Improved FTIR retrieval strategy for HCFC-22 ($CHClF_2$), comparisons with in situ and satellite datasets with the support of models, and determination of its long-term trend above Jungfraujoch

Prignon et al., 2019, ACPD
URSPHERES day
14-06-2019, Liège

Outline

- FTIR observations at **Jungfraujoch**
- FTIR observation **technique**
- **Context**: halogenated compound in the atmosphere
- **Results**
- **Conclusion**

FTIR observations at Jungfraujoch (Swiss Alps)

1970

1974:
1^{st} FTIR
Spectrometer
(homemade)

1984:
Observations on a regular basis
→ 10 000 spectra recorded in 800 days of observations until 2008

1990:
Bruker 120HR FTIR spectrometer installed
→ 45000 spectra recorded in more than 2700 days of observations until today

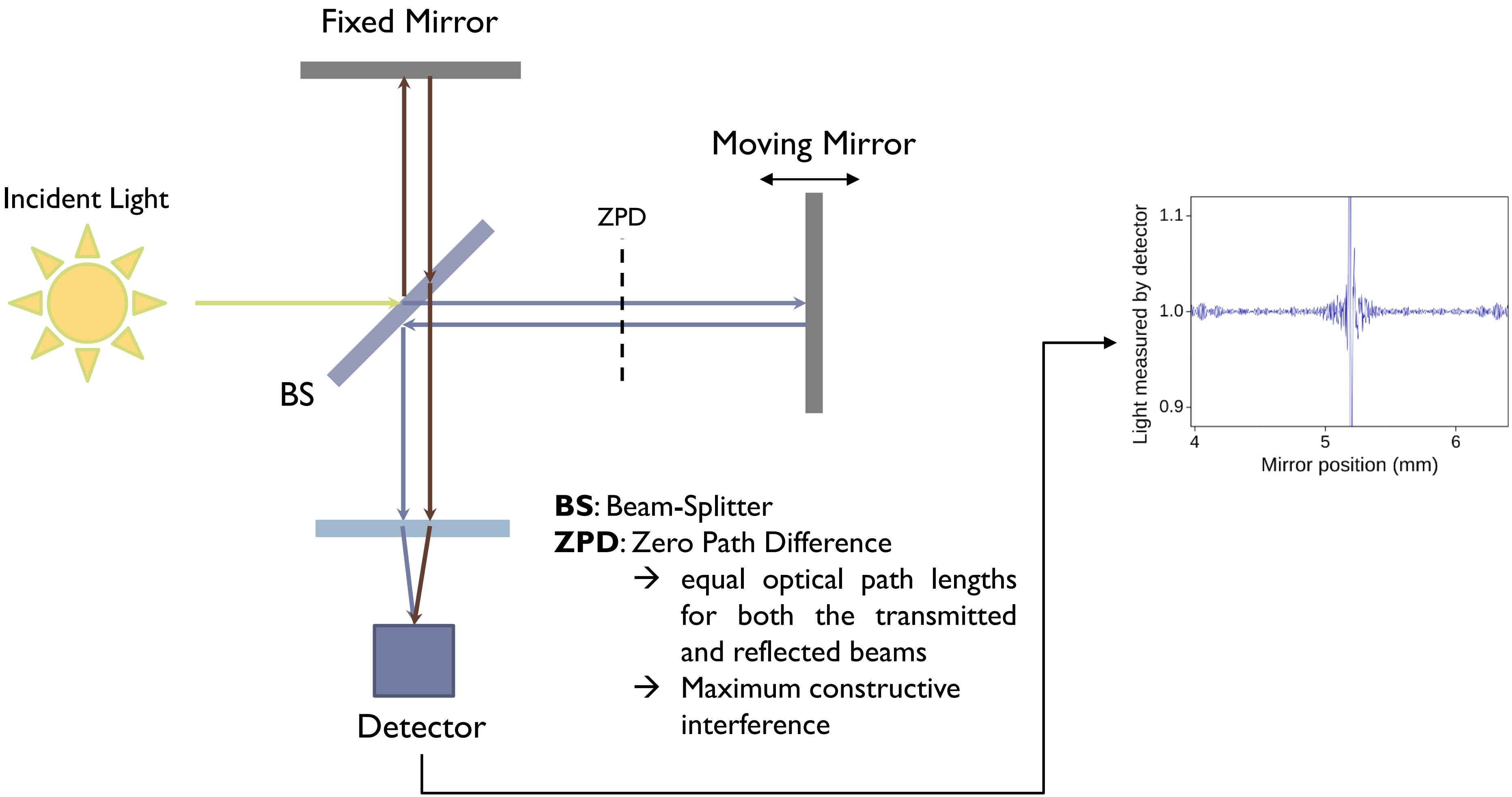
2008:
Bruker remotely controlled from Liège

Homemade instrument out of order

35 years of measurements with no significant interruption

➔ Longest FTIR time series in the world!

FTIR technique: Michelson interferometer

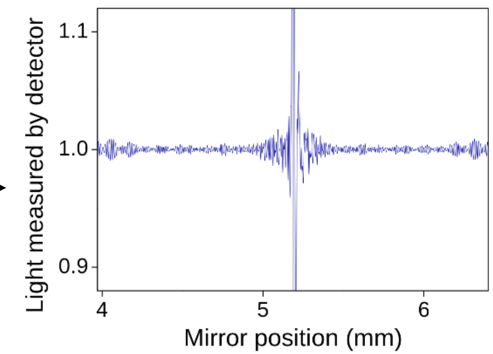


BS: Beam-Splitter
ZPD: Zero Path Difference
→ equal optical path lengths for both the transmitted and reflected beams
→ Maximum constructive interference

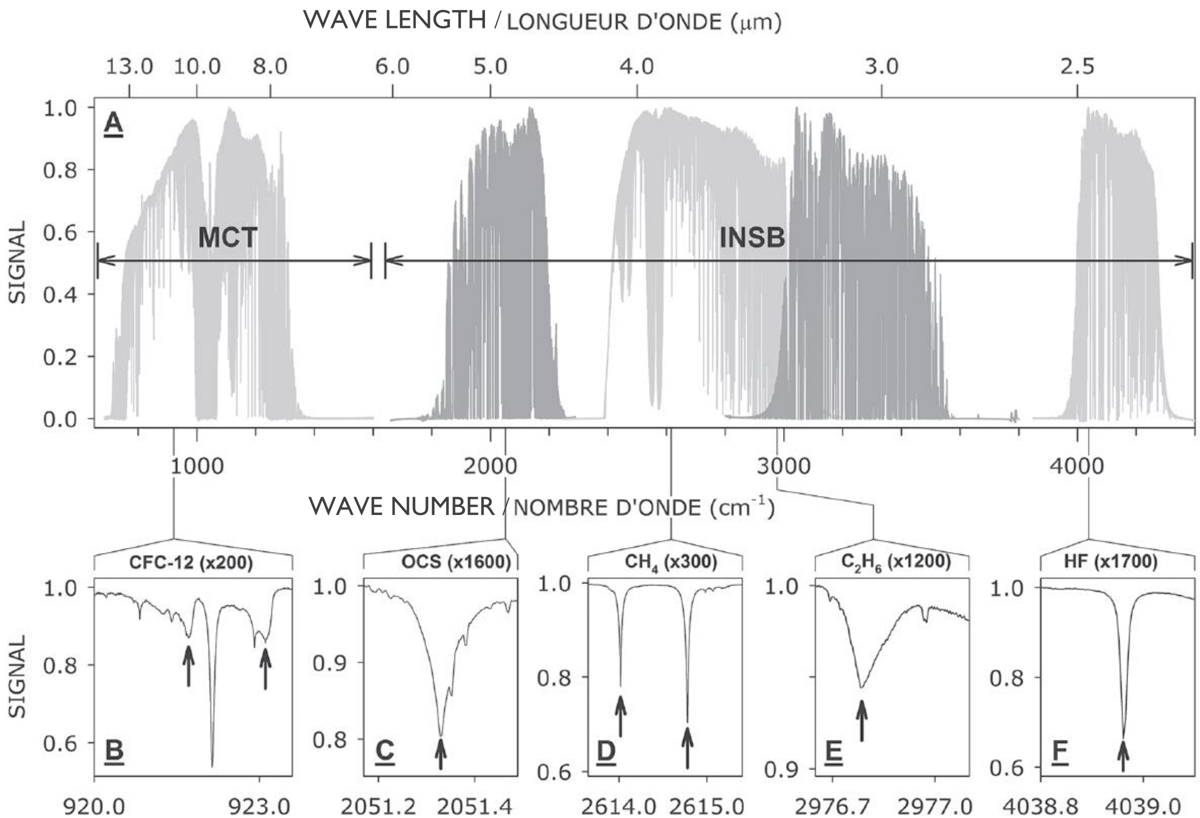
erometer

Inci

lengths
nsmitted
s
tive



FTIR technique: Spectra / Detectors / Filters



Mahieu et al., 2017, BSGLg

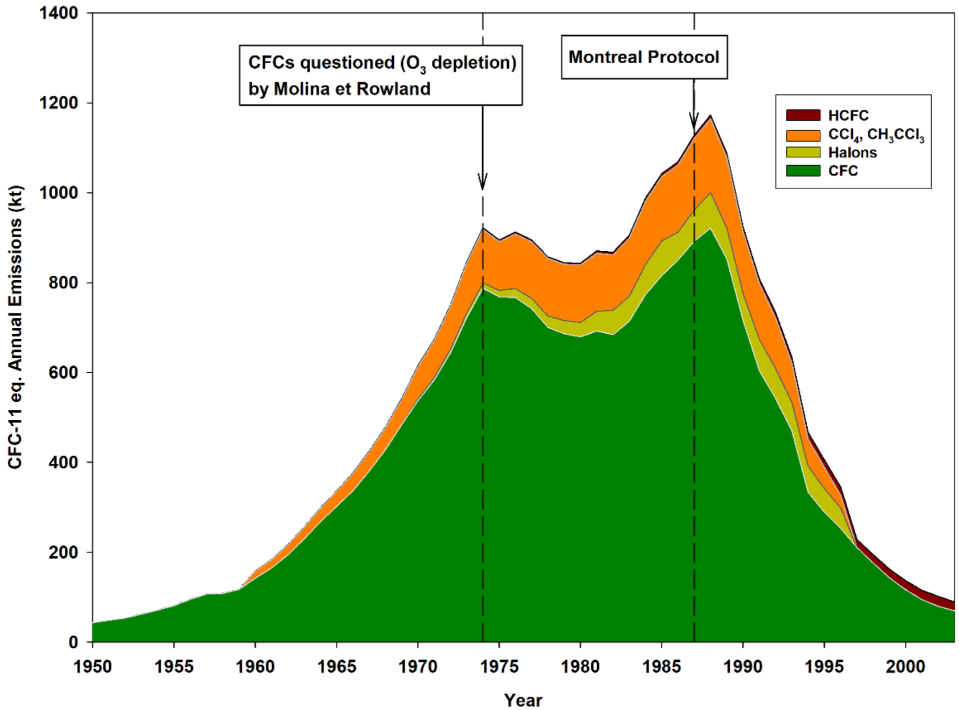
- 0.003 cm^{-1} of spectral resolution
- Absorption features associated to gases that we still have to discover
- Sensitivity from ground to ~50km (depending on the target species)

Halogenated Compound in the atmosphere

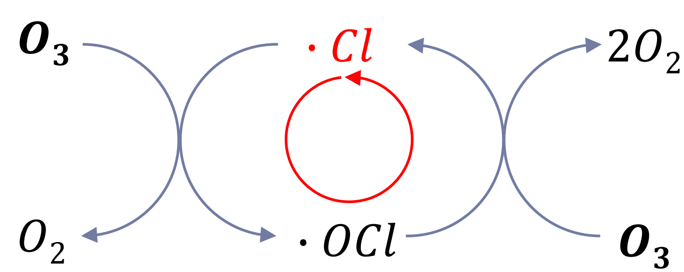
- CFCs (chlorofluorocarbons) intensively produced in the 20th century as refrigerant, blowing agent and propellant because of their low toxicity, reactivity and flammability.
- **1974**: Molina and Rowland + Stolarski and Cicerone:

  $CFC + UV \rightarrow X + Cl + F$

  → Cl leading to the catalytic destruction of stratospheric ozone (O_3)



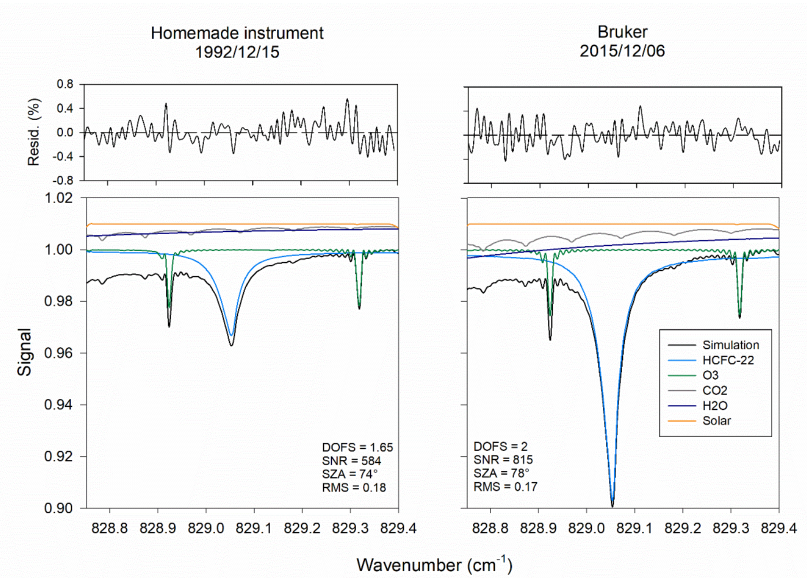
$$O_3 + \cdot Cl \rightarrow O_2 + \cdot OCl$$
$$O_3 + \cdot OCl \rightarrow 2O_2 + \cdot Cl$$



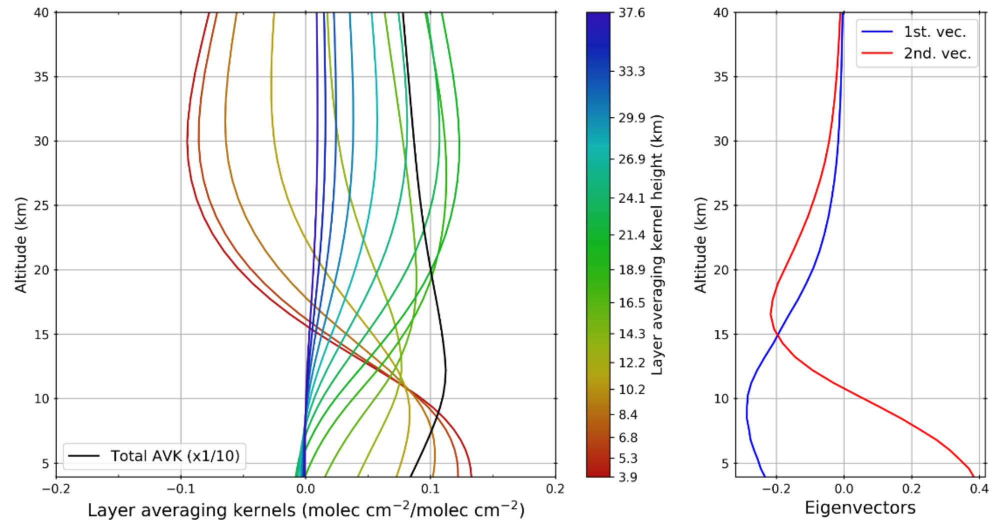
1987: Montreal Protocol on Substances that deplete the Ozone Layer (entered into force in 1989)

→ HCFCs (weaker ozone depleting potential than CFCs) on the rise in the late 80's

HCFC-22 Retrievals

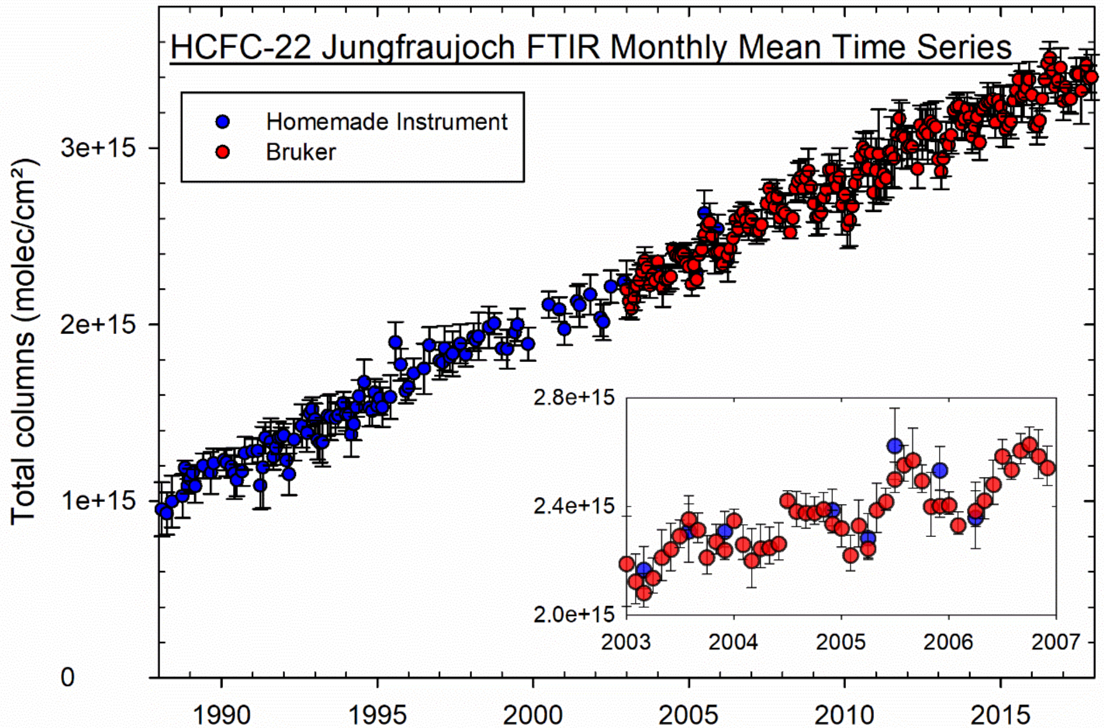


- Relatively good results from the homemade instrument despite the weaker absorption (less HCFC-22 in the atmosphere at the time it was operated) and nosier spectra



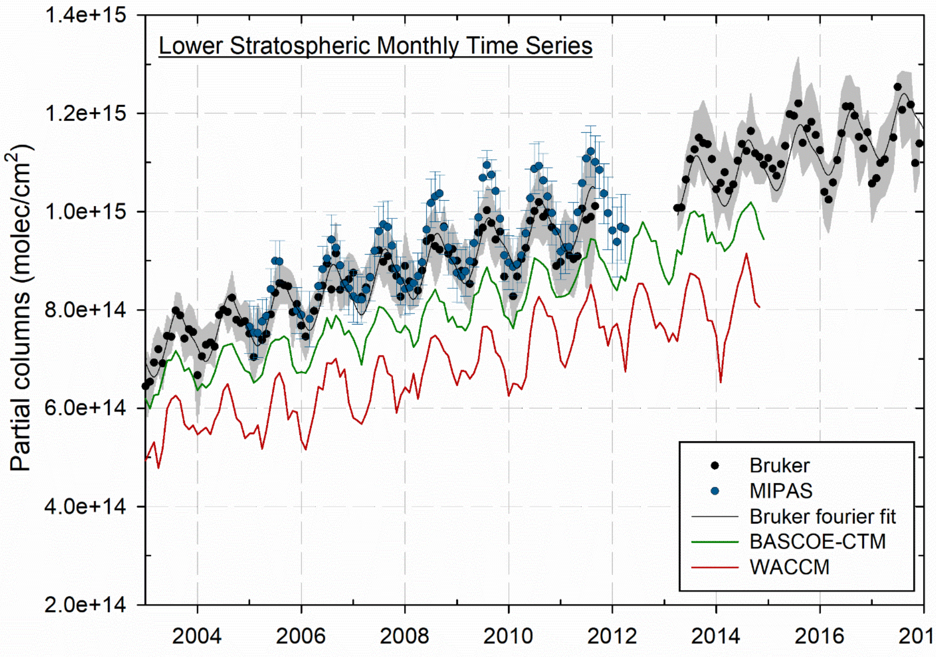
- Eigenvectors tell how independent is the retrieval from the HCFC-22 a priori profile assumed for the inversion.
- The intersection of a vector with the 0 value defines the partial columns limits
- 2nd vector has a value of 0.85, meaning that **tropospheric** and **lower stratospheric columns** (<11.21km<) can be extracted with 85% of information coming from the retrieval itself

Results: HCFC-22 Total Columns

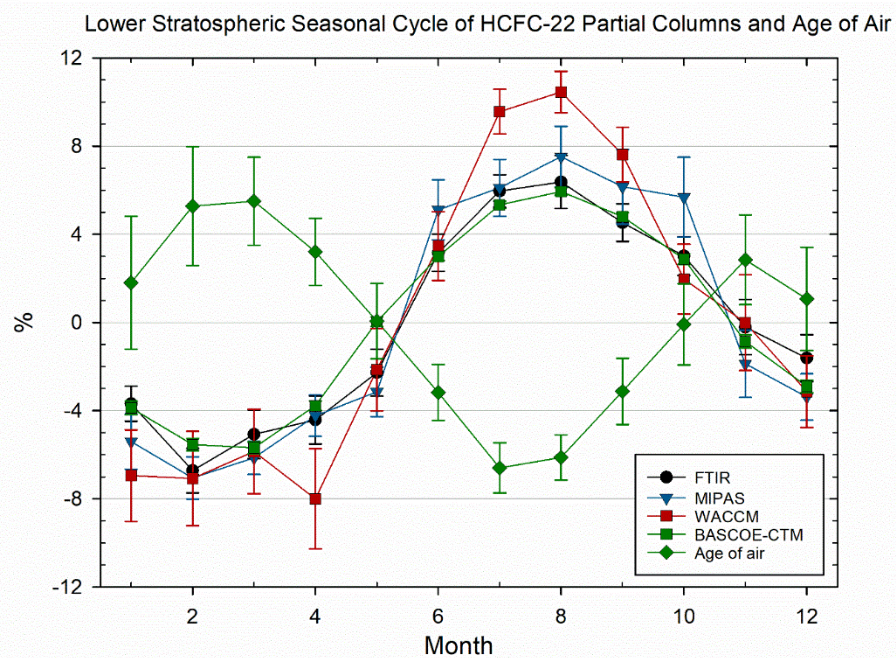


FTIR monthly time series of HCFC-22 above Jungfraujoch derived from spectra recorded by the homemade FTIR (blue) as well as by the Bruker IFS-120HR (red). Vertical bars are the standard deviations around the monthly means. Due to pollution events starting in 1996 and mainly influencing the Bruker instrument, observations retrieved from the Bruker spectra are discarded before 2003. Note the excellent agreement between the two instruments (insert frame).

Results: HCFC-22 Lower Stratospheric columns



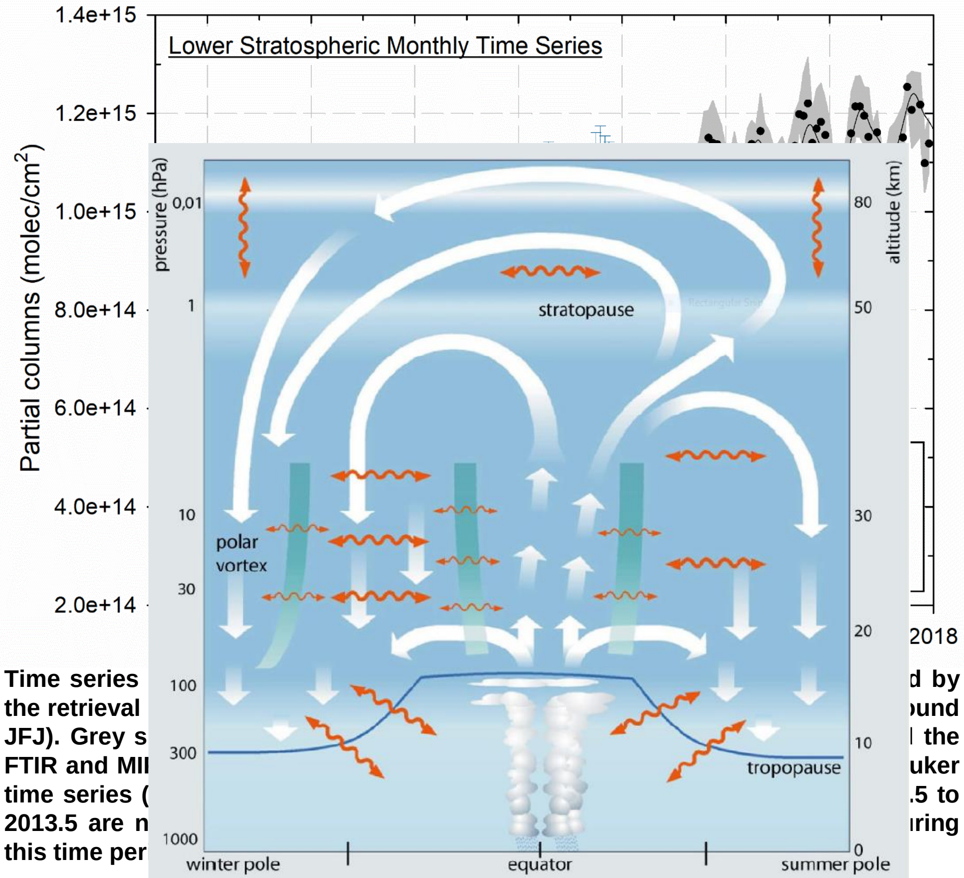
Time series of lower stratospheric partial columns (11.21 to 30 km, as defined by the retrieval information content) above Jungfraujoch (MIPAS at ±5° latitude around JFJ). Grey shade and blue vertical bars depict the standard deviation around the FTIR and MIPAS monthly means, respectively. A Fourier series fitted to the Bruker time series (black curve) is also represented. FTIR partial columns from 2011.5 to 2013.5 are not displayed because of the lower quality retrievals observed during this time period.



Seasonal cycle (see section 4.4 for method) in the lower stratosphere (11.21 to 30 km) based on measurements and model outputs (2005-2012). MIPAS measurements are at a maximum distance of 500 km from JFJ station. Vertical bars depict the 2σ standard error of the means. Age of air simulation is performed by BASCOE-CTM from ERA-Interim reanalysis. The peak-to-peak amplitude of the age of air cycle is 0.37 year and the mean age of air is 2.96 year.

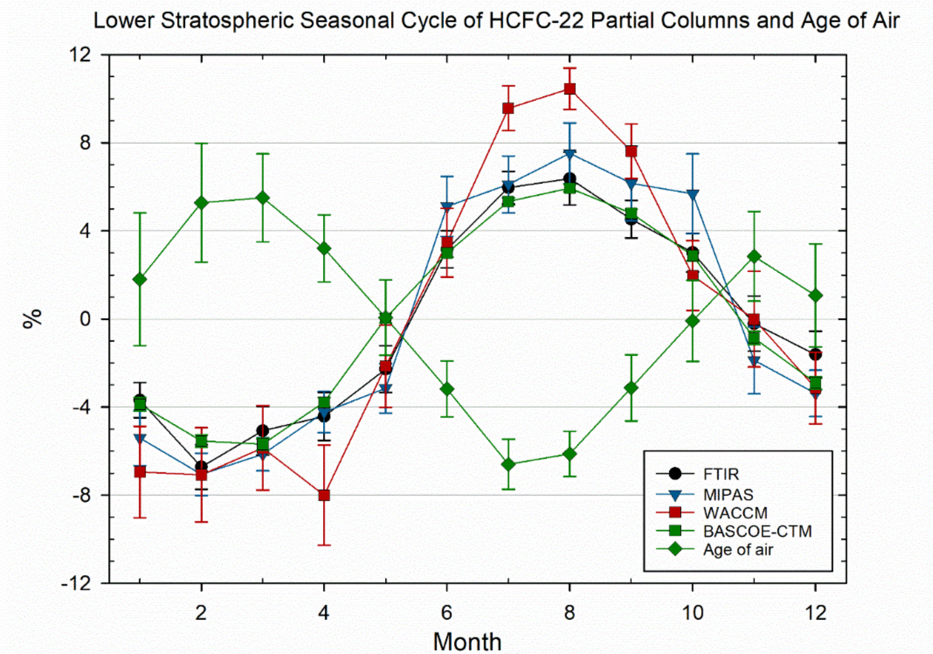
Bönisch et al., 2011, ACP

Results: HCFC-22 Lower Stratospheric columns



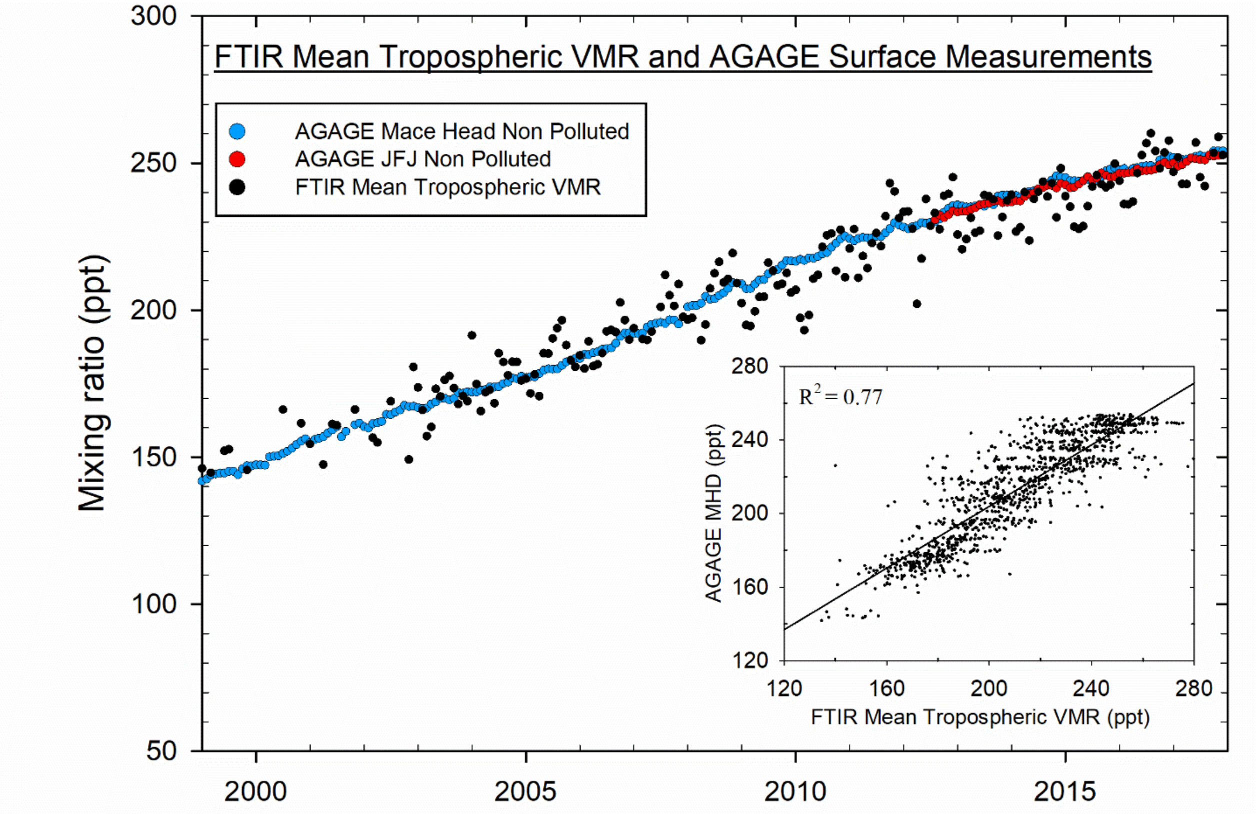
Time series ... d by
the retrieval ... ound
JFJ). Grey s ... l the
FTIR and MII ... uker
time series (... .5 to
2013.5 are n ... uring
this time per

Bönisch et al., 2011, ACP



Seasonal cycle (see section 4.4 for method) in the lower stratosphere (11.21 to 30 km) based on measurements and model outputs (2005-2012). MIPAS measurements are at a maximum distance of 500 km from JFJ station. Vertical bars depict the 2σ standard error of the means. Age of air simulation is performed by BASCOE-CTM from ERA-Interim reanalysis. The peak-to-peak amplitude of the age of air cycle is 0.37 year and the mean age of air is 2.96 year.

Results: HCFC-22 in the troposphere



Tropospheric monthly time series at Jungfraujoch. FTIR series (black) is constructed by taking the average of all the layers below 11.21 km, the altitude limit objectively defined by the retrieval information content. AGAGE in situ series from Mace Head (light blue) and JFJ (red) are baseline. Daily coincidences between Mace Head and FTIR are depicted in the lower right scatter plot. The coefficient of determination of the linear regression, R^2, is 0.77 (R = 0.88).

Relative trends:

- 1999-2008:
  - FTIR: (3.72±0.2)%
  - MHD: (3.66±0.06)%
- 2008-2017:
  - FTIR: (2.29±0.18)%
  - MHD: (2.27±0.05)%

➔ Confirms the recent slowing down of HCFC-22 accumulation rate in the atmosphere

Conclusion

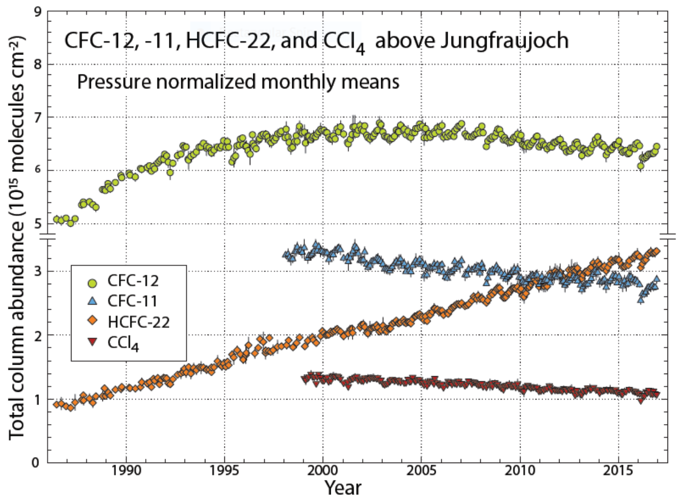


Figure 1-2. Monthly mean total vertical column abundances (in molecules per square centimeter) for CFC-12, CFC-11, CCl_4, and HCFC-22 above Jungfraujoch station, Switzerland, from 1986 to 2016 (updated from Zander et al., 2008 and Rinsland et al., 2012). The bootstrap resampling tool described by Gardiner et al. (2008) and Rinsland et al. (2012) was used for the trend evaluations (see **Table 1-2**). Note the discontinuity in the vertical scale.

WMO Ozone Assessment Report 2018

- Improvement of the retrieval strategy enables to retrieve reliable tropospheric and lower stratospheric time series
- Slowing down of the last decade HCFC-22 growth rates → fulfillment of the Montreal Protocol and its amendments
- These kind of strategy improvements could be applied to other ozone depleting substances (e.g., CFC-12)

Backup slides