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First measurements of CFC-12 in 1951 at Jungfraujoch and comparison to current measurements and atmospheric models

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Abstract

We prove results on unique continuation at the boundary for the solutions of real analytic elliptic partial differential equations of the form

$$\sum_{i,j=1}^{n} a_{ij}(x)\frac{\partial^2 u}{\partial x_i \partial x_j} + \sum_{k=1}^{n} b_k(x)\frac{\partial u}{\partial x_k} + c(x)u = 0$$

This work is motivated by and generalized the main results of , (missing citation),(missing citation), X.Huang et al in ,(missing citation),(missing citation) and M.S Baouendi and L.P. Rothschild in (missing citation) Key words: Elliptic partial differential equation ; Hopf Lemma ; Unique continuation principle ; Real analytic hypersurface ; Real analytic functions

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References

First measurements of CFC-12 in 1951 at Jungfraujoch and comparison to current measurements and atmospheric models

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Key Points:

- We retrieved total column values of CFC-12 in 1951, using historical solar absorption spectra.
- We show the evolution of atmospheric CFC-12 from 1951 to present using different measurement and model results.
- The surface value from the historical spectra is larger than the emission based forward model estimates, but agrees within error bars.

Abstract

Chlorofluorocarbons (CFCs) have played a major role in the depletion of the stratospheric ozone layer. Understanding the historical trend of emissions and atmospheric concentrations is crucial for quantifying their environmental impact. The first direct measurements of atmospheric CFCs were performed *in-situ* by Lovelock in 1970. We have analysed historical solar absorption infrared spectra from 1951, recorded at the high Alpine Jungfraujoch site to retrieve the CFC-12 surface mole fraction. The results are compared to routine solar absorption measurements at the site starting in 1984 and emission-based forward models. The surface value from the historical spectra is 26.1 ± 18.5 ppt. This measurement agrees with model results within the error bars although the model values may be biased low due to unreported emissions. Our result represents the earliest atmospheric CFC measurement, predating Lovelock's detection by over two decades.

Plain Language Summary

This paper presents the first worldwide measurement of CFC-12 surface mole fraction obtained at Jungfraujoch in 1951. The study was performed using digitized paper-based solar spectra produced by a Pfund-type grating spectrometer. The surface mole fraction was calculated using modern retrieval methods, and compared with model predictions.

An analysis of errors is presented and compared with the reconstructed histories and AGAGE models. The comparison gives a concentration three times higher than the one predicted by the model. However, the model result and the reconstructed histories lie within uncertainty of measurement.

1 Introduction

The compound CCl_2F_2, more commonly referred to as CFC-12 or Freon-12™, was first synthesised by Thomas Midgley (Midgley & Henne, 1930) and is one of the most important atmospheric ozone-depleting substances (ODSs) regulated by the Montreal Protocol. It has an ozone depletion potential (ODP) of 1.0 and contributes around 1000pptv of current stratospheric chlorine loading ((WMO) Scientific Assessment of Ozone Depletion, 2022). It is also a potent greenhouse gas (GHG) with a Global Warming Potential (GWP) relative to CO_2 of 10900 over a 100-year horizon, one of the highest of the regulated gases (The Kigali Amendment, 2016).

CFC-12 was used for many decades due to its chemical non-reactivity and stability as a refrigerant and as a propellant. Most of the reported production and emissions of CFC-12 (and other regulated ODS) were reported by the Chemical Manufacturers Association (CMA, 1980). In 2016, the global emissions of CFC-12 were estimated to be approximately 33 ± 21 Gg yr^{-1}, while in 2020, these emissions were estimated to have reduced to around 25 ± 20 Gg yr^{-1} ((WMO) Scientific Assessment of Ozone Depletion, 2022).

In July and August of 1970, James Lovelock discovered trace amounts of a similar chlorofluorocarbon, $CFCl_3$ (CFC-11) and SF_6, using a new and very sensitive Electron Capture Detector (ECD) device when studying tropospheric haze over Ireland. Lovelock also tried to measure CFC-12 in the atmosphere. However, this was not possible due to its relatively low rate

of reaction with thermal electrons in the electron capture detector (Lovelock, 1971). Later, Molina & Rowland, (1974) proposed that CFCs would dissociate in the stratosphere when exposed to ultraviolet radiation, freeing chlorine, and causing a catalytic reaction cycle that destroys stratospheric ozone. This discovery was one prime motivation for the Montreal Protocol (MP), which proposed a gradual phase-out of these gases (United Nations Treaty Collection, 1989). The Atmospheric Lifetime Experiment (ALE) started measuring multiple gases including CFC-11 and CFC-12 in 1978 based on theoretical work of Cunnold et al., (1978) using ECD-based chromatographs.

Following a continued increase in its atmospheric abundance, the first slowing in the growth rate of CFC-12 was reported by Engel et al., (1998) based on *in-situ* balloon measurements. The subsequent global decline in CFC-12, albeit slow due to the relatively long lifetime of CFC-12 of more than 100 years, has contributed to the slow recovery of the ozone layer ((WMO) Scientific Assessment of Ozone Depletion, 2022). This global picture of declining emissions of the major CFCs suffered a setback due to non-compliant production of CFC-11 from 2012, which was reported by Montzka et al., (2018) and later confirmed using FTIR observations at Jungfraujoch and Lauder (Cantos et al., 2022). However, CFC-11 emissions now seem to have returned to values consistent with expectations (Montzka et al., 2021).

In this work, we present the retrieval of CFC-12 from historical solar infrared absorption spectra from a Pfund-type grating spectrometer together with FTIR spectra from both a home-made (since 1984) and a commercial instrument (Bruker, since the 1990s) from the International Scientific Station at Jungfraujoch, as well as an estimation of the surface concentration of this gas. The retrieved CFC-12 values are compared to reconstructed histories estimation of CFC-12 in 1951 as well as the AGAGE 12-Box Model (Rigby et al., 2013). The digitization and calibration of the historical spectra has been published by Makkor et al., (2024).

2 Materials and Methods

2.1 Retrieval theory

The inverse model follows the optimal estimation or Tikhonov regularization formalism proposed by Rodgers, (2000). It is a semi-empirical method of inverse calculation of the state of the atmosphere. The optimal estimation method (following Bayesian statistics) constricts the unknown state of the atmosphere to an *apriori* state vector x_a,

$$\hat{x} = x_a + A(x_t - x_a) + \epsilon$$

Where $\hat{x}$ represents the retrieved state, x_t is the true state of the atmosphere and A represents the averaging kernels matrix, which provides an assessment of the sensitivity of the retrieval over a given altitude range for a smooth profile. The retrieval requires an *apriori* profile that is usually constructed to represent the general state of the atmosphere (layers, temperature, pressure and gas profiles). The tool used to perform the retrievals is SFIT4 v1.0.21 developed by the University of Colorado, Boulder, and maintained by the University of Bremen, Germany. It is often used within the solar absorption infrared community of the Network for the Detection of Atmospheric Composition Change (NDACC; ndacc.org).

101 2.1.1 The Spectroscopy

102 The line-by-line and pseudoline lists adopted in this work were provided by the AMT20
103 (additional missing lines can be compiled from https://mark4sun.jpl.nasa.gov/pseudo.html),
104 where for each gas, the line position, the strength and other parameters are compiled into a
105 line list that is then used to calculate the simulated spectrum based on radiative transfer
106 theory. In this work CFC-12 from the historical spectra is retrieved from the micro window 1
107 (MW1) [920.1-923.6 cm^{-1}] and the FTIR spectra from both the aforementioned and the micro
108 window 2 (MW2) [1160-1163 cm^{-1}] (Mahieu et al., 2008).

109 2.1.2 Instrumental parameters

110 The instrumental line shape (ILS) function used for the grating spectrometer can be described
111 by a $sinc^2$ function. SFIT4 requires the ILS in the interferogram domain. A $sinc^2$ function in the
112 spectral domain corresponds to a triangular apodization in the interferogram domain (Griffiths,
113 2006). It causes an artificial line widening and reduction in intensity. The overall resolution of
114 the Pfund-type grating spectrometer is usually between 0.1 and 0.4 cm^{-1} depending on the
115 wavenumber range and the grating used. However, in the retrieval, it must be considered that
116 the resolution of a grating spectrometer is wavelength dependent. The FTIR spectra from
117 Jungfraujoch are usually much lower in noise and higher in resolution than the grating-based
118 ones. The apodization function used during the retrieval is a boxcar function. The resolution is
119 0.012 cm^{-1} (OPD of 82 cm) for the home-made spectrometer and 0.008 cm^{-1} (OPD of 120 cm)
120 for the Bruker FTS-120HR.

121 The pressure, temperature and a priori water vapour profiles are obtained from the National
122 Centers for Environmental Prediction reanalysis (NCEP; (Kalnay et al., 1996)) and are extended
123 above 55 km using data from the Whole Atmosphere Community Climate Model version 7
124 (WACCMv7).

125 **3 Atmospheric Measurements at Jungfraujoch and models used**

126 3.1 Atmospheric measurements at Jungfraujoch

127 3.1.1 Grating measurements at Jungfraujoch

128 In 1949, Professor Migeotte and his team built and used a Pfund-type infrared grating
129 spectrometer with a 1-metre focal length, which was equipped with a PerkinElmer thermopile
130 detector, to record the solar spectrum at Jungfraujoch (46.55°N, 7.98°E, 3.58 km above sea
131 level) (Migeotte, 1949). The recorded spectra were saved on paper rolls using an electronically
132 guided pen. Altogether, these paper spectra covered a wavenumber range from 421 to 3571
133 cm^{-1} and contained handwritten atmospheric metadata such as surface temperature and
134 humidity (Migeotte et al., 1956). Spectra from 1951 have been scanned, digitized and calibrated
135 to the proper wavenumber range, and saved in a machine readable format (Makkor et al.,
136 2024). An example of the resulting digitized spectrum covering the retrieved region is shown in
137 Figure 1.

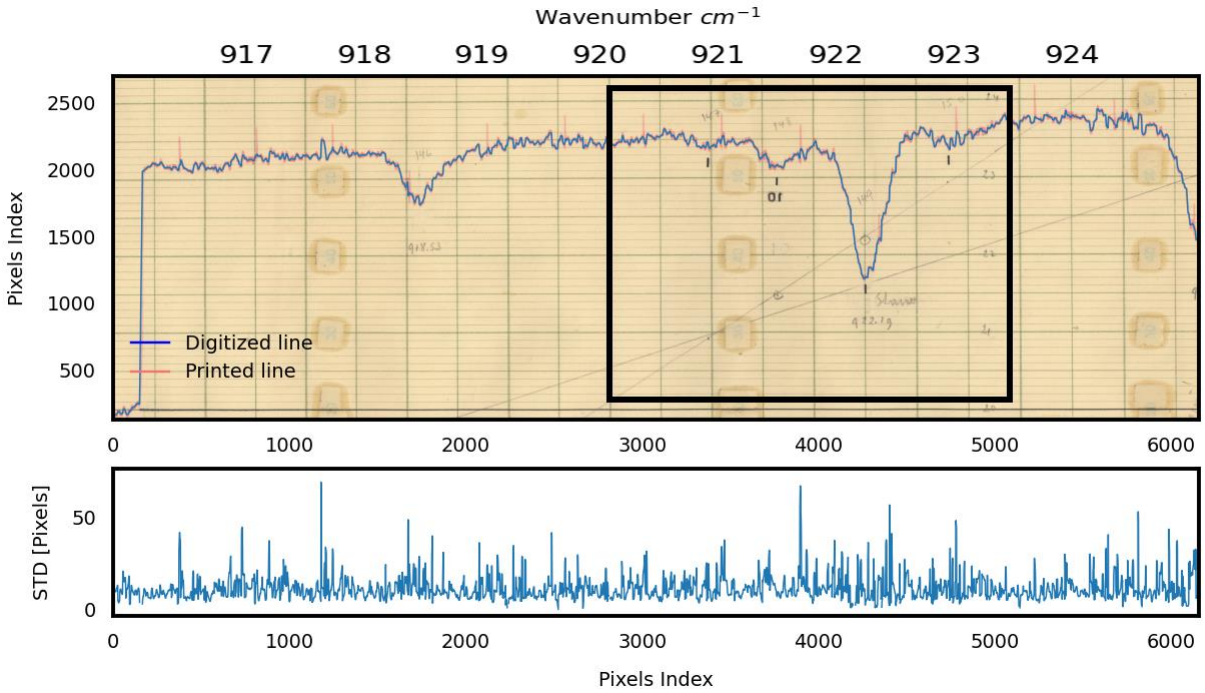


Figure 1: The original spectrum from which the CFC-12 surface value was retrieved. This spectrum was digitized and calibrated using the procedure described in Makkor et al (2024). The lower panel shows the standard digitization deviation of a 9.9 pixel average. The black rectangle shows the retrieval region used. 1

138

3.1.2 FTIR Measurements at Jungfraujoch

The first instrument (following the Pfund-type grating instruments at the same site) that allowed routine measurements of atmospheric spectra at Jungfraujoch was a redesign of an earlier Connes-type Fourier Transform Infrared (FTIR) spectrometer (Zander et al., 2008). By changing from a step-scan mode to a continuous scan mode, the scan speed increased by 100-fold. In addition to this, the instrument's optical length was doubled reaching a resolution of 0.0025 cm^{-1}. These changes allowed the start of recording atmospheric spectra regularly until the instrument was retired in 2008. In the early 1990s, the University of Liège team deployed and started to use a Bruker IFS 120HR instrument achieving a resolution of 0.001 cm^{-1}, and recorded spectra alongside the home-made. This instrument was also decommissioned in 2024, to be replaced with a Bruker IFS 125HR spectrometer.

3.1.3 Reconstructed histories of CFC-12 and AGAGE model

The first model-reconstructed histories of CFC-11 and CFC-12 were performed by Walker et al., (2000) from a two-box atmospheric model using estimates of annual atmospheric halocarbon release, following the work of Bullister & Weiss, (1983). These estimates extend from 1935 to 1998 for CFC-12.

The 12-Box AGAGE model is a simplified advection-convection model developed by the AGAGE group that uses emission estimates to calculate the atmospheric mole fractions of multiple gases such as CFC-11, CFC-12 and others (Rigby et al., 2013). The emissions for this model have been calculated up to the year 2024 using the approach provided by Lickley et al., (2020), which employs the so-called Bayesian melding method developed by Poole & Raftery, (2000). This technique enables the estimation of emissions by leveraging information from both observed concentrations and the mechanistic simulation model of the bank, emissions, and concentrations.

CFC-12 (as well as CFC-11) production data have been reported since 1975 by the CMA and the Alternative Fluorocarbons Environmental Acceptability Study (AFEAS), covering most major global producers. However, these reports exclude significant production from non-reporting companies, particularly in the former Soviet Union and Eastern Europe. Emission estimates incorporating these unreported sources were later provided by Mccarthy et al., (1977) who estimated the total world annual production and subsequent world atmospheric emission from 1930 onwards.

4 Results

4.1 Estimation of CFC-12 level in 1951

The MW1 was used in the retrieval of CFC-12 from the digitized historical spectra (see Figure 2). This microwindow is normally significantly influenced by water vapour, making it inappropriate for ground-based retrievals at sea level but suitable for sites located at higher altitudes like Jungfraujoch (3.58 km above sea level) where this influence is less critical. MW1 and MW2

showed the best agreement and minimum intra-day variability in the total column values when adopting 300 and 65 for the Tikhonov regularization parameter, respectively. In MW1 the spectral bands of CO_2 and CFC-12 overlap, which requires a careful retrieval of CO_2 to get CFC-12 correct. The figure shows the measured and calculated spectra with a relatively good fit, albeit with high noise.

Figure 3 shows all of the retrieved gases alongside CFC-12. The main contributions in this spectral interval come from H_2O and CO_2, but there is also a small and quantifiable contribution from CFC-12 in this spectrum (black dotted curve in Figure 3). This is not surprising since the production of CFC-12 started in the 1930s, and the compound was subsequently widely adopted by various industries as a foam agent and propellant. The early emissions of CFC-12 mostly came from the United States and various European countries. The CFC-12 was detected in particular a the less noisy spectrum with an absorption strength slightly below 1%. The retrieval of CFC-12 from MW2 using the grating spectra was not possible due to significant influence from the noise.

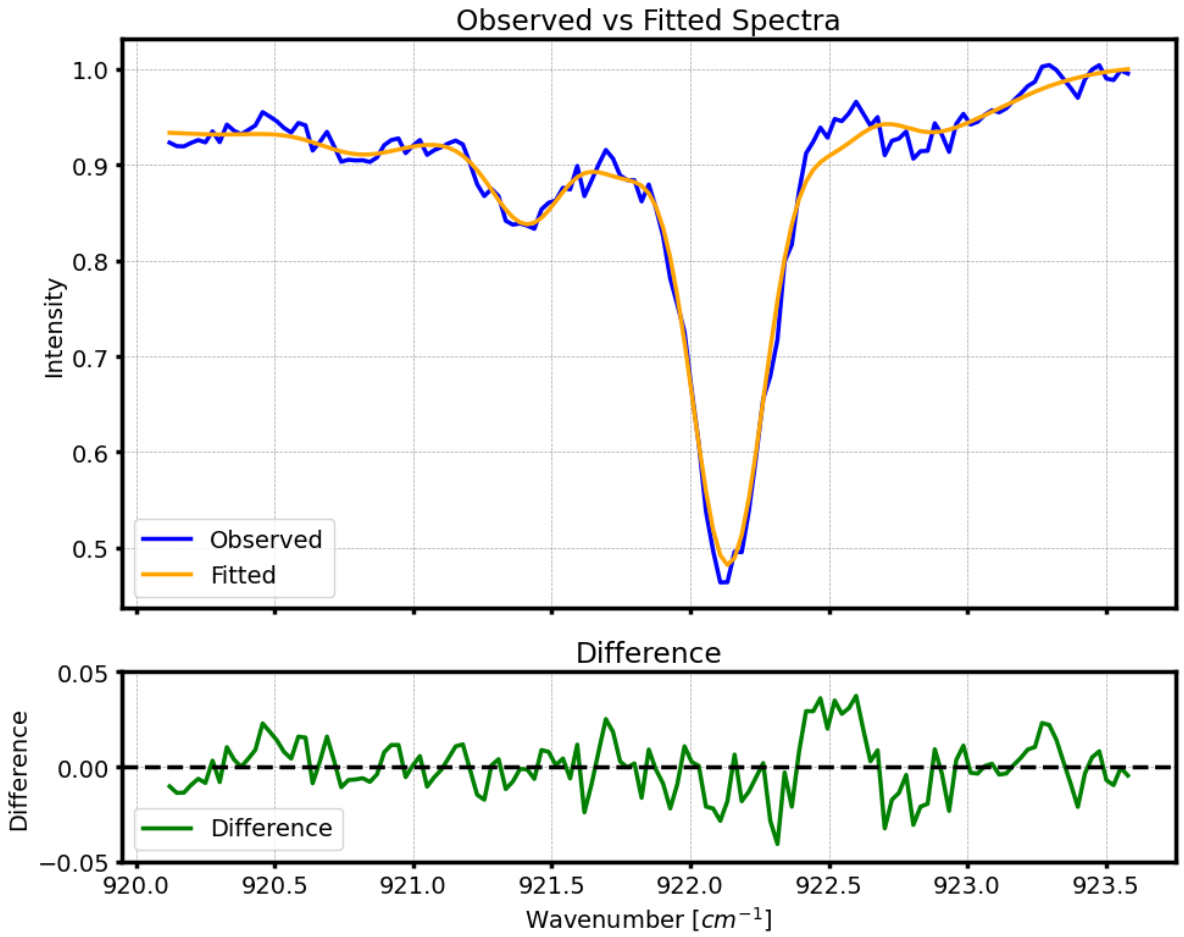
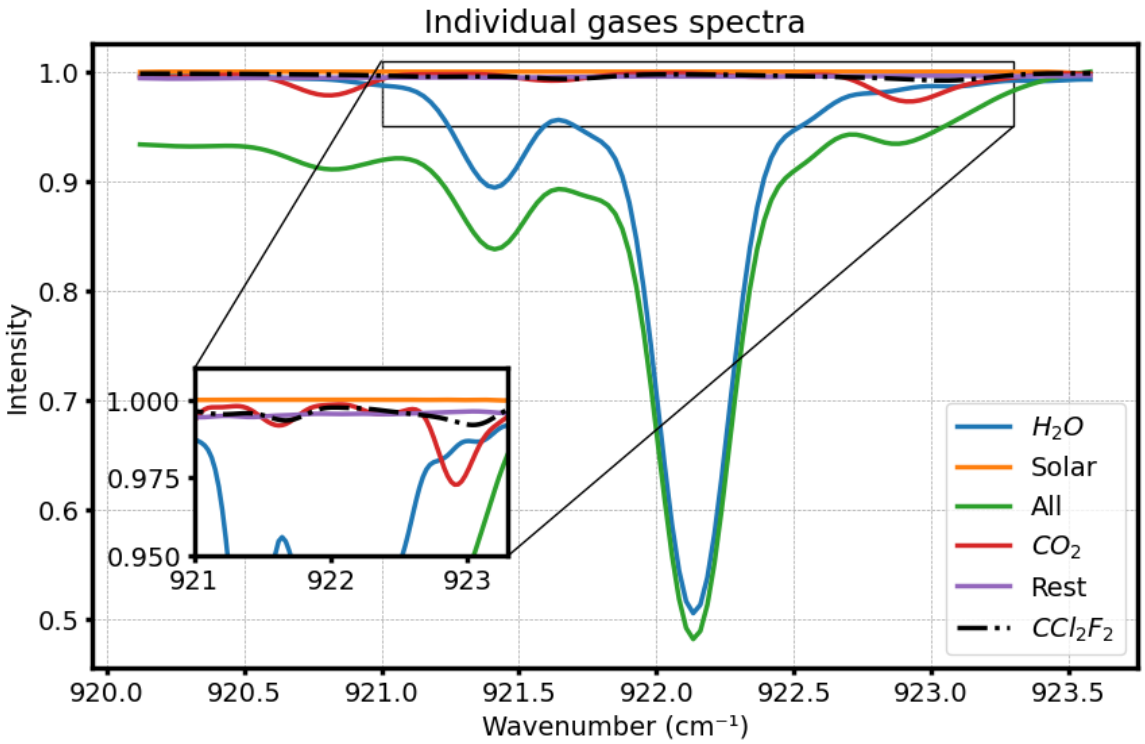


Figure 2: Fitting of the grating spectrum compared to the forward model calculated spectrum. This retrieval gives a CFC-12 surface value of 26.1 ppt. The lower panel shows the difference between the modeled and the observed spectrum. An optimal resolution of 0.277cm-1 and a Triangular apodization were used.



193 *Figure 3: All the retrieved spectral lines from the old spectra. CFC-12 absorption although weaker is still present even on spectra from 1951 with an absorption strength of about 1%. The small zoomed-in window allows a closer look at the CFC-12 line (dotted black). The retrieval was performed using Tikhonov regularization facilitated by SFIT4 spectral fit routines.*

194 ## 4.2 Error Analysis

195 ### 4.2.1 Error estimation of spectra from 1951

196 Quantifying the sources of errors in Pfund-type instrument from the 1950s involves considering
197 two main categories of errors: those originating from the instrument itself and those arising
198 from the paper spectra after processing (which includes digitization and calibration). One
199 source of random error associated with the instrument, the zero transmission level offset,
200 accounts for approximately 3% of the error (Zander et al., 1994). The resulting spectra have two
201 error sources: a digitization error of around 1.55% and a wavenumber shift error of about 1%.
202 The largest source of error comes from the retrieval itself. The total random error is about
203 48.3% (retrieval, digitization and instrumental errors) and total systematic error of about 21.6%
204 (see Table 1). The resolution of the historical spectra are not well known, but they are between
205 0.1 and 0.4 cm^{-1}. Multiple retrievals were therefore performed for different resolutions. The
206 resolution of 0.277 cm^{-1}, which has lowest calculated RMS was used in the retrieval.
207 Additionally, the bias arising from the low resolution and triangular apodization when using the
208 grating spectra was analysed. To achieve this, high-resolution FTIR spectra from the
209 Jungfraujoch site were artificially lowered to resolutions of 0.1, 0.25 and 0.4 cm^{-1} (minimum,
210 average and maximum resolutions) by truncating the Fourier transform of the high-resolution
211 (HR) spectrum using the Bruker OPUS software (OPUS - Spectroscopy Software, n.d.).
212 Subsequently, CFC-12 total columns were retrieved using these resolution values, then
213 compared to the original values from the HR FTIR spectra. The triangular apodization was also
214 applied to the lowered resolution FTIR spectra (See Table 2).

215

216 ### 4.2.2 Error estimation of Jungfraujoch spectra

217 The largest source of error for the FTIR spectra is from temperature with a random and
218 systematic error of 2.34% (in total). The random smoothing error $x_t - x_a$ is calculated from
219 WACCM covariance matrix. The uncertainty coming from the solar zenith calculation is about
220 0.1° for Jungfraujoch.

221

	Digitized 1951 grating spectrum	Bruker FTIR [1996 – present] spectra
	Random (%)	
Zero level offset	3.0	0.2
Temperature	0.72	1.34
Solar zenith angle	0.11	0.7
Phase function	-	0.28

Telluric species interference	35.6	0.02
Measurement	21.7	0.34
Digitization/Calibration	2.55	-
Total random	48.3	1.64
	Systematic (%)	
Zero level offset	3	0.2
Spectroscopy (ATM19)	2.0	2
Temperature	4.8	1.0
Interfering species	21.5	-
Solar zenith angle	1.19	0.21
Phase function	-	0.28
Total systematic	21.6	2.31

Table 1: Error summary of the retrieved CFC-12 values from the old 1951 grating data and the modern FTIR spectra.

Resolution [cm^{-1}]	0.1	0.25	0.4
Relative difference [%]	0.65	1.0	2.5

Table 2: Relative difference in the retrieved CFC-12 value between the original high resolution FTIR spectra and the resolution-degraded ones.

4.3 The full trend of CFC-12

Figure 4 shows the long-term trend of CFC-12 from 1951 to present using various datasets and model results. The reconstructed historical model values represent the assumed CFC concentrations for the selected period between 1949 and 1960 (represented in the region of interest by the blue markers in Figure 4) from estimates of annual industrial production and emissions (Walker et al., 2000). The calculated CFC-12 concentration from these histories in 1951 is 9.2 ± 0.2 ppt for the northern hemisphere. This reconstructed historical value have never be validated using actual measurements, because no measurements existed. The surface value we derived for 1951 of 26.1 ± 18.5 ppt from the spectra is 3 times larger than the reconstructed historical estimate for the northern hemisphere. These differences can be explained by inaccurate emissions in 1951, but the uncertainty of the measurement from the old instrument needs to also be considered. The concentration of CFC-12 from the old grating measurement was taken from the value given by the SFIT4 at the surface during the retrieval of the total column (which has a value of $3.36 \times 10^{14} \pm 1.58 \times 10^{14}$ molecule /cm^2). The dry air mole

fractions from the FTIR measurements were also plotted and their trend fitted using a Dynamic Linear Model facilitated by the tool provided by Hachmeister et al., (2024). Additionally, the whole time series was complemented with the 12-Box AGAGE model with a CFC-12 concentration value of 7.0 ± 0.28 ppt for 1951 to give a complete picture of the evolution of CFC-12 over the past decades and to account for the missing data between the 1950s and the start of FTIR measurements at Jungfraujoch.

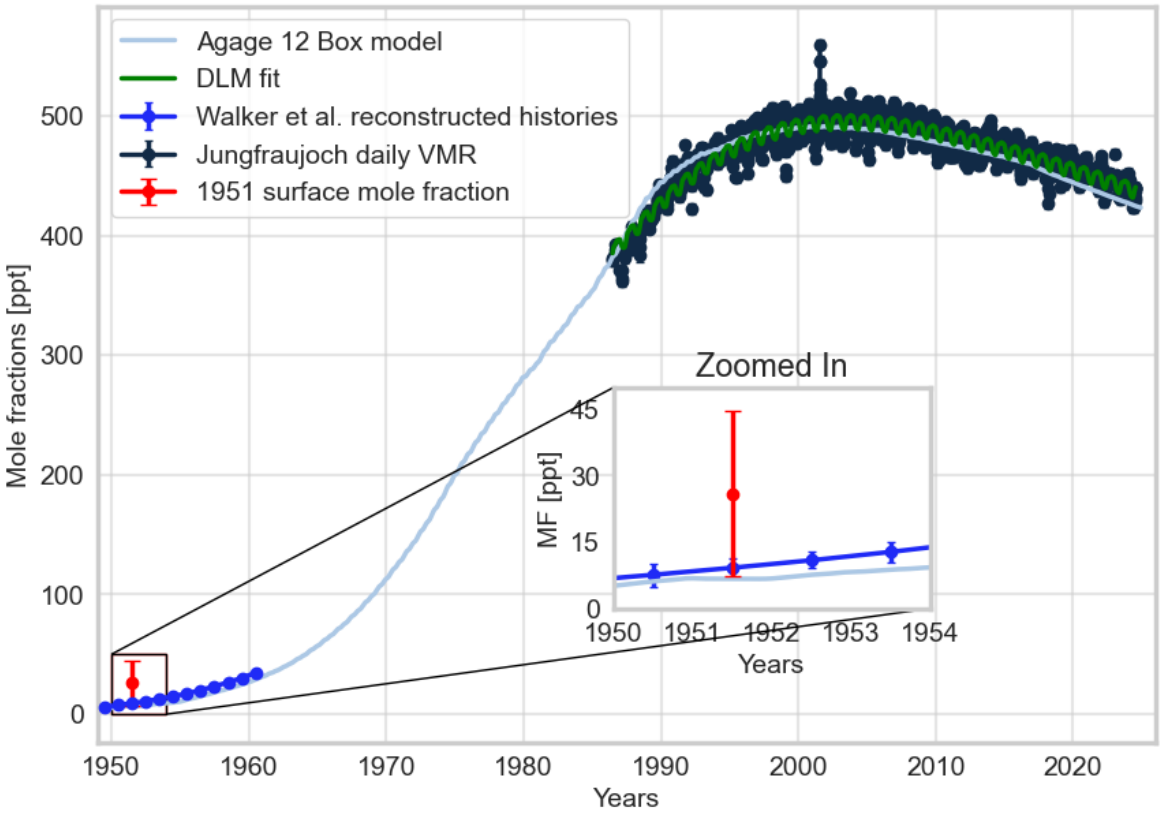


Figure 4: Atmospheric surface CFC-12 mole fraction (ppt) from 1951 to present. The measurement value in 1951 is retrieved in this work from solar measurements at Jungfraujoch in the same time period. It is compared to a 2-Box model reconstructed histories and 12-Box model from AGAGE. The retrieved mole fraction from FTIR measurements at Jungfraujoch are also represented and fitted using a Dynamic Linear Model (DLM).

5 Conclusions

A CFC-12 surface mole fraction of 26.1 ± 18.5 ppt for 1951 was derived from a grating spectrometer at Jungfraujoch. Using results from a home-made FTIR-spectrometer (from 1984 to 2008) and from a commercial FTIR-spectrometer (from the beginning of the 1990s to present), the interannual trend for the period 1951 – 2024 has been presented. The result for 1951 has been compared to estimates from reconstructed histories and AGAGE 12-Box models. The estimated error value includes the preprocessing error (digitization and calibration) of the historical paper spectra, error inherent to the instrument (finite S/N ratio) as well as the retrieval error. The largest error source comes from the retrieval due to the weak signal-to-noise ratio of the historical spectra.

The reconstructed histories and AGAGE 12-Box model estimates of CFC-12 for the northern hemisphere in 1951 of about 9.2 ± 0.2 and 7.0 ± 0.28 ppt, respectively, are about 3 times lower than the retrieved surface value from the historical grating instrument, but the error bars overlap. We note, however, these values could be biased low due to erroneous emission estimates in the 1950s.

The historical measurement has allowed us to retrieve the earliest atmospheric values of CFC-12, predating previous earliest values by over two decades. The historical spectra also cover other spectral regions, allowing the future retrieval of other trace gas concentrations in 1951.

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Author contributions

JN and MPC supervised the work, MP provided the retrieval software and supervised the work, MB and ZW edited the work. ZW provided advice on modelling. IPC and EM provided the CFC-12 data retrieved from the Jungfraujoch FTIR spectra. JM retrieved the grating spectra, ran the models/comparisons and authored the letter. All co-authors provided comments and suggestions.

Availability and data statement

The FTIR measurements are regularly provided and updated by the GIRPAS team and can be freely downloaded from https://www-air.larc.nasa.gov/pub/NDACC/PUBLIC/stations/jungfraujoch/hdf/ftir/ (last accessed 11/06/2025)

The digitized grating spectra can be freely downloaded under https://zenodo.org/records/14537672 (last accessed 11/06/2025)

SFIT4 retrieval program and related tools can be freely downloaded under https://wiki.ucar.edu/spaces/sfit4/pages/402424651/SFIT4+Version+1.0.xx+Release (last accessed 11/06/2025)

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