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Global stratospheric fluorine inventory for 2004–2009 from Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) measurements and SLIMCAT model simulations

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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 3-D chemical transport model. The budgets are calculated between 2004 and 2009 for a number of latitude bands: 70–30° N, 30–00° N, 00° N–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 hydrogen fluoride (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 calculation of radiative efficiency-weighted total fluorine allows the changes in radiative forcing between 2004 and 2009 to be calculated. These results show an increase in radiative forcing of between 0.23 ± 0.11 % per year and 0.45 ± 0.11 % per year, due to the increase in fluorine-containing species during this time. The decreasing trends in the mixing ratios of halons and chlorofluorocarbons (CFCs), due to their prohibition under the Montreal Protocol, have suppressed an increase in total fluorine caused by increasing mixing ratios of hydrofluorocarbons (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, nontoxic 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 therefore 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,

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Table 1. The global warming potential, on a 20 yr timescale (Solomon et al., 2007), the radiative efficiencies (Solomon et al., 2007) and the lifetimes (Montzka et al., 2011) of some of the species used in this work.

Species	Global warming potential	Radiative efficiency (Wm^{-2} ppb^{-1})	Stratospheric lifetime (years)
CF_4	5210	0.10	> 50 000
CFC-11 (CCl_3F)	6730	0.25	45
CFC-12 (CCl_2F_2)	11 000	0.32	100
CFC-113 ($C_2Cl_3F_3$)	6540	0.30	85
CFC-114 ($C_2Cl_2F_4$)	8040	0.31	190
CFC-115 (C_2ClF_5)	5310	0.18	1020
Halon-1301 ($CBrF_3$)	8480	0.32	65
Halon-1211 ($CBrClF_2$)	4750	0.30	–
HCFC-22 ($CHClF_2$)	5160	0.20	186
HCFC-141b ($C_2H_3Cl_2F$)	2250	0.14	64.9
HCFC-142b ($C_2H_3ClF_2$)	5490	0.20	160
HFC-23 (CHF_3)	12 000	0.19	2347
HFC-134a ($C_2H_2F_4$)	3830	0.16	232
HFC-152a ($C_2H_4F_2$)	437	0.09	45.4
SF_6	16 300	0.52	3200

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 et al., 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 20 yr global warming potential of 12 000 relative to that of CO_2 (Solomon et al., 2007). 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. Differences between observed and model budgets would suggest that there are additional species contributing to the total fluorine budget which have not been considered. 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 contain both chlorine and fluorine (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.

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 CF_4, CFC-11, CFC-12, COF_2, HCFC-22, HF and SF_6 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 CF_4, CFC-11, CFC-12, CFC-113, COF_2, HCFC-22, HF and SF_6 (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 CF_4, CFC-11, CFC-12, CFC-113, COClF, COF_2, HCFC-22, HCFC-141b, HCFC-142b, HFC-134a, HF and SF_6. When considered together these works show increasing stratospheric fluorine volume mixing ratios between 1985 and 2004. Total column measurements of COF_2 and HF from ground-based Fourier Transform Infrared 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). Additionally, FTIR measurements of total column abundances from the Network for the Detection of Atmospheric Composition Change (NDACC) have shown an increase in the HF column between 2000 and 2009 (Kohlhepp et al., 2012).

This paper presents fluorine budgets calculated from ACE-FTS measurements supplemented with output from the SLIMCAT 3-D 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 four 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 recent 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 three 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. In addition to this, the radiative efficiency-weighted trend in total fluorine is calculated showing the impact of the changing VMR of fluorine on radiative forcing between 2004 and 2009.

Section 2 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 3-D chemical transport model (CTM) (for a full description of the model readers are directed to Chipperfield, 2006) is given in Sect. 3 and stratospheric fluorine chemistry is discussed in Sect. 4. Section 5 outlines our method for calculating the fluorine budget. The results of this work are presented and discussed in Sect. 6.

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 ($0.02\,\mathrm{cm}^{-1}$) FTS which operates between 750 and $4400\,\mathrm{cm}^{-1}$. 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. 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 ninth year of operation with routine data being available from February 2004.

The ACE-FTS currently measures 8 fluorine-containing species, CF_4, CFC-12 (CCl_2F_2), CFC-11 (CCl_3F), COF_2, COClF, HCFC-22 (CHF_2Cl), HF and SF_6 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 ($C_2Cl_3F_3$), HCFC-142b ($C_2H_3ClF_2$), HCFC-141b ($C_2H_3Cl_2F$) (Brown et al., 2011) and HFC-23 (CHF_3) (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 3-D 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.

Table 2. The retrieval altitudes of the fluorine-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 ($CHClF_2$)	5–30	7–30
COClF	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

Fluorine-containing species retrieved by the Atmospheric Chemistry Experiment

Carbon tetrafluoride – CF_4

CF_4 is a powerful greenhouse gas (see Table 1) which has a lifetime of over 50 000 yr (Montzka et al., 2010), 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 MkIV balloon profiles. This study found that profiles from MkIV and ACE were within ±10 % (Velazco et al., 2011).

Sulfur hexafluoride – SF_6

SF_6 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 $950\,\mathrm{cm}^{-1}$ are responsible for making this molecule a potent greenhouse gas. ACE-FTS retrievals of SF_6 were validated by comparison with non-coincident MkIV balloon profiles. This study found an agreement within ±15 % between 12 and 19 km (Velazco et al., 2011).

CFC-11 – CCl_3F

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 MkIV profiles show agreement to 10 % above 12 km and 20 % below 12 km (Mahieu et al., 2008). Non-coincident MkIV balloon profiles were also used for validation, producing differences of less than ±20 % between 17 km and 24 km (Velazco et al., 2011).

CFC-12 – CCl_2F_2

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 MkIV instrument. This study also found differences of around ±10 % (Velazco et al., 2011).

HCFC-22 – $CHClF_2$

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.

Carbonyl chlorofluoride – COClF

COClF 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).

Carbonyl fluoride – COF_2

COF_2 is produced from the decomposition of molecules which contain two fluorine atoms. Comparisons with measurements from MkIV show an agreement to within ±20 % (Velazco et al., 2011).

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 MkIV and ground-based FTIR to ACE HF. These results showed a difference of ±10 % between ACE and MKIV above 19 km and an average difference of ±7.4 % for the average partial column averages from ground-based FTIRs.

3 SLIMCAT 3-D 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° × 5.6° 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 four latitude bins (70–30° N, 30–00° N, 00° N–30° S and 30–70° S) on a 1 km altitude grid.

Results from SLIMCAT have been included in many previous studies which have looked at the trends of stratospheric gases or have compared model profiles which depend on stratospheric loss rates or transport timescales. Monge-Sanz et al. (2007) showed that the model produced a realistic stratospheric Brewer–Dobson circulation and therefore could model the transport of tropospheric source gases through the stratosphere. Brown et al. (2011) showed that the model reproduced the observed profiles of fluorine-containing species in that stratosphere. Kohlhepp et al. (2012) showed that when forced with ERA-Interim analyses the model performed well in reproducing the ground-based observations on column HF, which is a measure of total fluorine in the atmosphere.

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.

4.1 CFCs and HCFCs which contain two fluorine atoms (COF_2 formation)

The source gas molecule will first be broken down into CF_2Cl and will then follow the following path (Tressaud, 2006):

$$CF_2Cl+O_2+M \rightarrow CF_2ClO_2+M$$

$$CF_2ClO_2+NO \rightarrow CF_2ClO+NO_2$$

$$CF_2ClO+O_2 \rightarrow COF_2+ClO_2$$

$$COF_2+hv \rightarrow FCO+F.$$

Above around 30 km the concentration of COF_2 decreases as photolysis becomes more effective. The primary source of COF_2 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 COF_2 has been increasing in recent years (Duchatelet et al., 2009; Brown et al., 2011).

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

The source gas molecule will first be broken down into $CFCl_2$ and will then follow the following path (Tressaud, 2006):

$$CFCl_2+O_2+M \rightarrow CFCl_2O_2+M \tag{R1}$$

$$CFCl_2O_2+NO \rightarrow CFCl_2O+NO_2 \tag{R2}$$

$$CFCl_2O+O_2 \rightarrow COClF+ClO_2 \tag{R3}$$

$$CFClO+hv \rightarrow FCO+Cl. \tag{R4}$$

Carbonyl chlorofluoride (COClF) 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 COClF is CFC-11.

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

$$FCO+O_2 \rightarrow FC(O)O_2 \rightarrow FO+CO_2 \tag{R5}$$

$$FO+O\left(^1D\right) \rightarrow F+O_2. \tag{R6}$$

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.

$$F+CH_4 \rightarrow HF+CH_3 \tag{R7}$$

$$F+H_2O \rightarrow HF+OH \tag{R8}$$

$$F+H_2 \rightarrow HF+H \tag{R9}$$

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.

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° N to 30° N – Northern Hemisphere extra-tropics, 30° N to 0° N – Northern Hemisphere tropics, 0° N to 30° S – Southern Hemisphere tropics – and 30° S to 70° S – Southern Hemisphere extra tropics). 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. SF_6 and CF_4 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 this calculation are given below, where square brackets indicate volume mixing ratio (VMR). Following the convention of previous

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

Year	70–30° N	30–00° N	00–30° 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

fluorine budgets (Zander et al., 1992; Nassar et al., 2006b), 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 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 SF_6).

$$[\mathrm{TotalFluorine}] = [\mathrm{TotalInorganicFluorine}]$$
$$+[\mathrm{TotalOrganicFluorine}]$$

$$[\mathrm{TotalInorganicFluorine}] = [\mathrm{HF}] + 2[\mathrm{COF_2}] + [\mathrm{COClF}]$$
$$+6[\mathrm{SF_6}]$$

$$[\mathrm{TotalOrganicFluorine}] = 4[\mathrm{CF_4}]+2[\mathrm{CFC-12}]+[\mathrm{CFC-11}]$$
$$+2[\mathrm{HCFC-22}] + 3[\mathrm{CFC-113}] + 4[\mathrm{CFC-114}]$$
$$+5[\mathrm{CFC-115}] + 2[\mathrm{HCFC-142b}] + [\mathrm{HCFC-141b}]$$
$$+3[\mathrm{HFC-23}] + 4[\mathrm{HFC-134a}] + 2[\mathrm{HFC-152a}]$$
$$+2[\mathrm{H-1211}] + 3[\mathrm{H-1301}]$$

6 Results and discussion

This section is divided into 5 subsections. Section 6.1 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. 6.2. The impacts of the individual species included in the total fluorine calculations on the total fluorine VMR are presented in Sect. 6.3. The final two sections present the changes in total fluorine between 2004 and 2009. Section 6.4 presents the changes in total stratospheric fluorine while Sect. 6.5 discusses the implication of these changes on climate forcing using a weighted total fluorine budget. The radiative impact of the total fluorine was evaluated by weighting the total fluorine VMR by the radiative efficiency of each species.

6.1 Vertical profiles

The results of the total fluorine calculations are shown in Figs. 1 and 2. As described in Section 6 each point on the mean vertical profile of a species retrieved by ACE has an error attached to it, which represents the standard deviation of the data at that altitude. The error bars shown on the plots represent the linear combination of the standard deviations of the contributing species for that particular altitude. It should be noted that the systematic retrieval errors associated with each ACE-FTS species are not known at this time and as such are not included in the error values presented in this work. 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 (Waugh and Hall, 2002), 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. 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.

Table 4. The slopes of the total fluorine profile and the mean stratospheric volume mixing ratio of the fluorine budgets for different latitude bands. The error quoted here is the 1-σ error of the fit. Systematic errors in the ACE-FTS retrieval of these species are not known at this time and are not included in these errors.

Year	70–30° N		30–00° 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	**00° N–30° S**		**30–70° 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

6.2 Fluorine source and reservoir species correlations

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, COF_2 and COClF are considered to be reservoir species; all other 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) are 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 SF_6) have been included in the study. Instead this method is a test whether the major source species which decompose in the stratosphere and reservoir species have been included in the budget. Correlations were calculated by summing the relevant species (see above) to produce source and reservoir species profiles. This produced a profile with a value for the VMR of reservoir and source species every 1 km between the tropopause and 53.5 km. These VMRs were plotted against one another for each altitude in order that a correlation could be calculated. 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. 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 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.

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.

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 COClF). Once more, the error given here is the 1-σ error of a least-squares fit.

Year	70–30° N	30–00° N	00° 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

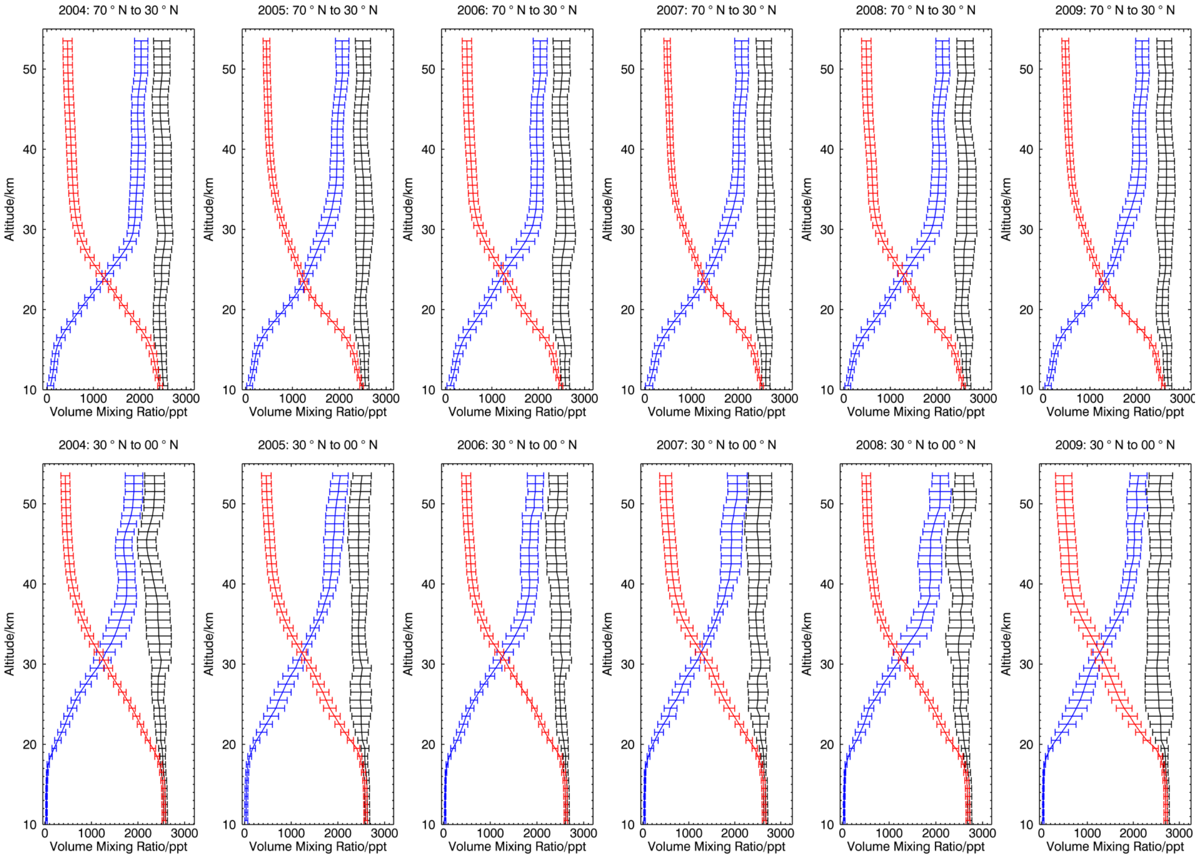

Fig. 1. The fluorine budgets from 2004 to 2009 in latitude band 30–70° N and 0–30° N (black). The inorganic (blue) and organic (red) fluorine profiles are also shown. The error bars are the result of combinations of standard deviations of the mean profiles of the relevant fluorine-containing species. Systematic retrieval errors are not known at this time and therefore are not included in these errors.

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.

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

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
	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

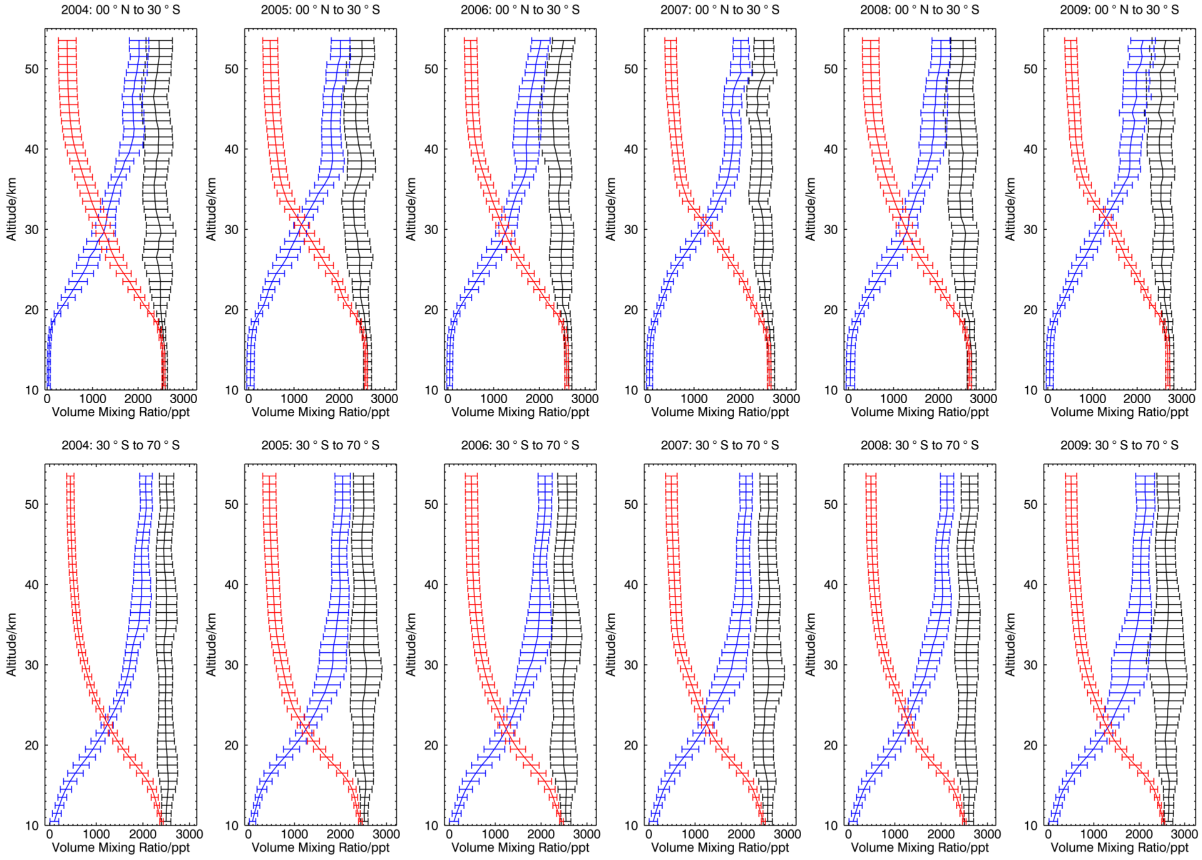

Fig. 2. As Fig. 1 but for latitude bands 0–30° S and 30–70° S.

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 four latitude bands and the results of the analysis can be seen in Table 7. The errors quoted here are the 1-σ fitting error of a linear least squares fit to the MAD filtered data. Systematic errors are not considered in this work as they have not been calculated for ACE-FTS at this time. Systematic 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 $1.12 \pm 0.11\,\%\,\mathrm{yr}^{-1}$ in the northern extra-tropical stratosphere, and $0.96 \pm 0.12\,\%\,\mathrm{yr}^{-1}$ in the southern extra-tropical stratosphere. The rate of increase in

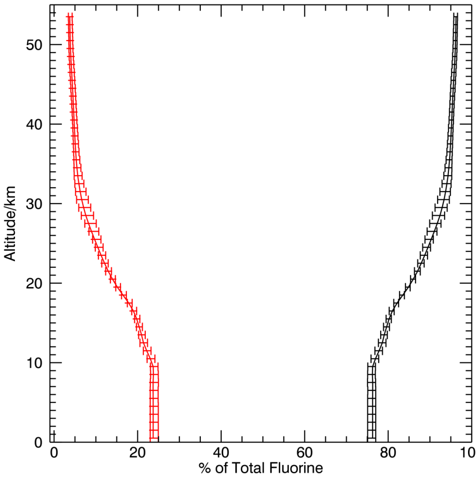

Fig. 3. The mean percentage contribution of ACE-FTS (black) and SLIMCAT (red) to the total fluorine VMR for all years between 70–30° N. The error bars are the maximum or minimum single contribution to the total fluorine percentage from these years.

stratospheric fluorine is also similar in the northern tropical stratosphere ($1.31 \pm 0.20\,\%\,\mathrm{yr}^{-1}$), and in the southern tropical ($1.21 \pm 0.22\,\%\,\mathrm{yr}^{-1}$). 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, $1.21 \pm 0.14\,\%\,\mathrm{yr}^{-1}$ in the Northern Hemisphere, and $1.07 \pm 0.21\,\%\,\mathrm{yr}^{-1}$ in the Southern Hemisphere. Once more the errors quoted here are the statistical 1-σ fitting error of a linear least squared fit to the MAD filtered data. Globally the VMR of fluorine in the stratosphere is increasing at a rate of $1.14 \pm 0.06\,\%\,\mathrm{yr}^{-1}$. 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 Radiative efficiency-weighted fluorine budget trends

The main environmental impact of the increased atmospheric fluorine will be on warming in the troposphere and lower stratosphere region. In order to evaluate this effect the total fluorine was weighted using the radiative efficiencies (Solomon et al., 2007) of the individual species to calculate the change in radiative forcing due to changes in fluorine VMR during this time. These changes in radiative forcing were calculated by removing VMR above the tropopause and multiplying the VMR (in ppb) of each species by its radiative efficiency giving the radiative forcing for each species. These values were summed together and the mean was calculated for each year. A least squares fit to the means then allowed the changes in radiative forcing to be calculated. The results of this analysis can be seen in Table 8 and Fig. 5. Both hemispheres exhibit a similar increase in radiative forcing due to fluorine species. In the Northern Hemisphere the rate are $0.23 \pm 0.11\,\%$ per year in the extra-tropics and $0.45 \pm 0.11\,\%$ per year in the tropics. Similarly in the Southern Hemisphere radiative forcing due to fluorine-containing species appeared to increase by $0.29 \pm 0.20\,\%$ per year in the tropics and $0.45 \pm 0.09\,\%$ per year in the extra-tropics. Once more the same calculations were carried out without including CFCs and halons. These results are shown for comparison in Table 8. Without CFCs and halons the increase in radiative forcing would be significantly higher ranging between $3.84 \pm 0.02\,\%$ per year in the extra-tropical Southern Hemisphere and $4.45 \pm 0.02\,\%$ per year in the extra-tropical Northern Hemisphere. Thus at the present time the climate impact of fluorine is increasing.

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. The error presented here is the 1-σ error of a least squares fit.

	$\mathrm{ppt\,yr}^{-1}$	$\%\,\mathrm{yr}^{-1}$
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

Table 8. The radiative efficiency – (RE) 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 radiative efficiency-weighted fluorine could be quantified. Once more the error presented here is the 1-σ error of a least squares fit.

Latitude band	RE-weighted total fluorine trend ($\%\,\mathrm{yr}^{-1}$)	RE-weighted total fluorine trend without CFCs and halons included ($\%\,\mathrm{yr}^{-1}$)
70–30° N	0.23 ± 0.11	4.45 ± 0.05
30–0° N	0.45 ± 0.11	4.33 ± 0.2
0° N–30° S	0.45 ± 0.09	3.94 ± 0.15
30–70° S	0.29 ± 0.2	3.84 ± 0.02

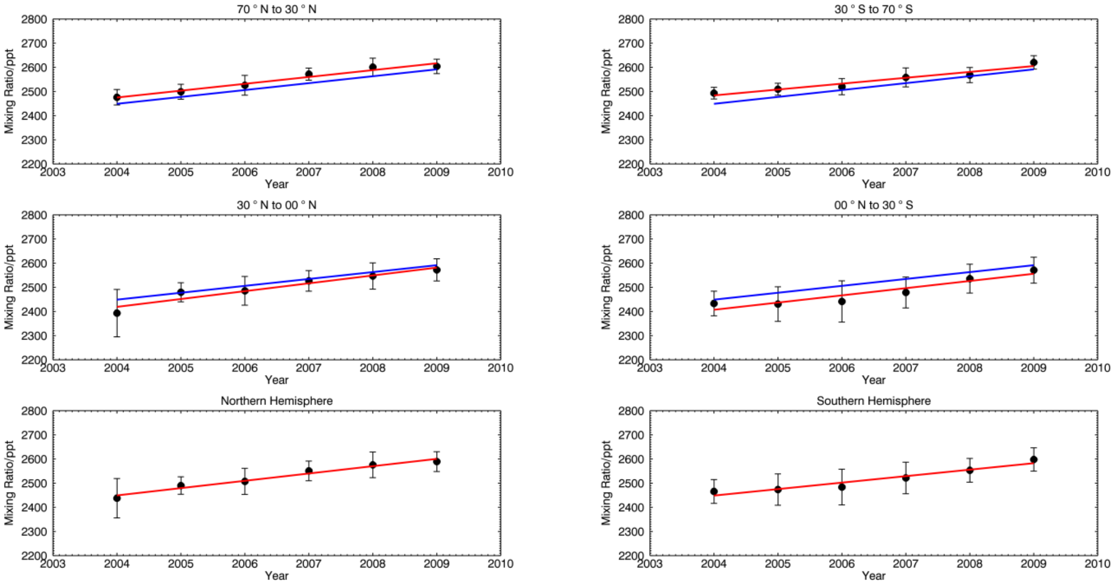

Fig. 4. The trends in the mean total stratospheric fluorine (ppt) between 2004 and 2009. The red line represents the line of best fit for the mean stratospheric total fluorine volume mixing ratio (the black circles). The blue line is the global trend in mean total stratospheric fluorine. The error bars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

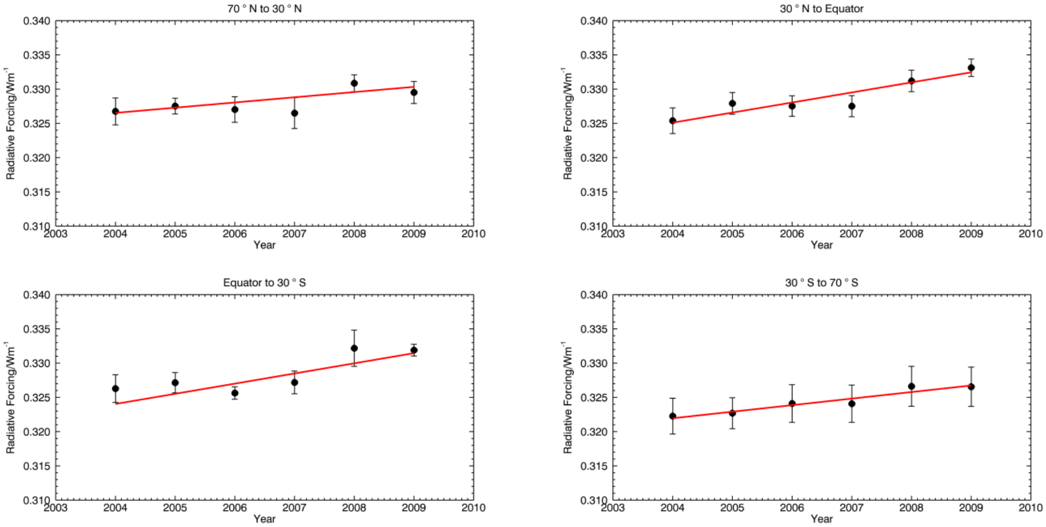

Fig. 5. The trends in the radiative efficiency-weighted total fluorine mean between 2004 and 2009. The red line represents the line of best fit for the radiative efficiency-weighted mean volume mixing ratio (the black circles). The error bars shown in the plots are the standard deviation of the data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

Weighting the total VMR does not allow the long term impact of these species to be evaluated as radiative efficiencies do not take the lifetime of species into account. It is likely that the replacement of CFCs and Halons with HFCs and HCFCs, which have significantly shorter tropospheric lifetimes, reduces the climatological effect of these species in the long term.

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 3-D 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 radiative efficiencies of the individual species used in this study. This allows the climatological implication of the increase in fluorine during this time to be quantified. Changes in climate forcing due to changes in the VMR of fluorine-containing species have also been calculated. These results show small increases of between 0.23 ± 0.11 % per year and 0.45 ± 0.11 % per year. It appears that in the short term the climate forcing effects atmospheric fluorine are increasing. However, the decrease in the VMR of CFCs and halons due to their phasing out under the Montreal Protocol has limited this increase, thus reducing the climatological effects of fluorine-containing species.

Appendix A

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

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

Altitude	ACE-FTS									SLIMCAT										
	HF	CF_4	CFC-12	CFC-11	COF_2	COClF	HCFC-22	SF_6	Total	CFC-113	CFC-114	CFC-115	HCFC-142b	HCFC-141b	HFC-23	HFC-134a	HFC-152a	H-1211	H-1301	Total
10.5	1.2	11.3	38.9	8.9	0.7	0.2	14.4	1.3	76.9	8.5	2.5	1.7	1.2	0.7	2.1	5.4	0.3	0.3	0.4	23.1
11.5	2.2	11.3	38.5	8.6	1.3	0.4	14.2	1.3	77.8	8.2	2.4	1.7	1.1	0.6	2.1	5.1	0.3	0.3	0.4	22.2
12.5	3.1	11.4	38.1	8.3	1.8	0.5	14.1	1.3	78.6	7.9	2.4	1.7	1.1	0.6	2.1	4.9	0.3	0.2	0.4	21.6
13.5	3.9	11.4	37.5	8.1	2.1	0.6	14.3	1.3	79.2	7.7	2.3	1.7	1.1	0.6	2	4.8	0.2	0.2	0.4	21
14.5	4.9	11.4	36.9	7.8	2.4	0.6	14.3	1.3	79.6	7.4	2.3	1.7	1	0.6	2	4.6	0.2	0.2	0.3	20.3
15.5	6.2	11.4	36.1	7.3	3.1	0.7	14.1	1.3	80.2	7.2	2.2	1.7	1	0.5	2	4.5	0.2	0.2	0.3	19.8
16.5	8.3	11.5	34.9	6.6	4.1	0.9	13.4	1.3	81	6.8	2.2	1.6	1	0.5	2	4.3	0.2	0.2	0.3	19.1
17.5	11.1	11.6	33	5.8	5.6	1.1	12.5	1.3	82	6.3	2.1	1.6	0.9	0.4	1.9	4	0.2	0.1	0.3	17.8
18.5	14.5	11.7	30.8	4.8	7.3	1.4	11.8	1.3	83.6	5.6	2	1.6	0.9	0.4	1.9	3.6	0.2	0.1	0.2	16.5
19.5	18	11.7	28.3	3.7	9	1.6	11.3	1.3	84.9	5	1.8	1.6	0.8	0.3	1.9	3.3	0.1	0.1	0.2	15.1
20.5	21.6	11.8	25.8	2.6	10.6	1.7	10.7	1.2	86	4.4	1.7	1.5	0.8	0.3	1.8	3.1	0.1	0	0.2	13.9
21.5	24.9	11.7	23.2	1.7	12.3	1.8	10.2	1.2	87	3.9	1.6	1.5	0.7	0.2	1.8	2.9	0.1	0	0.1	12.8
22.5	27.9	11.7	20.7	1.1	13.7	1.7	9.8	1.2	87.8	3.4	1.5	1.5	0.7	0.2	1.8	2.8	0.1	0	0.1	12.1
23.5	30.8	11.6	18.4	0.7	15.1	1.5	9.5	1.2	88.8	2.9	1.5	1.4	0.7	0.2	1.8	2.7	0.1	0	0.1	11.4
24.5	33.6	11.6	16	0.4	16.3	1.3	9.1	1.2	89.5	2.4	1.4	1.4	0.6	0.1	1.7	2.6	0.1	0	0.1	10.4
25.5	36.6	11.5	13.7	0.2	17.2	1	8.8	1.2	90.2	2	1.3	1.4	0.6	0.1	1.7	2.5	0.1	0	0	9.7
26.5	39.6	11.5	11.4	0.1	18	0.8	8.4	1.2	91	1.6	1.2	1.4	0.6	0.1	1.7	2.4	0.1	0	0	9.1
27.5	42.8	11.5	9.2	0.1	18.4	0.6	8	1.2	91.8	1.2	1.1	1.3	0.5	0.1	1.7	2.3	0	0	0	8.2
28.5	45.9	11.4	7.2	0	18.5	0.4	7.7	1.2	92.3	0.9	1	1.3	0.5	0	1.7	2.2	0	0	0	7.6
29.5	49	11.4	5.7	0	18.1	0.3	7.3	1.2	93	0.7	0.9	1.3	0.4	0	1.6	2.1	0	0	0	7
30.5	52	11.4	4.3	0	17.5	0.2	6.9	1.2	93.5	0.5	0.8	1.3	0.4	0	1.6	2	0	0	0	6.6
31.5	54.7	11.5	3.2	0	16.5	0.1	6.6	1.2	93.8	0.3	0.7	1.2	0.4	0	1.6	1.9	0	0	0	6.1
32.5	57.1	11.5	2.4	0	15.7	0.1	6.3	1.2	94.3	0.2	0.7	1.2	0.4	0	1.6	1.8	0	0	0	5.9
33.5	59.2	11.5	1.7	0	14.8	0	6	1.2	94.4	0.1	0.6	1.2	0.3	0	1.6	1.8	0	0	0	5.6
34.5	60.8	11.5	1.2	0	14.1	0	5.7	1.2	94.5	0.1	0.6	1.2	0.3	0	1.6	1.7	0	0	0	5.5
35.5	62.3	11.5	0.9	0	13.3	0	5.5	1.2	94.7	0.1	0.5	1.2	0.3	0	1.6	1.6	0	0	0	5.3
36.5	63.6	11.5	0.6	0	12.6	0	5.3	1.2	94.8	0	0.5	1.2	0.3	0	1.6	1.6	0	0	0	5.2
37.5	64.7	11.5	0.4	0	11.9	0	5.2	1.2	94.9	0	0.5	1.2	0.3	0	1.6	1.6	0	0	0	5.2
38.5	65.8	11.5	0.3	0	11.2	0	5	1.2	95	0	0.4	1.1	0.3	0	1.6	1.5	0	0	0	4.9
39.5	66.8	11.5	0.2	0	10.6	0	4.8	1.2	95.1	0	0.4	1.1	0.2	0	1.6	1.5	0	0	0	4.8
40.5	67.7	11.5	0.2	0	10	0	4.7	1.2	95.3	0	0.4	1.1	0.2	0	1.6	1.4	0	0	0	4.7
41.5	68.6	11.6	0.1	0	9.3	0	4.5	1.2	95.3	0	0.4	1.1	0.2	0	1.6	1.4	0	0	0	4.7
42.5	69.4	11.6	0.1	0	8.7	0	4.3	1.2	95.3	0	0.3	1.1	0.2	0	1.6	1.4	0	0	0	4.6
43.5	70.3	11.7	0.1	0	8.1	0	4.2	1.2	95.6	0	0.3	1.1	0.2	0	1.6	1.3	0	0	0	4.5
44.5	71.3	11.7	0	0	7.4	0	4	1.2	95.6	0	0.3	1.1	0.2	0	1.6	1.3	0	0	0	4.5
45.5	72.2	11.7	0	0	6.7	0	3.8	1.2	95.6	0	0.3	1.1	0.2	0	1.6	1.3	0	0	0	4.5
46.5	73.2	11.6	0	0	6.1	0	3.7	1.2	95.8	0	0.2	1.1	0.2	0	1.6	1.2	0	0	0	4.3
47.5	74.1	11.6	0	0	5.5	0	3.5	1.2	95.9	0	0.2	1	0.1	0	1.6	1.2	0	0	0	4.1
48.5	75	11.5	0	0	5	0	3.4	1.2	96.1	0	0.2	1	0.1	0	1.5	1.1	0	0	0	3.9
49.5	75.8	11.5	0	0	4.4	0	3.2	1.2	96.1	0	0.2	1	0.1	0	1.5	1.1	0	0	0	3.9
50.5	76.3	11.5	0	0	4	0	3.1	1.2	96.1	0	0.1	1	0.1	0	1.5	1.1	0	0	0	3.8
51.5	76.7	11.5	0	0	3.7	0	3.1	1.2	96.2	0	0.1	1	0.1	0	1.5	1.1	0	0	0	3.8
52.5	77	11.6	0	0	3.5	0	3	1.2	96.3	0	0.1	1	0.1	0	1.5	1	0	0	0	3.7
53.5	77.3	11.6	0	0	3.3	0	2.9	1.2	96.3	0	0.1	1	0.1	0	1.5	1	0	0	0	3.7

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

Altitude	ACE-FTS									SLIMCAT										
	HF	CF_4	CFC-12	CFC-11	COF_2	COClF	HCFC-22	SF_6	Total	CFC-113	CFC-114	CFC-115	HCFC-142b	HCFC-141b	HFC-23	HFC-134a	HFC-152a	H-1211	H-1301	Total
10.5	0	11.1	39.18	9.4	0.02	0.02	15.31	1.35	76.38	8.67	2.51	1.7	1.17	0.69	2.14	5.68	0.33	0.32	0.42	23.63
11.5	0	11.1	39.19	9.4	0.03	0.02	15.31	1.35	76.4	8.67	2.51	1.7	1.17	0.69	2.14	5.67	0.33	0.31	0.42	23.61
12.5	0	11.11	39.21	9.4	0.04	0.02	15.32	1.36	76.46	8.64	2.5	1.7	1.17	0.69	2.14	5.63	0.33	0.31	0.41	23.52
13.5	0	11.12	39.25	9.41	0.07	0.04	15.34	1.36	76.59	8.61	2.5	1.7	1.17	0.69	2.13	5.58	0.33	0.31	0.41	23.43
14.5	0.05	11.13	39.28	9.42	0.09	0.05	15.35	1.36	76.73	8.57	2.49	1.7	1.16	0.68	2.12	5.53	0.32	0.3	0.41	23.28
15.5	0.17	11.16	39.26	9.45	0.13	0.07	15.29	1.35	76.88	8.53	2.48	1.7	1.15	0.67	2.11	5.46	0.31	0.3	0.41	23.12
16.5	0.34	11.24	39.45	9.34	0.19	0.1	15.06	1.36	77.08	8.46	2.47	1.7	1.15	0.66	2.11	5.37	0.3	0.29	0.4	22.91
17.5	0.79	11.29	39.37	9.16	0.47	0.19	14.92	1.37	77.56	8.3	2.44	1.7	1.13	0.64	2.09	5.21	0.29	0.27	0.39	22.46
18.5	1.84	11.31	38.59	8.64	1.06	0.38	15.23	1.35	78.4	7.97	2.38	1.68	1.09	0.61	2.05	4.95	0.26	0.24	0.37	21.6
19.5	3.37	11.38	37.56	8.01	1.98	0.75	15.08	1.32	79.45	7.54	2.3	1.66	1.05	0.56	2.01	4.65	0.24	0.2	0.35	20.56
20.5	5.45	11.57	36.56	7.27	3.2	1.2	13.66	1.31	80.22	7.2	2.25	1.66	1.02	0.52	2	4.43	0.22	0.15	0.32	19.77
21.5	7.51	11.64	35.3	6.36	4.4	1.63	12.82	1.3	80.96	6.87	2.21	1.65	1	0.49	1.98	4.25	0.2	0.11	0.3	19.06
22.5	9.25	11.67	34.07	5.37	5.75	2.08	12.15	1.3	81.64	6.54	2.17	1.64	0.97	0.45	1.96	4.09	0.18	0.07	0.28	18.35
23.5	10.85	11.72	32.69	4.35	7.12	2.49	11.75	1.3	82.27	6.21	2.13	1.64	0.96	0.42	1.95	3.98	0.17	0.04	0.25	17.75
24.5	12.2	11.83	31.34	3.29	8.67	2.79	11.35	1.31	82.78	5.89	2.11	1.64	0.95	0.38	1.95	3.9	0.16	0.02	0.23	17.23
25.5	14.14	11.85	29.5	2.38	10.22	3.01	11.06	1.31	83.47	5.47	2.08	1.64	0.93	0.34	1.94	3.81	0.15	0.01	0.19	16.56
26.5	15.96	11.85	27.68	1.62	11.99	3.04	10.85	1.31	84.3	4.95	2.03	1.62	0.91	0.29	1.92	3.7	0.14	0	0.16	15.72
27.5	17.57	11.93	25.86	1	14.16	2.73	10.64	1.32	85.21	4.32	1.97	1.62	0.88	0.24	1.91	3.61	0.13	0	0.12	14.8
28.5	19.63	11.92	23.76	0.51	16.55	2.21	10.34	1.31	86.23	3.68	1.89	1.6	0.85	0.18	1.89	3.49	0.12	0	0.09	13.79
29.5	22.16	11.84	21.49	0.3	18.54	1.63	9.96	1.3	87.22	3.06	1.79	1.56	0.81	0.14	1.86	3.36	0.11	0	0.06	12.75
30.5	25.1	11.85	18.83	0.11	20.12	1.13	9.7	1.3	88.14	2.48	1.71	1.55	0.78	0.1	1.85	3.26	0.1	0	0.04	11.87
31.5	28.34	11.92	15.85	0.03	21.51	0.34	9.5	1.31	88.8	2.01	1.65	1.55	0.76	0.07	1.85	3.2	0.09	0	0.02	11.2
32.5	31.41	11.91	12.81	0.02	22.65	0.23	9.2	1.32	89.55	1.56	1.56	1.53	0.73	0.05	1.84	3.1	0.08	0	0.01	10.46
33.5	34.39	11.93	9.87	0.01	23.66	0.12	8.93	1.32	90.23	1.13	1.47	1.52	0.69	0.03	1.84	3.01	0.07	0	0.01	9.77
34.5	37	11.87	7.65	0.01	24.39	0.08	8.55	1.32	90.87	0.83	1.37	1.49	0.66	0.02	1.82	2.89	0.06	0	0	9.14
35.5	39.68	11.9	5.51	0	24.75	0.04	8.21	1.32	91.41	0.56	1.28	1.47	0.62	0.01	1.81	2.78	0.05	0	0	8.58
36.5	42.27	11.94	3.88	0	24.77	0.02	7.77	1.32	91.97	0.36	1.17	1.45	0.58	0.01	1.79	2.65	0.04	0	0	8.05
37.5	45.17	11.91	2.72	0	24.04	0.01	7.28	1.32	92.45	0.24	1.06	1.41	0.53	0	1.77	2.5	0.03	0	0	7.54
38.5	48.3	11.87	1.66	0	22.92	0.01	6.83	1.32	92.91	0.13	0.95	1.38	0.48	0	1.75	2.36	0.02	0	0	7.07
39.5	50.7	11.87	1.2	0	21.62	0	6.48	1.32	93.19	0.09	0.87	1.36	0.45	0	1.75	2.26	0.02	0	0	6.8
40.5	53.16	11.82	0.81	0	20.24	0	6.13	1.32	93.48	0.06	0.79	1.34	0.41	0	1.74	2.15	0.02	0	0	6.51
41.5	55.56	11.86	0.51	0	18.62	0	5.83	1.33	93.71	0.03	0.72	1.33	0.39	0	1.73	2.07	0.01	0	0	6.28
42.5	57.47	11.9	0.38	0	17.25	0	5.57	1.34	93.91	0.02	0.67	1.31	0.36	0	1.73	1.99	0.01	0	0	6.09
43.5	59.32	12.01	0.25	0	15.86	0	5.31	1.34	94.09	0.01	0.61	1.29	0.34	0	1.72	1.92	0.01	0	0	5.9
44.5	61.28	12.12	0.17	0	14.34	0	5.03	1.35	94.29	0.01	0.56	1.27	0.31	0	1.72	1.83	0.01	0	0	5.71
45.5	63.27	12.18	0.11	0	12.84	0	4.74	1.35	94.49	0.01	0.5	1.25	0.29	0	1.71	1.75	0.01	0	0	5.52
46.5	65.37	12.17	0.06	0	11.31	0	4.44	1.35	94.7	0	0.44	1.23	0.26	0	1.7	1.66	0	0	0	5.29
47.5	67.23	12.12	0.03	0	10.01	0	4.17	1.34	94.9	0	0.39	1.2	0.24	0	1.68	1.58	0	0	0	5.09
48.5	69.01	12	0.02	0	8.85	0	3.91	1.33	95.12	0	0.35	1.16	0.22	0	1.65	1.49	0	0	0	4.87
49.5	70.78	11.87	0.02	0	7.72	0	3.64	1.32	95.35	0	0.3	1.13	0.2	0	1.62	1.41	0	0	0	4.66
50.5	72.17	11.85	0.01	0	6.73	0	3.43	1.31	95.5	0	0.26	1.11	0.18	0	1.61	1.35	0	0	0	4.51
51.5	73.09	11.81	0.01	0	6.1	0	3.28	1.31	95.6	0	0.24	1.09	0.17	0	1.6	1.3	0	0	0	4.4
52.5	73.77	11.81	0	0	5.62	0	3.17	1.31	95.68	0	0.22	1.08	0.16	0	1.59	1.27	0	0	0	4.32
53.5	74.48	11.79	0	0	5.14	0	3.06	1.31	95.78	0	0.2	1.06	0.15	0	1.58	1.23	0	0	0	4.22

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

Altitude	ACE-FTS									SLIMCAT										
	HF	CF_4	CFC-12	CFC-11	COF_2	COClF	HCFC-22	SF_6	Total	CFC-113	CFC-114	CFC-115	HCFC-142b	HCFC-141b	HFC-23	HFC-134a	HFC-152a	H-1211	H-1301	Total
10.5	0.02	11.08	39.31	9.42	0.03	0.02	15.08	1.36	76.32	8.69	2.51	1.7	1.18	0.7	2.15	5.69	0.34	0.32	0.42	23.70
11.5	0.04	11.08	39.31	9.42	0.03	0.02	15.08	1.36	76.34	8.69	2.51	1.7	1.18	0.7	2.14	5.68	0.34	0.32	0.42	23.68
12.5	0.08	11.08	39.31	9.43	0.04	0.02	15.08	1.35	76.39	8.67	2.51	1.7	1.17	0.69	2.14	5.66	0.33	0.31	0.42	23.60
13.5	0.14	11.08	39.31	9.43	0.06	0.03	15.08	1.35	76.48	8.64	2.5	1.7	1.17	0.69	2.14	5.63	0.33	0.31	0.41	23.52
14.5	0.21	11.08	39.31	9.43	0.08	0.03	15.08	1.35	76.57	8.61	2.5	1.7	1.17	0.69	2.13	5.59	0.33	0.31	0.41	23.44
15.5	0.39	11.11	39.26	9.46	0.13	0.05	15.01	1.35	76.76	8.56	2.48	1.7	1.16	0.68	2.12	5.52	0.32	0.3	0.41	23.25
16.5	0.75	11.17	39.27	9.32	0.22	0.09	14.9	1.35	77.07	8.45	2.46	1.7	1.15	0.66	2.11	5.4	0.31	0.29	0.4	22.93
17.5	1.32	11.22	39.14	9.14	0.52	0.18	14.8	1.36	77.68	8.23	2.42	1.69	1.12	0.64	2.08	5.2	0.29	0.27	0.39	22.33
18.5	2.35	11.29	38.43	8.64	1.26	0.41	15	1.34	78.72	7.82	2.33	1.66	1.07	0.6	2.04	4.89	0.26	0.23	0.37	21.27
19.5	3.95	11.4	37.38	7.95	2.25	0.8	14.78	1.32	79.83	7.35	2.24	1.64	1.02	0.55	2.01	4.58	0.24	0.19	0.34	20.16
20.5	6.1	11.57	36.26	7.2	3.57	1.2	13.43	1.3	80.63	7.01	2.19	1.64	0.99	0.51	1.99	4.36	0.22	0.15	0.31	19.37
21.5	7.84	11.67	34.91	6.3	4.84	1.69	12.68	1.28	81.21	6.74	2.15	1.64	0.97	0.48	1.98	4.21	0.2	0.11	0.3	18.78
22.5	9.77	11.66	33.63	5.31	6.16	2.04	12.03	1.27	81.87	6.43	2.12	1.63	0.95	0.45	1.95	4.07	0.19	0.07	0.27	18.13
23.5	11.59	11.7	32.07	4.3	7.49	2.39	11.7	1.27	82.51	6.1	2.08	1.62	0.94	0.41	1.94	3.95	0.17	0.04	0.25	17.50
24.5	13.51	11.81	30.46	3.19	9.19	2.61	11.09	1.28	83.14	5.7	2.05	1.63	0.92	0.37	1.94	3.85	0.16	0.02	0.22	16.86
25.5	15.91	11.83	27.99	2.25	11.2	2.74	10.78	1.28	83.98	5.18	2	1.61	0.9	0.32	1.92	3.73	0.15	0.01	0.18	16.00
26.5	18.3	11.87	25.72	1.47	13.04	2.7	10.53	1.28	84.91	4.6	1.94	1.6	0.87	0.27	1.91	3.61	0.14	0	0.15	15.09
27.5	20.27	11.84	23.42	0.92	15.4	2.36	10.3	1.28	85.79	4.01	1.88	1.59	0.85	0.22	1.89	3.52	0.13	0	0.11	14.20
28.5	22.27	11.85	21.28	0.49	17.46	1.92	10.09	1.27	86.63	3.45	1.82	1.58	0.83	0.17	1.88	3.43	0.12	0	0.08	13.36
29.5	24.33	11.83	18.94	0.3	19.49	1.5	9.83	1.27	87.49	2.92	1.75	1.56	0.8	0.14	1.86	3.33	0.11	0	0.06	12.53
30.5	26.93	11.94	16.29	0.11	20.94	1.01	9.68	1.28	88.18	2.42	1.7	1.56	0.78	0.1	1.86	3.27	0.1	0	0.04	11.83
31.5	29.5	12.1	13.67	0.04	22.15	0.58	9.49	1.29	88.82	1.97	1.63	1.56	0.76	0.07	1.87	3.21	0.09	0	0.02	11.18
32.5	32.08	12.2	10.98	0.02	23.31	0.39	9.23	1.3	89.51	1.52	1.55	1.55	0.73	0.05	1.88	3.11	0.08	0	0.01	10.48
33.5	34.87	12.31	8.34	0.01	24.22	0.21	8.94	1.32	90.22	1.09	1.46	1.55	0.69	0.03	1.88	3.01	0.07	0	0.01	9.79
34.5	37.17	12.27	6.52	0	24.79	0.14	8.58	1.31	90.78	0.82	1.37	1.52	0.65	0.02	1.86	2.9	0.06	0	0	9.20
35.5	39.92	12.17	4.74	0	25.07	0.08	8.14	1.3	91.42	0.56	1.26	1.49	0.61	0.01	1.83	2.76	0.05	0	0	8.57
36.5	42.97	12.02	3.41	0	24.59	0.04	7.67	1.29	91.99	0.38	1.15	1.45	0.57	0.01	1.8	2.62	0.04	0	0	8.02
37.5	45.81	11.94	2.5	0	23.6	0.03	7.26	1.28	92.42	0.27	1.05	1.42	0.53	0	1.78	2.49	0.03	0	0	7.57
38.5	48.39	11.96	1.69	0	22.51	0.01	6.92	1.29	92.77	0.17	0.97	1.4	0.49	0	1.77	2.39	0.03	0	0	7.22
39.5	51.08	11.91	1.23	0	21.11	0.01	6.5	1.29	93.13	0.12	0.88	1.37	0.45	0	1.76	2.27	0.02	0	0	6.87
40.5	53.67	11.97	0.8	0	19.58	0.01	6.12	1.3	93.45	0.07	0.79	1.35	0.41	0	1.76	2.16	0.02	0	0	6.56
41.5	56.5	11.98	0.5	0	17.76	0	5.71	1.3	93.75	0.04	0.7	1.32	0.37	0	1.74	2.04	0.01	0	0	6.22
42.5	59.12	12.09	0.34	0	15.83	0	5.35	1.31	94.04	0.03	0.62	1.3	0.34	0	1.73	1.93	0.01	0	0	5.96
43.5	61.61	12.27	0.19	0	13.92	0	4.99	1.32	94.3	0.01	0.55	1.28	0.31	0	1.73	1.82	0.01	0	0	5.71
44.5	63.44	12.45	0.12	0	12.42	0	4.72	1.33	94.48	0.01	0.49	1.26	0.28	0	1.73	1.74	0.01	0	0	5.52
45.5	65.61	12.39	0.08	0	10.9	0	4.41	1.32	94.71	0	0.43	1.23	0.26	0	1.71	1.65	0	0	0	5.28
46.5	67.5	12.42	0.04	0	9.5	0	4.13	1.33	94.92	0	0.38	1.21	0.23	0	1.7	1.56	0	0	0	5.08
47.5	69.14	12.36	0.02	0	8.36	0	3.89	1.32	95.09	0	0.33	1.18	0.21	0	1.68	1.49	0	0	0	4.89
48.5	70.41	12.22	0.02	0	7.59	0	3.71	1.31	95.26	0	0.3	1.15	0.2	0	1.66	1.43	0	0	0	4.74
49.5	71.66	12.07	0.01	0	6.84	0	3.53	1.29	95.4	0	0.27	1.13	0.18	0	1.63	1.38	0	0	0	4.59
50.5	72.78	11.95	0.01	0	6.15	0	3.37	1.28	95.54	0	0.25	1.1	0.17	0	1.61	1.33	0	0	0	4.46
51.5	73.61	11.82	0	0	5.7	0	3.25	1.26	95.64	0	0.23	1.08	0.16	0	1.59	1.29	0	0	0	4.35
52.5	74.01	11.82	0	0	5.4	0	3.19	1.26	95.68	0	0.22	1.08	0.16	0	1.59	1.27	0	0	0	4.32
53.5	74.52	11.77	0	0	5.08	0	3.11	1.26	95.74	0	0.2	1.06	0.15	0	1.58	1.25	0	0	0	4.24

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

Altitude	ACE-FTS									SLIMCAT										
	HF	CF4	CFC-12	CFC-11	COF_2	COClF	HCFC-22	SF6	Total	CFC-113	CFC-114	CFC-115	HCFC-142b	HCFC-141b	HFC-23	HFC-134a	HFC-152a	H-1211	H-1301	Total
10.5	1.42	11.41	38.49	8.94	0.87	0.25	14.01	1.33	76.72	8.55	2.5	1.74	1.16	0.67	2.16	5.49	0.31	0.3	0.41	23.29
11.5	2.56	11.41	37.85	8.71	1.52	0.42	13.98	1.33	77.78	8.13	2.41	1.71	1.11	0.63	2.11	5.17	0.29	0.27	0.38	22.21
12.5	3.82	11.39	37.18	8.46	2.21	0.6	13.96	1.33	78.95	7.66	2.31	1.67	1.06	0.59	2.06	4.84	0.26	0.24	0.36	21.05
13.5	5.2	11.37	36.29	8.17	2.86	0.71	13.9	1.32	79.82	7.3	2.22	1.65	1.02	0.55	2.02	4.61	0.24	0.22	0.34	20.17
14.5	7.06	11.33	34.96	7.71	3.7	0.82	13.72	1.31	80.61	6.95	2.14	1.62	0.98	0.52	1.98	4.41	0.23	0.2	0.32	19.35
15.5	9.5	11.34	33.28	7.02	4.85	0.95	13.21	1.3	81.45	6.59	2.07	1.6	0.94	0.49	1.95	4.21	0.21	0.18	0.3	18.54
16.5	12.41	11.45	31.39	6.14	6.31	1.12	12.24	1.28	82.34	6.19	1.98	1.58	0.9	0.46	1.93	3.98	0.19	0.16	0.28	17.65
17.5	15.88	11.58	29.11	5.13	7.97	1.3	11.25	1.26	83.48	5.68	1.88	1.56	0.85	0.41	1.9	3.69	0.17	0.13	0.25	16.52
18.5	19.69	11.62	26.45	4	9.94	1.46	10.53	1.24	84.93	5.03	1.74	1.52	0.79	0.35	1.85	3.34	0.14	0.1	0.22	15.08
19.5	23.1	11.64	23.95	2.91	11.82	1.57	10.04	1.21	86.24	4.41	1.62	1.49	0.73	0.3	1.8	3.04	0.12	0.06	0.18	13.75
20.5	26.29	11.68	21.57	1.9	13.36	1.62	9.67	1.19	87.28	3.9	1.52	1.46	0.68	0.25	1.78	2.82	0.1	0.04	0.15	12.7
21.5	29.06	11.65	19.22	1.16	14.92	1.54	9.38	1.18	88.11	3.47	1.45	1.44	0.65	0.22	1.75	2.68	0.09	0.02	0.13	11.9
22.5	31.47	11.6	16.93	0.78	16.52	1.32	9.09	1.17	88.88	3.03	1.38	1.42	0.62	0.18	1.73	2.57	0.08	0.01	0.11	11.13
23.5	33.88	11.59	14.8	0.52	17.71	1.07	8.85	1.17	89.59	2.59	1.32	1.4	0.6	0.15	1.72	2.48	0.07	0	0.08	10.41
24.5	36.17	11.58	12.78	0.31	18.73	0.89	8.63	1.16	90.25	2.15	1.26	1.39	0.58	0.11	1.71	2.42	0.06	0	0.06	9.74
25.5	38.28	11.55	10.91	0.16	19.67	0.72	8.42	1.16	90.87	1.75	1.2	1.38	0.56	0.08	1.7	2.37	0.06	0	0.04	9.14
26.5	40.38	11.48	9.2	0.08	20.35	0.58	8.19	1.15	91.41	1.4	1.15	1.36	0.54	0.06	1.69	2.32	0.05	0	0.03	8.6
27.5	42.62	11.47	7.55	0.04	20.67	0.44	7.95	1.15	91.89	1.11	1.08	1.34	0.51	0.04	1.68	2.26	0.04	0	0.02	8.08
28.5	44.89	11.49	6.07	0.02	20.72	0.32	7.7	1.15	92.36	0.85	1.02	1.33	0.49	0.03	1.67	2.21	0.04	0	0.01	7.65
29.5	47.13	11.53	4.89	0.01	20.34	0.23	7.45	1.15	92.73	0.66	0.96	1.32	0.47	0.02	1.67	2.15	0.03	0	0.01	7.29
30.5	49.47	11.6	3.82	0	19.69	0.15	7.18	1.15	93.06	0.48	0.89	1.31	0.44	0.01	1.67	2.09	0.03	0	0	6.92
31.5	51.71	11.68	2.89	0	18.95	0.08	6.91	1.16	93.38	0.34	0.83	1.29	0.42	0.01	1.67	2.03	0.02	0	0	6.61
32.5	53.81	11.69	2.19	0	18.18	0.05	6.61	1.16	93.69	0.24	0.76	1.27	0.39	0.01	1.66	1.96	0.02	0	0	6.31
33.5	55.97	11.65	1.57	0	17.33	0.03	6.29	1.15	93.99	0.16	0.7	1.25	0.37	0	1.64	1.88	0.02	0	0	6.02
34.5	57.99	11.62	1.13	0	16.32	0.01	6.01	1.15	94.23	0.1	0.64	1.23	0.34	0	1.63	1.81	0.01	0	0	5.76
35.5	59.87	11.6	0.81	0	15.21	0.01	5.76	1.14	94.4	0.07	0.59	1.21	0.32	0	1.62	1.76	0.01	0	0	5.58
36.5	61.64	11.57	0.57	0	14.15	0.01	5.51	1.14	94.59	0.05	0.55	1.19	0.3	0	1.61	1.7	0.01	0	0	5.41
37.5	63.26	11.54	0.42	0	13.13	0	5.26	1.14	94.75	0.03	0.51	1.18	0.28	0	1.6	1.64	0.01	0	0	5.25
38.5	64.73	11.53	0.29	0	12.17	0	5.03	1.14	94.89	0.02	0.47	1.16	0.27	0	1.6	1.59	0.01	0	0	5.12
39.5	66.07	11.55	0.2	0	11.25	0	4.81	1.14	95.02	0.01	0.43	1.15	0.25	0	1.6	1.53	0.01	0	0	4.98
40.5	67.28	11.59	0.15	0	10.38	0	4.6	1.15	95.15	0.01	0.39	1.14	0.23	0	1.6	1.48	0	0	0	4.85
41.5	68.44	11.64	0.1	0	9.52	0	4.4	1.16	95.26	0.01	0.36	1.13	0.22	0	1.6	1.44	0	0	0	4.76
42.5	69.44	11.69	0.06	0	8.76	0	4.22	1.16	95.33	0	0.33	1.12	0.2	0	1.6	1.39	0	0	0	4.64
43.5	70.51	11.75	0.04	0	7.97	0	4.03	1.16	95.46	0	0.3	1.11	0.19	0	1.6	1.34	0	0	0	4.54
44.5	71.53	11.79	0.03	0	7.2	0	3.85	1.17	95.57	0	0.27	1.09	0.18	0	1.59	1.3	0	0	0	4.43
45.5	72.48	11.77	0.02	0	6.55	0	3.69	1.16	95.67	0	0.25	1.08	0.17	0	1.58	1.26	0	0	0	4.34
46.5	73.48	11.72	0.01	0	5.89	0	3.52	1.16	95.78	0	0.22	1.06	0.15	0	1.57	1.21	0	0	0	4.21
47.5	74.41	11.65	0.01	0	5.3	0	3.37	1.15	95.89	0	0.2	1.04	0.14	0	1.55	1.17	0	0	0	4.1
48.5	75.02	11.57	0	0	4.94	0	3.28	1.14	95.95	0	0.19	1.03	0.14	0	1.54	1.15	0	0	0	4.05
49.5	75.59	11.49	0	0	4.6	0	3.2	1.13	96.01	0	0.17	1.01	0.13	0	1.53	1.13	0	0	0	3.97
50.5	75.92	11.52	0	0	4.32	0	3.15	1.14	96.05	0	0.16	1.01	0.13	0	1.53	1.12	0	0	0	3.95
51.5	76.17	11.54	0	0	4.11	0	3.11	1.14	96.07	0	0.15	1.01	0.13	0	1.53	1.11	0	0	0	3.93
52.5	76.38	11.55	0	0	3.94	0	3.08	1.14	96.09	0	0.15	1	0.12	0	1.53	1.1	0	0	0	3.9
53.5	76.63	11.55	0	0	3.76	0	3.04	1.14	96.12	0	0.14	1	0.12	0	1.53	1.09	0	0	0	3.88

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