

Geophysical Research Letters®



RESEARCH LETTER

10.1029/2024GL112470

Key Points:

- Implemented a harmonized NDACC-IRWG FTIR HCFC-22 retrieval strategy and provided a new global long-term HCFC-22 data set
- A decrease in the growth rate of the atmospheric HCFC-22 column has been observed by FTIR measurements since 2009
- The FTIR, NOAA flask samplings, and ACE-FTS satellite observations show a good agreement in the HCFC-22 trend

Supporting Information:

Supporting Information may be found in the online version of this article.

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Citation:

Zhou, M., Langerock, B., Vigouroux, C., Smale, D., Toon, G., Polyakov, A., et al. (2024). Recent decreases in the growth rate of atmospheric HCFC-22 column derived from the ground-based FTIR harmonized retrievals at 16 NDACC sites. *Geophysical Research Letters*, 51, e2024GL112470. <https://doi.org/10.1029/2024GL112470>

Received 12 SEP 2024
Accepted 27 OCT 2024

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Recent Decreases in the Growth Rate of Atmospheric HCFC-22 Column Derived From the Ground-Based FTIR Harmonized Retrievals at 16 NDACC Sites

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Abstract HCFC-22 is an ozone-depleting substance with a greenhouse effect. The atmospheric mole fractions of HCFC-22 have been increasing since the 1950s. Within the NDACC-IRWG network, HCFC-22 mol fractions can be retrieved from solar absorption spectra measured by ground-based FTIR. However, only a few sites have provided HCFC-22 data sets. Here, we demonstrate a harmonized FTIR HCFC-22 retrieval strategy and generate a new global NDACC-IRWG HCFC-22 data set at 16 FTIR sites. The systematic and random uncertainties are 5.3%–8.7% and 3.2%–8.0%, respectively. A maximum HCFC-22 column annual growth rate was observed in 2009 with a mean of 7.65 ± 1.39 ppt/year, and the HCFC-22 annual growth rate decreased to 3.57 ± 1.39 ppt/year (2016–2020) and 2.15 ± 2.09 ppt/year (2021–2023). The annual growth rates derived from the FTIR measurements are compared to the ones derived from NOAA surface flask samplings and ACE-FTS satellite measurements, and the three independent data sets show a good agreement.

Plain Language Summary Monitoring the atmospheric HCFC-22 mol fraction and its long-term trend is important to the stratospheric ozone layer and climate change. Ground-based FTIR measurements within the NDACC-IRWG community provide a powerful technique for observing atmospheric trace gases. However, due to different retrieval software and procedures among the sites, the record was too heterogeneous for monitoring the global evolution of HCFC-22 over time. In this study, we propose a harmonized FTIR HCFC-22 retrieval strategy and generate a global NDACC-IRWG HCFC-22 data set at 16 FTIR sites. The retrieval uncertainty of the FTIR HCFC-22 is well presented and discussed. Based on the new FTIR HCFC-22 measurements, the HCFC-22 annual growth rates between 1990 and 2023 are evaluated. The results are compared with two independent data sets: NOAA flask samplings and ACE-FTS satellite measurements. Good agreement among the three data sets is found, with a clear decrease in the growth rate of atmospheric HCFC-22 in recent years. According to the latest Montreal Protocol, HCFC-22 should be phased out within the next 5–6 years. The global FTIR observations will assure continuity into the next years and decades when HCFC-22 mol fractions should start decreasing after the official phase-out.

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1. Introduction

Chlorodifluoromethane (HCFC-22, R-22 or CHClF_2) is a stratospheric Ozone Depletion Substance (ODS) as well as a greenhouse gas, affecting the Earth's environment (IPCC, 2021; Solomon et al., 1992). The global warming potential (GWP) of HCFC-22 is about 1,760 times greater than CO_2 over a 100-year time period, and its effective radiative forcing is 0.053 W/m^2 in 2019 with respect to the pre-industrial era (Forster et al., 2021).

HCFC-22 is an anthropogenic species, which is used as a refrigerant fluid and emitted by cooling, air-conditioning equipment, and foam production. The sink of HCFC-22 is the reaction with the hydroxyl radical (OH) in the troposphere, and in the stratosphere its photochemical destruction and reaction with $\text{O}(^1\text{D})$ and chlorine (Moore & Remedios, 2008; Saikawa et al., 2012). The lifetime of atmospheric HCFC-22 is estimated to be about 12 years (Forster et al., 2021).

To protect the stratospheric ozone layer, the Montreal Protocol signed in 1987 introduced a series of control measures to phase out different groups of ODSs, with an initial focus on chlorofluorocarbons (CFCs) (Murdoch & Sandler, 1997). As a result, the mole fractions of CFC-11, CFC-12, and CFC-113 started to decline in the 1990s and 2000s (Walker et al., 2000). As hydrochlorofluorocarbons (HCFCs) have shorter lifetimes than CFCs, HCFC-22 was then widely used as a substitute for CFCs as a propellant and refrigerant, resulting in an accelerated increase in atmospheric HCFC-22 mol fraction (Montzka et al., 2009). Currently, HCFC-22 has become the most abundant HCFC, surpassing CFC-11 in recent years. Due to its impact on stratospheric ozone and large GWP, the Montreal Protocol Parties started to speed up the phase out of HCFCs in 2007. Developed and developing countries agreed to start their mitigation controls with a target of zero-emission of HCFCs by 2020 and 2030, respectively.

Long-term atmospheric HCFC-22 monitoring has been carried out by the NOAA Halocarbons and other Atmospheric Trace Species (HATS) flask gas chromatography mass spectrometry detection (GC-MSD) program, the NOAA Chromatograph for Atmospheric Trace Species (CATS) in situ Halocarbons Program, and the Advanced Global Atmospheric Gases Experiment (AGAGE) (Montzka et al., 1993; Prinn et al., 2018). By utilizing the infrared absorptions of HCFC-22, HCFC-22 columns or profiles can also be detected by both space-based and ground-based spectrometers via the remote sensing technique, such as the space-based Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) and the Atmospheric Chemistry Experiment Fourier transform spectrometer (ACE-FTS), and the ground-based Fourier transform infrared (FTIR) spectrometers affiliated to the Network for the Detection of Atmospheric Composition Change-Infrared Working Group (NDACC-IRWG) community (Chirkov et al., 2016; De Mazière et al., 2018; Kolonjari et al., 2024). The NDACC-IRWG measurements started in the early 1990s and at many sites the measurement time coverage is longer than that of any satellite.

Within the NDACC-IRWG community, several groups have carried out independent studies to retrieve HCFC-22 columns or profiles. HCFC-22 absorption lines allow us to derive its mole fraction from ground-based FTIR spectra recorded at mountain (Jungfraujoch and Kitt Peak), urban (St. Petersburg and Hefei), countryside (Lauder) and relatively humid (Reunion St-Denis) sites (Polyakov et al., 2021; Prignon et al., 2019; Rinsland et al., 2005; Vanessa et al., 1997; Zeng et al., 2023; Zhou et al., 2016). However, the NDACC-IRWG HCFC-22 retrieval strategy has not been harmonized, in particular regarding the applied spectroscopy, regularization, and uncertainty estimation. Moreover, until now, only a few NDACC-IRWG sites have implemented routine HCFC-22 retrievals, which do not represent the spatio-temporal variation of HCFC-22 globally.

The objectives of this study are to (a) provide a harmonized FTIR HCFC-22 retrieval strategy and a global NDACC-IRWG HCFC-22 data set, and (b), investigate the long-term trend of HCFC-22 based on the FTIR measurements together with other independent data sets. Section 2 gives a brief introduction to the data sets, including FTIR, NOAA flask, and ACE-FTS satellite observations. Section 3 presents the FTIR harmonized retrieval strategy and a trend analysis tool. In Section 4, the time series and the annual growth rate of the HCFC-22 measurements are discussed. Finally, conclusions are drawn in Section 5.

Table 1

Characteristics of NDACC FTIR Sites and Data Sets: Location, Altitude (in km a.s.l.), Time Coverage, Retrieval Code, Retrieval Microwindows (MW1: 828.75–829.4 cm^{-1} ; MW2: 1115.5–1116.1 cm^{-1}), Retrieval Uncertainty and Mean of the DOF

Site	Latitude	Longitude	Altitude [km a.s.l.]	Time coverage	Code	Retrieval window	Retrieval uncertainty (sys/ran)	DOF
Eureka	80.05°N	86.42°W	0.61	2006–2020	SFIT4	MW1	7.3/4.6%	1.11
Thule	76.90°N	68.77°W	0.02	1999–2022	SFIT4	MW1	7.3/3.5%	1.05
Harestua	60.22°N	10.75°E	0.59	1994–2022	SFIT4	MW1	7.2/3.2%	1.27
St.Petersburg	59.88°N	29.83°E	0.02	2009–2023	SFIT4	MW1 ^a	7.9/7.4%	1.06
Bremen	53.10°N	8.85°E	0.03	2004–2022	SFIT4	MW1	7.2/4.2%	1.07
Jungfrauoch	46.55°N	7.98°E	3.58	2003–2023	SFIT4	MW1	5.3/3.7%	1.40
Toronto	43.60°N	79.36°W	0.17	2013–2022	SFIT4	MW1	7.4/7.8%	1.03
Rikubetsu	43.46°N	143.77°E	0.38	1995–2023	SFIT4	MW1	5.3/5.6%	1.15
Boulder	39.99°N	105.26°W	1.63	2010–2022	SFIT4	MW1 + MW2	7.4/6.7%	1.01
Tsukuba	36.05°N	140.12°E	0.03	2014–2023	SFIT4	MW1 + MW2	7.5/8.0%	1.04
Izaña	28.30°N	16.50°W	2.37	1999–2022	PROFFIT9	MW1	8.7/6.6%	1.29
Reunion St-Denis	20.90°S	55.49°E	0.09	2004–2011	SFIT4	MW1 + MW2	7.9/5.3%	1.09
Reunion Maïdo	21.08°S	55.38°E	2.16	2013–2022	SFIT4	MW1 + MW2	7.4/3.6%	1.09
Wollongong	34.41°S	150.88°E	0.03	2000–2022	SFIT4	MW1 + MW2	6.6/4.4%	1.08
Lauder	45.04°S	169.68°E	0.37	1990–2023	SFIT4	MW1	7.1/3.2%	1.15
Arrival Heights	77.83°S	166.67°E	0.18	1997–2022	SFIT4	MW1	7.3/4.5%	1.06

Note. See details in Section 3.1. ^aThe retrieval window at St. Petersburg uses 828.75–829.38 cm^{-1} that slightly differs from the MW1.

2. Data Sets

2.1. Ground-Based FTIR

Sixteen NDACC-IRWG FTIR sites are involved in this HCFC-22 harmonization study, providing a good global coverage with latitudes ranging from 80.05°N to 77.83°S (Table 1 and Figure S1 in Supporting Information S1). Ground-based FTIR records the solar absorption spectra in the infrared region with a high spectral resolution of 0.0035–0.0075 cm^{-1} . The spectra are recorded under direct clear line-of-sight conditions. Therefore, no data are available for the high-latitude sites during the polar night. The HCFC-22 column can be retrieved from FTIR spectra by fitting specific absorption lines of HCFC-22 between 800 and 1,200 cm^{-1} . Most sites in this study have been recording spectra since the 1990s or 2000s, enabling the retrieval of a long-term HCFC-22 time series.

2.2. Flask Measurements

HCFC-22 mol fractions near the Earth's surface are measured by the NOAA HATS flask gas GC-MSD program, started in the early 1990s (Montzka et al., 1993; Vimont et al., 2022). It has a high precision and the reported standard deviation (SD) is about 0.5 ppt. The locations and time coverages of the NOAA 14 flask sites used are shown in Figure S1 and Table S1 in Supporting Information S1.

2.3. ACE-FTS Satellite Observations

The solar occultation measurements by the ACE-FTS instrument have been used to retrieve HCFC-22 vertical profiles with a vertical resolution of ~ 2 km in the upper troposphere—lower stratosphere (UTLS) region (5–25 km) globally since 2004 (Bernath, 2017; Boone et al., 2023). The latest ACE-FTS HCFC-22 v5.2 data sets, with a 1-km altitude grid, are used. Five microwindows between 804 and 830 cm^{-1} are adopted for the HCFC-22 retrieval, with an additional six microwindows are included between 802 and 2,977 cm^{-1} for interfering species. The ACE-FTS measurements has been validated recently, and the bias between the ACE-FTS HCFC-22 observations and MIPAS satellite, aircraft, and balloon data is about 0%–10% between 5 and 15 km and within 15% from 15 to 25 km (Kolonjari et al., 2024). In this study, we select the co-located ACE-FTS measurement above each FTIR site within $\pm 10^\circ$ latitude and $\pm 30^\circ$ longitude, for which at least 17 altitudes should be available among

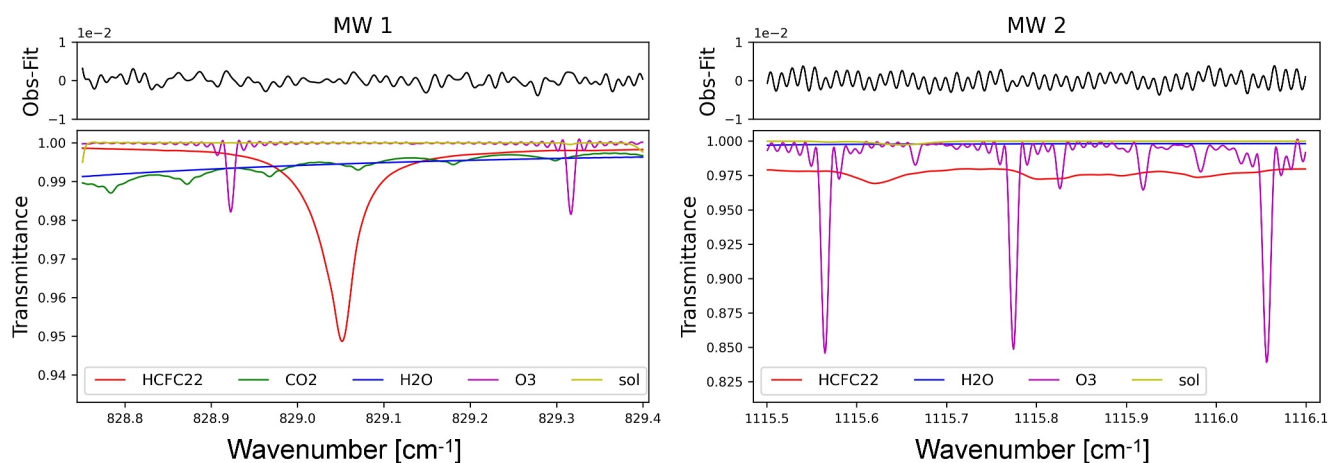


Figure 1. The transmittance of the fitted spectra, together with the fitting residuals in micro-windows 1 and 2 at Maido on 1st July 2018 with a solar zenith angle of 61.1°. For the fitted spectra, the transmittances from all species and the solar lines are plotted separately.

the 20 observed altitudes between 5 and 25 km after filtering out the poor-quality retrievals (Sheese & Walker, 2023).

3. Materials and Methods

3.1. FTIR Harmonized Retrieval Strategy

We use SFIT4 or PROFFIT9 Bayesian inversion codes (Rodgers, 2000) to retrieve HCFC-22 column. Both codes are widely used within the NDACC-IRWG, and their results show good consistency (Hase et al., 2004). The retrieval theory of the ground-based FTIR is illustrated Text S1 in Supporting Information S1. Two micro-windows (MWs) were selected (Figure 1), which include relatively strong HCFC-22 absorption lines and minimize interfering species: 828.75–829.4 cm^{-1} (MW1) and 1115.5–1116.1 cm^{-1} (MW2). The HCFC-22 retrieval at each NDACC site is either performed using only MW1 or combining two windows (MW1+MW2), depending on the spectral characteristics. In general, all sites tested MW1 and MW1 + MW2, and selected the better one in terms of fitting residual, number of spectral fits converging, and degree of freedom for signal (DOF). The retrieval window selection used at each NDACC-IRWG site is given in Table 1. Nevertheless, the mean difference between HCFC-22 columns at all sites derived from only MW1 and MW1+MW2 is less than 0.5%, which is negligible as compared to its retrieval uncertainty.

The spectroscopic parameters, that is, line intensity, air-broadened parameter, and self-broadened parameter, are the key elements in calculating the gas absorption cross-section. We generated a new pseudo HCFC-22 linelist (https://mark4sun.jpl.nasa.gov/data/spec/Pseudo/HCFC_22_Pseudo_Line_List.pdf) based on the cross-section spectra provided by Harrison (2016) and Clerbaux et al. (1993). Regarding interfering species, we tested several line lists, including HITRAN2016, HITRAN2020, ATM2012, ATM2016, and ATM2020, and finally used HITRAN2020 (Gordon et al., 2022) for CO_2 and H_2O , and ATM2020 (created by Geoff Toon) for O_3 , as these line lists allow us to get the lowest root-mean-square error (RMSE) of the fitting residual.

Mean profiles from the WACCMv6 (Gettelman et al., 2019) 61-year' simulations (1980–2040) are used to generate a priori profiles of HCFC-22, O_3 and CO_2 for each site. More information about the a priori profile of HCFC-22 is presented Text S2 in Supporting Information S1. As H_2O is extremely variable in the atmosphere, the NCEP reanalysis data (Kalnay et al., 1996) is applied to create the water vapor a priori profile based on the measurement location and time.

The vertical profile shape of HCFC-22 in the atmosphere is relatively stable. For chemically-inactive gases with weak absorptions, for example, CFC-11, CFC-12, HCFC-22, Polyakov et al. (2021) have tested both the optimal estimation method (OEM) and Tikhonov (TIK) for the FTIR retrieval at St. Petersburg. In case of the OEM regularization $\mathbf{R} = \mathbf{S}_a^{-1}$. Here, \mathbf{S}_a is the covariance of the a priori state vector. In case of TIK regularization, $\mathbf{R} = \alpha \mathbf{L}_1^T \mathbf{L}_1$ (Tikhonov, 1963). They found that the retrieved vertical profiles are more stable with the TIK

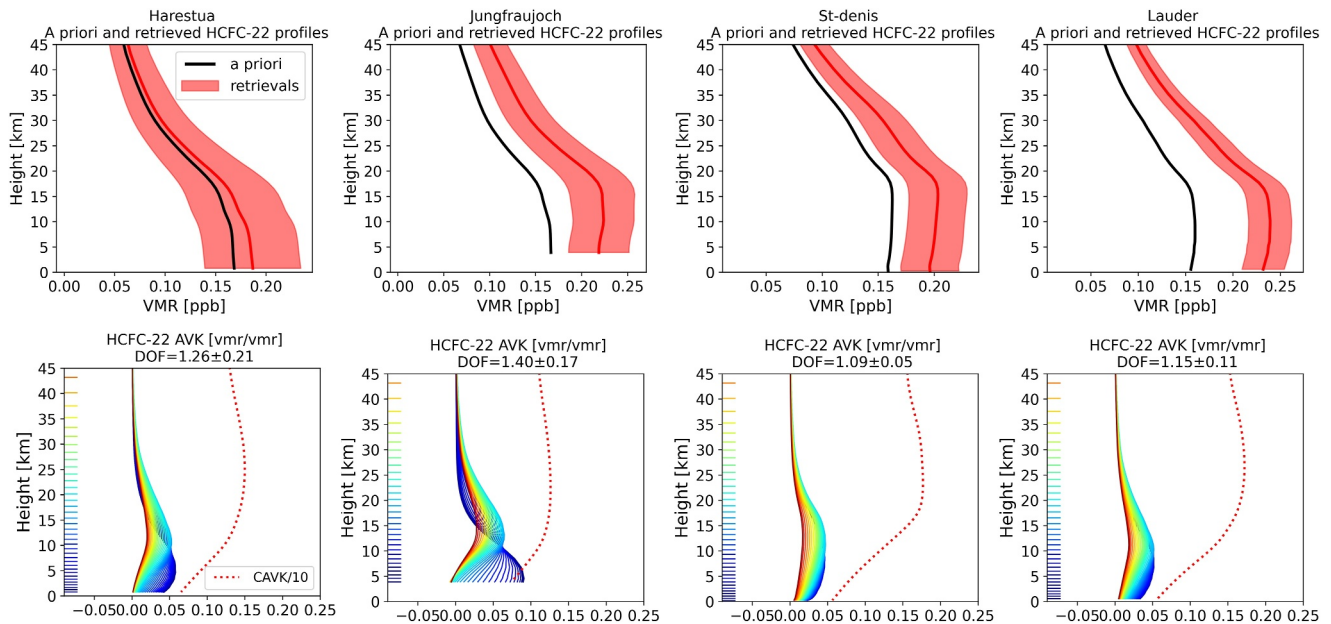


Figure 2. Top panels: The a priori and retrieved HCFC-22 profiles (mean and SD) at Harestua, Jungfraujoch, St-Denis and Lauder. Bottom panels: The profile averaging kernel matrix (AVK) and column averaging kernel vector (CAVK).

method. Similar experiments have been carried out at St-Denis, Maïdo and Lauder with the same results. Consequently, all NDACC sites here use the TIK regularization.

The key parameters applied for the NDACC FTIR HCFC-22 harmonized retrieval are summarized Table S2 in Supporting Information S1. To harmonize the data sets and reduce the impact of the surface pressure variation at each FTIR site, we derived the dry-air column-averaged mole fraction of HCFC-22 (XHCFC-22).

3.2. Trend Analysis Tool

The curve fitting method (Thoning et al., 1989) is applied to derive the annual growth rate of XHCFC-22 from the FTIR measurements and other reference data sets. The method is widely used for trend analysis. Here, we give a brief introduction.

The observation is fitted using the following function:

$$Y(t) = a_0 + a_1t + a_2t^2 + \sum_{n=1}^4 c_n [\sin(2n\pi t + \varphi_n)], \quad (1)$$

where $Y(t)$ is the fitted function, and t is the measurement time. The term $a_0 + a_1t + a_2t^2$ represents the polynomial regression part, and the term $\sum_{n=1}^4 c_n [\sin(2n\pi t + \varphi_n)]$ represents the yearly harmonics. After fitting the data with Equation 1, a Fast Fourier Transform (FFT) is applied to the residual to filter based on the short-term cutoff and long-term cutoff. The filtered data are then transformed back to the time domain with an inverse FFT. In this study, we set the short-term and long-term cutoffs to 10 and 4,000 days, respectively. More information is given in Text S3 in Supporting Information S1.

4. Results and Discussion

4.1. FTIR Retrieval and Its Uncertainty

As an example, Figure 2 shows the HCFC-22 a priori and retrieved profiles at Harestua, Jungfraujoch, St-Denis, and Lauder, together with their averaging kernels. The shape of the retrieved HCFC-22 vertical profiles is close to that of the a priori profile. The averaging kernels at all FTIR sites have a similar pattern, and the retrieved profile is

mainly sensitive to the troposphere and lower stratosphere. The mean DOFs of the HCFC-22 retrieved profile are between 1.0 and 1.4 (Table 1). Due to relatively low DOFs, we mainly focus on the retrieved column.

According to Rodgers (2000), the retrieval uncertainty of the FTIR HCFC-22 column is composed of 3 parts: (a) The smoothing error; (b) the forward model parameter error, and (c) the measurement error. Table S3 and S4 in Supporting Information S1 list the overview of the uncertainty estimates based on HCFC-22 retrievals at Reunion Maïdo (SFIT4) and at Izaña (PROFFIT9). The systematic and random uncertainties of HCFC-22 retrieved columns are 7.4% and 3.6% at Maïdo, and 8.7% and 6.6% at Izaña. The line intensity dominates the systematic uncertainty, and the random uncertainty mainly comes from the measurement error.

Uncertainty estimation has been carried out for all FTIR sites, with the same forward model parameter uncertainty inputs given in Table S3 in Supporting Information S1. The systematic and random uncertainties at all NDACC sites are 5.3%–8.7%, and 3.2%–8.0%, respectively.

4.2. Time Series and Annual Growth

The time series of the XHCFC-22 at 16 NDACC FTIR sites together with the co-located ACE-FTS satellite observations are shown in Figure 3. The XHCFC-22 level increases from below 100 ppt in the early 1990s to ~260 ppt in 2020. The scatter in the FTIR HCFC-22 retrievals at Toronto, Boulder, and Tsukuba are slightly larger than other sites, which is consistent with their relatively large random uncertainty estimates (>6%; Table 1). The Reunion data is the combination from Reunion St-Denis and Maïdo, as they are nearby (Zhou et al., 2018). The two data sets present a similar temporal pattern: an increase in XHCFC-22 is observed at all the FTIR sites for the whole time series, and the annual growth rate of XHCFC-22 has started to decline in recent years. We have also extracted the FTIR partial column between 5 and 25 km to compare with ACE-FTS in Figure S2 in Supporting Information S1 and got a similar result. In addition, the FTIR vertical retrievals can observe HCFC-22 vertical transport between the upper troposphere and lower stratosphere. The seasonal variations of XHCFC-22 between 8 and 25 km observed by FTIR are generally consistent with XN₂O, as well as ACE-FTS satellite XHCFC-22 measurements at Jungfraujoch and Wollongong (Figure S3 in Supporting Information S1).

Figure 4 shows the global annual growth rate of the XHCFC-22 derived from FTIR sites. Here, Toronto, Boulder and Tsukuba are not used due to their relatively short time coverages (Table 1). Similarly, the HCFC-22 annual growth rates are also derived from the NOAA flask measurements (all 14 sites) and co-located ACE-FTS satellite measurements. As the FTIR observes the whole column, it is not straightforward to compare it to the flask or ACE-FTS measurements directly. Nevertheless, it is assumed that the annual growth rates of HCFC-22 are consistent at the surface, in the UTLS region, and in the total column, given the long lifetime of HCFC-22 in the atmosphere (Kolonjari et al., 2024).

The three independent data sets observe similar time series of the annual growth rate. The 5-year growth rates of HCFC-22 are listed in Table S5 in Supporting Information S1. The annual growth rate of HCFC-22 varies with time, and is relatively large before 2016 with median values larger than 4.0 ppt/year. A reduction in the HCFC-22 annual growth rates has been observed in all the data sets in recent years. The mean and SD of annual growth rates are 5.70 ± 1.27 ppt/year, 5.28 ± 0.94 ppt/year, and 5.05 ± 0.67 ppt/year (2011–2015), 3.57 ± 1.39 ppt/year, 2.48 ± 0.72 ppt/year, and 3.19 ± 0.46 ppt/year (2016–2020), 2.15 ± 2.09 ppt/year, 1.42 ± 0.92 ppt/year, and 1.61 ± 0.39 ppt/year (2021–2023) from the FTIR, flask, and ACE-FTS measurements, respectively. A maximum HCFC-22 annual growth rate is observed by the NOAA flask data in 2008 with a mean value of 7.28 ± 0.52 ppt/year, and by the ground-based FTIR measurements in 2009 with a mean of 7.65 ± 1.39 ppt/year. The breakpoint in the annual growth rate around 2008 and 2009 is consistent with Graziosi et al. (2015).

Kolonjari et al. (2024) have derived the linear trend of the HCFC-22 mol fraction at 5.5 km altitude from the ACE-FTS satellite measurements between 2004 and 2018, and compared to the NOAA surface samplings and the Canadian Middle Atmosphere Model (CMAM39) simulations. They calculated the HCFC-22 annual growth rate before and after 2012. The HCFC-22 annual growth rate was found to be 6.9–7.8 ppt/year (2004–2012), decreasing to 3.1–4.7 ppt/year (2012–2018). The mean and SD of annual growth rates of XHCFC-22 derived from FTIR measurements are 7.14 ± 1.36 ppt/year (2004–2012) and 4.40 ± 1.94 ppt/year (2012–2018), respectively. Our harmonized NDACC-IWRG data set confirms the decreasing growth rate that was shown recently in these ACE-FTS satellite observations up to 2018, and also examines the most recent years, obtaining an annual growth rate of 2.31 ± 1.92 ppt/year (2019–2023).

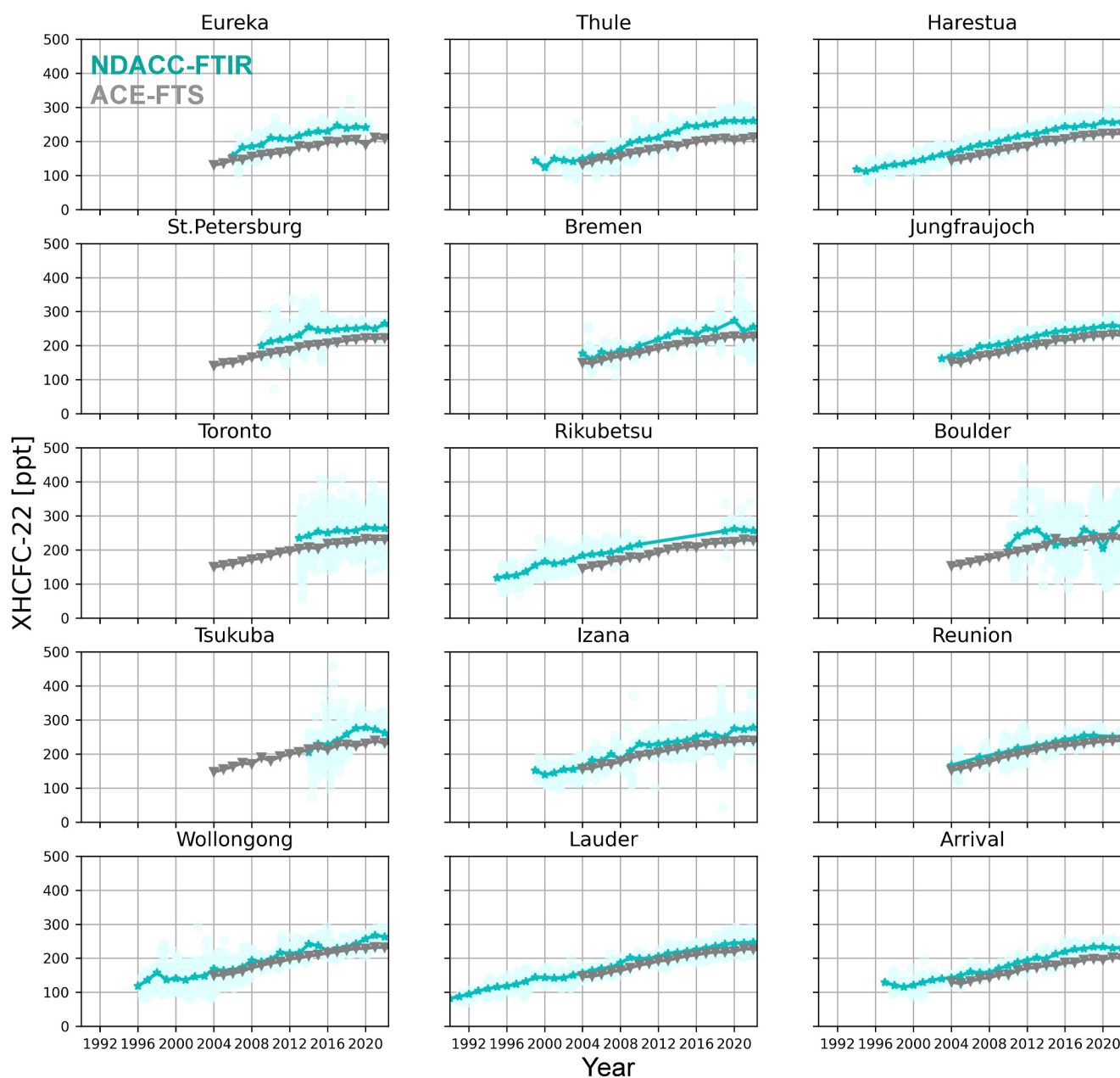


Figure 3. The time series of the individual retrieved XHCFC-22 (lightcyan dots) together with the FTIR (between the surface and 100 km) annual means (cyan symbols and line) and ACE-FTS satellite (between 5 and 25 km) annual means (gray symbols and line).

We further select four FTIR sites together with NOAA flasks at similar latitudes: Thule and Barrow, Jungfrauoch and Trinidad Head, Reunion and American Samoa, and Arrival Heights and South Pole. The HCFC-22 annual growth rates derived from the flask measurements are in good agreement with the XHCFC-22 annual growth rates derived from the FTIR measurements at all these sites (Figure S4 in Supporting Information S1), with a clear decrease in the annual growth rate in recent years. Moreover, as the sampling of the FTIR and flask in situ measurements are not the same, we also calculate the HCFC-22 annual growth rate based on their co-temporal weekly means. The results with and without temporal selection are consistent at these sites: the recent decrease in HCFC-22 annual growth rate is not affected by changing the temporal sampling of the data sets.

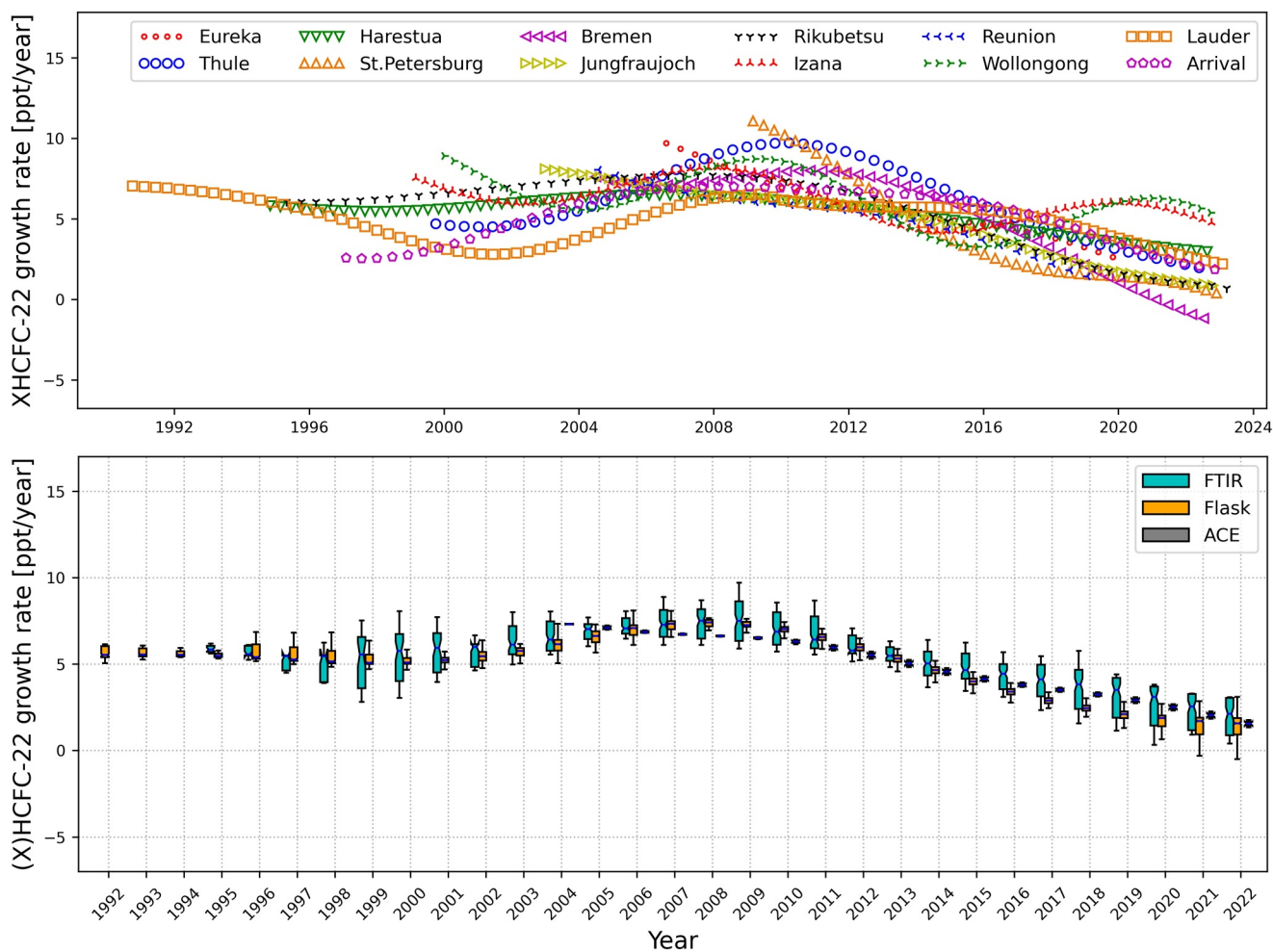


Figure 4. The annual growth rates of XHCFC-22 derived from the FTIR measurements at each site (upper panel) and a box plot of the HCFC-22 annual growth rates from FTIR sites, co-located ACE-FTS satellite measurements, and all NOAA surface flask observations in each year (bottom panel). The boxplot only shows the FTIR results after 1995, where at least 3 FTIR sites are available.

5. Conclusions

A long-term harmonized HCFC-22 column data set is generated at 16 NDACC-IRWG sites. The FTIR HCFC-22 retrieved column shows a good vertical sensitivity in the troposphere and stratosphere. The retrieval uncertainties are 5.3%–8.7% for the systematic component and 3.2%–8.0% for the random component across all FTIR sites. The annual growth rate of HCFC-22 is derived from the FTIR together with two other independent data sets (NOAA flask samplings and ACE-FTS satellite measurements). Excellent agreement among the three data sets is found, with a clear decrease in the growth rate of atmospheric HCFC-22 in recent years. According to the FTIR measurements, the maximum HCFC-22 annual growth rate was observed in 2009 with a mean of 7.65 ± 1.39 ppt/year, decreasing to 3.57 ± 1.39 ppt/year (2016–2020) and 2.15 ± 2.09 ppt/year (2021–2023). The harmonized NDACC FTIR HCFC-22 measurements, together with NOAA flask surface measurements and ACE-FTS satellite HCFC-22 measurements, confirm the decrease in the atmospheric HCFC-22 annual growth rate in recent years, which reveals another success of the Montreal Protocol. While the FTIR data are less precise than the flask samples, comparisons between the derived trends show that FTIR observations can capture the same changes as the flask samplings do. Moreover, the FTIR measurement technique offers column observations including both tropospheric and stratospheric information, which is complementarity to the surface measurements. NDACC-IRWG will be continued into the coming years and are demonstrated here to provide an excellent source to monitor the trend in HCFC-22 column. The unique long-term FTIR data set will provide new and critical constraints on the global HCFC-22 budget.

Data Availability Statement

The NOAA flask in situ measurements are publicly available via <https://gml.noaa.gov/hats/gases/HCFC22.html>. The ACE-FTS satellite HCFC-22 v5.2 observations are available by registration via <https://database.scisat.ca/level2>. The ACE-FTS data quality flags for v5.2 are available via Sheese and Walker (2023). The NDACC-IRWG FTIR XHCFC-22 data sets used in this study are publicly available via Zhou & NDACC-IRWG (2024). The code of the curve fitting method is publicly available via <https://gml.noaa.gov/aftp/user/thoning/ccgcrv/>.

Acknowledgments

This study is supported by the National key R&D program (2023YFC3705202). Measurements at Lauder and Arrival Heights are funded by NIWA through New Zealand's Ministry of Business, Innovation and Employment Strategic Science Investment Fund. Arrival Heights is supported by Antarctica New Zealand. ULiège team was supported by the Fonds de la Recherche Scientifique (F.R.S. - FNRS, Brussels, Belgium) [J.0126.21] and the GAW-CH program of MeteoSwiss. EM is a senior research associate with F.R.S. - FNRS. We thank the HFSJG, Bern, CH for supporting FTIR observations at Jungfraujoch. Eureka FTIR measurements were made at the PEARL by the CANDAC, and Toronto FTIR measurements were made at TAO; both are primarily supported by the Natural Sciences and Engineering Research Council of Canada, the Canadian Space Agency (CSA), and Environment and Climate Change Canada. The ACE-FTS is a Canadian-led mission mainly supported by the CSA. NDACC operations in Reunion are supported by the Université de la Réunion as well as the French regional and national organizations. Harestua FTIR measurements are supported by the Swedish Environmental Protection Agency. Rikubetsu and Tsukuba sites are supported by the GOSAT projects. The Rikubetsu site is funded by the joint research program of the Institute for Space-Earth Environmental Research, Nagoya University.

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