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

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Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Abstract

Fluorine-containing species can be extremely effective atmospheric greenhouse gases. We present fluorine budgets using organic and inorganic species retrieved by the ACE-FTS satellite instrument supplemented with output from the SLIMCAT 3D chemical transport model. The budgets are calculated between 2004 and 2009 for a number of latitude bands: 70–30° N, 30–0° N, 0–30° S, and 30–70° S. At lower altitudes total fluorine profiles are dominated by the contribution from CFC-12, up to an altitude of 20 km in the extra-tropics and 29 km in the tropics, above these altitudes the profiles are dominated by HF. Our data show that total fluorine profiles at all locations have a negative slope with altitude, providing evidence that overall fluorine emissions (measured by their F content) have been increasing with time. Total stratospheric fluorine is increasing at a similar rate in the tropics; 32.5 ± 4.9 ppt yr⁻¹ (1.31 ± 0.20 % per year) in the Northern Hemisphere (NH) and 29.8 ± 5.3 ppt yr⁻¹ (1.21 ± 0.22 % per year) in the Southern Hemisphere (SH). Extra-tropical total stratospheric fluorine is also increasing at a similar rate in both the NH and SH; 28.3 ± 2.7 ppt per year (1.12 ± 0.11 % per year) in the NH and 24.3 ± 3.1 ppt per year (0.96 ± 0.12 % per year) in the SH. The volume mixing ratio of each species used in this study was weighted by its global warming potential (GWP), relative molecular mass and the atmospheric pressure to produce a GWP-weighted total fluorine trend. These trends show mean changes of 0.02 ± 0.08 % per year in the NH, and 0.07 ± 0.05 % per year in the SH. Overall, GWP-weighted fluorine remains roughly constant globally. However, the decreasing trends in the mixing ratios of halons and CFCs, due to their prohibition under the Montreal Protocol, have suppressed an increase in total fluorine caused by increasing mixing ratios of HFCs. This has reduced the impact of fluorine containing species on global warming.

1 Introduction

Many fluorine-containing chemicals are widely used in industry and elsewhere because they are chemically inert, non-toxic and odourless. A number of fluorine-containing species are controlled under the Montreal Protocol (UNEP, 2009) because they are ozone depleting substances. Whilst fluorine atoms do not play a direct part in ozone loss, many of the species, such as chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs) also contain chlorine and so do contribute to ozone loss. The C–F bonds in these molecules typically absorb infrared radiation between 1000 and 1300 cm^{-1} (Lide, 1990); a “window” region in which the atmosphere is almost transparent. At wavenumbers below 1000 cm^{-1} incoming radiation is absorbed by CO_2 and H_2O , at wavenumbers above 1400 cm^{-1} incoming radiation is absorbed by CH_4 and H_2O . Fluorine-containing species are therefore very powerful greenhouse gases and as such their emissions are limited under the Kyoto Protocol (Solomon, 2007). The global warming potentials (GWPs) (on a 20 yr timeframe) of some of the species used in this work are shown in Table 1. Many fluorine containing species such as CFCs and hydrofluorocarbons (HFCs) are very stable and inert, thus they have extremely long atmospheric lifetimes. For example, HFC-23 has a 100 yr global warming potential of 14 800 relative to that of CO_2 . Long-term monitoring of these species is therefore important for climate prediction. Fluorine budgets are useful metrics for checking the atmospheric chemistry of fluorine containing species. Previously fluorine budgets have used changes in the VMR of total fluorine as a proxy for changes in VMR of chlorine since many of these species contained both fluorine and chlorine containing species (Nassar et al., 2006b). In recent years, with the increased emissions of HFCs and decreases in purely chlorine containing species such as CCl_4 and CH_3CCl_3 , this approach can no longer be used. However, coupled with stratospheric chlorine budgets the total fluorine calculation allows the effects of the Montreal Protocol to be quantified, with chlorine-containing species being replaced by fluorine-containing species.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Global stratospheric
fluorine inventories
for 2004–2009**

A. T. Brown et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

A number of fluorine budgets have been calculated previously. Measurements made by the Atmospheric Trace MOlecule Spectroscopy (ATMOS) instrument were used to calculate a stratospheric fluorine budget using VMR profiles of CF₄, CFC-11, CFC-12, COF₂, HCFC-22, HF and SF₆ at 30° N for 1985 (Zander et al., 1992). Further work was carried out using the Jet Propulsion Laboratory's MkIV balloon-borne Fourier Transform Spectrometer retrievals of CF₄, CFC-11, CFC-12, CFC-113, COF₂, HCFC-22, HF and SF₆ (Sen et al., 1996). The most recent fluorine budget was carried out using data from the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) (Nassar et al., 2006b). This budget used version 2.2 ACE-FTS retrievals of CF₄, CFC-11, CFC-12, CFC-113, COF₂, HCFC-22, HCFC-142b, HFC-134a, HF and SF₆. When considered together these works show increasing stratospheric fluorine volume mixing ratios between 1985 and 2004. Total column measurements of COF₂ and HF from ground-based Fourier Transform Infra-red Spectrometers (FTIR) showed stratospheric fluorine increased at a rate of 0.4 % per year between 2005 and 2008 (WMO, 2011). Ground-based FTIR measurements from Kiruna, Sweden show that the HF column increased at a rate of 1.0 ± 0.3 % per year between 1996 and 2008 (Mikuteit, 2008).

This paper presents fluorine budgets calculated from ACE-FTS measurements supplemented with output from the SLIMCAT 3D Chemical Transport Model (CTM). The budgets have been calculated using 18 fluorine-containing species (lists of which are given in Sect. 3), and represents the most comprehensive fluorine budget in the upper atmosphere to date. These budgets were calculated in 4 latitude bands between 70° N and 70° S. The ACE-FTS instrument has been active since 2004 and we are therefore able to chart the long-term changes in atmospheric volume mixing ratios (VMR) of fluorine-containing species during this time. As such an individual fluorine budget has been calculated for every year from 2004 to 2009. The large number of fluorine-containing species used in this work allows the calculation of the total fluorine VMR. Previous studies from ACE data from this period have shown that the rate of decrease in the VMRs of CFC-11, CFC-12 and CFC-113 is significantly smaller than the rate

of increase in the VMRs of the 3 most common HCFCs, HCFC-22, HCFC-141b and HCFC-142b (Brown et al., 2011). The effect that these changes in the atmospheric VMRs of particular species will have on the total fluorine budget will be dependent on the number of fluorine atoms contained by each species. The GWP weighted trends in total fluorine are also presented and illustrate how changes in atmospheric fluorine have affected climate forcing.

Section 3 gives a brief description of ACE and a discussion on the validation of fluorine-containing species retrieved by ACE. A brief description of the SLIMCAT 3D Chemical Transport Model (CTM) (for a full description of the model readers are directed to Chipperfield, 2006) is given in Sect. 4 and stratospheric fluorine chemistry is discussed in Sect. 5. Section 6 outlines our method for calculating the fluorine budget. The results of this work are presented and discussed in Sect. 7.

2 The Atmospheric Chemistry Experiment

Designed to study “the chemical and dynamical processes that control the distribution of ozone in the stratosphere and upper troposphere” (Bernath, 2006), the Atmospheric Chemistry Experiment (ACE) Fourier Transform Spectrometer (FTS) was launched on board the satellite SCISAT-1 in August 2003. SCISAT-1’s orbit, a low circular orbit with an inclination of 74° (Bernath et al., 2005), gives ACE-FTS almost global coverage from the Antarctic to the Arctic. ACE’s primary instrument is a high-resolution (0.02 cm^{-1}) FTS which operates between 750 and 4400 cm^{-1} . The sun acts as a source of infra-red radiation allowing ACE-FTS to record transmission spectra of the limb of the atmosphere at a series of tangent heights during sunrise and sunset. Using the retrieval methods outlined by Boone et al. (2005), version 3.0 of the ACE retrieval currently produces atmospheric profiles of over 30 molecules from ACE-FTS spectra (<http://www.ace.uwaterloo.ca>). The ACE-FTS is in its tenth year of operation with routine data being available from February 2004.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



The ACE-FTS currently measures 8 fluorine-containing species, CF₄, CFC-12 (CCl₂F₂), CFC-11 (CCl₃F), COF₂, COClF, HCFC-22 (CHF₂Cl), HF and SF₆ with version 3.0 retrievals and 9 yr of data (2004–2012; version 3.0 has some problems after September 2010). The retrieval limits of these species are shown in Table 2. Version 3.0 also has retrievals for CFC-113 (C₂Cl₃F₃), HCFC-142b (C₂H₃ClF₂), HCFC-141b (C₂H₃Cl₂F) (Brown et al., 2011) and HFC-23 (CHF₃) (Harrison et al., 2012), but the retrieved concentrations have substantial biases so it was decided to use model data instead. ACE data was supplemented with output from the SLIMCAT 3D chemical transport model for CFC-113, CFC-114, CFC-115, H-1211, H-1301, HCFC-141b, HCFC-142b, HFC-23, HFC-134a and HFC-152a.

2.1 Fluorine containing species retrieved by the Atmospheric Chemistry Experiment

2.1.1 Carbon Tetrafluoride – CF₄

CF₄ is a powerful greenhouse gas (see Table 1) which has a lifetime of over 2300 yr, the emissions of which are primarily from aluminium production. First retrieved from ACE-FTS data for the stratospheric fluorine budget (Nassar et al., 2006b), the ACE-FTS retrieval was validated by comparison with non-coincident Mk-IV balloon profiles. This study found that profiles from Mk-IV and ACE were within ±10% (Velazco et al., 2011).

2.1.2 Sulphur Hexafluoride – SF₆

SF₆ is used as an electrical insulating gas in power distribution equipment and as an inert chemical tracer. The long lifetime of 3200 yr and strong infrared absorption cross sections in an atmospheric window region near 950 cm⁻¹, are responsible for making this molecule a potent greenhouse gas. ACE-FTS retrievals of SF₆ were validated by

ACPD

13, 16885–16924, 2013

Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



comparison with non-coincident Mk-IV balloon profiles. This study found that an agreement within $\pm 15\%$ between 12 and 19 km (Velazco et al., 2011).

2.1.3 CFC-11 – CCl₃F

CFC-11 is banned under the Montreal Protocol and is the second most abundant CFC in the atmosphere. Validation of ACE-FTS retrievals were carried out using the FIRS-2 instrument. These comparisons showed an agreement to within 10% below 16 km (Mahieu et al., 2008). Comparisons between ACE and Mk-IV profiles 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 $\pm 20\%$ between 17 km and 24 km (Velazco et al., 2011).

2.1.4 CFC-12 – CCl₂F₂

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

2.1.5 HCFC-22 – CHClF₂

HCFCs are transitional replacement compounds for CFCs under the Montreal Protocol. HCFC-22 is the most abundant HCFC in the atmosphere as it has been widely used since the 1950s. The HCFC-22 retrieval used in this paper is new; a paper is currently under preparation which discusses this new retrieval.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



2.1.6 Carbonyl Chlorofluoride – COClF

COCIF is produced from the decomposition of molecules which contain single fluorine atoms, 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 Carbonyl Fluoride – COF₂

COF₂ is produced from the decomposition of molecules which contain two fluorine atoms. Comparisons with measurements from Mk-IV show an agreement to within ±20 % (Velazco et al., 2011).

2.1.8 Hydrogen Fluoride – HF

HF is the main fluorine reservoir in the stratosphere. Retrievals of ACE-FTS HF have been compared to retrievals from the HALOE instrument. The results of a first analysis showed that the VMR of retrievals from ACE were around 10–20 % larger than HALOE (McHugh et al., 2005; Mahieu et al., 2008). ACE-FTS retrievals were again compared once more to HALOE, these results showed that ACE-FTS retrievals were larger by between 5–20 % (Mahieu et al., 2008). HF from ACE-FTS has also been compared to measurements from FIRS-2. These results showed significant differences between ACE and FIRS-2 with differences between 60 % (below 16 km where the VMR of HF is at its lowest) to 20 % (Mahieu et al., 2008). This study also compared Mk-IV and ground-based FTIR to ACE HF. These results showed a difference of ±10 % between ACE and MK-IV above 19 km and an average difference of ±7.4 % for the average partial column averages from ground-based FTIRs.

ACPD

13, 16885–16924, 2013

Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



3 SLIMCAT 3D chemical transport model

We have supplemented ACE observations 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). The model uses winds from meteorological analyses to specify horizontal transport while 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 $5.6^\circ \times 5.6^\circ$ and 32 levels from the surface to about 60 km. The model uses a $\sigma - \theta$ 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) (WMO/UNEP, 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 ($70-30^\circ N$, $30-00^\circ N$, $00^\circ N-30^\circ S$ and $30-70^\circ S$) on a 1 km altitude grid.

4 Fluorine chemistry

There are two principal reaction paths for the breakdown of fluorine-containing species in the atmosphere. The reaction path for a particular compound is determined by the number of fluorine atoms in the molecule.

Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

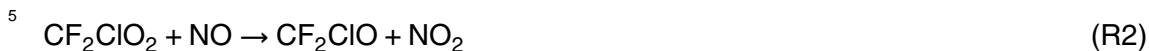
Printer-friendly Version

Interactive Discussion



4.1 CFCs and HCFCs which contain two fluorine atoms (COF₂ formation)

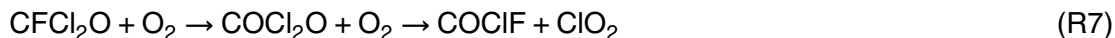
The source gas molecule will first be broken down into CF₂Cl and will then follow the following path (Tressaud, 2006):



Above around 30 km the concentration of COF₂ decreases as photolysis becomes more effective. The primary source of COF₂ in the atmosphere is the decomposition of HCFC-22 and CFC-12. Contributions from HFCs, whilst smaller, are significant ensuring that the concentration of stratospheric COF₂ has been increasing since 2004 (Brown et al., 2011).

4.2 CFCs which contain a single fluorine atom (COCIF formation)

The source gas molecule will first be broken down into CFCI₂ and will then follow the following path (Tressaud, 2006):



16894

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





5 Carbonyl chlorofluoride (COCIF) is less stable than COF_2 and can be photolyzed more easily. It is therefore present in far smaller concentrations and peaks at lower altitudes than COF_2 . The primary source for atmospheric COCIF is CFC-11.

The production of FCO from the decomposition of COCIF and COF_2 leads to the formation of fluorine atoms, along the following reaction path:



The fluorine produced in both series of reactions will react with methane (CH_4), water (H_2O) or molecular hydrogen (H_2) to form HF.



20 Photolysis of HF does not readily occur in the stratosphere, making HF an almost permanent reservoir of stratospheric fluorine (Tressaud, 2006). This means that the atmospheric concentrations of F and FO are very small, preventing fluorine from causing significant ozone loss. HF is ultimately removed from the stratosphere by slow transport to, and rainout in, the troposphere or by upward transport to the mesosphere where its mixing ratio remains constant up to high altitudes.

Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



5 Fluorine budget method

In total 16 186 ACE-FTS occultations were used in this study. For this analysis the globe was divided into 4 latitude bands (70–30° N, 30–0° N, 0° N–30° S and 30–70° S) chosen to represent the different transport regimes in the stratosphere. Outside of the tropics and the polar regions the stratosphere is well mixed by breaking planetary scale waves. Within the tropics powerful convective transport can penetrate the tropopause, mixing the lower layers of the stratosphere. Occultations were divided by latitude and year; the number of data in each band is shown in Table 3. Data in each band were filtered using the median absolute distribution (MAD) of the data. The MAD is a robust statistic which allows the variability of a data set to be calculated without giving large significance to outliers. For this reason it is ideal for filtering profiles retrieved from ACE which may contain a small number of large outliers due to problems with retrievals (such as problems with retrievals in occultations with high lying clouds or aerosols). Any value which was greater than 2.5 times the MAD from the median of the raw data was discarded. The 2.5 MAD filter was used since this includes between 80 and 98 %, depending on the distribution of the data around the median, of the ACE-FTS data ensuring that only outliers were removed. Once the outliers had been removed a mean profile of each species was produced by calculating the mean mixing ratio at each altitude. The error on this profile is calculated using the standard deviation of the data used to calculate each mean mixing ratio. This produces a profile with errors which vary with altitude, dependent on the variation of the data 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. SF₆ and CF₄ did not have corresponding SLIMCAT data and so were extended vertically upwards at a constant VMR from the final retrieved altitude. All profiles were extended up to 54.5 km corresponding to the maximum altitude to which HF is retrieved by ACE-FTS.

The total fluorine volume mixing ratio was calculated at 54 levels between 0.5 km and 53.5 km corresponding to the ACE retrieval altitude grid. The equations used for

ACPD

13, 16885–16924, 2013

Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



this calculation are given below, where square brackets indicate volume mixing ratio (VMR). Following the convention of previous fluorine budgets (Nassar et al., 2006b; Zander et al., 1992), the total fluorine volume mixing ratio can be expressed as the sum of the total inorganic fluorine and the total organic fluorine. The terms inorganic and organic are not used in the conventional sense in this work (and the fluorine budgets previous to this one). Instead the term organic fluorine can be loosely interpreted as those fluorine species which are emitted from the surface. Inorganic fluorine species are fluorine-containing molecules which are produced from the decomposition of the organic fluorine species (with the exception of SF₆).

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

$$[\text{Total Inorganic Fluorine}] = [\text{HF}] + 2[\text{COF}_2] + [\text{COCIF}] + 6[\text{SF}_6]$$

$$\begin{aligned} [\text{Total Organic Fluorine}] = & 4[\text{CF}_4] + 2[\text{CFC-12}] + [\text{CFC-11}] + 2[\text{HCFC-22}] \\ & + 3[\text{CFC-113}] + 4[\text{CFC-114}] + 5[\text{CFC-115}] + 2[\text{HCFC-142b}] + [\text{HCFC-141b}] \\ & + 3[\text{HFC-23}] + 4[\text{HFC-134a}] + 2[\text{HFC-152a}] + 2[\text{H-1211}] + 3[\text{H-1301}] \end{aligned}$$

6 Results and discussion

This section is divided into 5 subsections. Section 7.1 presents the vertical profiles of the total fluorine calculations, with a focus on the gradient on the profile and its implications for the total fluorine VMR in the atmosphere. Subsequently the slopes of the correlation plots between source fluorine species and reservoir fluorine species are presented and discussed in Sect. 7.2. The impacts of the individual species included in the total fluorine calculations on the total fluorine VMR are presented in Sect. 7.3. The final two sections present the changes in total fluorine between 2004 and 2009. Section 7.4 presents the changes in total stratospheric fluorine while Sect. 7.5 discusses the implications of these changes on climate forcing using a weighted total fluorine

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



budget. These values were calculated using the mean total fluorine VMR weighted by the relative molecular mass, pressure and 20 yr GWP.

6.1 Vertical profiles

The results of the total fluorine calculations are shown in Figs. 1 and 2. As described in Sect. 6 each point on the mean vertical profile of a species retrieved by ACE has an error, which represents the standard deviation of the data at that altitude, attached to it. The error bars shown on the plots represent the linear combination of the standard deviations of the contributing species for that particular altitude. The SLIMCAT profiles have been given a 5 % error on their VMR. This value was chosen as it is an overestimation of the error on the surface measurements used to force SLIMCAT, and allows for some error in model transport. The total fluorine profiles seem to follow a straight line with the majority of the deviations from this line coming from the HF retrieval. The mean total stratospheric fluorine and the slope of the total fluorine profile can be seen in Table 4. There does not appear to be any statistically significant difference between extra-tropical and tropical total fluorine VMR. The mean stratospheric fluorine in the extra-tropical region is generally larger than in the tropics. The differences between corresponding tropical and extra-tropical stratospheric means are generally smaller than the error on the values. This suggests that these differences are likely due to fluctuations in the retrieval of HF (the main source of atmospheric fluorine at higher altitudes) as opposed to a true feature. The slopes of the total fluorine profiles are smaller in the tropical stratosphere than in the extra-tropical stratosphere. Since the age-of-air increases with altitude, the slope of the total fluorine profile represents the changes in the emissions of fluorine-containing species over time. The negative slopes of the total fluorine profiles show that the mixing ratios of fluorine-containing species have been increasing. Additionally, the slopes suggest that the rate of increase is faster in the tropics than in the extra-tropics. Figure 3 shows the mean percentage contribution of ACE-FTS and SLIMCAT VMR to the total fluorine profile for all years in the extra-tropical northern latitudes. The error bars represent the largest and smallest percentage contributions.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



All latitude bands show very similar altitude dependent percentage contributions. ACE-FTS data accounts for between around 77 % of the total fluorine, in the lower altitudes, to around 96 % at the highest altitudes.

6.2 Fluorine source and reservoir species correlations

5 The fluorine-containing species used in this analysis can be divided into source and reservoir species, reservoir species being produced from the decomposition of source species. In this work HF, COF₂ and COClF are considered to be reservoir species; all other species (CF₄, CFC-12, CFC-11, HCFC-22, SF₆, CFC-113, CFC-114, CFC-115, H-1211, H-1301, HCFC-141b, HCFC-142b, HFC-23, HFC-134a and HFC-152a) are
10 source species. Correlating the source and reservoir species gives an indication as to whether the major fluorine-containing species have been considered; a slope close to one would show that most of the major fluorine containing species were included in the budget. It should be noted that this method cannot be used to check if all very long lived fluorine species (such as SF₆) have been included in the study. Instead this
15 method is a test whether the major source species which decompose in the stratosphere and reservoir species have been included in the budget. The results of these plots can be seen in Table 5. All calculated slopes are greater than 0.9 with the lowest values occurring in the tropical regions. The mean of the slope in the extra-tropics is -0.97 ± 0.01 in the Northern Hemisphere and -0.98 ± 0.01 Southern Hemisphere.
20 In the northern tropics the mean slope of the correlation is -0.91 ± 0.02 , and in the southern tropics the mean slope is -0.90 ± 0.01 . These values suggest that the most important fluorine-containing species have indeed been included in this budget. Whilst many short-lived species are not included their overall contribution to stratospheric fluorine is small, and so their addition to the budget calculations would not significantly
25 affect the final fluorine budget. The slopes from the tropical latitudes are smaller than those from extra-tropical latitudes. This is likely an artefact from the smaller number of occultations which are available at tropical latitudes.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



6.3 The contributions of species to the total fluorine budget

The contributions of individual species to the total fluorine budget were calculated by producing a mean fluorine budget for each latitude band from annual data. Once these budgets had been produced the impact of each species on the fluorine budget could be calculated. The contribution of individual species to the total fluorine budgets can be found in the appendix. The species which contribute the largest VMR to the total fluorine budget (F_{tot}) for different altitudes and latitude bands are shown in Table 6. At the lower altitudes of this study, between 11 and 20 km in the extra-tropics (ET) and 11 and 29 km in the tropics (T), the total fluorine VMR is dominated by CFC-12. At its peak CFC-12 accounts for almost 40 % of atmospheric fluorine. This occurs despite a decrease in the VMR of atmospheric CFC-12 during this time. At altitudes of up to 34 km in the tropics (29 km in ET), the VMR of total fluorine from CFCs and halons is larger than that from HFCs and HCFCs. The contribution of HCFCs and HFCs to atmospheric fluorine is dominated by HCFC-22 which, at its peak at 10.5 km, accounts for between 14 % and 15 % of the total atmospheric fluorine depending on the latitude band. The combined contribution of the HCFCs (excluding HCFC-22) and HFCs used in this study peaks between 9.8 % (ET) and 10.1 % (T). These values are significantly smaller than the combined contribution of the CFCs and halons (excluding CFC-12) which peak between 22 % (ET) and 23 % (T). Above 20 km in the ET and 29 km in the T, HF dominates reaching between 74 % (ET) and 77 % (T) of total fluorine at 53.5 km.

6.4 Trends in stratospheric fluorine

Since the fluorine budget has been calculated for the years between 2004 and 2009 it is possible to calculate trends in the annual changes of the mean total stratospheric fluorine. Figure 4 shows the trend plots for each of the 4 latitude bands and the results of the analysis can be seen in Table 7. The errors quoted here are the $1-\sigma$ fitting error of a linear least squares fit to the data. Systematic errors are not considered in this work as they have not been calculated for ACE-FTS at this time. Systematic

Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



errors should not have a significant effect on the trends as they are consistent annually for each species used in this budget. In addition, altitude dependent systematic errors will be identical annually, thus as long as the same altitudes are used to calculate an average they will cancel for the annual trends. The VMR of total stratospheric fluorine is increasing at a similar rate at all latitudes. The VMR of total stratospheric fluorine is increasing at a rate of $1.12 \pm 0.11 \text{ \% yr}^{-1}$ in the northern extra-tropical stratosphere, and $0.96 \pm 0.12 \text{ \% yr}^{-1}$ in the southern extra-tropical stratosphere. The rate of increase in stratospheric fluorine is also similar in the northern tropical stratosphere ($1.31 \pm 0.20 \text{ \% yr}^{-1}$), and in the southern tropical ($1.21 \pm 0.22 \text{ \% yr}^{-1}$). Northern and southern hemispheric trends were calculated using the averages of the fluorine budgets of the relevant latitude bands. This method was preferred to calculating a budget from all the individual occultations within each hemisphere so that the budget is more representative of the whole hemisphere. As can be seen from Table 3 there are significantly more measurements at higher latitudes than in the tropics. A budget calculated from all the occultations in a particular hemisphere would therefore produce an average budget which was biased towards the higher latitudes. Total fluorine in the Northern Hemisphere and Southern Hemisphere is increasing at a similar rate, $1.21 \pm 0.14 \text{ \% yr}^{-1}$ in the Northern Hemisphere, and $1.07 \pm 0.21 \text{ \% yr}^{-1}$ in the Southern Hemisphere. Once more the errors quoted here are the statistical $1-\sigma$ fitting error of a linear least squared fit to the data. Globally the VMR of fluorine in the stratosphere is increasing at a rate of $1.14 \pm 0.06 \text{ \% yr}^{-1}$. Despite the lower altitudes of the budget being dominated by CFC-12, a strong increase in the VMR of total fluorine is still seen. This is due to increases in the VMR of HCFCs and HFCs being significantly larger than the decreases in CFCs and halons during this time.

6.5 Global Warming Potential weighted fluorine budget trends

The main environmental impact of the increased atmospheric fluorine will be on warming in the troposphere and lower stratosphere region. The different species analysed for this work have different Global Warming Potentials (GWPs) (see Table 1) and therefore

**Global stratospheric
fluorine inventories
for 2004–2009**

A. T. Brown et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



have different effects on climate. A more useful quantity is therefore the trend in the atmospheric fluorine-weighted GWP. Since the GWP is a measure of the global warming potential of 1 kg of substance compared to 1 kg of CO₂, the GWP total fluorine cannot be calculated by simply weighting the VMR of the fluorine source species by their GWP. Instead, the total fluorine 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 effect of these species is concentrated in the troposphere, data above the tropopause was not included in the calculations of the GWP-weighted total fluorine. The time-series of these means can be seen in Fig. 5. GWP-weighted fluorine appears to be stable in all regions. Similar statistically insignificant increases are seen in both hemispheres with increases of 0.10 ± 0.10 % per year in the Northern Hemisphere tropics, and 0.11 ± 0.13 % per year in the Southern Hemisphere tropics. Once more the error on these values was calculated using a $1-\sigma$ fitting error of a linear least squares fit to the data. Extra-tropical latitudes also show statistically insignificant trends. In the Northern Hemisphere extra-tropical region the GWP-weighted fluorine appears to be decreasing at a rate of -0.06 ± 0.09 % per year. Whilst in the Southern Hemisphere hemisphere extra-tropical region GWP-weighted fluorine is increasing at a rate of 0.02 ± 0.03 % per year. The errors on all of these GWP-weighted trends are larger than the reported GWP-weighted trends; these results suggest that it is likely that the GWP-weighted fluorine has remained roughly constant during this time. These trends are significantly smaller than the trends in the total fluorine presented in Sect. 7.4. The reason for this difference is the importance of CFCs and halons in these calculations. As can be seen in Table 1 if the species used in this study are ranked by GWP only 2 out of the largest 7 are not CFCs or halons. Since use of these species is prohibited under the Montreal Protocol the VMR of these species has been decreasing during the time frame of this study (Brown et al., 2011). HCFCs and, increasingly, HFCs have been replacing the species banned under the Montreal Protocol. The de-

**Global stratospheric
fluorine inventories
for 2004–2009**

A. T. Brown et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



crease in CFCs and halons has, in effect, suppressed the GWP-weighted fluorine trend which would otherwise have risen during this time. Carrying out the same calculations without including CFCs and halons shows the effects of this suppression. The trends in the weighted total fluorine range between $3.51 \pm 0.16\%$ and $3.92 \pm 0.19\%$ per year when CFCs and halons are not included (see Table 8). These results suggest that the Montreal Protocol has had a positive effect on reducing global warming. By banning the use of a number of ozone-depleting substances which were also potent greenhouse gases the Montreal Protocol has inadvertently slowed the rate of increase in the VMR of greenhouse gases in the atmosphere. This agrees with the work of Velders et al. (2007), which stated that the Montreal Protocol had had a significant effect in reducing the emission of greenhouse gases.

7 Conclusions

This paper presents the stratospheric fluorine budget calculated for the years 2004 to 2009 for a number of latitude bands representing the extra-tropical and tropical latitudes. The fluorine budgets were calculated using species retrieved by ACE-FTS. ACE-FTS measurements were supplemented with data from the SLIMCAT 3D chemical transport model. The fluorine budgets, therefore, include all the major fluorine-containing species currently in the atmosphere. In the lower altitudes these budgets are dominated by the large mixing ratio of CFC-12. At its peak CFC-12 contributes around 39 % of total fluorine. Other species are much less important. HCFC-22 contributes 15 % at its maximum and combined contribution of the other HCFCs and HFCs used in this study peaks around 10 %. The remaining CFCs and halons used in this study contribute a maximum of about 23 %. As altitude increases HF overtakes CFC-12, between 21 km in the extra-tropics and 28 km in the tropics as the most dominant species in the total fluorine budget.

The gradients of the total fluorine profiles offer an indication of the long-term changes in total fluorine. All budgets exhibit a negative slope with altitude. Since air at higher

altitudes in the stratosphere is older than air at lower altitudes, higher concentrations observed at lower altitudes suggest that fluorine emissions have been increasing with time. Stratospheric fluorine is increasing in all the latitude bands used in this study.

Total tropospheric fluorine trends have been produced which have been weighted by the GWPs of the individual species used in this study. These results allow the climatological implication of the increase in fluorine during this time to be quantified. Analysis carried out without using data for the CFCs or halons show GWP-weighted fluorine increasing by a mean of $3.85 \pm 0.07\%$ per year in the Northern Hemisphere and $3.58 \pm 0.07\%$ per year in the Southern Hemisphere. When CFCs and halons are included in the calculations it appears that the VMR of GWP-weighted fluorine has not changed significantly during this time. It appears that the decrease in species banned under the Montreal Protocol has limited the climatological effects of a general increase in the VMR of fluorine-containing species, in particular HCFCs and HFCs, during this time.

The mean contribution of individual species to the total fluorine VMR in the different latitude bands.

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16904

ACPD

13, 16885–16924, 2013

Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Global stratospheric
fluorine inventories
for 2004–2009**

A. T. Brown et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



J.: Atmospheric Chemistry Experiment (ACE): Mission overview, *Geophys. Res. Lett.*, 32, L15S01, doi:10.1029/2005gl022386, 2005.

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Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Global stratospheric
fluorine inventories
for 2004–2009**

A. T. Brown et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Table 1. The global warming potential, on a 20 yr timescale, of some of the species used in this work (Solomon, 2007).

Species	Global Warming Potential
CF ₄	5210
CFC-11 (CCl ₃ F)	6730
CFC-12 (CCl ₂ F ₂)	11 000
CFC-113 (C ₂ Cl ₃ F ₃)	6540
CFC-114 (C ₂ Cl ₂ F ₄)	8040
CFC-115 (C ₂ ClF ₅)	5310
Halon-1301 (CBrF ₃)	8480
Halon-1211 (CBrClF ₂)	4750
HCFC-22 (CHClF ₂)	5160
HCFC-141b (C ₂ H ₃ Cl ₂ F)	2250
HCFC-142b (C ₂ H ₃ ClF ₂)	5490
HFC-23 (CHF ₃)	12 000
HFC-134a (C ₂ H ₂ F ₄)	3830
HFC-152a (C ₂ H ₄ F ₂)	437
SF ₆	16 300

**Global stratospheric
fluorine inventories
for 2004–2009**

A. T. Brown et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Table 2.** The retrieval altitudes of the fluorine-containing species used in this study.

Species	Altitude (km)	
	Polar	Equatorial
CFC-11 (CCl ₃ F)	5–23	6–28
CFC-12 (CCl ₂ F ₂)	5–28	5–36
HCFC-22 (CHF ₂ Cl)	5–30	7–30
COCIF	13–25	15–32
COF ₂	12–34	12–45
SF ₆	8–32	12–32
CF ₄	15–55	15–55
HF	12–52	12–57

Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

Table 3. The number of ACE-FTS occultations used for each latitude band and year.

Year	70–30° N	30–0° N	0–30° S	30–70° 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

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

Table 4. The slopes of the total fluorine profile and the mean stratospheric volume mixing ratio of the fluorine budgets for different latitude bands.

Year	70–30° N		30–0° N	
	Slope of total fluorine profile [ppt km ⁻¹]	Mean Stratospheric Fluorine [ppt]	Slope of total fluorine profile [ppt km ⁻¹]	Mean Stratospheric Fluorine [ppt]
2004	-1.76 ± 0.38	2478 ± 33	-6.94 ± 0.45	2429 ± 115
2005	-2.21 ± 0.36	2501 ± 33	-4.29 ± 0.31	2508 ± 68
2006	-2.68 ± 0.34	2529 ± 43	-5.41 ± 0.28	2513 ± 82
2007	-1.46 ± 0.27	2573 ± 25	-3.84 ± 0.39	2551 ± 65
2008	-1.78 ± 0.41	2603 ± 38	-4.51 ± 0.53	2577 ± 82
2009	-2.03 ± 0.35	2607 ± 32	-4.37 ± 0.47	2604 ± 78
Year	0° N–30° S		30° S–70° S	
	Slope of total fluorine profile [ppt km ⁻¹]	Mean Stratospheric Fluorine [ppt]	Slope of total fluorine profile [ppt km ⁻¹]	Mean Stratospheric Fluorine [ppt]
2004	-4.54 ± 0.39	2455 ± 62	-1.28 ± 0.22	2494 ± 24
2005	-5.17 ± 0.62	2451 ± 89	-1.42 ± 0.23	2510 ± 25
2006	-6.23 ± 0.51	2488 ± 76	-0.95 ± 0.38	2521 ± 33
2007	-5.49 ± 0.46	2508 ± 71	-0.29 ± 0.39	2559 ± 38
2008	-5.90 ± 0.40	2569 ± 69	-1.42 ± 0.38	2569 ± 31
2009	-4.53 ± 0.42	2592 ± 64	-1.21 ± 0.26	2621 ± 28

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

Table 5. The slopes of the correlation between fluorine source species (CF_4 , CFC-12, CFC-11, HCFC-22, SF_6 , CFC-113, CFC-114, CFC-115, H-1211, H-1301, HCFC-141b, HCFC-142b, HFC-23, HFC-134a and HFC-152a) and fluorine reservoir species (HF, COF_2 and COCIF).

Year	70–30° N	30–0° N	0° N–30° S	30–70° S
2004	-0.97 ± 0.01	-0.88 ± 0.01	-0.92 ± 0.01	-0.97 ± 0.01
2005	-0.96 ± 0.01	-0.92 ± 0.01	-0.90 ± 0.01	-0.97 ± 0.01
2006	-0.96 ± 0.01	-0.91 ± 0.01	-0.89 ± 0.01	-0.97 ± 0.01
2007	-0.97 ± 0.01	-0.93 ± 0.01	-0.90 ± 0.01	-0.99 ± 0.01
2008	-0.97 ± 0.01	-0.92 ± 0.01	-0.90 ± 0.01	-0.97 ± 0.01
2009	-0.96 ± 0.01	-0.92 ± 0.01	-0.92 ± 0.01	-0.98 ± 0.01
Mean	-0.97 ± 0.01	-0.91 ± 0.02	-0.90 ± 0.01	-0.98 ± 0.01

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)


Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Table 6. The species and its mean percentage contribution which contribute the largest VMR to the total fluorine budget (F_{tot}) for different altitudes and latitude bands.

Species	Altitude Range	% F_{tot} range	Species	Altitude Range	% F_{tot} range
70–30° N			30–0° N		
CFC-12	10.5–20.5 km	38.9–25.8	CFC-12	10.5–28.5 km	39.5–23.8
HF	21.5–53.5 km	24.9–77.3	HF	29.5–53.5 km	22.2–74.5
Species	Altitude Range	% F_{tot} range	Species	Altitude Range	% F_{tot} range
0° N–30° S			30–70° S		
CFC-12	10.5–27.5 km	39.3–23.4	CFC-12	10.5–19.5 km	38.5–24.0
HF	28.5–53.5 km	22.3–74.5	HF	20.5–53.5 km	26.3–76.6

Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

Table 7. The trends in the mean stratospheric total fluorine volume mixing ratio between 2004 and 2009 for different latitude bands and the NH and SH.

	ppt yr ⁻¹	% yr ⁻¹
70–30° N	28.3 ± 2.7	1.12 ± 0.11
30–0° N	32.5 ± 4.9	1.31 ± 0.20
0° N–30° S	29.8 ± 5.3	1.21 ± 0.22
30–70° S	24.3 ± 3.1	0.96 ± 0.12
Northern Hemisphere	30.3 ± 2.7	1.21 ± 0.11
Southern Hemisphere	26.8 ± 3.7	1.07 ± 0.15

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)


Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Table 8. The global warming potential-weighted trends in the total fluorine volume mixing ratio between 2004 and 2009. The weighted total fluorine budgets were also calculated without including CFCs and halons so that the impact of the Montreal Protocol on global warming potential weighted fluorine could be quantified.

Latitude band	Weighted total fluorine trend (% yr ⁻¹)	Weighted total halons fluorine trend without CFCs and included (% yr ⁻¹)
70–30° N	-0.06 ± 0.09	3.78 ± 0.34
30–0° N	0.10 ± 0.10	3.92 ± 0.19
0° N–30° S	0.11 ± 0.13	3.65 ± 0.27
30–70° S	0.02 ± 0.03	3.51 ± 0.16

Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

Table A1. The percentage contribution of each species to the total fluorine budget in the latitude bands between 70° N and 30° N.

Altitude	HF	CF ₂	CFC-12	CFC-11	COF ₂	COClF	HCFC-22	SF ₆	CFC-113	CFC-114	CFC-115	HCFC-142b	HCFC-141b	HFC-23	HFC-134a	HFC-152a	H-1211	H-1301
10.5	1.2	11.3	38.9	8.9	0.7	0.2	14.4	1.3	8.5	2.5	1.7	1.2	0.7	2.1	5.4	0.3	0.3	0.4
11.5	2.2	11.3	38.5	8.6	1.3	0.4	14.2	1.3	8.2	2.4	1.7	1.1	0.6	2.1	5.1	0.3	0.3	0.4
12.5	3.1	11.4	38.1	8.3	1.8	0.5	14.1	1.3	7.9	2.4	1.7	1.1	0.6	2.1	4.9	0.3	0.2	0.4
13.5	3.9	11.4	37.5	8.1	2.1	0.6	14.3	1.3	7.7	2.3	1.7	1.1	0.6	2.0	4.8	0.2	0.2	0.4
14.5	4.9	11.4	36.9	7.8	2.4	0.6	14.3	1.3	7.4	2.3	1.7	1.0	0.6	2.0	4.6	0.2	0.2	0.3
15.5	6.2	11.4	36.1	7.3	3.1	0.7	14.1	1.3	7.2	2.2	1.7	1.0	0.5	2.0	4.5	0.2	0.2	0.3
16.5	8.3	11.5	34.9	6.6	4.1	0.9	13.4	1.3	6.8	2.2	1.6	1.0	0.5	2.0	4.3	0.2	0.2	0.3
17.5	11.1	11.6	33.0	5.8	5.6	1.1	12.5	1.3	6.3	2.1	1.6	0.9	0.4	1.9	4.0	0.2	0.1	0.3
18.5	14.5	11.7	30.8	4.8	7.3	1.4	11.8	1.3	5.6	2.0	1.6	0.9	0.4	1.9	3.6	0.2	0.1	0.2
19.5	18.0	11.7	28.3	3.7	9.0	1.6	11.3	1.3	5.0	1.8	1.6	0.8	0.3	1.9	3.3	0.1	0.1	0.2
20.5	21.6	11.8	25.8	2.6	10.6	1.7	10.7	1.2	4.4	1.7	1.5	0.8	0.3	1.8	3.1	0.1	0.0	0.2
21.5	24.9	11.7	23.2	1.7	12.3	1.8	10.2	1.2	3.9	1.6	1.5	0.7	0.2	1.8	2.9	0.1	0.0	0.1
22.5	27.9	11.7	20.7	1.1	13.7	1.7	9.8	1.2	3.4	1.5	1.5	0.7	0.2	1.8	2.8	0.1	0.0	0.1
23.5	30.8	11.6	18.4	0.7	15.1	1.5	9.5	1.2	2.9	1.5	1.4	0.7	0.2	1.8	2.7	0.1	0.0	0.1
24.5	33.6	11.6	16.0	0.4	16.3	1.3	9.1	1.2	2.4	1.4	1.4	0.6	0.1	1.7	2.6	0.1	0.0	0.1
25.5	36.6	11.5	13.7	0.2	17.2	1.0	8.8	1.2	2.0	1.3	1.4	0.6	0.1	1.7	2.5	0.1	0.0	0.0
26.5	39.6	11.5	11.4	0.1	18.0	0.8	8.4	1.2	1.6	1.2	1.4	0.6	0.1	1.7	2.4	0.1	0.0	0.0
27.5	42.8	11.5	9.2	0.1	18.4	0.6	8.0	1.2	1.2	1.1	1.3	0.5	0.1	1.7	2.3	0.0	0.0	0.0
28.5	45.9	11.4	7.2	0.0	18.5	0.4	7.7	1.2	0.9	1.0	1.3	0.5	0.0	1.7	2.2	0.0	0.0	0.0
29.5	49.0	11.4	5.7	0.0	18.1	0.3	7.3	1.2	0.7	0.9	1.3	0.4	0.0	1.6	2.1	0.0	0.0	0.0
30.5	52.0	11.4	4.3	0.0	17.5	0.2	6.9	1.2	0.5	0.8	1.3	0.4	0.0	1.6	2.0	0.0	0.0	0.0
31.5	54.7	11.5	3.2	0.0	16.5	0.1	6.6	1.2	0.3	0.7	1.2	0.4	0.0	1.6	1.9	0.0	0.0	0.0
32.5	57.1	11.5	2.4	0.0	15.7	0.1	6.3	1.2	0.2	0.7	1.2	0.4	0.0	1.6	1.8	0.0	0.0	0.0
33.5	59.2	11.5	1.7	0.0	14.8	0.0	6.0	1.2	0.1	0.6	1.2	0.3	0.0	1.6	1.8	0.0	0.0	0.0
34.5	60.8	11.5	1.2	0.0	14.1	0.0	5.7	1.2	0.1	0.6	1.2	0.3	0.0	1.6	1.7	0.0	0.0	0.0
35.5	62.3	11.5	0.9	0.0	13.3	0.0	5.5	1.2	0.1	0.5	1.2	0.3	0.0	1.6	1.6	0.0	0.0	0.0
36.5	63.6	11.5	0.6	0.0	12.6	0.0	5.3	1.2	0.0	0.5	1.2	0.3	0.0	1.6	1.6	0.0	0.0	0.0
37.5	64.7	11.5	0.4	0.0	11.9	0.0	5.2	1.2	0.0	0.5	1.2	0.3	0.0	1.6	1.6	0.0	0.0	0.0
38.5	65.8	11.5	0.3	0.0	11.2	0.0	5.0	1.2	0.0	0.4	1.1	0.3	0.0	1.6	1.5	0.0	0.0	0.0
39.5	66.8	11.5	0.2	0.0	10.6	0.0	4.8	1.2	0.0	0.4	1.1	0.2	0.0	1.6	1.5	0.0	0.0	0.0
40.5	67.7	11.5	0.2	0.0	10.0	0.0	4.7	1.2	0.0	0.4	1.1	0.2	0.0	1.6	1.4	0.0	0.0	0.0
41.5	68.6	11.6	0.1	0.0	9.3	0.0	4.5	1.2	0.0	0.4	1.1	0.2	0.0	1.6	1.4	0.0	0.0	0.0
42.5	69.4	11.6	0.1	0.0	8.7	0.0	4.3	1.2	0.0	0.3	1.1	0.2	0.0	1.6	1.4	0.0	0.0	0.0
43.5	70.3	11.7	0.1	0.0	8.1	0.0	4.2	1.2	0.0	0.3	1.1	0.2	0.0	1.6	1.3	0.0	0.0	0.0
44.5	71.3	11.7	0.0	0.0	7.4	0.0	4.0	1.2	0.0	0.3	1.1	0.2	0.0	1.6	1.3	0.0	0.0	0.0
45.5	72.2	11.7	0.0	0.0	6.7	0.0	3.8	1.2	0.0	0.3	1.1	0.2	0.0	1.6	1.3	0.0	0.0	0.0
46.5	73.2	11.6	0.0	0.0	6.1	0.0	3.7	1.2	0.0	0.2	1.1	0.2	0.0	1.6	1.2	0.0	0.0	0.0
47.5	74.1	11.6	0.0	0.0	5.5	0.0	3.5	1.2	0.0	0.2	1.0	0.1	0.0	1.6	1.2	0.0	0.0	0.0
48.5	75.0	11.5	0.0	0.0	5.0	0.0	3.4	1.2	0.0	0.2	1.0	0.1	0.0	1.5	1.1	0.0	0.0	0.0
49.5	75.8	11.5	0.0	0.0	4.4	0.0	3.2	1.2	0.0	0.2	1.0	0.1	0.0	1.5	1.1	0.0	0.0	0.0
50.5	76.3	11.5	0.0	0.0	4.0	0.0	3.1	1.2	0.0	0.1	1.0	0.1	0.0	1.5	1.1	0.0	0.0	0.0
51.5	76.7	11.5	0.0	0.0	3.7	0.0	3.1	1.2	0.0	0.1	1.0	0.1	0.0	1.5	1.1	0.0	0.0	0.0
52.5	77.0	11.6	0.0	0.0	3.5	0.0	3.0	1.2	0.0	0.1	1.0	0.1	0.0	1.5	1.0	0.0	0.0	0.0
53.5	77.3	11.6	0.0	0.0	3.3	0.0	2.9	1.2	0.0	0.1	1.0	0.1	0.0	1.5	1.0	0.0	0.0	0.0

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

Table A2. The percentage contribution of each species to the total fluorine budget in the latitude bands between 30° N and 0° N.

Altitude	HF	CF ₂	CFC-12	CFC-11	COF ₂	COClF	HCFC-22	SF ₆	CFC-113	CFC-114	CFC-115	HCFC-142b	HCFC-141b	HFC-23	HFC-134a	HFC-152a	H-1211	H-1301
10.5	0.00	11.10	39.18	9.40	0.02	0.02	15.31	1.35	8.67	2.51	1.70	1.17	0.69	2.14	5.68	0.33	0.32	0.42
11.5	0.00	11.10	39.19	9.40	0.03	0.02	15.31	1.35	8.67	2.51	1.70	1.17	0.69	2.14	5.67	0.33	0.31	0.42
12.5	0.00	11.11	39.21	9.40	0.04	0.02	15.32	1.36	8.64	2.50	1.70	1.17	0.69	2.14	5.63	0.33	0.31	0.41
13.5	0.00	11.12	39.25	9.41	0.07	0.04	15.34	1.36	8.61	2.50	1.70	1.17	0.69	2.13	5.58	0.33	0.31	0.41
14.5	0.05	11.13	39.28	9.42	0.09	0.05	15.35	1.36	8.57	2.49	1.70	1.16	0.68	2.12	5.53	0.32	0.30	0.41
15.5	0.17	11.16	39.26	9.45	0.13	0.07	15.29	1.35	8.53	2.48	1.70	1.15	0.67	2.11	5.46	0.31	0.30	0.41
16.5	0.34	11.24	39.45	9.34	0.19	0.10	15.06	1.36	8.46	2.47	1.70	1.15	0.66	2.11	5.37	0.30	0.29	0.40
17.5	0.79	11.29	39.37	9.16	0.47	0.19	14.92	1.37	8.30	2.44	1.70	1.13	0.64	2.09	5.21	0.29	0.27	0.39
18.5	1.84	11.31	38.59	8.64	1.06	0.38	15.23	1.35	7.97	2.38	1.68	1.09	0.61	2.05	4.95	0.26	0.24	0.37
19.5	3.37	11.38	37.56	8.01	1.98	0.75	15.08	1.32	7.54	2.30	1.66	1.05	0.56	2.01	4.65	0.24	0.20	0.35
20.5	5.45	11.57	36.56	7.27	3.20	1.20	13.66	1.31	7.20	2.25	1.66	1.02	0.52	2.00	4.43	0.22	0.15	0.32
21.5	7.51	11.64	35.30	6.36	4.40	1.63	12.82	1.30	6.87	2.21	1.65	1.00	0.49	1.98	4.25	0.20	0.11	0.30
22.5	9.25	11.67	34.07	5.37	5.75	2.08	12.15	1.30	6.54	2.17	1.64	0.97	0.45	1.96	4.09	0.18	0.07	0.28
23.5	10.85	11.72	32.69	4.35	7.12	2.49	11.75	1.30	6.21	2.13	1.64	0.96	0.42	1.95	3.98	0.17	0.04	0.25
24.5	12.20	11.83	31.34	3.29	8.67	2.79	11.35	1.31	5.89	2.11	1.64	0.95	0.38	1.95	3.90	0.16	0.02	0.23
25.5	14.14	11.85	29.50	2.38	10.22	3.01	11.06	1.31	5.47	2.08	1.64	0.93	0.34	1.94	3.81	0.15	0.01	0.19
26.5	15.96	11.85	27.68	1.62	11.99	3.04	10.85	1.31	4.95	2.03	1.62	0.91	0.29	1.92	3.70	0.14	0.00	0.16
27.5	17.57	11.93	25.86	1.00	14.16	2.73	10.64	1.32	4.32	1.97	1.62	0.88	0.24	1.91	3.61	0.13	0.00	0.12
28.5	19.63	11.92	23.76	0.51	16.55	2.21	10.34	1.31	3.68	1.89	1.60	0.85	0.18	1.89	3.49	0.12	0.00	0.09
29.5	22.16	11.84	21.49	0.30	18.54	1.63	9.96	1.30	3.06	1.79	1.56	0.81	0.14	1.86	3.36	0.11	0.00	0.06
30.5	25.10	11.85	18.83	0.11	20.12	1.13	9.70	1.30	2.48	1.71	1.55	0.78	0.10	1.85	3.26	0.10	0.00	0.04
31.5	29.34	11.92	15.85	0.03	21.51	0.34	9.50	1.31	2.01	1.65	1.55	0.76	0.07	1.85	3.20	0.09	0.00	0.02
32.5	31.41	11.91	12.81	0.02	22.65	0.23	9.20	1.32	1.56	1.53	1.53	0.73	0.05	1.84	3.10	0.08	0.00	0.01
33.5	34.39	11.93	9.87	0.01	23.66	0.12	8.93	1.32	1.13	1.47	1.52	0.69	0.03	1.84	3.01	0.07	0.00	0.01
34.5	37.00	11.87	7.65	0.01	24.39	0.08	8.55	1.32	0.83	1.37	1.49	0.66	0.02	1.82	2.89	0.06	0.00	0.00
35.5	39.68	11.90	5.51	0.00	24.75	0.04	8.21	1.32	0.56	1.28	1.47	0.62	0.01	1.81	2.78	0.05	0.00	0.00
36.5	42.27	11.94	3.88	0.00	24.77	0.02	7.77	1.32	0.36	1.17	1.45	0.58	0.01	1.79	2.65	0.04	0.00	0.00
37.5	45.17	11.91	2.72	0.00	24.04	0.01	7.28	1.32	0.24	1.06	1.41	0.53	0.00	1.77	2.50	0.03	0.00	0.00
38.5	48.30	11.87	1.66	0.00	22.92	0.01	6.83	1.32	0.13	0.95	1.38	0.48	0.00	1.75	2.36	0.02	0.00	0.00
39.5	50.70	11.82	1.20	0.00	21.62	0.00	6.48	1.32	0.09	0.87	1.36	0.45	0.00	1.75	2.26	0.02	0.00	0.00
40.5	53.16	11.82	0.81	0.00	20.24	0.00	6.13	1.32	0.06	0.79	1.34	0.41	0.00	1.74	2.15	0.02	0.00	0.00
41.5	55.56	11.86	0.51	0.00	18.62	0.00	5.83	1.33	0.03	0.72	1.33	0.39	0.00	1.73	2.07	0.01	0.00	0.00
42.5	57.47	11.90	0.38	0.00	17.25	0.00	5.57	1.34	0.02	0.67	1.31	0.36	0.00	1.73	1.99	0.01	0.00	0.00
43.5	59.32	12.01	0.25	0.00	15.86	0.00	5.31	1.34	0.01	0.61	1.29	0.34	0.00	1.72	1.92	0.01	0.00	0.00
44.5	61.28	12.02	0.17	0.00	14.34	0.00	5.03	1.35	0.01	0.56	1.27	0.31	0.00	1.72	1.83	0.01	0.00	0.00
45.5	63.27	12.18	0.11	0.00	12.84	0.00	4.74	1.35	0.01	0.50	1.25	0.29	0.00	1.71	1.75	0.01	0.00	0.00
46.5	65.37	12.17	0.06	0.00	11.31	0.00	4.44	1.35	0.00	0.44	1.23	0.26	0.00	1.70	1.66	0.00	0.00	0.00
47.5	67.23	12.12	0.03	0.00	10.01	0.00	4.17	1.34	0.00	0.39	1.20	0.24	0.00	1.68	1.58	0.00	0.00	0.00
48.5	69.01	12.00	0.02	0.00	8.85	0.00	3.91	1.33	0.00	0.35	1.16	0.22	0.00	1.65	1.49	0.00	0.00	0.00
49.5	70.78	11.87	0.02	0.00	7.72	0.00	3.64	1.32	0.00	0.30	1.13	0.20	0.00	1.62	1.41	0.00	0.00	0.00
50.5	72.17	11.85	0.01	0.00	6.73	0.00	3.43	1.31	0.00	0.26	1.11	0.18	0.00	1.61	1.35	0.00	0.00	0.00
51.5	73.09	11.81	0.01	0.00	6.10	0.00	3.28	1.31	0.00	0.24	1.09	0.17	0.00	1.60	1.30	0.00	0.00	0.00
52.5	73.77	11.81	0.00	0.00	5.62	0.00	3.17	1.31	0.00	0.22	1.08	0.16	0.00	1.59	1.27	0.00	0.00	0.00
53.5	74.48	11.79	0.00	0.00	5.14	0.00	3.06	1.31	0.00	0.20	1.06	0.15	0.00	1.58	1.23	0.00	0.00	0.00

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

⏴

⏵

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Global stratospheric
fluorine inventories
for 2004–2009

A. T. Brown et al.

Table A3. The percentage contribution of each species to the total fluorine budget in the latitude bands between 0° N and 30° S.

Altitude	HF	CF ₂	CFC-12	CFC-11	COF ₂	COClF	HCFC-22	SF ₆	CFC-113	CFC-114	CFC-115	HCFC-142b	HCFC-141b	HFC-23	HFC-134a	HFC-152a	H-1211	H-1301
10.5	0.02	11.08	39.31	9.42	0.03	0.02	15.08	1.36	8.69	2.51	1.70	1.18	0.70	2.15	5.69	0.34	0.32	0.42
11.5	0.04	11.08	39.31	9.42	0.03	0.02	15.08	1.36	8.69	2.51	1.70	1.18	0.70	2.14	5.68	0.34	0.32	0.42
12.5	0.08	11.08	39.31	9.43	0.04	0.02	15.08	1.35	8.67	2.51	1.70	1.17	0.69	2.14	5.66	0.33	0.31	0.42
13.5	0.14	11.08	39.31	9.43	0.06	0.03	15.08	1.35	8.64	2.50	1.70	1.17	0.69	2.14	5.63	0.33	0.31	0.41
14.5	0.21	11.08	39.31	9.43	0.08	0.03	15.08	1.35	8.61	2.50	1.70	1.17	0.69	2.13	5.59	0.33	0.31	0.41
15.5	0.39	11.11	39.26	9.46	0.13	0.05	15.01	1.35	8.56	2.48	1.70	1.16	0.68	2.12	5.52	0.32	0.30	0.41
16.5	0.75	11.17	39.27	9.32	0.22	0.09	14.90	1.35	8.45	2.46	1.70	1.15	0.66	2.11	5.40	0.31	0.29	0.40
17.5	1.32	11.22	39.14	9.14	0.52	0.18	14.80	1.36	8.23	2.42	1.69	1.12	0.64	2.08	5.20	0.29	0.27	0.39
18.5	2.35	11.29	38.43	8.64	1.26	0.41	15.00	1.34	7.82	2.33	1.66	1.07	0.60	2.04	4.89	0.26	0.23	0.37
19.5	3.95	11.40	37.38	7.95	2.25	0.80	14.78	1.32	7.35	2.24	1.64	1.02	0.55	2.01	4.58	0.24	0.19	0.34
20.5	6.10	11.57	36.26	7.20	3.57	1.20	13.43	1.30	7.01	2.19	1.64	0.99	0.51	1.99	4.36	0.22	0.15	0.31
21.5	7.84	11.87	34.91	6.30	4.84	1.69	12.68	1.28	6.74	2.15	1.64	0.97	0.48	1.98	4.21	0.20	0.11	0.30
22.5	9.77	11.66	33.63	5.31	6.16	2.04	12.03	1.27	6.43	2.12	1.63	0.95	0.45	1.95	4.07	0.19	0.07	0.27
23.5	11.59	11.70	32.07	4.30	7.49	2.39	11.70	1.27	6.10	2.08	1.62	0.94	0.41	1.94	3.95	0.17	0.04	0.25
24.5	13.51	11.81	30.46	3.19	9.19	2.61	11.09	1.28	5.70	2.05	1.63	0.92	0.37	1.94	3.85	0.16	0.02	0.22
25.5	15.91	11.83	27.99	2.25	11.20	2.74	10.78	1.28	5.18	2.00	1.61	0.90	0.32	1.92	3.73	0.15	0.01	0.18
26.5	18.30	11.87	25.72	1.47	13.04	2.70	10.53	1.28	4.60	1.94	1.60	0.87	0.27	1.91	3.61	0.14	0.00	0.15
27.5	20.27	11.84	23.42	0.92	15.40	2.36	10.30	1.28	4.01	1.88	1.59	0.85	0.22	1.89	3.52	0.13	0.00	0.11
28.5	22.27	11.85	21.28	0.49	17.46	1.92	10.09	1.27	3.45	1.82	1.58	0.83	0.17	1.88	3.43	0.12	0.00	0.08
29.5	24.33	11.83	18.94	0.30	19.49	1.50	9.83	1.27	2.92	1.75	1.56	0.80	0.14	1.86	3.33	0.11	0.00	0.06
30.5	26.93	11.94	16.29	0.11	20.94	1.01	9.68	1.28	2.42	1.70	1.56	0.78	0.10	1.86	3.27	0.10	0.00	0.04
31.5	29.50	12.10	13.67	0.04	22.15	0.58	9.49	1.29	1.97	1.63	1.56	0.76	0.07	1.87	3.21	0.09	0.00	0.02
32.5	32.08	12.20	10.98	0.02	23.31	0.39	9.23	1.30	1.52	1.55	1.55	0.73	0.05	1.88	3.11	0.08	0.00	0.01
33.5	34.87	12.31	8.34	0.01	24.22	0.21	8.94	1.32	1.09	1.46	1.55	0.69	0.03	1.88	3.01	0.07	0.00	0.01
34.5	37.17	12.27	6.52	0.00	24.79	0.14	8.58	1.31	0.82	1.37	1.52	0.65	0.02	1.86	2.90	0.06	0.00	0.00
35.5	39.92	12.17	4.74	0.00	25.07	0.08	8.14	1.30	0.56	1.26	1.49	0.61	0.01	1.83	2.76	0.05	0.00	0.00
36.5	42.97	12.02	3.41	0.00	24.59	0.04	7.67	1.29	0.38	1.15	1.45	0.57	0.01	1.80	2.62	0.04	0.00	0.00
37.5	45.81	11.94	2.50	0.00	23.60	0.03	7.26	1.28	0.27	1.05	1.42	0.53	0.00	1.78	2.49	0.03	0.00	0.00
38.5	48.39	11.96	1.69	0.00	22.51	0.01	6.92	1.29	0.17	0.97	1.40	0.49	0.00	1.77	2.39	0.03	0.00	0.00
39.5	51.08	11.91	1.23	0.00	21.11	0.01	6.50	1.29	0.12	0.88	1.37	0.45	0.00	1.76	2.27	0.02	0.00	0.00
40.5	53.67	11.97	0.80	0.00	19.58	0.01	6.12	1.30	0.07	0.79	1.35	0.41	0.00	1.76	2.16	0.02	0.00	0.00
41.5	56.50	11.98	0.50	0.00	17.76	0.00	5.71	1.30	0.04	0.70	1.32	0.37	0.00	1.74	2.04	0.01	0.00	0.00
42.5	59.12	12.09	0.34	0.00	15.83	0.00	5.35	1.31	0.03	0.62	1.30	0.34	0.00	1.73	1.93	0.01	0.00	0.00
43.5	61.51	12.27	0.19	0.00	13.92	0.00	4.99	1.32	0.01	0.55	1.28	0.31	0.00	1.73	1.82	0.01	0.00	0.00
44.5	63.44	12.45	0.12	0.00	12.42	0.00	4.72	1.33	0.01	0.49	1.26	0.28	0.00	1.73	1.74	0.01	0.00	0.00
45.5	65.61	12.39	0.08	0.00	10.90	0.00	4.41	1.32	0.00	0.43	1.23	0.26	0.00	1.71	1.65	0.00	0.00	0.00
46.5	67.50	12.42	0.04	0.00	9.50	0.00	4.13	1.33	0.00	0.38	1.21	0.23	0.00	1.70	1.56	0.00	0.00	0.00
47.5	69.14	12.36	0.02	0.00	8.36	0.00	3.89	1.32	0.00	0.33	1.18	0.21	0.00	1.68	1.49	0.00	0.00	0.00
48.5	70.41	12.22	0.02	0.00	7.59	0.00	3.71	1.31	0.00	0.30	1.15	0.20	0.00	1.66	1.43	0.00	0.00	0.00
49.5	71.66	12.07	0.01	0.00	6.84	0.00	3.53	1.29	0.00	0.27	1.13	0.18	0.00	1.63	1.38	0.00	0.00	0.00
50.5	72.78	11.95	0.01	0.00	6.15	0.00	3.37	1.28	0.00	0.25	1.10	0.17	0.00	1.61	1.33	0.00	0.00	0.00
51.5	73.61	11.82	0.00	0.00	5.70	0.00	3.25	1.26	0.00	0.23	1.08	0.16	0.00	1.59	1.29	0.00	0.00	0.00
52.5	74.01	11.82	0.00	0.00	5.40	0.00	3.19	1.26	0.00	0.22	1.08	0.16	0.00	1.59	1.27	0.00	0.00	0.00
53.5	74.52	11.77	0.00	0.00	5.08	0.00	3.11	1.26	0.00	0.20	1.06	0.15	0.00	1.58	1.25	0.00	0.00	0.00



Global stratospheric
fluorine inventories
for 2004–2009

A. T. Brown et al.

Table A4. The percentage contribution of each species to the total fluorine budget in the latitude bands between 30° S and 70° S.

Altitude	HF	CF4	CFC-12	CFC-11	COF ₂	COClF	HCFC-22	SF6	CFC-113	CFC-114	CFC-115	HCFC-142b	HCFC-141b	HFC-23	HFC-134a	HFC-152a	H-1211	H-1301
10.5	1.42	11.41	38.49	8.94	0.87	0.25	14.01	1.33	8.55	2.50	1.74	1.16	0.67	2.16	5.49	0.31	0.30	0.41
11.5	2.56	11.41	37.85	8.71	1.52	0.42	13.98	1.33	8.13	2.41	1.71	1.11	0.63	2.11	5.17	0.29	0.27	0.38
12.5	3.82	11.39	37.18	8.46	2.21	0.60	13.96	1.33	7.66	2.31	1.67	1.06	0.59	2.06	4.84	0.26	0.24	0.36
13.5	5.20	11.37	36.29	8.17	2.86	0.71	13.90	1.32	7.30	2.22	1.65	1.02	0.55	2.02	4.61	0.24	0.22	0.34
14.5	7.06	11.33	34.96	7.71	3.70	0.82	13.72	1.31	6.95	2.14	1.62	0.98	0.52	1.98	4.41	0.23	0.20	0.32
15.5	9.50	11.34	33.28	7.02	4.85	0.95	13.21	1.30	6.59	2.07	1.60	0.94	0.49	1.95	4.21	0.21	0.18	0.30
16.5	12.41	11.45	31.39	6.14	6.31	1.12	12.24	1.28	6.19	1.98	1.58	0.90	0.46	1.93	3.98	0.19	0.16	0.28
17.5	15.88	11.58	29.11	5.13	7.97	1.30	11.25	1.26	5.68	1.88	1.56	0.85	0.41	1.90	3.69	0.17	0.13	0.25
18.5	19.69	11.62	26.45	4.00	9.94	1.46	10.53	1.24	5.03	1.74	1.52	0.79	0.35	1.85	3.34	0.14	0.10	0.22
19.5	23.10	11.64	23.95	2.91	11.82	1.57	10.04	1.21	4.41	1.62	1.49	0.73	0.30	1.80	3.04	0.12	0.06	0.18
20.5	26.29	11.68	21.57	1.90	13.36	1.62	9.67	1.19	3.90	1.52	1.46	0.68	0.25	1.78	2.82	0.10	0.04	0.15
21.5	29.06	11.65	19.22	1.16	14.92	1.54	9.38	1.18	3.47	1.45	1.44	0.65	0.22	1.75	2.68	0.09	0.02	0.13
22.5	31.47	11.60	16.93	0.78	16.52	1.32	9.09	1.17	3.03	1.38	1.42	0.62	0.18	1.73	2.57	0.08	0.01	0.11
23.5	33.88	11.59	14.80	0.52	17.71	1.07	8.85	1.17	2.59	1.32	1.40	0.60	0.15	1.72	2.48	0.07	0.00	0.08
24.5	36.17	11.58	12.78	0.31	18.73	0.89	8.63	1.16	2.15	1.26	1.39	0.58	0.11	1.71	2.42	0.06	0.00	0.06
25.5	38.28	11.55	10.91	0.16	19.67	0.72	8.42	1.16	1.75	1.20	1.38	0.56	0.08	1.70	2.37	0.06	0.00	0.04
26.5	40.38	11.48	9.20	0.08	20.35	0.58	8.19	1.15	1.40	1.15	1.36	0.54	0.06	1.69	2.32	0.05	0.00	0.03
27.5	42.62	11.47	7.55	0.04	20.67	0.44	7.95	1.15	1.11	1.08	1.34	0.51	0.04	1.68	2.26	0.04	0.00	0.02
28.5	44.89	11.49	6.07	0.02	20.72	0.32	7.70	1.15	0.85	1.02	1.33	0.49	0.03	1.67	2.21	0.04	0.00	0.01
29.5	47.13	11.53	4.89	0.01	20.34	0.23	7.45	1.15	0.66	0.96	1.32	0.47	0.02	1.67	2.15	0.03	0.00	0.01
30.5	49.47	11.60	3.82	0.00	19.69	0.15	7.18	1.15	0.48	0.89	1.31	0.44	0.01	1.67	2.09	0.03	0.00	0.00
31.5	51.71	11.68	2.89	0.00	18.95	0.08	6.91	1.16	0.34	0.83	1.29	0.42	0.01	1.67	2.03	0.02	0.00	0.00
32.5	53.81	11.69	2.19	0.00	18.18	0.05	6.61	1.16	0.24	0.76	1.27	0.39	0.01	1.66	1.96	0.02	0.00	0.00
33.5	55.97	11.65	1.57	0.00	17.33	0.03	6.29	1.15	0.16	0.70	1.25	0.37	0.00	1.64	1.88	0.02	0.00	0.00
34.5	57.99	11.62	1.13	0.00	16.32	0.01	6.01	1.15	0.10	0.64	1.23	0.34	0.00	1.63	1.81	0.01	0.00	0.00
35.5	59.87	11.60	0.81	0.00	15.21	0.01	5.76	1.14	0.07	0.59	1.21	0.32	0.00	1.62	1.76	0.01	0.00	0.00
36.5	61.64	11.57	0.57	0.00	14.15	0.01	5.51	1.14	0.05	0.55	1.19	0.30	0.00	1.61	1.70	0.01	0.00	0.00
37.5	63.26	11.54	0.42	0.00	13.13	0.00	5.26	1.14	0.03	0.51	1.18	0.28	0.00	1.60	1.64	0.01	0.00	0.00
38.5	64.73	11.53	0.29	0.00	12.17	0.00	5.03	1.14	0.02	0.47	1.16	0.27	0.00	1.60	1.59	0.01	0.00	0.00
39.5	66.07	11.55	0.20	0.00	11.25	0.00	4.81	1.14	0.01	0.43	1.15	0.25	0.00	1.60	1.53	0.01	0.00	0.00
40.5	67.28	11.59	0.15	0.00	10.38	0.00	4.60	1.15	0.01	0.39	1.14	0.23	0.00	1.60	1.48	0.00	0.00	0.00
41.5	68.44	11.64	0.10	0.00	9.52	0.00	4.40	1.16	0.01	0.36	1.13	0.22	0.00	1.60	1.44	0.00	0.00	0.00
42.5	69.44	11.69	0.06	0.00	8.76	0.00	4.22	1.16	0.00	0.33	1.12	0.20	0.00	1.60	1.39	0.00	0.00	0.00
43.5	70.51	11.75	0.04	0.00	7.97	0.00	4.03	1.16	0.00	0.30	1.11	0.19	0.00	1.60	1.34	0.00	0.00	0.00
44.5	71.53	11.79	0.03	0.00	7.20	0.00	3.85	1.17	0.00	0.27	1.09	0.18	0.00	1.59	1.30	0.00	0.00	0.00
45.5	72.48	11.77	0.02	0.00	6.55	0.00	3.69	1.16	0.00	0.25	1.08	0.17	0.00	1.58	1.26	0.00	0.00	0.00
46.5	73.48	11.72	0.01	0.00	5.89	0.00	3.52	1.16	0.00	0.22	1.06	0.15	0.00	1.57	1.21	0.00	0.00	0.00
47.5	74.41	11.65	0.01	0.00	5.30	0.00	3.37	1.15	0.00	0.20	1.04	0.14	0.00	1.55	1.17	0.00	0.00	0.00
48.5	75.02	11.57	0.00	0.00	4.94	0.00	3.28	1.14	0.00	0.19	1.03	0.14	0.00	1.54	1.15	0.00	0.00	0.00
49.5	75.59	11.49	0.00	0.00	4.60	0.00	3.20	1.13	0.00	0.17	1.01	0.13	0.00	1.53	1.13	0.00	0.00	0.00
50.5	75.92	11.52	0.00	0.00	4.32	0.00	3.15	1.14	0.00	0.16	1.01	0.13	0.00	1.53	1.12	0.00	0.00	0.00
51.5	76.17	11.54	0.00	0.00	4.11	0.00	3.11	1.14	0.00	0.15	1.01	0.13	0.00	1.53	1.11	0.00	0.00	0.00
52.5	76.38	11.55	0.00	0.00	3.94	0.00	3.08	1.14	0.00	0.15	1.00	0.12	0.00	1.53	1.10	0.00	0.00	0.00
53.5	76.63	11.55	0.00	0.00	3.76	0.00	3.04	1.14	0.00	0.14	1.00	0.12	0.00	1.53	1.09	0.00	0.00	0.00

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

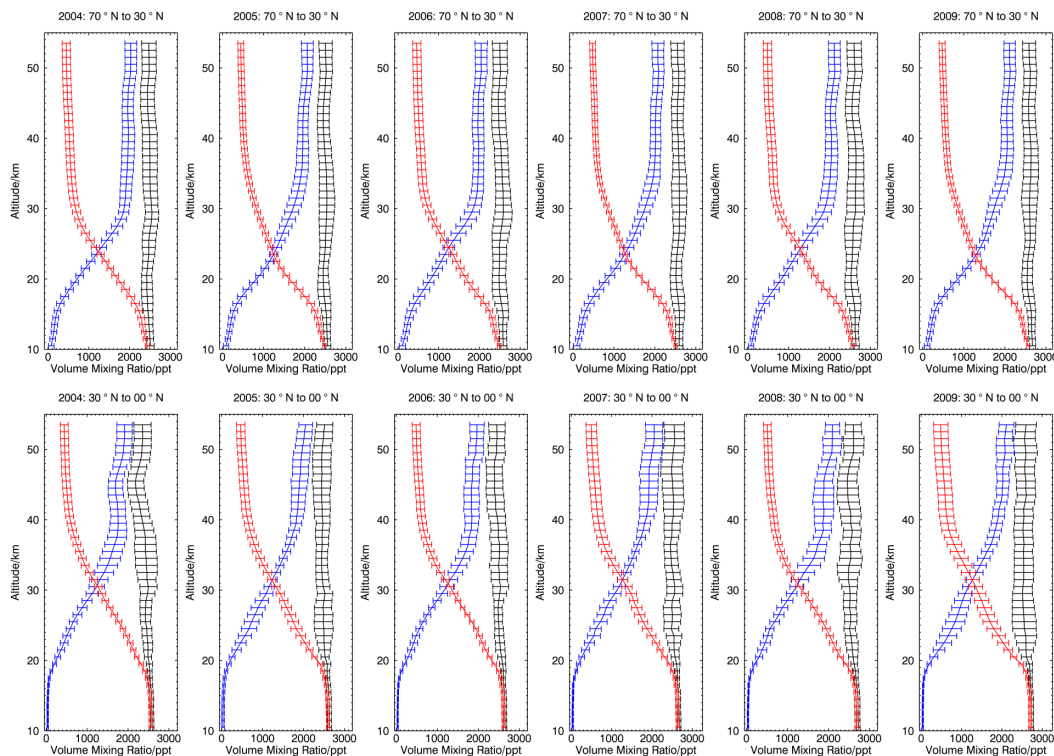


Fig. 1. The fluorine budgets from 2004 to 2009 in latitude band 30–70° N and 0–30° N (black). The inorganic (blue) and organic (red) fluorine profiles are also shown. The error bars are the result of combinations of standard deviations of the mean profiles of the relevant fluorine containing species.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

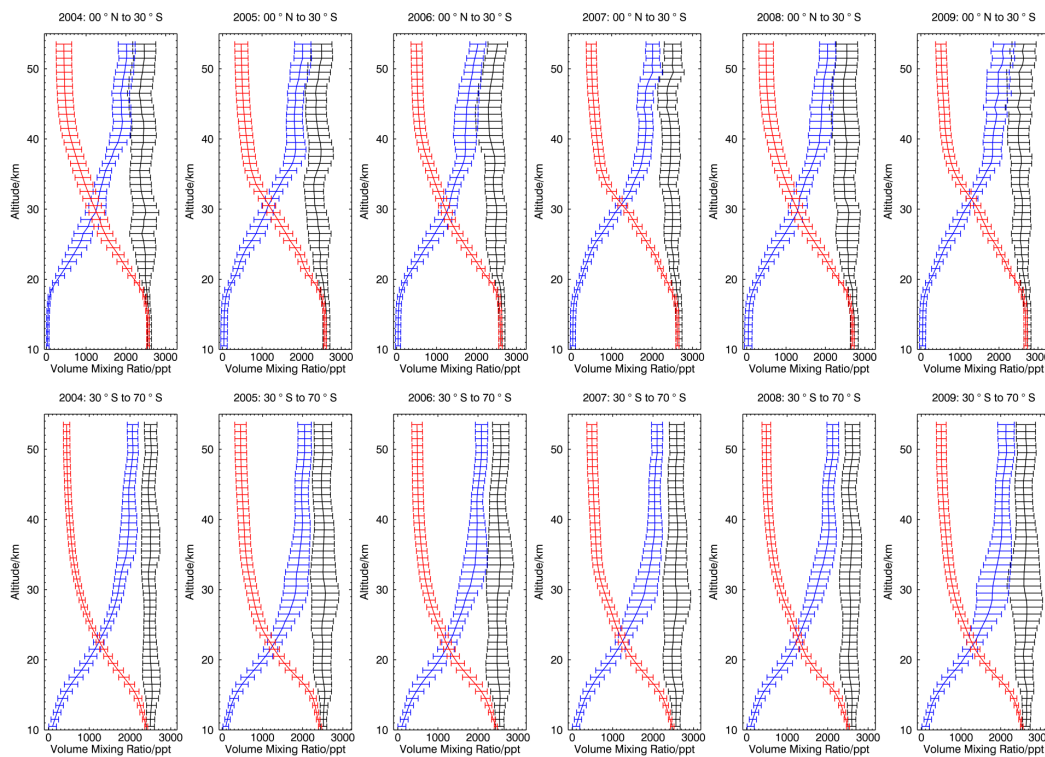


Fig. 2. As Fig. 1 but for latitude bands 0–30° S and 30–70° S.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

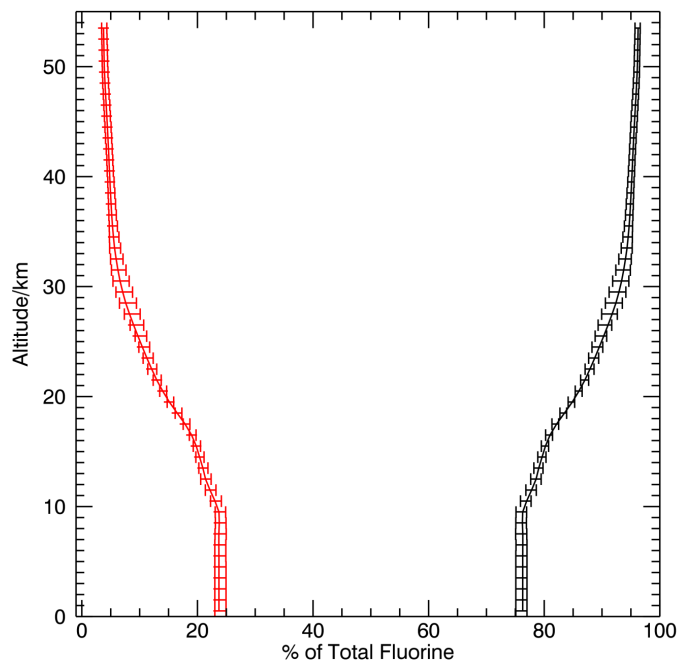


Fig. 3. The mean percentage contribution of ACE-FTS (black) and SLIMCAT (red) to the total fluorine VMR for all years between 70–30°N. The error bars are the maximum or minimum single contribution to the total fluorine percentage from these years.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Global stratospheric fluorine inventories for 2004–2009

A. T. Brown et al.

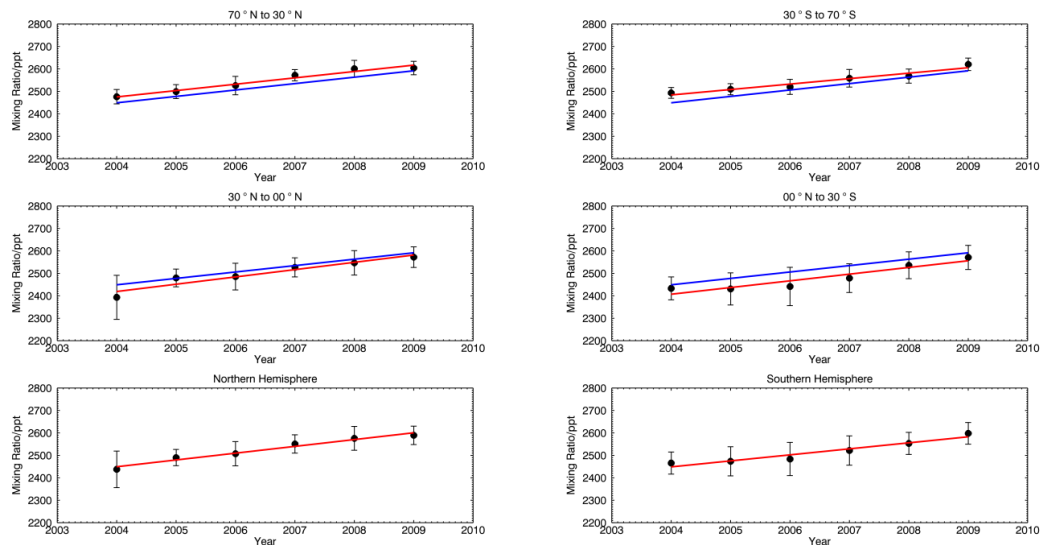


Fig. 4. The trends in the mean total stratospheric fluorine (ppt) between 2004 and 2009. The red line represents the line of best fit for the mean stratospheric total fluorine volume mixing ratio (the black circles). The blue line is the global trend in mean total stratospheric fluorine. The error bars shown in the plots are calculated from the standard deviation of the data used to calculate the mean total fluorine VMRs.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Global stratospheric
fluorine inventories
for 2004–2009

A. T. Brown et al.

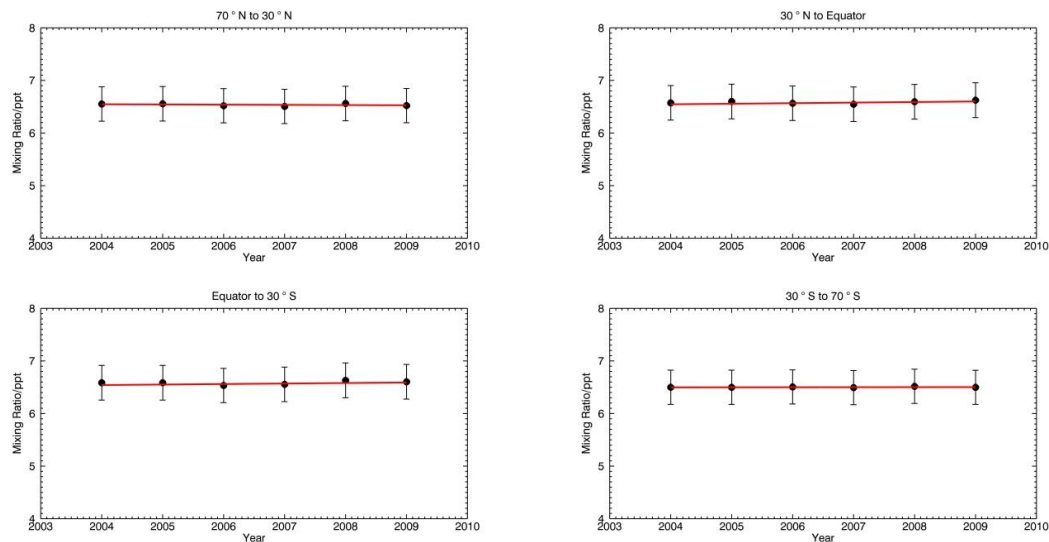


Fig. 5. The trends in the GWP-weighted total fluorine mean between 2004 and 2009. The red line represents the line of best fit for the GWP-weighted mean volume mixing ratio (the black circles). The error bars shown in the plots are calculated from the weighted standard deviation of the data used to calculate the means.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)