluorine inventories in the stratosphere based on ATMOS observations at 30° north latitude, Journal of Atmospheric Chemistry, 15, 171-186, doi:10.1007/bf00053758, 1992.

Zander, R., Mahieu, E., Gunson, M. R., Abrams, M. C., Chang, A. Y., Abbas, M., Aellig, C., Engel, A., Goldman, A., Irion, F. W., Kämpfer, N., Michelson, H. A., Newchurch, M. J., Rinsland, C. P., Salawitch, R. J., Stiller, G. P., and Toon, G. C.: The 1994 northern midlatitude budget of stratospheric chlorine derived from ATMOS/ATLAS‐3 observations, Geophys. Res. Lett., 23, 2357–2360, doi:10.1029/96gl01792, 1996.

Table 1. The ACE retrieval altitude ranges of the chlorine-containing species used in this study.

Species	Altitude (km)	
	Polar	Equatorial
CFC-11 (CCl_3F)	5–23	6–28
CFC-12 (CCl_2F_2)	5–28	5–36
HCFC-22 (CHF_2Cl)	5–30	7–30
COClF	13–25	15–32
$COCl_2$	8–24	10–29
CH_3Cl	9–40	12–40
CCl_4	6–25	7–30
HCl	6–57	7–63
$ClONO_2$	10–41	10–36

Table 2. The number of occultations used in this study.

Year	70° N to 30° N	30° N to 0° N	0° N to 30° S	30° S to 70° S	Total
2004	788	171	153	960	2072
2005	1631	278	301	1592	3802
2006	1166	164	178	1038	2546
2007	887	116	129	804	1936
2008	1449	134	197	1427	3207
2009	1162	168	202	1091	2623

Table 3. The mean stratospheric total chlorine volume mixing ratio (ppb).

Morning				
Year	70° N to 30° N	30° N to 0° N	0° N to 30° S	30° S to 70° S
2004	3.53 ± 0.18	3.46 ± 0.15	3.54 ± 0.13	3.60 ± 0.09
2005	3.54 ± 0.14	3.46 ± 0.12	3.47 ± 0.14	3.53 ± 0.11
2006	3.52 ± 0.13	3.44 ± 0.12	3.42 ± 0.14	3.51 ± 0.19
2007	3.51 ± 0.10	3.43 ± 0.12	3.42 ± 0.14	3.47 ± 0.19
2008	3.52 ± 0.12	3.41 ± 0.13	3.41 ± 0.14	3.43 ± 0.16
2009	3.44 ± 0.18	3.40 ± 0.13	3.47 ± 0.08	3.46 ± 0.22
Evening				
Year	70° N to 30° N	30° N to 0° N	0° N to 30° S	30° S to 70° S
2004	3.55 ± 0.18	3.46 ± 0.14	3.53 ± 0.13	3.57 ± 0.15
2005	3.55 ± 0.18	3.49 ± 0.10	3.49 ± 0.15	3.50 ± 0.17
2006	3.47 ± 0.13	3.43 ± 0.12	3.44 ± 0.13	3.46 ± 0.15
2007	3.51 ± 0.15	3.46 ± 0.12	3.44 ± 0.11	3.45 ± 0.14
2008	3.47 ± 0.11	3.39 ± 0.13	3.41 ± 0.11	3.43 ± 0.18
2009	3.50 ± 0.13	3.44 ± 0.10	3.42 ± 0.14	3.44 ± 0.22

Title Page
Abstract | Introduction
Conclusions | References
Tables | Figures
◀| |▶
◀ ▶
Back | Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

Table 4. The vertical slopes of the total chlorine profiles (km/ppb) for both evening and morning occultations in the 4 latitude bins used in this study.

Year	70° N to 30° N	30° N to 0° N	0° N to 30° S	30° S to 70° S
Morning				
2004	0.010 ± 0.001	0.004 ± 0.002	0.006 ± 0.001	0.003 ± 0.001
2005	0.009 ± 0.001	0.004 ± 0.001	0.006 ± 0.001	0.007 ± 0.001
2006	0.007 ± 0.001	0.005 ± 0.001	0.005 ± 0.001	0.010 ± 0.002
2007	0.004 ± 0.001	0.005 ± 0.001	0.003 ± 0.002	0.011 ± 0.002
2008	0.006 ± 0.001	0.004 ± 0.002	0.004 ± 0.002	0.010 ± 0.001
2009	0.011 ± 0.002	0.005 ± 0.001	0.006 ± 0.000	0.013 ± 0.002
Evening				
Year	70° N to 30° N	30° N to 0° N	0° N to 30° S	30° S to 70° S
2004	0.011 ± 0.001	0.006 ± 0.001	0.009 ± 0.001	0.007 ± 0.001
2005	0.013 ± 0.001	0.006 ± 0.001	0.008 ± 0.001	0.010 ± 0.001
2006	0.008 ± 0.001	0.006 ± 0.001	0.006 ± 0.001	0.011 ± 0.001
2007	0.010 ± 0.001	0.008 ± 0.001	0.006 ± 0.001	0.010 ± 0.001
2008	0.005 ± 0.001	0.004 ± 0.001	0.004 ± 0.001	0.012 ± 0.001
2009	0.004 ± 0.001	0.006 ± 0.001	0.006 ± 0.001	0.014 ± 0.002

Table 5. The correlation between organic chlorine species (CCl_4, CFC-12, CFC-11, HCFC-22, CH_3Cl, CFC-113, CFC-114, CFC-115, H-1211, HCFC-141b and HCFC-142b) and inorganic chlorine (HCl, $COCl_2$, $ClONO_2$, ClO, HOCl and COClF).

Morning				
Year	70° N to 30° N	30° N to 0° N	0° N to 30° S	30° S to 70° S
2004	−1.07 ± 0.01	−1.02 ± 0.01	−1.05 ± 0.01	−1.06 ± 0.01
2005	−1.08 ± 0.01	−1.02 ± 0.01	−1.07 ± 0.02	−1.08 ± 0.01
2006	−1.07 ± 0.01	−1.03 ± 0.01	−1.03 ± 0.01	−1.11 ± 0.01
2007	−1.03 ± 0.01	−1.03 ± 0.01	−1.00 ± 0.01	−1.10 ± 0.01
2008	−1.06 ± 0.01	−1.01 ± 0.01	−1.01 ± 0.01	−1.09 ± 0.01
2009	−1.07 ± 0.01	−1.01 ± 0.01	−1.05 ± 0.00	−1.11 ± 0.02
Mean	−1.06 ± 0.02	−1.02 ± 0.01	−1.04 ± 0.03	−1.09 ± 0.02

Evening				
Year	70° N to 30° N	30° N to 0° N	0° N to 30° S	30° S to 70° S
2004	−1.08 ± 0.01	−1.03 ± 0.01	−1.06 ± 0.01	−1.05 ± 0.01
2005	−1.09 ± 0.01	−1.04 ± 0.01	−1.09 ± 0.02	−1.07 ± 0.01
2006	−1.06 ± 0.01	−1.03 ± 0.01	−1.04 ± 0.01	−1.08 ± 0.01
2007	−1.05 ± 0.01	−1.05 ± 0.01	−1.04 ± 0.01	−1.08 ± 0.01
2008	−1.03 ± 0.01	−1.01 ± 0.01	−1.01 ± 0.01	−1.10 ± 0.01
2009	−1.04 ± 0.01	−1.03 ± 0.01	−1.03 ± 0.01	−1.11 ± 0.02
Mean	−1.06 ± 0.02	−1.03 ± 0.01	−1.05 ± 0.03	−1.08 ± 0.02

Title Page
Abstract | Introduction
Conclusions | References
Tables | Figures
◀| |▶
◀ ▶
Back | Close
Full Screen / Esc
Printer-friendly Version
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Table 6. The trends in the mean stratospheric total chlorine volume mixing ratio (ppb), between 2004 and 2009 for different regions and for morning, evening and combined morning and evening occultations. The errors on the morning and evening values are the 1-σ fitting error of a linear least squared fit to the data. The errors on the mean values are the standard deviations of the corresponding calculations.

	Morning					
	ppb yr^{-1}			% yr^{-1}		
70° N–30° N	−0.014	±	0.006	−0.40	±	0.17
30° N–0° N	−0.014	±	0.002	−0.40	±	0.05
0° N–30° S	−0.015	±	0.011	−0.43	±	0.31
30° S–70° S	−0.022	±	0.006	−0.62	±	0.18
Northern Hemisphere	−0.014	±	0.003	−0.40	±	0.09
Southern Hemisphere	−0.026	±	0.006	−0.75	±	0.18
Global	−0.016	±	0.004	−0.47	±	0.10
	Evening					
	ppb yr^{-1}			% yr^{-1}		
70° N–30° N	−0.013	±	0.007	−0.36	±	0.21
30° N–0° N	−0.010	±	0.008	−0.30	±	0.22
0° N–30° S	−0.023	±	0.005	−0.65	±	0.14
30° S–70° S	−0.015	±	0.005	−0.44	±	0.13
Northern Hemisphere	−0.012	±	0.007	−0.33	±	0.20
Southern Hemisphere	−0.022	±	0.004	−0.64	±	0.11
Global	−0.015	±	0.004	−0.44	±	0.12
	Mean (Morning & Evening)					
	ppb yr^{-1}			% yr^{-1}		
70° N–30° N	−0.013	±	0.001	−0.38	±	0.03
30° N–0° N	−0.012	±	0.002	−0.35	±	0.07
0° N–30° S	−0.019	±	0.005	−0.54	±	0.16
30° S–70° S	−0.018	±	0.004	−0.53	±	0.12
Northern Hemisphere	−0.013	±	0.002	−0.37	±	0.05
Southern Hemisphere	−0.024	±	0.003	−0.70	±	0.08
Global	−0.016	±	0.001	−0.46	±	0.02

Table 7. The trends in the mean stratospheric total chlorine volume mixing ratio (ppb), between 2004 and 2009 for different regions and for morning, evening and combined morning and evening occultations. These values correspond to the changes in the total chlorine between the altitudes of 48.5 and 53.5 km. The errors on the morning and evening values are the 1-σ fitting error of a linear least squared fit to the data. The errors on the mean values are the standard deviations of the corresponding calculations.

	ppb yr^{-1}	% yr^{-1}
Mean (Morning)		
70° N–30° N	−0.022 ± 0.006	−0.59 ± 0.17
30° N–0° N	−0.017 ± 0.004	−0.47 ± 0.10
0° N–30° S	−0.031 ± 0.004	−0.84 ± 0.11
30° S–70° S	−0.019 ± 0.011	−0.52 ± 0.30
Northern Hemisphere	−0.020 ± 0.003	−0.53 ± 0.09
Southern Hemisphere	−0.025 ± 0.008	−0.68 ± 0.22
Global	−0.022 ± 0.006	−0.60 ± 0.16
Mean (Evening)		
70° N–30° N	−0.034 ± 0.014	−0.89 ± 0.36
30° N–0° N	−0.019 ± 0.004	−0.50 ± 0.10
0° N–30° S	−0.030 ± 0.007	−0.80 ± 0.19
30° S–70° S	−0.035 ± 0.016	−0.95 ± 0.43
Northern Hemisphere	−0.026 ± 0.011	−0.70 ± 0.27
Southern Hemisphere	−0.033 ± 0.004	−0.88 ± 0.10
Global	−0.029 ± 0.008	−0.79 ± 0.20
Mean (Morning & Evening)		
70° N–30° N	−0.028 ± 0.008	−0.74 ± 0.21
30° N–0° N	−0.018 ± 0.001	−0.49 ± 0.02
0° N–30° S	−0.030 ± 0.001	−0.82 ± 0.02
30° S–70° S	−0.027 ± 0.011	−0.73 ± 0.30
Northern Hemisphere	−0.023 ± 0.007	−0.61 ± 0.18
Southern Hemisphere	−0.029 ± 0.002	−0.78 ± 0.06
Global	−0.026 ± 0.004	−0.70 ± 0.12

Table 8. The global warming potential-weighted trends in the total chlorine volume mixing ratio (% yr^{-1}) between 2004 and 2009. The errors quoted here are the 1-σ fitting error of a linear least squared fit to the data. The weighted total chlorine budgets were also calculated without including HCFCs so that the impact of the replacement of CFCs by HCFCs, under the Montreal Protocol, on global warming potential weighted chlorine could be quantified.

Latitude band	Weighted total chlorine trend (% yr^{-1})	Weighted total chlorine trend without HCFCs (% yr^{-1})
70° N–30° N	−0.37 ± 0.09	−0.72 ± 0.06
30° N–0° N	−0.19 ± 0.11	−0.58 ± 0.12
0° N–30° S	−0.19 ± 0.13	−0.51 ± 0.11
30° S–70° S	−0.27 ± 0.12	−0.57 ± 0.03
Northern Hemisphere	−0.31 ± 0.08	−0.68 ± 0.02
Southern Hemisphere	−0.23 ± 0.08	−0.56 ± 0.11

Table 9. The ozone depletion potential-weighted trends in the total chlorine volume mixing ratio between 2004 and 2009. The error on the trends is the 1-σ error on the least squared fit.

Latitude band	Weighted total chlorine trend (% yr^{-1}))		
70° N–30° N	−0.82	±	0.09
30° N–0° N	−0.66	±	0.08
0° N–30° S	−0.49	±	0.08
30° S–70° S	−1.0	±	0.19
Northern Hemisphere	−0.74	±	0.05
Southern Hemisphere	−0.73	±	0.13

Table A1. Chlorine Budgets.

The percentage contribution of HCl, CCl_4, CFC-12, CFC-11 COClF, $COCl_2$, HCFC-22, CH_3Cl and CFC-113 to the total chlorine budget in the latitude bands between 70° N and 30° N (evening occultations.

Altitude	HCl	CCl_4	CFC-12	CFC-11	COClF	$COCl_2$	HCFC-22	CH_3Cl	CFC-113
10.5	3.96	12.08	30.09	20.65	1.07	0.18	5.55	16.72	6.56
11.5	6.29	11.20	29.62	19.94	1.14	0.29	5.48	16.27	6.29
12.5	8.52	10.38	29.15	19.16	1.19	0.39	5.40	15.79	6.02
13.5	10.53	9.79	28.46	18.38	1.21	0.44	5.41	15.58	5.81
14.5	12.63	9.19	27.77	17.51	1.21	0.49	5.40	15.37	5.58
15.5	15.65	8.53	26.95	16.28	1.22	0.56	5.26	14.44	5.35
16.5	19.80	7.71	25.74	14.66	1.24	0.67	4.94	12.91	5.01
17.5	24.88	6.65	24.15	12.72	1.27	0.83	4.58	11.20	4.59
18.5	30.33	5.58	22.42	10.50	1.31	0.99	4.31	9.60	4.11
19.5	35.41	4.50	20.56	8.07	1.30	1.14	4.10	8.13	3.63
20.5	39.88	3.56	18.72	5.65	1.20	1.24	3.90	6.88	3.20
21.5	43.49	2.77	16.91	3.82	1.04	1.28	3.73	5.78	2.81
22.5	46.66	1.83	15.15	2.49	0.86	1.22	3.59	4.90	2.46
23.5	49.34	1.18	13.46	1.51	0.70	1.10	3.47	4.07	2.11
24.5	51.89	0.62	11.72	0.92	0.63	0.94	3.34	3.25	1.78
25.5	54.42	0.28	10.00	0.52	0.50	0.77	3.22	2.50	1.46
26.5	56.97	0.10	8.32	0.26	0.38	0.61	3.09	1.83	1.16
27.5	59.44	0.05	6.77	0.14	0.28	0.45	2.96	1.31	0.90
28.5	62.05	0.01	5.36	0.05	0.20	0.32	2.83	0.92	0.68
29.5	64.61	0.01	4.22	0.03	0.14	0.22	2.72	0.62	0.52
30.5	67.07	0.00	3.20	0.01	0.08	0.13	2.58	0.46	0.37
31.5	69.28	0.00	2.42	0.00	0.05	0.08	2.46	0.38	0.26
32.5	71.42	0.00	1.81	0.00	0.03	0.05	2.36	0.30	0.18
33.5	73.26	0.00	1.26	0.00	0.01	0.02	2.26	0.23	0.11
34.5	75.59	0.00	0.94	0.00	0.01	0.01	2.20	0.19	0.08
35.5	77.00	0.00	0.66	0.00	0.01	0.01	2.12	0.14	0.05
36.5	77.72	0.00	0.47	0.00	0.00	0.00	2.04	0.11	0.03
37.5	77.60	0.00	0.33	0.00	0.00	0.00	1.95	0.09	0.02
38.5	77.56	0.00	0.23	0.00	0.00	0.00	1.86	0.06	0.01
39.5	77.78	0.00	0.17	0.00	0.00	0.00	1.77	0.05	0.01
40.5	78.07	0.00	0.12	0.00	0.00	0.00	1.69	0.04	0.01
41.5	78.68	0.00	0.08	0.00	0.00	0.00	1.60	0.03	0.00
42.5	79.49	0.00	0.06	0.00	0.00	0.00	1.52	0.02	0.00
43.5	80.60	0.00	0.04	0.00	0.00	0.00	1.45	0.02	0.00
44.5	81.98	0.00	0.03	0.00	0.00	0.00	1.37	0.01	0.00
45.5	83.53	0.00	0.02	0.00	0.00	0.00	1.31	0.01	0.00
46.5	85.10	0.00	0.01	0.00	0.00	0.00	1.25	0.01	0.00
47.5	86.78	0.00	0.01	0.00	0.00	0.00	1.20	0.01	0.00
48.5	88.32	0.00	0.01	0.00	0.00	0.00	1.15	0.00	0.00
49.5	89.71	0.00	0.00	0.00	0.00	0.00	1.10	0.00	0.00
50.5	90.95	0.00	0.00	0.00	0.00	0.00	1.06	0.00	0.00
51.5	91.95	0.00	0.00	0.00	0.00	0.00	1.03	0.00	0.00
52.5	92.82	0.00	0.00	0.00	0.00	0.00	1.00	0.00	0.00
53.5	93.61	0.00	0.00	0.00	0.00	0.00	0.98	0.00	0.00

Table A2. The percentage contribution of CFC-114, CFC-115, HCFC-142b, HCFC-141b, $ClONO_2$, ClO, HOCl and H-1211 to the total chlorine budget in the latitude bands between 70° N and 30° N (evening occultations).

Altitude	CFC-114	CFC-115	HCFC-142b	HCFC-141b	$ClONO_2$	ClO	HOCl	H-1211
10.5	0.96	0.27	0.45	1.03	0.30	0.01	0.04	0.11
11.5	0.93	0.26	0.43	0.97	0.75	0.01	0.03	0.10
12.5	0.91	0.26	0.42	0.91	1.37	0.01	0.03	0.09
13.5	0.88	0.25	0.40	0.88	1.85	0.01	0.03	0.09
14.5	0.86	0.25	0.39	0.84	2.41	0.01	0.03	0.08
15.5	0.83	0.25	0.38	0.79	3.39	0.02	0.03	0.07
16.5	0.80	0.24	0.36	0.73	5.08	0.02	0.03	0.06
17.5	0.76	0.24	0.34	0.66	7.03	0.03	0.04	0.05
18.5	0.71	0.23	0.32	0.57	8.86	0.05	0.06	0.04
19.5	0.66	0.23	0.30	0.48	11.34	0.08	0.07	0.03
20.5	0.63	0.22	0.28	0.41	14.02	0.12	0.09	0.02
21.5	0.59	0.22	0.27	0.34	16.67	0.15	0.11	0.01
22.5	0.57	0.22	0.25	0.29	19.20	0.18	0.14	0.00
23.5	0.54	0.21	0.24	0.23	21.45	0.22	0.16	0.00
24.5	0.50	0.21	0.23	0.19	23.30	0.28	0.19	0.00
25.5	0.47	0.20	0.22	0.14	24.70	0.39	0.21	0.00
26.5	0.43	0.20	0.20	0.11	25.50	0.57	0.25	0.00
27.5	0.40	0.20	0.19	0.08	25.71	0.82	0.31	0.00
28.5	0.36	0.19	0.18	0.05	25.37	1.07	0.37	0.00
29.5	0.33	0.19	0.16	0.04	24.36	1.39	0.45	0.00
30.5	0.30	0.19	0.15	0.02	22.79	2.05	0.59	0.00
31.5	0.27	0.19	0.14	0.01	20.58	3.06	0.80	0.00
32.5	0.25	0.18	0.13	0.01	17.87	4.34	1.04	0.00
33.5	0.23	0.18	0.12	0.00	14.91	6.05	1.33	0.00
34.5	0.22	0.18	0.12	0.00	10.78	8.01	1.67	0.00
35.5	0.20	0.18	0.11	0.00	7.63	9.99	1.88	0.00
36.5	0.19	0.18	0.11	0.00	4.75	12.40	2.00	0.00
37.5	0.17	0.17	0.10	0.00	2.92	14.61	2.03	0.00
38.5	0.16	0.17	0.09	0.00	1.83	16.06	1.95	0.00
39.5	0.15	0.17	0.09	0.00	1.02	16.97	1.81	0.00
40.5	0.14	0.16	0.08	0.00	0.56	17.48	1.65	0.00
41.5	0.13	0.16	0.08	0.00	0.31	17.47	1.46	0.00
42.5	0.12	0.16	0.07	0.00	0.16	17.15	1.25	0.00
43.5	0.11	0.15	0.07	0.00	0.07	16.44	1.05	0.00
44.5	0.10	0.15	0.06	0.00	0.04	15.39	0.87	0.00
45.5	0.09	0.15	0.06	0.00	0.02	14.12	0.70	0.00
46.5	0.08	0.14	0.05	0.00	0.01	12.78	0.56	0.00
47.5	0.07	0.14	0.05	0.00	0.00	11.29	0.44	0.00
48.5	0.06	0.14	0.05	0.00	0.00	9.92	0.34	0.00
49.5	0.06	0.14	0.04	0.00	0.00	8.68	0.26	0.00
50.5	0.05	0.14	0.04	0.00	0.00	7.55	0.21	0.00
51.5	0.05	0.13	0.04	0.00	0.00	6.63	0.17	0.00
52.5	0.04	0.13	0.04	0.00	0.00	5.82	0.13	0.00
53.5	0.04	0.13	0.04	0.00	0.00	5.09	0.11	0.00

Table A3. The percentage contribution of HCl, CCl_4, CFC-12, CFC-11 COClF, $COCl_2$, HCFC-22, CH_3Cl & CFC-113 to the total chlorine budget in the latitude bands between 70° N and 30° N (morning occultations).

Altitude	HCl	CCl_4	CFC-12	CFC-11	COClF	$COCl_2$	HCFC-22	CH_3Cl	CFC-113
10.5	3.96	12.09	30.11	20.66	1.07	0.18	5.56	16.73	6.57
11.5	6.31	11.23	29.69	19.99	1.15	0.29	5.49	16.31	6.30
12.5	8.56	10.43	29.28	19.25	1.20	0.39	5.43	15.87	6.04
13.5	10.60	9.85	28.65	18.50	1.22	0.44	5.44	15.68	5.85
14.5	12.74	9.27	28.00	17.65	1.22	0.49	5.44	15.50	5.63
15.5	15.84	8.64	27.27	16.47	1.23	0.56	5.32	14.62	5.41
16.5	20.14	7.84	26.18	14.91	1.26	0.69	5.02	13.13	5.10
17.5	25.28	6.75	24.54	12.93	1.29	0.84	4.65	11.38	4.66
18.5	30.78	5.67	22.75	10.65	1.33	1.01	4.38	9.74	4.17
19.5	35.84	4.55	20.81	8.17	1.31	1.15	4.15	8.23	3.67
20.5	40.12	3.58	18.83	5.68	1.21	1.25	3.92	6.92	3.22
21.5	43.40	2.77	16.87	3.81	1.03	1.27	3.72	5.77	2.81
22.5	46.15	1.81	14.99	2.46	0.85	1.21	3.55	4.85	2.43
23.5	48.52	1.16	13.24	1.49	0.69	1.08	3.41	4.01	2.08
24.5	50.86	0.61	11.49	0.90	0.62	0.93	3.28	3.19	1.74
25.5	53.24	0.27	9.78	0.51	0.49	0.75	3.15	2.44	1.43
26.5	55.70	0.10	8.13	0.26	0.37	0.60	3.03	1.79	1.13
27.5	58.08	0.04	6.62	0.13	0.28	0.44	2.89	1.28	0.88
28.5	60.57	0.01	5.23	0.05	0.19	0.31	2.77	0.90	0.66
29.5	62.95	0.01	4.12	0.03	0.14	0.22	2.65	0.60	0.51
30.5	65.15	0.00	3.11	0.01	0.08	0.13	2.51	0.45	0.36
31.5	67.22	0.00	2.35	0.00	0.05	0.08	2.39	0.36	0.25
32.5	69.22	0.00	1.75	0.00	0.03	0.05	2.29	0.29	0.18
33.5	71.28	0.00	1.23	0.00	0.01	0.02	2.20	0.22	0.11
34.5	73.70	0.00	0.92	0.00	0.01	0.01	2.15	0.18	0.08
35.5	75.57	0.00	0.64	0.00	0.01	0.01	2.08	0.14	0.05
36.5	77.16	0.00	0.46	0.00	0.00	0.00	2.03	0.11	0.03
37.5	78.29	0.00	0.33	0.00	0.00	0.00	1.97	0.09	0.02
38.5	79.43	0.00	0.23	0.00	0.00	0.00	1.90	0.07	0.01
39.5	79.95	0.00	0.17	0.00	0.00	0.00	1.82	0.05	0.01
40.5	80.36	0.00	0.12	0.00	0.00	0.00	1.74	0.04	0.01
41.5	80.79	0.00	0.09	0.00	0.00	0.00	1.64	0.03	0.00
42.5	81.32	0.00	0.06	0.00	0.00	0.00	1.56	0.02	0.00
43.5	82.06	0.00	0.04	0.00	0.00	0.00	1.47	0.02	0.00
44.5	83.12	0.00	0.03	0.00	0.00	0.00	1.39	0.01	0.00
45.5	84.29	0.00	0.02	0.00	0.00	0.00	1.33	0.01	0.00
46.5	85.55	0.00	0.01	0.00	0.00	0.00	1.26	0.01	0.00
47.5	86.99	0.00	0.01	0.00	0.00	0.00	1.21	0.01	0.00
48.5	88.52	0.00	0.01	0.00	0.00	0.00	1.16	0.00	0.00
49.5	89.89	0.00	0.00	0.00	0.00	0.00	1.11	0.00	0.00
50.5	91.11	0.00	0.00	0.00	0.00	0.00	1.06	0.00	0.00
51.5	92.17	0.00	0.00	0.00	0.00	0.00	1.03	0.00	0.00
52.5	93.13	0.00	0.00	0.00	0.00	0.00	1.01	0.00	0.00
53.5	93.97	0.00	0.00	0.00	0.00	0.00	0.99	0.00	0.00

Table A4. The percentage contribution of CFC-114, CFC-115, HCFC-142b, HCFC-141b, $ClONO_2$, ClO, HOCl & H-1211 to the total chlorine budget in the latitude bands between 70° N and 30° N (morning occultations).

Altitude	CFC-114	CFC-115	HCFC-142b	HCFC-141b	$ClONO_2$	ClO	HOCl	H-1211
10.5	0.96	0.27	0.45	1.03	0.19	0.01	0.05	0.11
11.5	0.94	0.26	0.43	0.97	0.48	0.01	0.04	0.10
12.5	0.91	0.26	0.42	0.92	0.88	0.02	0.04	0.09
13.5	0.89	0.26	0.41	0.88	1.20	0.03	0.03	0.09
14.5	0.86	0.25	0.39	0.84	1.56	0.04	0.02	0.08
15.5	0.84	0.25	0.38	0.80	2.21	0.05	0.03	0.08
16.5	0.81	0.25	0.37	0.75	3.40	0.08	0.03	0.07
17.5	0.77	0.24	0.35	0.67	5.48	0.11	0.03	0.05
18.5	0.72	0.24	0.32	0.58	7.41	0.17	0.04	0.04
19.5	0.67	0.23	0.30	0.49	10.12	0.23	0.06	0.03
20.5	0.63	0.22	0.28	0.41	13.29	0.33	0.08	0.02
21.5	0.59	0.22	0.27	0.34	16.52	0.48	0.11	0.01
22.5	0.56	0.21	0.25	0.28	19.66	0.61	0.13	0.00
23.5	0.53	0.21	0.24	0.23	22.16	0.79	0.17	0.00
24.5	0.49	0.20	0.23	0.18	24.12	0.95	0.21	0.00
25.5	0.46	0.20	0.21	0.14	25.57	1.09	0.25	0.00
26.5	0.42	0.20	0.20	0.10	26.44	1.23	0.29	0.00
27.5	0.39	0.19	0.18	0.07	26.77	1.40	0.34	0.00
28.5	0.35	0.19	0.17	0.05	26.61	1.55	0.39	0.00
29.5	0.32	0.19	0.16	0.04	25.89	1.75	0.45	0.00
30.5	0.29	0.18	0.15	0.02	24.96	2.03	0.56	0.00
31.5	0.27	0.18	0.14	0.01	23.52	2.45	0.71	0.00
32.5	0.24	0.18	0.13	0.01	21.77	2.99	0.86	0.00
33.5	0.22	0.18	0.12	0.00	19.69	3.67	1.03	0.00
34.5	0.21	0.18	0.12	0.00	16.61	4.62	1.21	0.00
35.5	0.20	0.18	0.11	0.00	13.87	5.78	1.36	0.00
36.5	0.19	0.18	0.10	0.00	11.06	7.23	1.45	0.00
37.5	0.18	0.18	0.10	0.00	8.45	8.89	1.49	0.00
38.5	0.16	0.17	0.10	0.00	5.77	10.65	1.49	0.00
39.5	0.15	0.17	0.09	0.00	3.71	12.43	1.44	0.00
40.5	0.14	0.17	0.08	0.00	2.04	13.92	1.37	0.00
41.5	0.13	0.16	0.08	0.00	1.12	14.70	1.26	0.00
42.5	0.12	0.16	0.07	0.00	0.56	15.00	1.12	0.00
43.5	0.11	0.16	0.07	0.00	0.27	14.84	0.96	0.00
44.5	0.10	0.15	0.06	0.00	0.13	14.20	0.79	0.00
45.5	0.09	0.15	0.06	0.00	0.06	13.35	0.65	0.00
46.5	0.08	0.14	0.05	0.00	0.03	12.33	0.53	0.00
47.5	0.07	0.14	0.05	0.00	0.01	11.08	0.43	0.00
48.5	0.06	0.14	0.05	0.00	0.01	9.71	0.34	0.00
49.5	0.06	0.14	0.04	0.00	0.00	8.48	0.26	0.00
50.5	0.05	0.14	0.04	0.00	0.00	7.38	0.21	0.00
51.5	0.05	0.13	0.04	0.00	0.00	6.39	0.17	0.00
52.5	0.04	0.13	0.04	0.00	0.00	5.51	0.14	0.00
53.5	0.04	0.13	0.04	0.00	0.00	4.73	0.11	0.00

Table A5. The percentage contribution of HCl, CCl_4, CFC-12, CFC-11 COClF, $COCl_2$, HCFC-22, CH_3Cl & CFC-113 to the total chlorine budget in the latitude bands between 30° N and 0° N (evening occultations).

Altitude	HCl	CCl_4	CFC-12	CFC-11	COClF	$COCl_2$	HCFC-22	CH_3Cl	CFC-113
10.5	0.11	13.07	30.56	21.98	0.13	0.01	5.97	18.43	6.77
11.5	0.20	12.96	30.54	21.97	0.15	0.01	5.97	18.42	6.75
12.5	0.17	12.73	30.60	22.01	0.22	0.02	5.98	18.46	6.75
13.5	0.22	12.55	30.60	22.01	0.32	0.03	5.98	18.46	6.71
14.5	0.22	12.54	30.55	21.99	0.43	0.04	5.97	18.43	6.66
15.5	0.46	12.25	30.42	21.98	0.59	0.05	5.92	18.41	6.61
16.5	1.03	12.01	30.51	21.66	0.80	0.08	5.82	18.10	6.54
17.5	2.64	11.49	30.36	21.19	0.97	0.14	5.75	17.38	6.40
18.5	5.83	10.75	29.78	20.00	1.10	0.29	5.88	16.31	6.15
19.5	10.67	9.57	28.91	18.49	1.30	0.58	5.80	14.72	5.80
20.5	16.08	8.28	27.78	16.56	1.54	0.91	5.19	13.03	5.47
21.5	20.68	6.85	26.59	14.38	1.75	1.23	4.83	11.41	5.17
22.5	24.04	5.47	25.33	11.97	1.88	1.55	4.52	10.76	4.86
23.5	26.55	4.19	24.10	9.61	1.93	1.83	4.33	10.21	4.58
24.5	29.28	3.15	23.01	7.25	1.98	2.05	4.17	9.10	4.32
25.5	31.77	1.93	21.87	5.28	1.87	2.23	4.10	8.18	4.05
26.5	34.51	0.97	20.66	3.62	1.66	2.27	4.05	7.37	3.69
27.5	37.90	0.51	19.37	2.24	1.32	2.04	3.98	6.33	3.24
28.5	41.74	0.17	17.90	1.15	1.09	1.67	3.89	5.72	2.77
29.5	46.28	0.10	16.31	0.69	0.87	1.24	3.78	4.74	2.32
30.5	52.19	0.03	14.24	0.25	0.65	0.85	3.67	3.83	1.87
31.5	57.87	0.00	11.91	0.08	0.48	0.26	3.57	3.20	1.51
32.5	62.59	0.01	9.55	0.05	0.32	0.17	3.43	2.64	1.16
33.5	67.18	0.01	7.30	0.03	0.17	0.09	3.30	2.07	0.83
34.5	71.02	0.01	5.67	0.02	0.11	0.06	3.17	1.70	0.62
35.5	74.22	0.00	4.05	0.01	0.05	0.03	3.02	1.35	0.41
36.5	77.06	0.00	2.86	0.01	0.03	0.01	2.86	1.05	0.27
37.5	78.99	0.00	1.99	0.01	0.02	0.01	2.67	0.79	0.18
38.5	80.71	0.00	1.22	0.01	0.01	0.00	2.51	0.56	0.10
39.5	81.84	0.00	0.88	0.01	0.01	0.00	2.37	0.43	0.07
40.5	83.23	0.00	0.59	0.00	0.00	0.00	2.23	0.31	0.04
41.5	84.56	0.00	0.37	0.00	0.00	0.00	2.10	0.22	0.02
42.5	85.83	0.00	0.27	0.00	0.00	0.00	1.99	0.18	0.02
43.5	87.17	0.00	0.17	0.00	0.00	0.00	1.87	0.13	0.01
44.5	88.43	0.00	0.11	0.00	0.00	0.00	1.75	0.09	0.01
45.5	89.52	0.00	0.08	0.00	0.00	0.00	1.63	0.07	0.00
46.5	90.61	0.00	0.04	0.00	0.00	0.00	1.51	0.05	0.00
47.5	91.50	0.00	0.02	0.00	0.00	0.00	1.41	0.03	0.00
48.5	92.38	0.00	0.02	0.00	0.00	0.00	1.33	0.02	0.00
49.5	93.02	0.00	0.01	0.00	0.00	0.00	1.24	0.02	0.00
50.5	93.50	0.00	0.01	0.00	0.00	0.00	1.16	0.01	0.00
51.5	93.94	0.00	0.00	0.00	0.00	0.00	1.11	0.01	0.00
52.5	94.36	0.00	0.00	0.00	0.00	0.00	1.07	0.01	0.00
53.5	94.69	0.00	0.00	0.00	0.00	0.00	1.03	0.01	0.00

Table A6. The percentage contribution of CFC-114, CFC-115, HCFC-142b, HCFC-141b, $ClONO_2$, ClO, HOCl and H-1211 to the total chlorine budget in the latitude bands between 30° N and 0° N (evening occultations).

Altitude	CFC-114	CFC-115	HCFC-142b	HCFC-141b	$ClONO_2$	ClO	HOCl	H-1211
10.5	0.98	0.27	0.46	1.08	0.03	0.00	0.03	0.12
11.5	0.98	0.27	0.46	1.08	0.08	0.00	0.03	0.12
12.5	0.98	0.27	0.46	1.08	0.15	0.00	0.03	0.12
13.5	0.97	0.26	0.45	1.07	0.20	0.01	0.03	0.12
14.5	0.97	0.26	0.45	1.06	0.26	0.02	0.05	0.12
15.5	0.96	0.26	0.45	1.04	0.37	0.04	0.07	0.12
16.5	0.96	0.26	0.44	1.03	0.57	0.02	0.05	0.11
17.5	0.94	0.26	0.43	0.99	0.92	0.01	0.01	0.10
18.5	0.92	0.26	0.42	0.94	1.27	0.00	0.01	0.09
19.5	0.89	0.26	0.40	0.87	1.65	0.01	0.01	0.08
20.5	0.86	0.25	0.39	0.80	2.76	0.01	0.02	0.06
21.5	0.83	0.25	0.38	0.74	4.78	0.04	0.06	0.04
22.5	0.80	0.24	0.36	0.67	7.33	0.08	0.11	0.03
23.5	0.79	0.24	0.35	0.61	10.30	0.15	0.19	0.02
24.5	0.78	0.24	0.35	0.56	13.17	0.27	0.31	0.01
25.5	0.77	0.24	0.34	0.50	15.98	0.40	0.48	0.00
26.5	0.76	0.24	0.34	0.43	18.18	0.56	0.69	0.00
27.5	0.74	0.24	0.33	0.35	19.86	0.70	0.86	0.00
28.5	0.71	0.24	0.32	0.28	20.56	0.83	0.97	0.00
29.5	0.68	0.24	0.31	0.22	20.17	0.99	1.08	0.00
30.5	0.65	0.23	0.30	0.15	18.63	1.27	1.20	0.00
31.5	0.62	0.23	0.29	0.11	16.84	1.69	1.34	0.00
32.5	0.58	0.23	0.27	0.08	14.84	2.62	1.46	0.00
33.5	0.54	0.22	0.26	0.04	12.61	3.80	1.54	0.00
34.5	0.51	0.22	0.24	0.03	10.28	4.72	1.62	0.00
35.5	0.47	0.22	0.23	0.01	8.22	6.04	1.66	0.00
36.5	0.43	0.21	0.21	0.01	6.37	6.96	1.67	0.00
37.5	0.39	0.21	0.19	0.00	4.84	8.05	1.66	0.00
38.5	0.35	0.20	0.18	0.00	3.35	9.28	1.52	0.00
39.5	0.32	0.20	0.16	0.00	2.10	10.25	1.37	0.00
40.5	0.29	0.20	0.15	0.00	1.17	10.57	1.21	0.00
41.5	0.26	0.19	0.14	0.00	0.65	10.44	1.03	0.00
42.5	0.24	0.19	0.13	0.00	0.33	9.98	0.85	0.00
43.5	0.22	0.18	0.12	0.00	0.16	9.27	0.69	0.00
44.5	0.19	0.18	0.11	0.00	0.08	8.49	0.56	0.00
45.5	0.17	0.17	0.10	0.00	0.04	7.80	0.43	0.00
46.5	0.15	0.17	0.09	0.00	0.02	7.03	0.34	0.00
47.5	0.13	0.16	0.08	0.00	0.01	6.36	0.28	0.00
48.5	0.12	0.16	0.07	0.00	0.00	5.68	0.22	0.00
49.5	0.10	0.15	0.07	0.00	0.00	5.20	0.18	0.00
50.5	0.09	0.15	0.06	0.00	0.00	4.86	0.15	0.00
51.5	0.08	0.15	0.06	0.00	0.00	4.52	0.12	0.00
52.5	0.07	0.15	0.05	0.00	0.00	4.19	0.10	0.00
53.5	0.07	0.14	0.05	0.00	0.00	3.94	0.08	0.00

Title Page
Abstract | Introduction
Conclusions | References
Tables | Figures
◀| |▶
◀ ▶
Back | Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

Table A7. The percentage contribution of HCl, CCl_4, CFC-12, CFC-11 COClF, $COCl_2$, HCFC-22, CH_3Cl and CFC-113 to the total chlorine budget in the latitude bands between 30° N and 0° N (morning occultations).

Altitude	HCl	CCl_4	CFC-12	CFC-11	COClF	$COCl_2$	HCFC-22	CH_3Cl	CFC-113
10.5	0.11	13.07	30.55	21.98	0.13	0.01	5.97	18.43	6.76
11.5	0.20	12.96	30.54	21.97	0.15	0.01	5.97	18.42	6.75
12.5	0.17	12.73	30.60	22.02	0.22	0.02	5.98	18.46	6.75
13.5	0.22	12.56	30.61	22.02	0.32	0.03	5.98	18.46	6.71
14.5	0.22	12.55	30.57	22.00	0.43	0.04	5.97	18.44	6.67
15.5	0.46	12.27	30.46	22.01	0.59	0.05	5.93	18.44	6.62
16.5	1.03	12.02	30.55	21.69	0.80	0.08	5.83	18.13	6.55
17.5	2.64	11.51	30.41	21.22	0.97	0.14	5.76	17.41	6.41
18.5	5.84	10.77	29.85	20.05	1.10	0.29	5.89	16.35	6.17
19.5	10.70	9.60	28.99	18.54	1.31	0.58	5.82	14.77	5.82
20.5	16.07	8.28	27.77	16.56	1.54	0.91	5.19	13.02	5.47
21.5	20.64	6.84	26.54	14.36	1.74	1.23	4.82	11.39	5.16
22.5	23.93	5.44	25.22	11.91	1.87	1.54	4.50	10.71	4.84
23.5	26.32	4.16	23.90	9.53	1.92	1.82	4.30	10.12	4.54
24.5	28.98	3.12	22.78	7.17	1.96	2.03	4.12	9.00	4.28
25.5	31.38	1.90	21.59	5.22	1.85	2.20	4.05	8.07	4.00
26.5	33.94	0.96	20.32	3.56	1.63	2.23	3.98	7.25	3.63
27.5	37.18	0.50	19.00	2.19	1.29	2.00	3.91	6.21	3.18
28.5	40.76	0.17	17.48	1.12	1.06	1.63	3.80	5.59	2.71
29.5	45.14	0.09	15.91	0.68	0.85	1.21	3.69	4.62	2.27
30.5	50.91	0.03	13.89	0.25	0.64	0.83	3.58	3.73	1.83
31.5	56.43	0.00	11.62	0.08	0.47	0.25	3.48	3.12	1.48
32.5	61.55	0.01	9.40	0.05	0.31	0.17	3.38	2.59	1.14
33.5	66.73	0.01	7.25	0.03	0.17	0.09	3.28	2.06	0.83
34.5	71.12	0.01	5.68	0.02	0.11	0.06	3.17	1.70	0.62
35.5	75.33	0.00	4.12	0.01	0.05	0.03	3.07	1.37	0.41
36.5	79.00	0.00	2.93	0.01	0.03	0.01	2.93	1.07	0.27
37.5	81.88	0.00	2.07	0.01	0.02	0.01	2.77	0.82	0.18
38.5	84.38	0.00	1.28	0.01	0.01	0.00	2.62	0.59	0.10
39.5	85.90	0.00	0.92	0.01	0.01	0.00	2.48	0.45	0.07
40.5	87.07	0.00	0.62	0.00	0.00	0.00	2.33	0.33	0.04
41.5	87.55	0.00	0.38	0.00	0.00	0.00	2.18	0.23	0.02
42.5	87.99	0.00	0.28	0.00	0.00	0.00	2.04	0.18	0.02
43.5	88.67	0.00	0.18	0.00	0.00	0.00	1.90	0.13	0.01
44.5	89.46	0.00	0.12	0.00	0.00	0.00	1.77	0.10	0.01
45.5	90.09	0.00	0.08	0.00	0.00	0.00	1.64	0.07	0.00
46.5	90.77	0.00	0.04	0.00	0.00	0.00	1.51	0.05	0.00
47.5	91.56	0.00	0.02	0.00	0.00	0.00	1.42	0.03	0.00
48.5	92.29	0.00	0.02	0.00	0.00	0.00	1.33	0.02	0.00
49.5	92.91	0.00	0.01	0.00	0.00	0.00	1.24	0.02	0.00
50.5	93.54	0.00	0.01	0.00	0.00	0.00	1.16	0.01	0.00
51.5	94.12	0.00	0.00	0.00	0.00	0.00	1.12	0.01	0.00
52.5	94.73	0.00	0.00	0.00	0.00	0.00	1.08	0.01	0.00
53.5	95.20	0.00	0.00	0.00	0.00	0.00	1.04	0.01	0.00

Table A8. The percentage contribution of CFC-114, CFC-115, HCFC-142b, HCFC-141b, $ClONO_2$, ClO, HOCl and H-1211 to the total chlorine budget in the latitude bands between 30° N and 0° N (morning occultations).

Altitude	CFC-114	CFC-115	HCFC-142b	HCFC-141b	$ClONO_2$	ClO	HOCl	H-1211
10.5	0.98	0.27	0.46	1.08	0.03	0.01	0.04	0.12
11.5	0.98	0.27	0.46	1.08	0.06	0.01	0.04	0.12
12.5	0.98	0.27	0.46	1.08	0.12	0.01	0.04	0.12
13.5	0.97	0.26	0.45	1.07	0.16	0.01	0.04	0.12
14.5	0.97	0.26	0.45	1.06	0.21	0.02	0.04	0.12
15.5	0.96	0.26	0.45	1.04	0.30	0.02	0.03	0.12
16.5	0.96	0.26	0.44	1.03	0.46	0.04	0.02	0.11
17.5	0.94	0.26	0.44	0.99	0.74	0.03	0.01	0.10
18.5	0.92	0.26	0.42	0.94	1.02	0.03	0.01	0.09
19.5	0.89	0.26	0.41	0.87	1.33	0.03	0.01	0.08
20.5	0.86	0.25	0.39	0.80	2.73	0.08	0.03	0.06
21.5	0.83	0.25	0.37	0.73	4.85	0.15	0.05	0.04
22.5	0.80	0.24	0.36	0.67	7.57	0.27	0.09	0.03
23.5	0.78	0.24	0.35	0.61	10.81	0.44	0.16	0.02
24.5	0.77	0.24	0.34	0.55	13.75	0.62	0.28	0.01
25.5	0.76	0.24	0.34	0.49	16.63	0.85	0.42	0.00
26.5	0.74	0.24	0.33	0.42	19.07	1.09	0.58	0.00
27.5	0.72	0.24	0.32	0.35	20.91	1.29	0.72	0.00
28.5	0.69	0.23	0.31	0.27	21.90	1.46	0.81	0.00
29.5	0.66	0.23	0.30	0.21	21.68	1.57	0.89	0.00
30.5	0.63	0.23	0.29	0.15	20.32	1.72	0.98	0.00
31.5	0.60	0.23	0.28	0.11	18.98	1.81	1.07	0.00
32.5	0.57	0.22	0.27	0.08	17.14	1.97	1.16	0.00
33.5	0.54	0.22	0.25	0.04	15.16	2.13	1.21	0.00
34.5	0.51	0.22	0.24	0.03	13.03	2.22	1.26	0.00
35.5	0.48	0.22	0.23	0.02	10.80	2.58	1.29	0.00
36.5	0.44	0.22	0.22	0.01	8.46	3.11	1.29	0.00
37.5	0.40	0.22	0.20	0.00	6.50	3.66	1.26	0.00
38.5	0.37	0.21	0.19	0.00	4.53	4.52	1.19	0.00
39.5	0.33	0.21	0.17	0.00	2.86	5.46	1.12	0.00
40.5	0.30	0.20	0.16	0.00	1.58	6.35	1.00	0.00
41.5	0.27	0.20	0.14	0.00	0.87	7.28	0.87	0.00
42.5	0.24	0.19	0.13	0.00	0.43	7.75	0.74	0.00
43.5	0.22	0.19	0.12	0.00	0.21	7.74	0.63	0.00
44.5	0.20	0.18	0.11	0.00	0.10	7.43	0.53	0.00
45.5	0.17	0.17	0.10	0.00	0.05	7.21	0.42	0.00
46.5	0.15	0.17	0.09	0.00	0.02	6.85	0.34	0.00
47.5	0.13	0.16	0.08	0.00	0.01	6.30	0.29	0.00
48.5	0.12	0.16	0.07	0.00	0.01	5.75	0.23	0.00
49.5	0.10	0.15	0.07	0.00	0.00	5.30	0.19	0.00
50.5	0.09	0.15	0.06	0.00	0.00	4.82	0.15	0.00
51.5	0.08	0.15	0.06	0.00	0.00	4.34	0.13	0.00
52.5	0.08	0.15	0.05	0.00	0.00	3.81	0.10	0.00
53.5	0.07	0.14	0.05	0.00	0.00	3.41	0.08	0.00

Table A9. The percentage contribution of HCl, CCl_4, CFC-12, CFC-11 COClF, $COCl_2$, HCFC-22, CH_3Cl and CFC-113 to the total chlorine budget in the latitude bands between 0° N and 30° S (evening occultations).

Altitude	HCl	CCl_4	CFC-12	CFC-11	COClF	$COCl_2$	HCFC-22	CH_3Cl	CFC-113
10.5	0.11	12.70	30.75	22.12	0.21	0.01	5.90	18.42	6.80
11.5	0.20	12.61	30.72	22.10	0.24	0.01	5.89	18.40	6.79
12.5	0.18	12.47	30.74	22.11	0.29	0.02	5.90	18.41	6.78
13.5	0.21	12.37	30.72	22.10	0.38	0.02	5.89	18.40	6.76
14.5	0.24	12.32	30.68	22.07	0.50	0.03	5.88	18.38	6.72
15.5	0.48	12.11	30.54	22.07	0.63	0.04	5.84	18.36	6.65
16.5	1.14	11.90	30.54	21.75	0.87	0.07	5.80	17.89	6.57
17.5	2.79	11.45	30.34	21.24	0.99	0.14	5.73	17.16	6.38
18.5	6.57	10.56	29.67	20.01	1.10	0.32	5.79	15.91	6.04
19.5	11.77	9.34	28.65	18.29	1.26	0.62	5.67	14.45	5.64
20.5	17.58	8.00	27.37	16.30	1.52	0.90	5.07	12.54	5.29
21.5	22.01	6.65	26.03	14.08	1.74	1.26	4.73	10.84	5.02
22.5	25.18	5.09	24.78	11.74	1.85	1.51	4.43	10.15	4.74
23.5	27.95	3.87	23.22	9.33	1.84	1.73	4.24	9.57	4.41
24.5	30.83	2.78	22.08	6.95	1.81	1.90	4.02	8.45	4.13
25.5	33.92	1.62	20.49	4.95	1.65	2.01	3.95	7.25	3.80
26.5	37.13	0.81	18.88	3.24	1.45	1.98	3.86	6.32	3.38
27.5	40.32	0.43	17.45	2.06	0.99	1.76	3.84	5.59	2.99
28.5	44.06	0.16	15.92	1.11	0.84	1.44	3.77	5.01	2.58
29.5	48.40	0.09	14.26	0.67	0.68	1.13	3.70	4.41	2.20
30.5	53.83	0.02	12.24	0.25	0.53	0.76	3.64	3.58	1.82
31.5	58.91	0.01	10.19	0.09	0.38	0.44	3.54	2.92	1.47
32.5	64.07	0.01	8.13	0.05	0.25	0.29	3.42	2.40	1.13
33.5	68.16	0.01	6.09	0.01	0.13	0.16	3.26	1.93	0.80
34.5	71.58	0.01	4.74	0.01	0.09	0.10	3.12	1.59	0.60
35.5	73.85	0.00	3.43	0.01	0.05	0.06	2.95	1.26	0.41
36.5	75.82	0.00	2.47	0.00	0.02	0.03	2.78	0.99	0.28
37.5	77.48	0.00	1.81	0.00	0.02	0.02	2.63	0.78	0.19
38.5	79.07	0.00	1.22	0.00	0.01	0.01	2.49	0.59	0.12
39.5	80.49	0.00	0.88	0.00	0.00	0.01	2.34	0.45	0.08
40.5	81.88	0.00	0.57	0.00	0.00	0.00	2.18	0.32	0.05
41.5	83.14	0.00	0.35	0.00	0.00	0.00	2.02	0.22	0.03
42.5	84.47	0.00	0.24	0.00	0.00	0.00	1.87	0.16	0.02
43.5	85.83	0.00	0.13	0.00	0.00	0.00	1.72	0.10	0.01
44.5	87.16	0.00	0.08	0.00	0.00	0.00	1.59	0.07	0.01
45.5	88.31	0.00	0.05	0.00	0.00	0.00	1.48	0.05	0.00
46.5	89.39	0.00	0.03	0.00	0.00	0.00	1.37	0.03	0.00
47.5	90.45	0.00	0.01	0.00	0.00	0.00	1.29	0.02	0.00
48.5	91.37	0.00	0.01	0.00	0.00	0.00	1.24	0.02	0.00
49.5	92.26	0.00	0.01	0.00	0.00	0.00	1.18	0.01	0.00
50.5	92.95	0.00	0.00	0.00	0.00	0.00	1.13	0.01	0.00
51.5	93.55	0.00	0.00	0.00	0.00	0.00	1.10	0.01	0.00
52.5	94.19	0.00	0.00	0.00	0.00	0.00	1.08	0.01	0.00
53.5	94.66	0.00	0.00	0.00	0.00	0.00	1.06	0.00	0.00

Table A10. The percentage contribution of CFC-114, CFC-115, HCFC-142b, HCFC-141b, $ClONO_2$, ClO, HOCl and H-1211 to the total chlorine budget in the latitude bands between 0° N and 30° S (evening occultations).

Altitude	CFC-114	CFC-115	HCFC-142b	HCFC-141b	$ClONO_2$	ClO	HOCl	H-1211
10.5	0.98	0.27	0.46	1.09	0.04	0.00	0.02	0.12
11.5	0.98	0.27	0.46	1.09	0.09	0.00	0.02	0.12
12.5	0.98	0.27	0.46	1.08	0.17	0.00	0.02	0.12
13.5	0.98	0.27	0.46	1.08	0.23	0.00	0.02	0.12
14.5	0.97	0.27	0.45	1.07	0.29	0.00	0.01	0.12
15.5	0.97	0.26	0.45	1.06	0.42	0.00	0.01	0.12
16.5	0.96	0.26	0.45	1.03	0.64	0.00	0.01	0.11
17.5	0.94	0.26	0.43	0.99	1.04	0.00	0.01	0.10
18.5	0.90	0.26	0.41	0.92	1.43	0.01	0.01	0.09
19.5	0.86	0.25	0.39	0.85	1.86	0.01	0.01	0.07
20.5	0.82	0.25	0.37	0.78	3.10	0.02	0.03	0.06
21.5	0.80	0.24	0.36	0.72	5.35	0.06	0.06	0.04
22.5	0.78	0.24	0.35	0.66	8.23	0.12	0.12	0.03
23.5	0.75	0.24	0.34	0.60	11.45	0.23	0.22	0.01
24.5	0.74	0.24	0.33	0.54	14.44	0.40	0.35	0.01
25.5	0.73	0.24	0.33	0.47	17.47	0.62	0.51	0.00
26.5	0.71	0.24	0.32	0.39	19.80	0.80	0.68	0.00
27.5	0.70	0.24	0.32	0.33	21.20	0.97	0.83	0.00
28.5	0.68	0.24	0.31	0.26	21.56	1.16	0.89	0.00
29.5	0.66	0.23	0.30	0.21	20.70	1.34	1.02	0.00
30.5	0.64	0.23	0.29	0.15	19.06	1.79	1.16	0.00
31.5	0.61	0.23	0.28	0.11	16.94	2.58	1.30	0.00
32.5	0.57	0.23	0.27	0.08	14.52	3.13	1.46	0.00
33.5	0.53	0.23	0.25	0.04	12.11	4.66	1.62	0.00
34.5	0.50	0.22	0.24	0.03	9.12	6.26	1.80	0.00
35.5	0.46	0.22	0.22	0.02	7.40	7.80	1.88	0.00
36.5	0.42	0.21	0.21	0.01	5.68	9.20	1.88	0.00
37.5	0.38	0.21	0.19	0.01	4.31	10.12	1.85	0.00
38.5	0.35	0.20	0.18	0.00	2.98	11.02	1.76	0.00
39.5	0.31	0.20	0.16	0.00	1.87	11.62	1.57	0.00
40.5	0.28	0.19	0.15	0.00	1.04	11.92	1.40	0.00
41.5	0.25	0.19	0.13	0.00	0.58	11.89	1.19	0.00
42.5	0.22	0.18	0.12	0.00	0.29	11.43	1.00	0.00
43.5	0.19	0.18	0.11	0.00	0.14	10.78	0.82	0.00
44.5	0.17	0.17	0.10	0.00	0.07	9.93	0.67	0.00
45.5	0.15	0.17	0.09	0.00	0.03	9.15	0.52	0.00
46.5	0.13	0.16	0.08	0.00	0.01	8.39	0.41	0.00
47.5	0.11	0.16	0.07	0.00	0.01	7.55	0.33	0.00
48.5	0.10	0.15	0.07	0.00	0.00	6.78	0.26	0.00
49.5	0.09	0.15	0.06	0.00	0.00	6.03	0.20	0.00
50.5	0.08	0.15	0.06	0.00	0.00	5.44	0.17	0.00
51.5	0.08	0.15	0.06	0.00	0.00	4.91	0.14	0.00
52.5	0.07	0.15	0.05	0.00	0.00	4.35	0.10	0.00
53.5	0.07	0.14	0.05	0.00	0.00	3.93	0.08	0.00

Table A11. The percentage contribution of HCl, CCl_4, CFC-12, CFC-11 COClF, $COCl_2$, HCFC-22, CH_3Cl and CFC-113 to the total chlorine budget in the latitude bands between 0° N and 30° S (morning occultations).

Altitude	HCl	CCl_4	CFC-12	CFC-11	COClF	$COCl_2$	HCFC-22	CH_3Cl	CFC-113
10.5	0.11	12.70	30.74	22.11	0.21	0.01	5.89	18.41	6.80
11.5	0.20	12.61	30.71	22.09	0.24	0.01	5.89	18.39	6.79
12.5	0.18	12.46	30.72	22.10	0.29	0.02	5.89	18.40	6.78
13.5	0.21	12.36	30.70	22.08	0.38	0.02	5.89	18.39	6.75
14.5	0.24	12.30	30.65	22.04	0.50	0.03	5.88	18.36	6.72
15.5	0.48	12.10	30.49	22.04	0.63	0.04	5.83	18.33	6.65
16.5	1.14	11.88	30.47	21.70	0.87	0.07	5.78	17.86	6.56
17.5	2.78	11.41	30.23	21.17	0.99	0.14	5.71	17.10	6.35
18.5	6.54	10.51	29.53	19.91	1.09	0.32	5.76	15.84	6.01
19.5	11.69	9.28	28.47	18.17	1.25	0.61	5.63	14.36	5.60
20.5	17.41	7.92	27.10	16.14	1.50	0.89	5.02	12.42	5.24
21.5	21.74	6.56	25.71	13.91	1.72	1.24	4.67	10.70	4.96
22.5	24.86	5.03	24.46	11.59	1.83	1.49	4.38	10.02	4.68
23.5	27.64	3.83	22.96	9.23	1.82	1.71	4.19	9.46	4.37
24.5	30.44	2.74	21.80	6.86	1.78	1.87	3.97	8.34	4.08
25.5	33.49	1.60	20.24	4.88	1.63	1.98	3.90	7.16	3.75
26.5	36.56	0.80	18.59	3.19	1.43	1.95	3.80	6.22	3.32
27.5	39.58	0.43	17.12	2.03	0.97	1.72	3.76	5.49	2.93
28.5	43.13	0.16	15.58	1.08	0.82	1.41	3.69	4.91	2.53
29.5	47.11	0.09	13.88	0.66	0.66	1.10	3.60	4.29	2.14
30.5	52.26	0.02	11.88	0.24	0.51	0.74	3.53	3.48	1.77
31.5	57.16	0.00	9.89	0.08	0.37	0.42	3.44	2.84	1.42
32.5	61.96	0.01	7.86	0.04	0.25	0.28	3.30	2.32	1.09
33.5	66.65	0.01	5.96	0.01	0.13	0.15	3.19	1.88	0.78
34.5	70.57	0.01	4.67	0.01	0.09	0.10	3.07	1.57	0.59
35.5	74.27	0.00	3.45	0.01	0.05	0.06	2.97	1.27	0.41
36.5	77.43	0.00	2.52	0.00	0.03	0.03	2.84	1.01	0.28
37.5	79.89	0.00	1.87	0.00	0.02	0.02	2.71	0.81	0.20
38.5	81.98	0.00	1.27	0.00	0.01	0.01	2.58	0.61	0.12
39.5	83.67	0.00	0.92	0.00	0.01	0.01	2.43	0.47	0.09
40.5	85.04	0.00	0.59	0.00	0.00	0.00	2.26	0.33	0.05
41.5	86.13	0.00	0.37	0.00	0.00	0.00	2.09	0.23	0.03
42.5	86.82	0.00	0.25	0.00	0.00	0.00	1.92	0.17	0.02
43.5	87.67	0.00	0.13	0.00	0.00	0.00	1.75	0.11	0.01
44.5	88.47	0.00	0.08	0.00	0.00	0.00	1.62	0.07	0.01
45.5	89.20	0.00	0.05	0.00	0.00	0.00	1.49	0.05	0.00
46.5	89.92	0.00	0.03	0.00	0.00	0.00	1.38	0.03	0.00
47.5	90.69	0.00	0.01	0.00	0.00	0.00	1.29	0.02	0.00
48.5	91.35	0.00	0.01	0.00	0.00	0.00	1.23	0.02	0.00
49.5	92.07	0.00	0.01	0.00	0.00	0.00	1.18	0.01	0.00
50.5	92.81	0.00	0.00	0.00	0.00	0.00	1.13	0.01	0.00
51.5	93.49	0.00	0.00	0.00	0.00	0.00	1.10	0.01	0.00
52.5	94.18	0.00	0.00	0.00	0.00	0.00	1.08	0.01	0.00
53.5	94.77	0.00	0.00	0.00	0.00	0.00	1.06	0.00	0.00

Table A12. The percentage contribution of CFC-114, CFC-115, HCFC-142b, HCFC-141b, $ClONO_2$, ClO, HOCl and H-1211 to the total chlorine budget in the latitude bands between 0° N and 30° S (morning occultations).

Altitude	CFC-114	CFC-115	HCFC-142b	HCFC-141b	$ClONO_2$	ClO	HOCl	H-1211
10.5	0.98	0.27	0.46	1.09	0.05	0.01	0.04	0.12
11.5	0.98	0.27	0.46	1.09	0.12	0.01	0.04	0.12
12.5	0.98	0.27	0.46	1.08	0.22	0.01	0.03	0.12
13.5	0.98	0.27	0.46	1.08	0.30	0.01	0.03	0.12
14.5	0.97	0.26	0.45	1.07	0.39	0.01	0.02	0.12
15.5	0.96	0.26	0.45	1.05	0.55	0.01	0.01	0.12
16.5	0.96	0.26	0.44	1.03	0.85	0.01	0.01	0.11
17.5	0.93	0.26	0.43	0.99	1.37	0.02	0.01	0.10
18.5	0.90	0.26	0.41	0.92	1.88	0.03	0.01	0.09
19.5	0.85	0.25	0.39	0.84	2.45	0.05	0.01	0.07
20.5	0.82	0.25	0.37	0.77	3.98	0.09	0.03	0.06
21.5	0.79	0.24	0.36	0.71	6.41	0.18	0.05	0.04
22.5	0.77	0.24	0.35	0.65	9.26	0.28	0.09	0.03
23.5	0.75	0.23	0.34	0.59	12.33	0.39	0.16	0.01
24.5	0.73	0.23	0.33	0.53	15.47	0.55	0.27	0.01
25.5	0.72	0.23	0.32	0.47	18.41	0.80	0.41	0.00
26.5	0.70	0.23	0.32	0.39	20.87	1.06	0.57	0.00
27.5	0.69	0.23	0.31	0.32	22.51	1.22	0.69	0.00
28.5	0.67	0.23	0.30	0.26	23.11	1.37	0.76	0.00
29.5	0.64	0.23	0.29	0.20	22.77	1.51	0.83	0.00
30.5	0.62	0.23	0.28	0.15	21.68	1.68	0.93	0.00
31.5	0.59	0.23	0.27	0.11	20.28	1.83	1.06	0.00
32.5	0.56	0.22	0.26	0.07	18.62	1.98	1.17	0.00
33.5	0.52	0.22	0.25	0.04	16.71	2.21	1.28	0.00
34.5	0.49	0.22	0.23	0.03	14.51	2.46	1.39	0.00
35.5	0.46	0.22	0.22	0.02	12.25	2.90	1.46	0.00
36.5	0.43	0.21	0.21	0.01	9.82	3.68	1.49	0.00
37.5	0.39	0.21	0.20	0.01	7.75	4.46	1.47	0.00
38.5	0.36	0.21	0.18	0.00	5.65	5.62	1.38	0.00
39.5	0.33	0.21	0.17	0.00	3.81	6.64	1.27	0.00
40.5	0.29	0.20	0.15	0.00	2.39	7.54	1.13	0.00
41.5	0.26	0.19	0.14	0.00	1.45	8.14	0.97	0.00
42.5	0.22	0.19	0.12	0.00	0.81	8.65	0.83	0.00
43.5	0.19	0.18	0.11	0.00	0.41	8.72	0.70	0.00
44.5	0.17	0.17	0.10	0.00	0.22	8.51	0.59	0.00
45.5	0.15	0.17	0.09	0.00	0.10	8.20	0.48	0.00
46.5	0.13	0.16	0.08	0.00	0.05	7.83	0.40	0.00
47.5	0.11	0.16	0.07	0.00	0.03	7.28	0.33	0.00
48.5	0.10	0.15	0.07	0.00	0.01	6.80	0.26	0.00
49.5	0.09	0.15	0.06	0.00	0.01	6.21	0.22	0.00
50.5	0.08	0.15	0.06	0.00	0.00	5.57	0.18	0.00
51.5	0.08	0.15	0.06	0.00	0.00	4.97	0.15	0.00
52.5	0.07	0.15	0.05	0.00	0.00	4.34	0.12	0.00
53.5	0.07	0.14	0.05	0.00	0.00	3.81	0.09	0.00

Table A13. The percentage contribution of HCl, CCl_4, CFC-12, CFC-11 COClF, $COCl_2$, HCFC-22, CH_3Cl and CFC-113 to the total chlorine budget in the latitude bands between 30° S and 70° S (evening occultations).

Altitude	HCl	CCl_4	CFC-12	CFC-11	COClF	$COCl_2$	HCFC-22	CH_3Cl	CFC-113
10.5	5.53	11.60	29.44	20.51	1.06	0.19	5.35	16.63	6.54
11.5	7.51	11.07	29.01	20.02	1.13	0.32	5.35	15.93	6.23
12.5	9.01	10.49	28.71	19.61	1.19	0.46	5.39	15.34	5.92
13.5	10.76	9.99	28.18	19.03	1.22	0.55	5.40	14.86	5.67
14.5	13.35	9.36	27.26	18.04	1.23	0.64	5.35	14.15	5.42
15.5	17.44	8.51	26.04	16.47	1.25	0.74	5.17	12.69	5.16
16.5	23.46	7.41	24.40	14.32	1.28	0.87	4.76	10.85	4.81
17.5	30.15	6.14	22.34	11.81	1.28	0.99	4.32	8.95	4.36
18.5	36.37	4.85	20.19	9.17	1.27	1.12	4.02	7.34	3.84
19.5	41.39	3.73	18.13	6.61	1.17	1.19	3.80	6.01	3.34
20.5	45.38	2.82	16.17	4.27	0.97	1.21	3.61	4.96	2.93
21.5	48.49	2.10	14.31	2.59	0.75	1.15	3.46	4.07	2.58
22.5	51.08	1.37	12.57	1.74	0.56	0.98	3.34	3.23	2.25
23.5	53.52	0.79	10.95	1.15	0.42	0.79	3.23	2.53	1.91
24.5	55.75	0.39	9.42	0.68	0.37	0.66	3.14	1.96	1.59
25.5	58.01	0.16	8.03	0.36	0.30	0.53	3.06	1.53	1.29
26.5	60.30	0.07	6.79	0.18	0.24	0.43	2.98	1.20	1.03
27.5	62.82	0.03	5.56	0.10	0.18	0.33	2.89	0.96	0.82
28.5	65.42	0.01	4.46	0.04	0.13	0.23	2.79	0.81	0.63
29.5	67.93	0.00	3.59	0.02	0.09	0.17	2.69	0.65	0.48
30.5	70.17	0.00	2.80	0.01	0.06	0.11	2.60	0.54	0.36
31.5	72.11	0.00	2.13	0.00	0.03	0.06	2.51	0.44	0.25
32.5	73.59	0.00	1.63	0.00	0.02	0.04	2.42	0.36	0.18
33.5	74.83	0.00	1.17	0.00	0.01	0.02	2.32	0.28	0.12
34.5	76.57	0.00	0.85	0.00	0.01	0.01	2.24	0.23	0.08
35.5	77.41	0.00	0.61	0.00	0.00	0.01	2.14	0.18	0.06
36.5	77.75	0.00	0.42	0.00	0.00	0.00	2.03	0.14	0.04
37.5	77.96	0.00	0.31	0.00	0.00	0.00	1.92	0.11	0.03
38.5	78.47	0.00	0.21	0.00	0.00	0.00	1.83	0.09	0.02
39.5	78.85	0.00	0.15	0.00	0.00	0.00	1.73	0.06	0.01
40.5	79.44	0.00	0.10	0.00	0.00	0.00	1.63	0.05	0.01
41.5	80.26	0.00	0.07	0.00	0.00	0.00	1.54	0.03	0.00
42.5	81.27	0.00	0.04	0.00	0.00	0.00	1.46	0.02	0.00
43.5	82.51	0.00	0.03	0.00	0.00	0.00	1.38	0.02	0.00
44.5	84.01	0.00	0.02	0.00	0.00	0.00	1.31	0.01	0.00
45.5	85.58	0.00	0.01	0.00	0.00	0.00	1.25	0.01	0.00
46.5	87.22	0.00	0.01	0.00	0.00	0.00	1.20	0.01	0.00
47.5	88.79	0.00	0.00	0.00	0.00	0.00	1.15	0.00	0.00
48.5	90.20	0.00	0.00	0.00	0.00	0.00	1.12	0.00	0.00
49.5	91.40	0.00	0.00	0.00	0.00	0.00	1.10	0.00	0.00
50.5	92.42	0.00	0.00	0.00	0.00	0.00	1.08	0.00	0.00
51.5	93.23	0.00	0.00	0.00	0.00	0.00	1.06	0.00	0.00
52.5	93.86	0.00	0.00	0.00	0.00	0.00	1.04	0.00	0.00
53.5	94.37	0.00	0.00	0.00	0.00	0.00	1.03	0.00	0.00

Table A14. The percentage contribution of CFC-114, CFC-115, HCFC-142b, HCFC-141b, $ClONO_2$, ClO, HOCl and H-1211 to the total chlorine budget in the latitude bands between 30° S and 70° S (evening occultations).

Altitude	CFC-114	CFC-115	HCFC-142b	HCFC-141b	$ClONO_2$	ClO	HOCl	H-1211
10.5	0.96	0.27	0.44	1.03	0.27	0.02	0.05	0.11
11.5	0.92	0.26	0.43	0.97	0.66	0.03	0.05	0.10
12.5	0.89	0.26	0.41	0.91	1.22	0.04	0.06	0.09
13.5	0.86	0.26	0.40	0.86	1.76	0.06	0.06	0.09
14.5	0.84	0.25	0.38	0.82	2.68	0.09	0.07	0.08
15.5	0.81	0.25	0.37	0.77	4.03	0.13	0.09	0.07
16.5	0.77	0.25	0.35	0.71	5.46	0.15	0.11	0.06
17.5	0.72	0.24	0.33	0.63	7.34	0.22	0.13	0.05
18.5	0.67	0.23	0.30	0.54	9.58	0.32	0.16	0.04
19.5	0.61	0.22	0.27	0.45	12.44	0.43	0.18	0.02
20.5	0.57	0.22	0.26	0.38	15.49	0.55	0.19	0.01
21.5	0.54	0.21	0.24	0.32	18.36	0.60	0.22	0.01
22.5	0.51	0.21	0.23	0.27	20.81	0.63	0.23	0.00
23.5	0.49	0.21	0.22	0.22	22.74	0.60	0.24	0.00
24.5	0.46	0.20	0.21	0.17	24.09	0.69	0.23	0.00
25.5	0.44	0.20	0.20	0.12	24.74	0.78	0.24	0.00
26.5	0.42	0.20	0.20	0.09	24.69	0.93	0.25	0.00
27.5	0.40	0.20	0.19	0.07	24.07	1.13	0.28	0.00
28.5	0.38	0.20	0.18	0.05	22.91	1.46	0.32	0.00
29.5	0.35	0.19	0.17	0.03	21.40	1.84	0.38	0.00
30.5	0.33	0.19	0.16	0.02	19.70	2.47	0.50	0.00
31.5	0.30	0.19	0.15	0.01	17.55	3.56	0.69	0.00
32.5	0.28	0.19	0.15	0.01	15.08	5.12	0.92	0.00
33.5	0.26	0.19	0.14	0.00	12.37	7.12	1.16	0.00
34.5	0.24	0.19	0.13	0.00	9.10	8.99	1.37	0.00
35.5	0.22	0.18	0.12	0.00	6.44	11.11	1.51	0.00
36.5	0.20	0.18	0.11	0.00	4.27	13.23	1.62	0.00
37.5	0.19	0.17	0.11	0.00	2.78	14.76	1.66	0.00
38.5	0.17	0.17	0.10	0.00	1.54	15.79	1.62	0.00
39.5	0.16	0.17	0.09	0.00	0.89	16.38	1.51	0.00
40.5	0.14	0.16	0.08	0.00	0.49	16.51	1.37	0.00
41.5	0.13	0.16	0.08	0.00	0.27	16.25	1.20	0.00
42.5	0.12	0.16	0.07	0.00	0.14	15.69	1.02	0.00
43.5	0.10	0.15	0.07	0.00	0.07	14.81	0.86	0.00
44.5	0.09	0.15	0.06	0.00	0.03	13.61	0.69	0.00
45.5	0.09	0.15	0.06	0.00	0.02	12.29	0.55	0.00
46.5	0.08	0.15	0.05	0.00	0.01	10.85	0.43	0.00
47.5	0.07	0.14	0.05	0.00	0.00	9.45	0.33	0.00
48.5	0.06	0.14	0.05	0.00	0.00	8.16	0.25	0.00
49.5	0.06	0.14	0.05	0.00	0.00	7.07	0.18	0.00
50.5	0.06	0.14	0.04	0.00	0.00	6.12	0.14	0.00
51.5	0.05	0.14	0.04	0.00	0.00	5.36	0.11	0.00
52.5	0.05	0.14	0.04	0.00	0.00	4.78	0.09	0.00
53.5	0.05	0.14	0.04	0.00	0.00	4.30	0.07	0.00

Table A15. The percentage contribution of HCl, CCl_4, CFC-12, CFC-11 COClF, $COCl_2$, HCFC-22, CH_3Cl and CFC-113 to the total chlorine budget in the latitude bands between 30° S and 70° S (morning occultations).

Altitude	HCl	CCl_4	CFC-12	CFC-11	COClF	$COCl_2$	HCFC-22	CH_3Cl	CFC-113
10.5	5.54	11.61	29.46	20.53	1.06	0.19	5.36	16.64	6.54
11.5	7.53	11.09	29.06	20.05	1.13	0.32	5.36	15.95	6.24
12.5	9.04	10.52	28.80	19.67	1.19	0.46	5.40	15.39	5.94
13.5	10.82	10.04	28.31	19.12	1.23	0.55	5.42	14.93	5.69
14.5	13.47	9.44	27.51	18.21	1.24	0.65	5.40	14.28	5.47
15.5	17.71	8.64	26.44	16.72	1.27	0.75	5.25	12.89	5.24
16.5	23.86	7.53	24.81	14.57	1.30	0.89	4.84	11.03	4.89
17.5	30.65	6.24	22.70	12.00	1.30	1.01	4.39	9.10	4.43
18.5	36.89	4.92	20.49	9.30	1.29	1.13	4.08	7.45	3.90
19.5	41.88	3.77	18.35	6.69	1.18	1.20	3.85	6.08	3.38
20.5	45.53	2.83	16.23	4.28	0.97	1.22	3.62	4.98	2.93
21.5	48.09	2.08	14.19	2.57	0.75	1.14	3.43	4.03	2.56
22.5	49.83	1.33	12.26	1.70	0.54	0.96	3.26	3.15	2.19
23.5	51.32	0.75	10.50	1.10	0.41	0.76	3.10	2.43	1.84
24.5	53.22	0.37	8.99	0.65	0.35	0.63	2.99	1.87	1.52
25.5	55.21	0.15	7.64	0.34	0.29	0.50	2.91	1.45	1.22
26.5	57.40	0.06	6.46	0.18	0.23	0.41	2.84	1.14	0.98
27.5	60.08	0.03	5.32	0.09	0.17	0.31	2.76	0.92	0.78
28.5	62.89	0.01	4.29	0.04	0.12	0.22	2.68	0.78	0.60
29.5	65.41	0.00	3.46	0.02	0.09	0.16	2.59	0.63	0.47
30.5	67.61	0.00	2.70	0.01	0.06	0.10	2.50	0.52	0.34
31.5	69.45	0.00	2.05	0.00	0.03	0.06	2.42	0.42	0.24
32.5	71.01	0.00	1.57	0.00	0.02	0.04	2.34	0.35	0.17
33.5	72.37	0.00	1.14	0.00	0.01	0.02	2.24	0.27	0.11
34.5	73.77	0.00	0.82	0.00	0.01	0.01	2.16	0.22	0.08
35.5	75.09	0.00	0.59	0.00	0.00	0.01	2.07	0.18	0.05
36.5	76.32	0.00	0.42	0.00	0.00	0.00	1.99	0.14	0.04
37.5	77.58	0.00	0.31	0.00	0.00	0.00	1.91	0.11	0.02
38.5	78.84	0.00	0.21	0.00	0.00	0.00	1.84	0.09	0.02
39.5	80.02	0.00	0.15	0.00	0.00	0.00	1.75	0.06	0.01
40.5	81.11	0.00	0.11	0.00	0.00	0.00	1.67	0.05	0.01
41.5	82.20	0.00	0.07	0.00	0.00	0.00	1.58	0.03	0.00
42.5	83.29	0.00	0.05	0.00	0.00	0.00	1.50	0.02	0.00
43.5	84.47	0.00	0.03	0.00	0.00	0.00	1.42	0.02	0.00
44.5	85.83	0.00	0.02	0.00	0.00	0.00	1.34	0.01	0.00
45.5	87.20	0.00	0.01	0.00	0.00	0.00	1.28	0.01	0.00
46.5	88.57	0.00	0.01	0.00	0.00	0.00	1.22	0.01	0.00
47.5	89.89	0.00	0.00	0.00	0.00	0.00	1.16	0.00	0.00
48.5	91.07	0.00	0.00	0.00	0.00	0.00	1.13	0.00	0.00
49.5	92.06	0.00	0.00	0.00	0.00	0.00	1.11	0.00	0.00
50.5	92.93	0.00	0.00	0.00	0.00	0.00	1.08	0.00	0.00
51.5	93.69	0.00	0.00	0.00	0.00	0.00	1.06	0.00	0.00
52.5	94.41	0.00	0.00	0.00	0.00	0.00	1.05	0.00	0.00
53.5	95.03	0.00	0.00	0.00	0.00	0.00	1.03	0.00	0.00

Table A16. The percentage contribution of CFC-114, CFC-115, HCFC-142b, HCFC-141b, $ClONO_2$, ClO, HOCl & H-1211 to the total chlorine budget in the latitude bands between 30° S and 70° S (morning occultations).

Altitude	CFC-114	CFC-115	HCFC-142b	HCFC-141b	$ClONO_2$	ClO	HOCl	H-1211
10.5	0.96	0.27	0.45	1.03	0.20	0.02	0.04	0.11
11.5	0.93	0.26	0.43	0.97	0.50	0.04	0.04	0.10
12.5	0.89	0.26	0.41	0.91	0.92	0.06	0.03	0.09
13.5	0.87	0.26	0.40	0.87	1.24	0.14	0.03	0.09
14.5	0.84	0.26	0.39	0.83	1.65	0.25	0.04	0.08
15.5	0.82	0.25	0.37	0.78	2.38	0.38	0.04	0.07
16.5	0.78	0.25	0.36	0.72	3.51	0.55	0.06	0.06
17.5	0.73	0.24	0.33	0.64	5.32	0.78	0.07	0.05
18.5	0.67	0.24	0.30	0.55	7.76	0.94	0.08	0.04
19.5	0.62	0.23	0.28	0.46	10.94	0.98	0.09	0.02
20.5	0.57	0.22	0.26	0.38	14.69	1.17	0.12	0.01
21.5	0.54	0.21	0.24	0.32	18.33	1.37	0.14	0.01
22.5	0.50	0.21	0.22	0.26	21.92	1.49	0.18	0.00
23.5	0.47	0.20	0.21	0.21	24.88	1.64	0.20	0.00
24.5	0.44	0.20	0.20	0.16	26.52	1.68	0.23	0.00
25.5	0.42	0.19	0.19	0.12	27.27	1.82	0.26	0.00
26.5	0.40	0.19	0.19	0.09	27.36	1.78	0.28	0.00
27.5	0.38	0.19	0.18	0.06	26.79	1.61	0.31	0.00
28.5	0.36	0.19	0.17	0.04	25.59	1.67	0.35	0.00
29.5	0.34	0.19	0.17	0.03	24.19	1.87	0.40	0.00
30.5	0.31	0.18	0.16	0.02	22.80	2.21	0.46	0.00
31.5	0.29	0.18	0.15	0.01	21.42	2.68	0.58	0.00
32.5	0.27	0.18	0.14	0.01	19.75	3.41	0.72	0.00
33.5	0.25	0.18	0.13	0.00	17.99	4.39	0.88	0.00
34.5	0.23	0.18	0.13	0.00	15.54	5.83	1.04	0.00
35.5	0.22	0.18	0.12	0.00	12.81	7.52	1.16	0.00
36.5	0.20	0.17	0.11	0.00	10.05	9.29	1.26	0.00
37.5	0.19	0.17	0.10	0.00	7.51	10.76	1.31	0.00
38.5	0.17	0.17	0.10	0.00	5.13	12.13	1.30	0.00
39.5	0.16	0.17	0.09	0.00	3.14	13.18	1.26	0.00
40.5	0.14	0.17	0.09	0.00	1.75	13.74	1.18	0.00
41.5	0.13	0.16	0.08	0.00	0.97	13.72	1.05	0.00
42.5	0.12	0.16	0.07	0.00	0.49	13.40	0.89	0.00
43.5	0.11	0.16	0.07	0.00	0.23	12.75	0.74	0.00
44.5	0.10	0.15	0.06	0.00	0.12	11.76	0.60	0.00
45.5	0.09	0.15	0.06	0.00	0.05	10.66	0.48	0.00
46.5	0.08	0.15	0.05	0.00	0.02	9.51	0.39	0.00
47.5	0.07	0.15	0.05	0.00	0.01	8.35	0.31	0.00
48.5	0.07	0.14	0.05	0.00	0.01	7.29	0.24	0.00
49.5	0.06	0.14	0.05	0.00	0.00	6.39	0.19	0.00
50.5	0.06	0.14	0.04	0.00	0.00	5.60	0.15	0.00
51.5	0.05	0.14	0.04	0.00	0.00	4.90	0.11	0.00
52.5	0.05	0.14	0.04	0.00	0.00	4.22	0.09	0.00
53.5	0.05	0.14	0.04	0.00	0.00	3.64	0.07	0.00

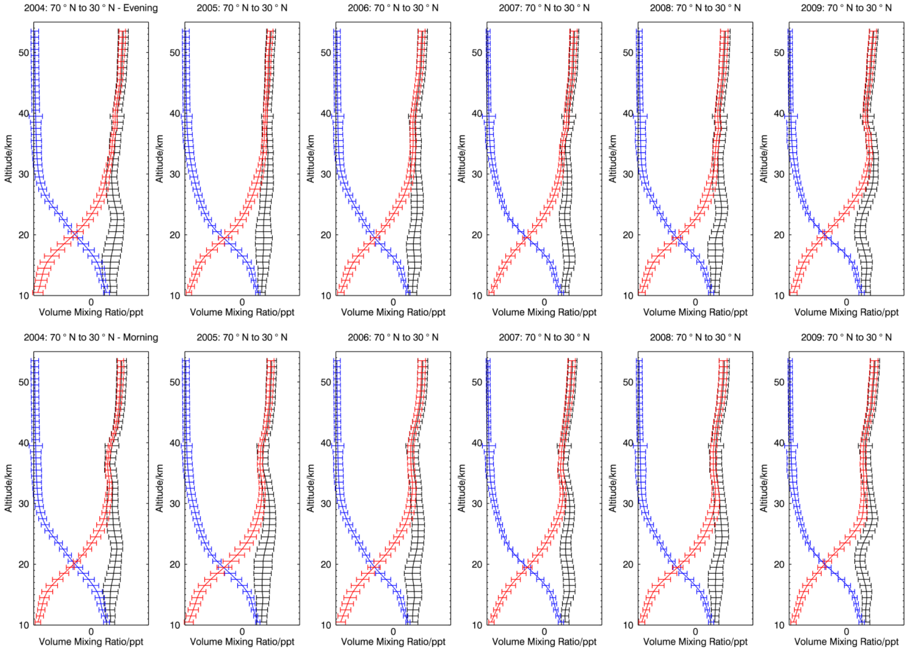

Fig. 1. The total chlorine (black), inorganic chlorine (blue) and organic chlorine (red) profiles for measurements made by the ACE-FTS between 70° N and 30° N. The error bars on the total, inorganic and organic chlorine profiles are a linear combination of the standard deviations of the data which was used to calculate these profiles. The profiles in the top panels are calculated using evening (local time) profiles of ClO, $ClONO_2$ and HOCl while the profiles in the bottom panels are for morning (local time) profiles.

Title Page
Abstract Introduction
Conclusions References
Tables Figures
◀| |▶
◀ ▶
Back Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

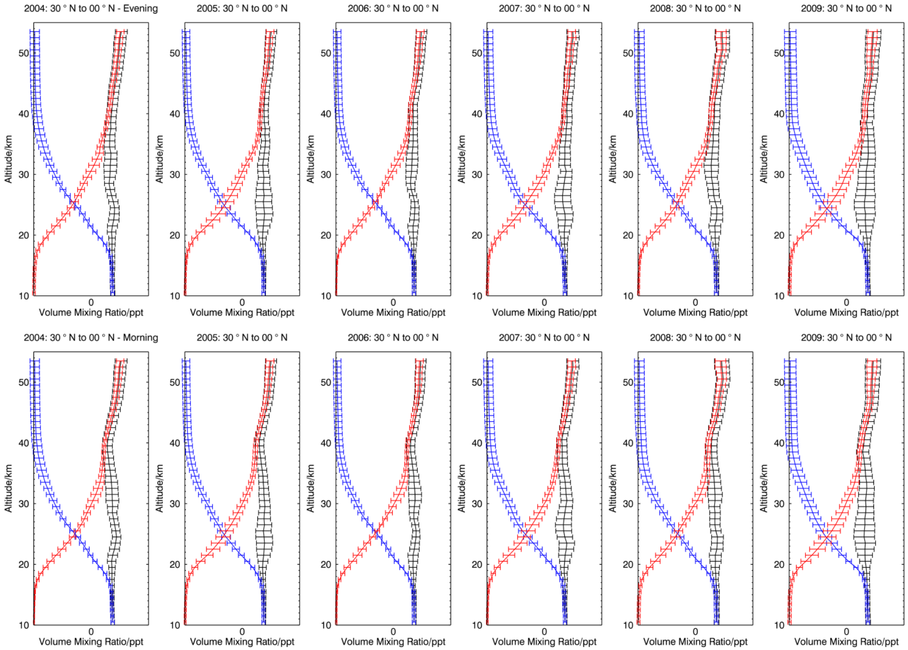

Fig. 2. As Fig. 1 but for 30° N–0° N.

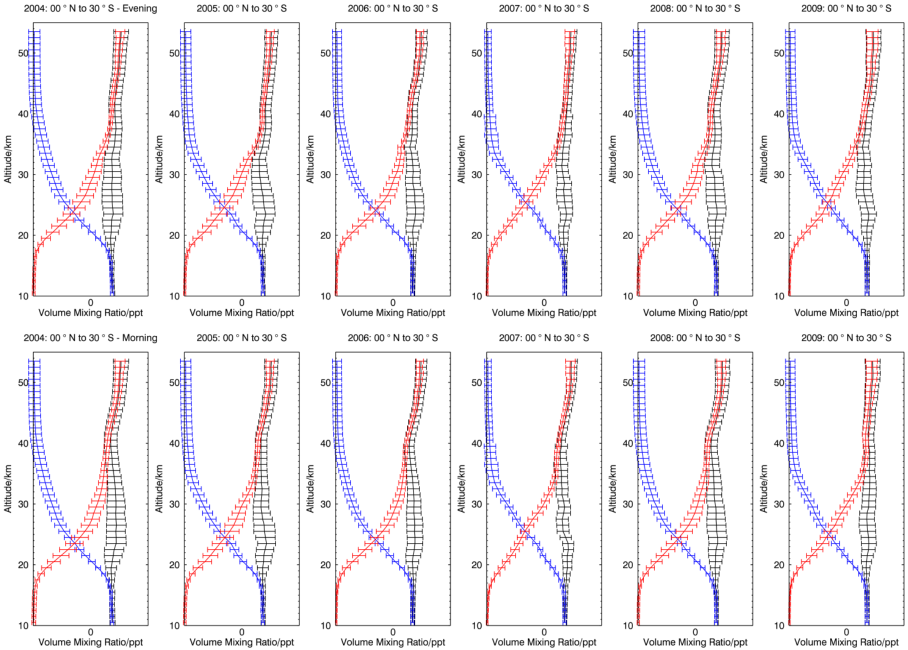

Fig. 3. As Fig. 1 but for 0° N–30° S.

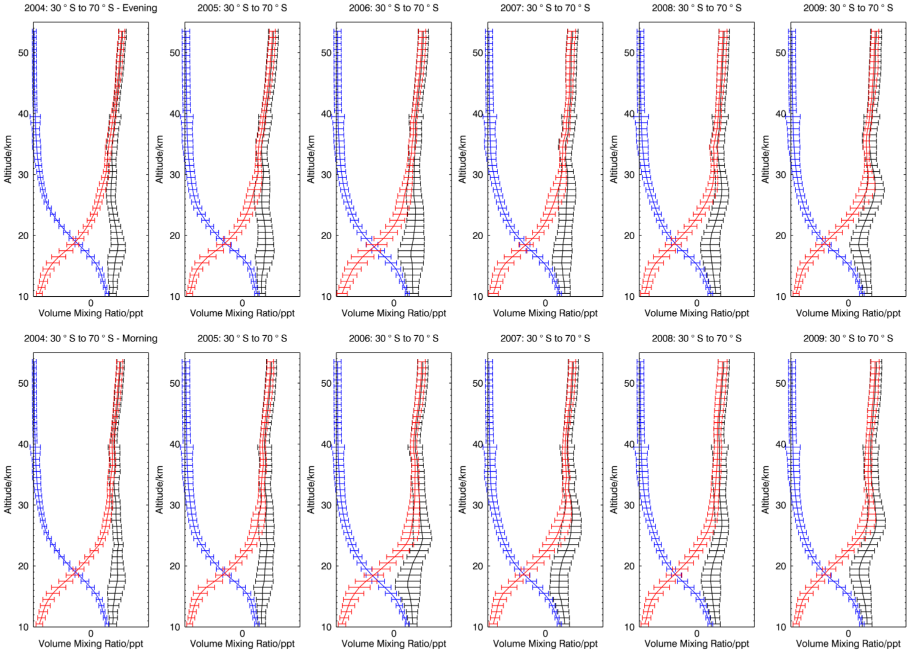

Fig. 4. As Figure 1 but for 30° S–70° S.

ACPD
13, 23491–23548, 2013

Title Page
Abstract Introduction
Conclusions References
Tables Figures
I◀ ▶I
◀ ▶
Back Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

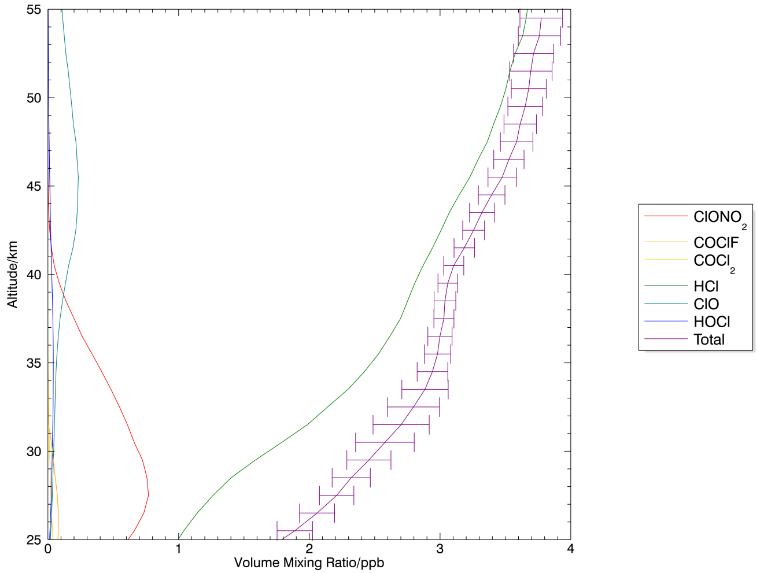

Fig. 5. The mean morning profiles of inorganic chlorine species ($ClONO_2$, COClF, $COCl_2$, HCl, ClO and HOCl) between 30° N and the equator.

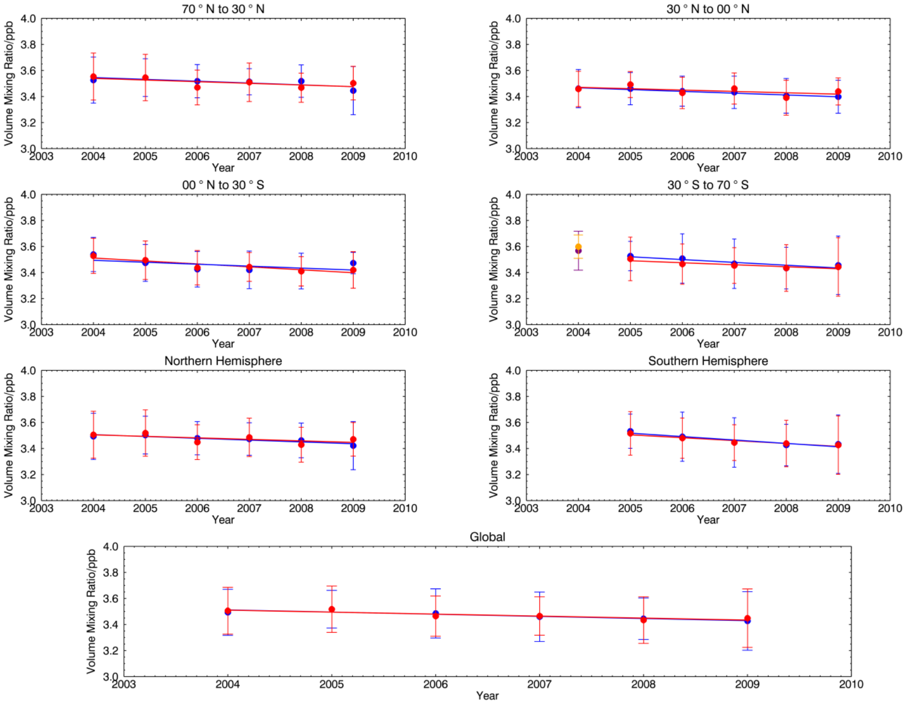

Fig. 6. The trends in the mean total stratospheric chlorine between 2004 and 2009 for different regions. The red line represents the line of best fit for the evening occultations mean stratospheric total chlorine volume mixing ratio (the red circles). The blue line represents the line of best fit for the morning occultations mean stratospheric total chlorine volume mixing ratio (the blue circles). The error bars shown in the plots are calculated from the standard deviation of the data used to calculate the mean total chlorine VMRs.

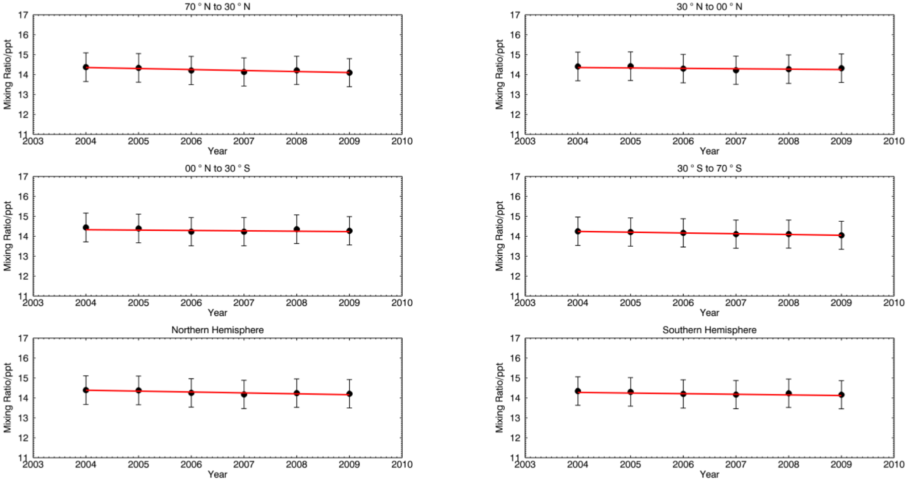

Fig. 7. The trends in the GWP-weighted total chlorine (ppb) between 2004 and 2009. The red line represents the line of best fit for the GWP weighted mean volume mixing ratio (the black circles). The error bars shown in the plots are calculated from the weighted standard deviation of the data used to calculate the means.

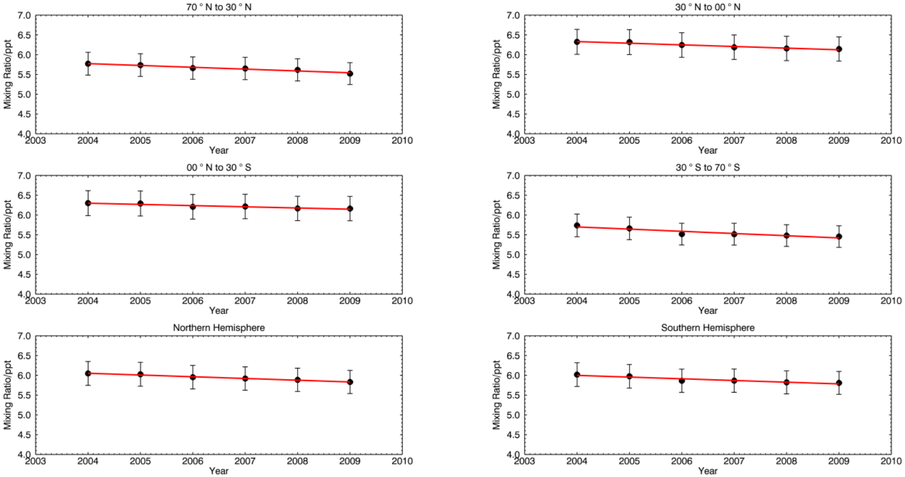

Fig. 8. The trends in the ODP-weighted total chlorine between 2004 and 2009. The red line represents the line of best fit for the ODP weighted mean volume mixing ratio (the black circles). The error bars shown in the plots are calculated from the weighted standard deviation of the data used to calculate the means.