Recent results derived from regular ground-based FTIR observations at the Jungfraujoch and other NDACC stations

Manu Mahieu, Whitney Bader, Bruno Franco
with the GIRPAS-team, University of Liège, BE
and the ACE-team, University of Waterloo, CA

emmanuel.mahieu@ulg.ac.be

The Jungfraujoch observational program

- **Ground-based FTIR** year-round measurements are performed under clear-sky conditions at the Jungfraujoch station [Swiss Alps, 46.5ºN, 8.0ºE, 3580m a.s.l., a site of the NDACC network, see www.ndacc.org], allowing to retrieve total and partial column abundances of more than two dozen atmospheric species, important minor and trace constituents of the troposphere and/or stratosphere
- A high-resolution Fourier Transform Infrared (FTIR) spectrometers (Bruker 120HR) is operated since the mid-1990s under clear-sky conditions at the Jungfraujoch (a homemade FTS has been in operation over 1984-2008)
- FTIR regular observations are available since 1984, with a high density of measurements (~120 days/yr on average over the last 20 years)
- Remote operation of the Bruker instrument is operational since late 2008 [design and implementation by *Ch. Servais*, ULg] => opt. obs. statistics
- In addition, grating mid-resolution spectra recorded essentially from 1976 to 1989 are also available, they cover narrow IR intervals specifically selected to include lines of HCl, HF, CH_4, N_2O…
- Altogether : now **more than 39 years of uninterrupted IR monitoring** (unique worldwide!)

emmanuel.mahieu@ulg.ac.be

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Retrieval algorithm and ancillary data

- For most species, the SFIT-2 algorithm (v3.91) is being used for the retrievals, this code is maintained and developed mainly at NCAR, (PROFFIT (v9.6) developed at Karlsruhe (KIT) is also used for some applications, e.g. for water vapor retrievals)
- SFIT-2 implements the Optimal Estimation Method of Rodgers, allowing to derive information on the vertical distribution of most species accessible to the ground-based FTIR technique
- HITRAN-2012 (or earlier versions whenever needed) are assumed in the retrievals, zpt info from NCEP daily data
- Cross sections are not handled by SFIT-2, hence pseudolines produced by G.C. Toon (NASA-JPL) from X-section laboratory spectra are used for numerous target or interfering species (e.g. CFCs, CCl_4, $ClONO_2$, C_2H_6)

emmanuel.mahieu@ulg.ac.be

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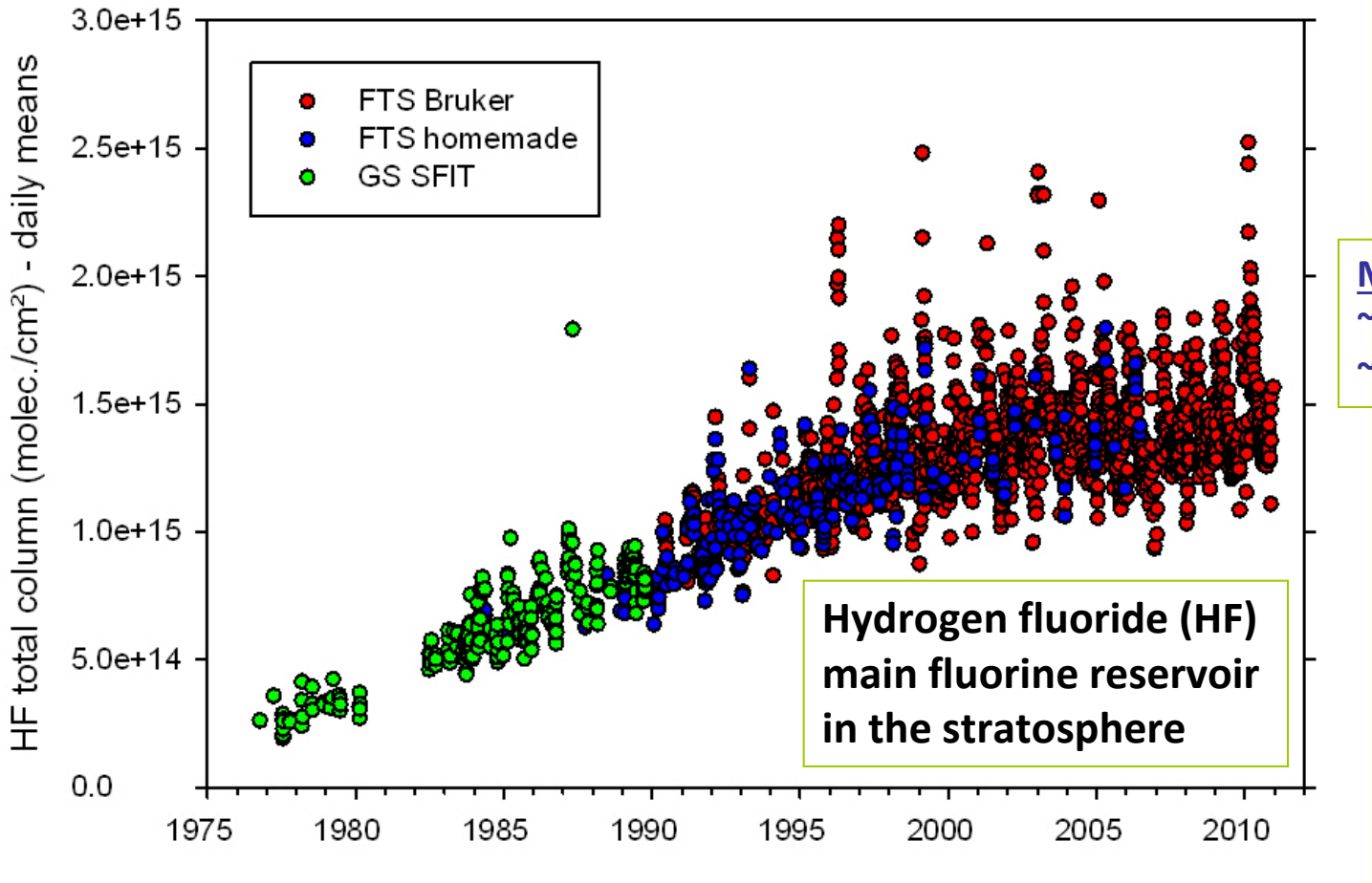
Harmonized time series: HF, since 1977

Nearly 40 years of continuous measurements, with overlaps!



Mean trends:
~5%/yr for 1977-1999
~1%/yr for 2000-2010

emmanuel.mahieu@ulg.ac.be

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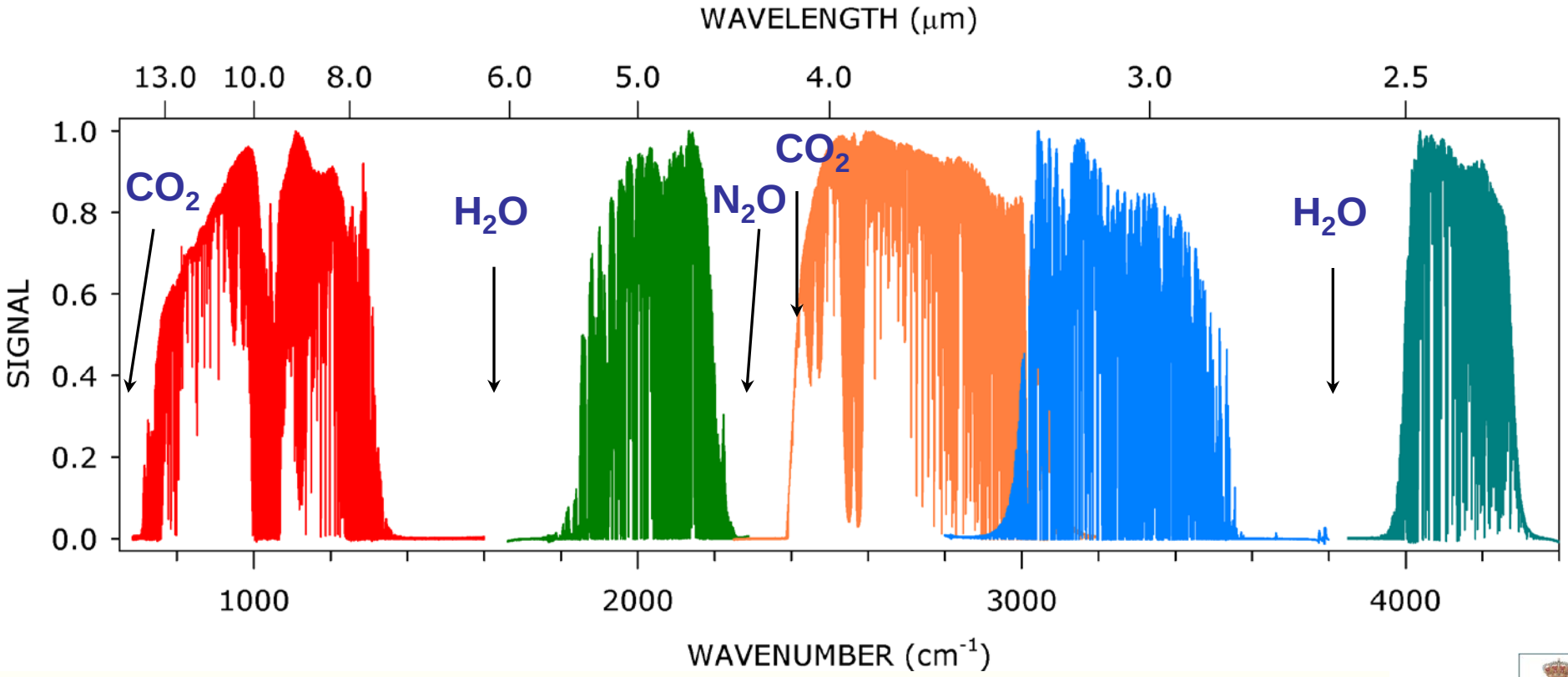
FTIR: 2 detectors + 5 optical filters

=> Broadband high-resolution spectra, with high S/N



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Current targets (>25)

Climate-relevant (GHGs)	H_2O, CO_2, **CH_4***, N_2O, CF_4, SF_6	Support to the **Kyoto Protocol**
Ozone-relevant	O_3, NO, NO_2, HNO_3, $ClONO_2$, HCl, HF, COF_2, **CFC-11**, **-12**, HCFC-22, -142b, CCl_4	Support to the **Montreal Protocol**
Air quality, biomass burning…	CO, **CH_3OH**, **C_2H_6**, C_2H_2, C_2H_4, HCN, HCHO, HCOOH, NH_3	Support e.g. to 
Others	OCS, N_2, many isotopic forms (HDO, CH_3D, $^{13}CH_4$, ^{13}CO…)	

*** See Whitney's talk tomorrow!**

emmanuel.mahieu@ulg.ac.be

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Methanol (CH_3OH)

Development of a retrieval strategy for methanol (CH_3OH) for Jungfraujoch: see Bader et al., *AMT*, *7*, 2014

Contact: w.bader@ulg.ac.be

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Methanol – Optimized retrieval strategy

Mahieu *et al.*, 2012

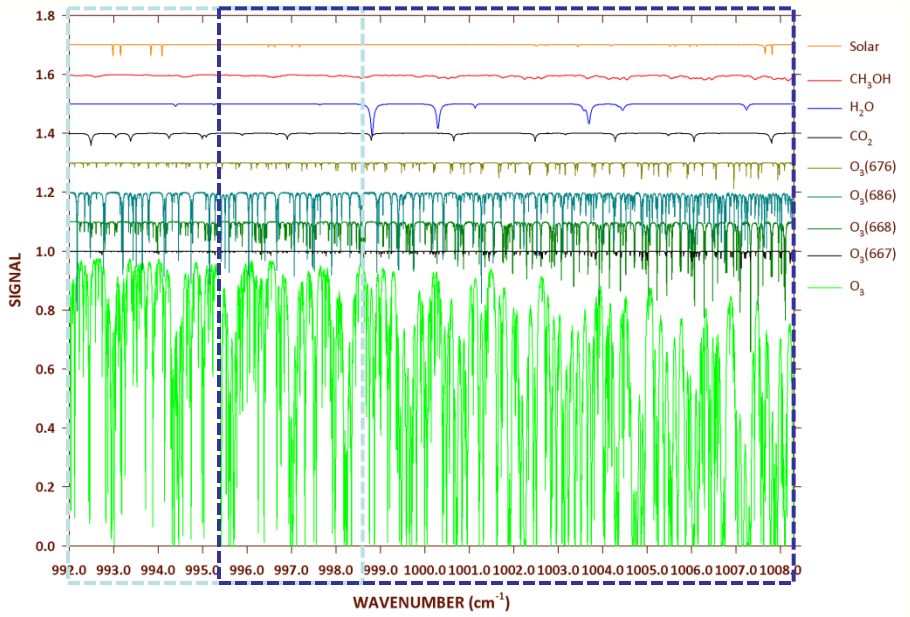
Rinsland *et al.*, 2009 + Atmospheric Chemistry Experiment, 2011

Stavrakou *et al.*, 2011

992 – 998.7 cm^{-1}

995.5 – 1008.3 cm^{-1}

1029 – 1037 cm^{-1}



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Simulations for Jungfraujoch, 80°, 6.1 mK using HITRAN08

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Methanol – Optimized retrieval strategy

Mahieu *et al.*, 2012

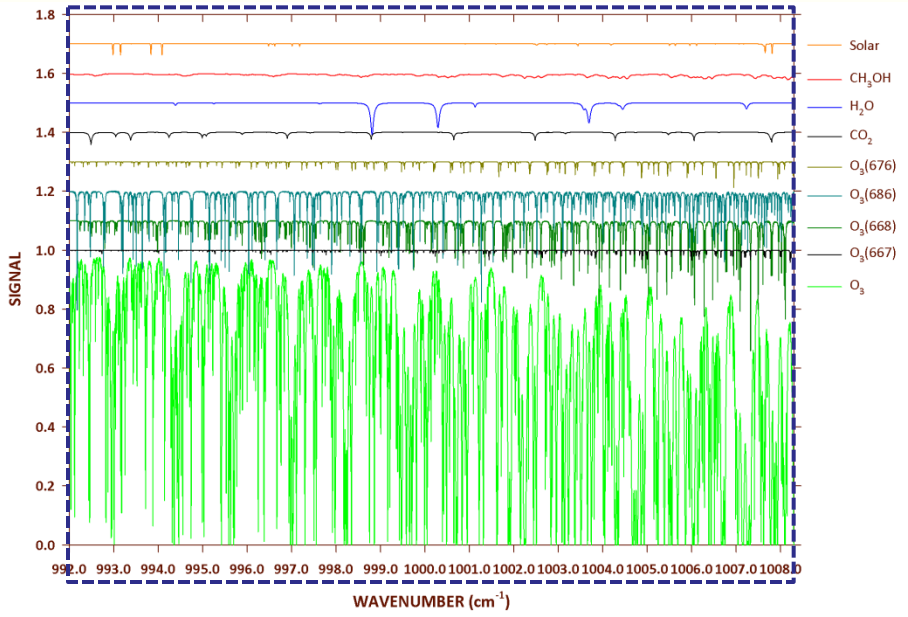
Rinsland *et al.*, 2009 + Atmospheric Chemistry Experiment, 2011

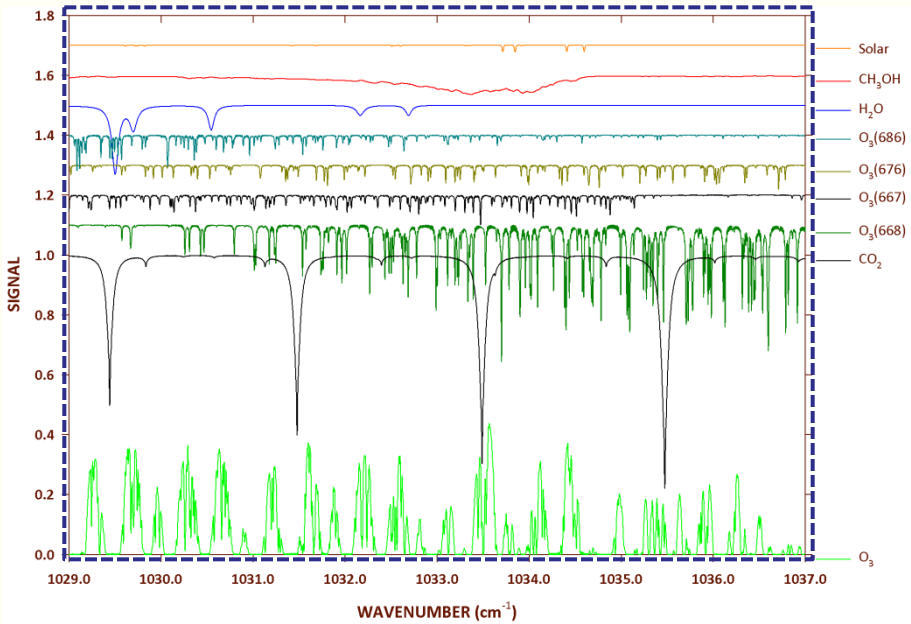
Stavrakou *et al.*, 2011

992 – 1008.3 cm^{-1}

Improvement of the DOFS from 1.5 to 1.8 (Bader et al., 2014)

1029 - 1037 cm^{-1}





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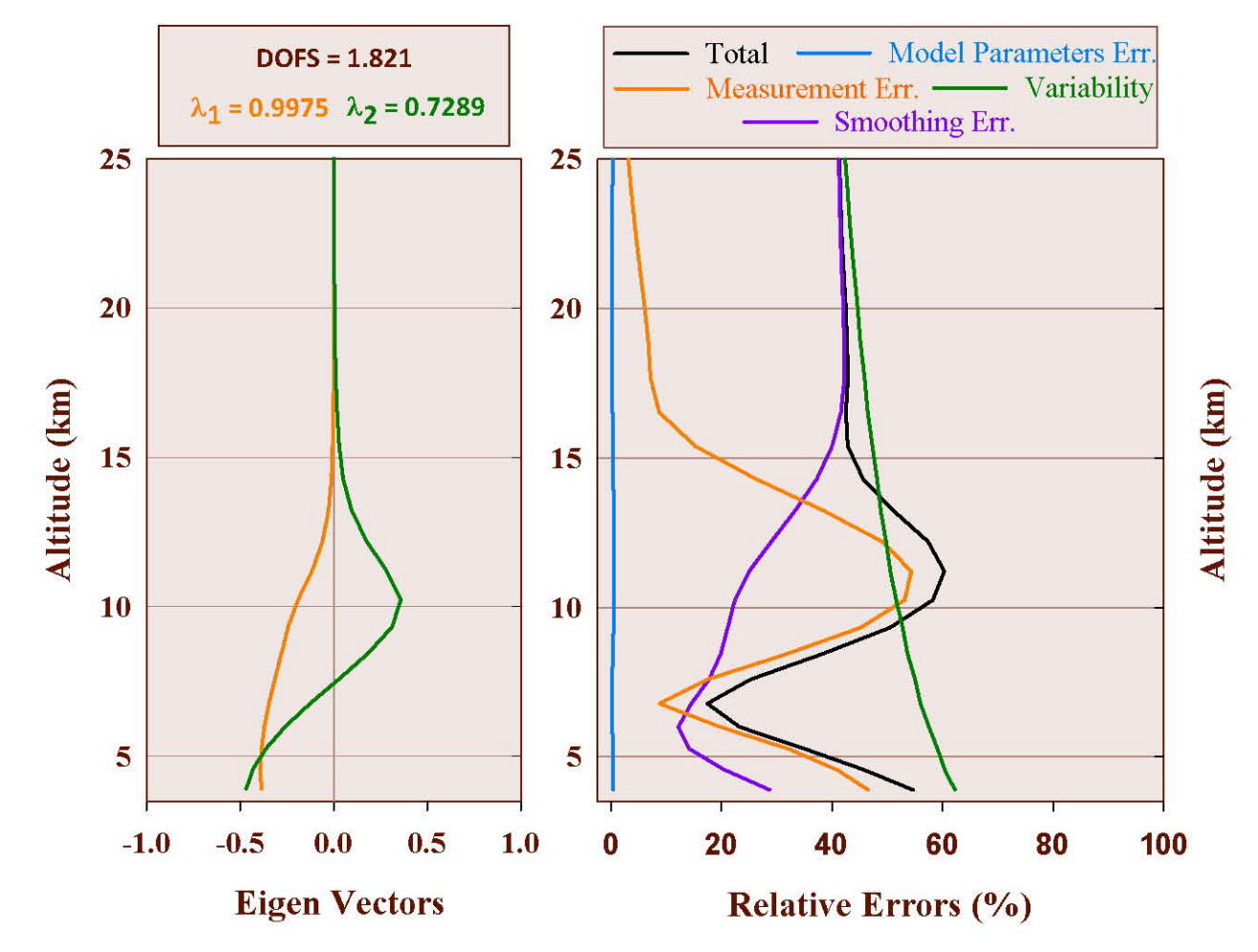
CH_3OH: typical information content



- DOFS ≈ 1.8
- 1st Eigen vector: tropospheric column
- 2nd Eigen vector: some vertical resolution (LT & UTLS)
- Random error on the total columns: ~5%

Contact: w.bader@ulg.ac.be

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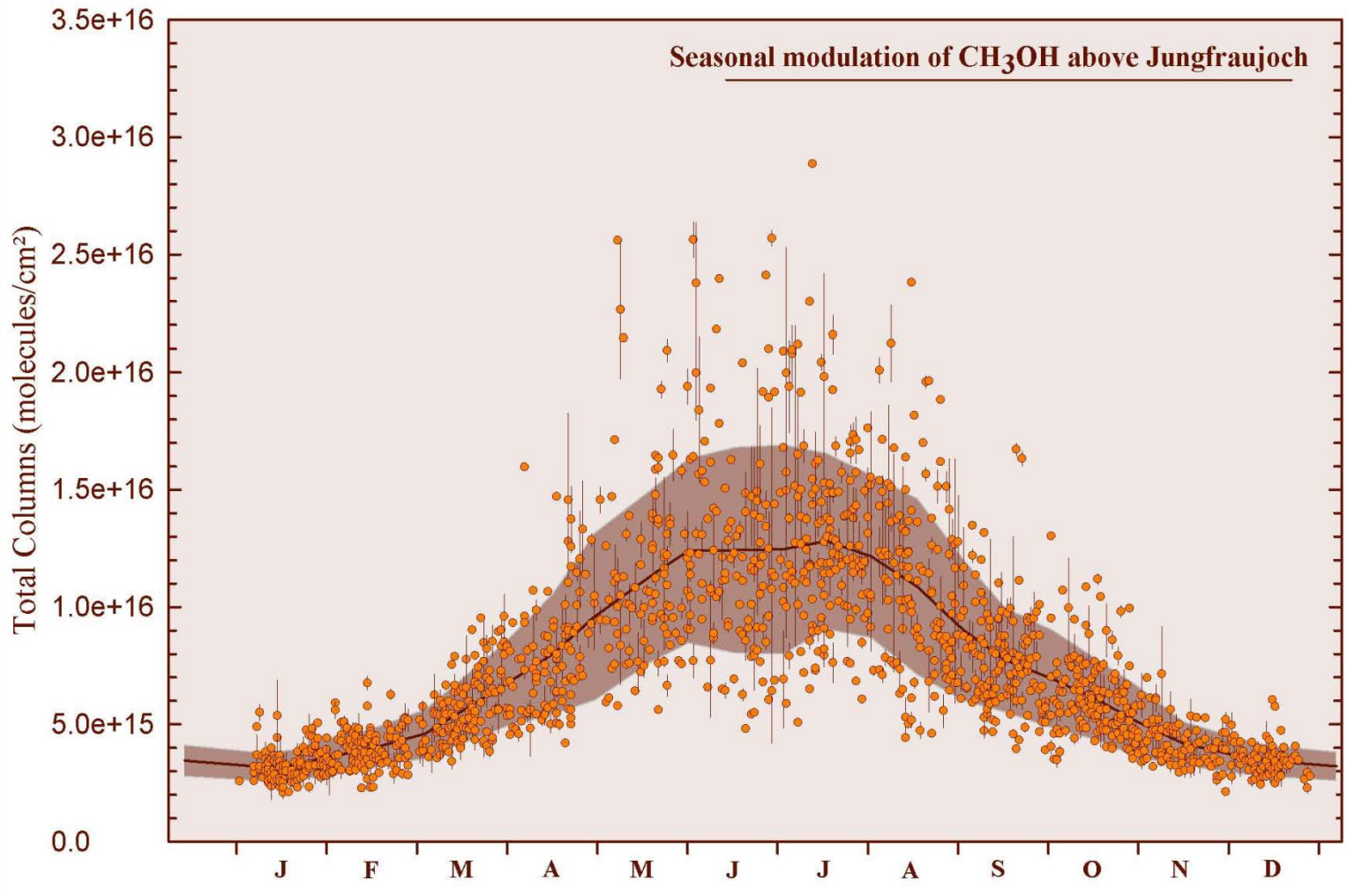
Methanol Seasonal Modulation and Variability

- No long-term trend over 1995-2012
- Minimum column & variability: December to February
- Maximum abundance & variability: June-July (plant growth is the major source)
- Large peak-to-peak amplitude: ≈130 % of the CH_3OH yearly mean



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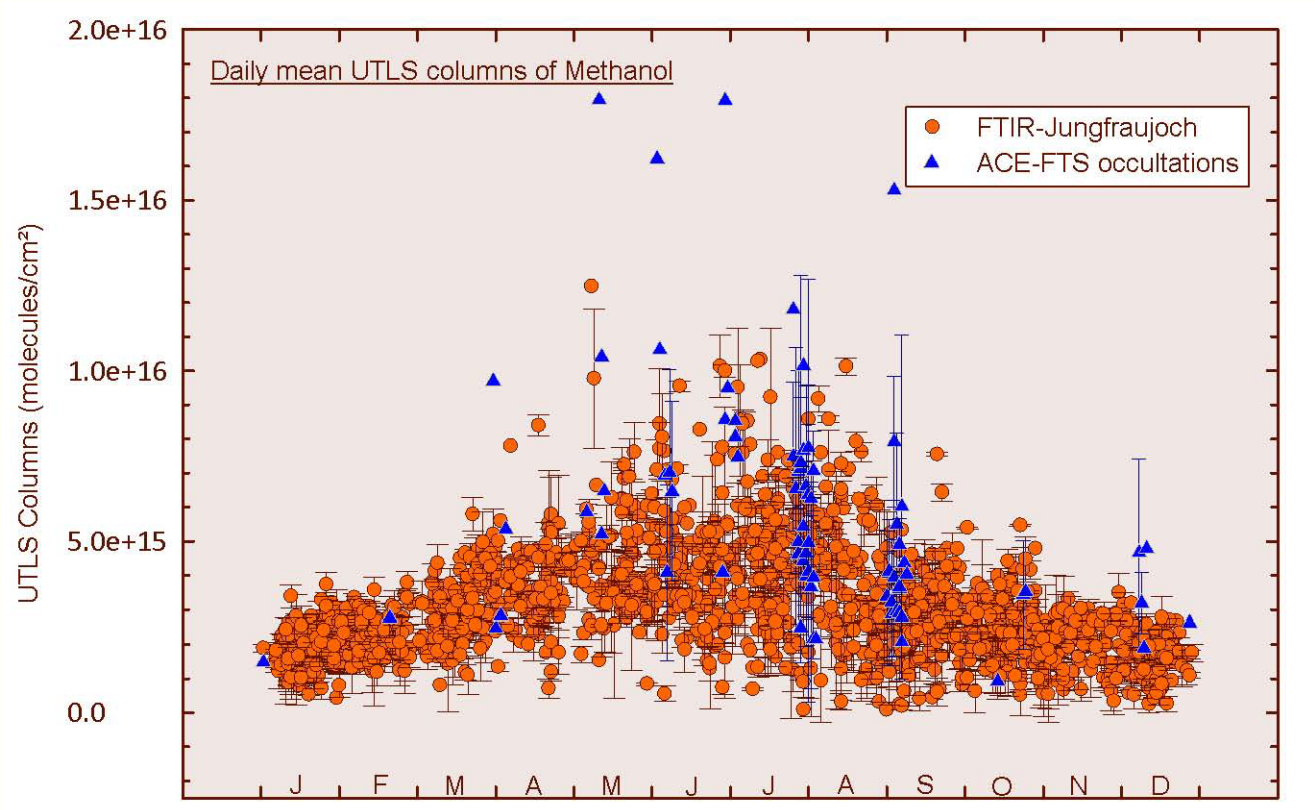
Comparison with ACE-FTS (UTLS, v3.5)



- Intercomparison with occultation measurements by ACE-FTS (7-14 km) between 41 and 51°N
- Time period: 2004-2012
- No significant bias
- ACE zonal bins may capture local events out of reach for the Jungfraujoch

w.bader@ulg.ac.be

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Ethane (C_2H_6)

Recent increase of ethane (C_2H_6) in the Northern Hemisphere

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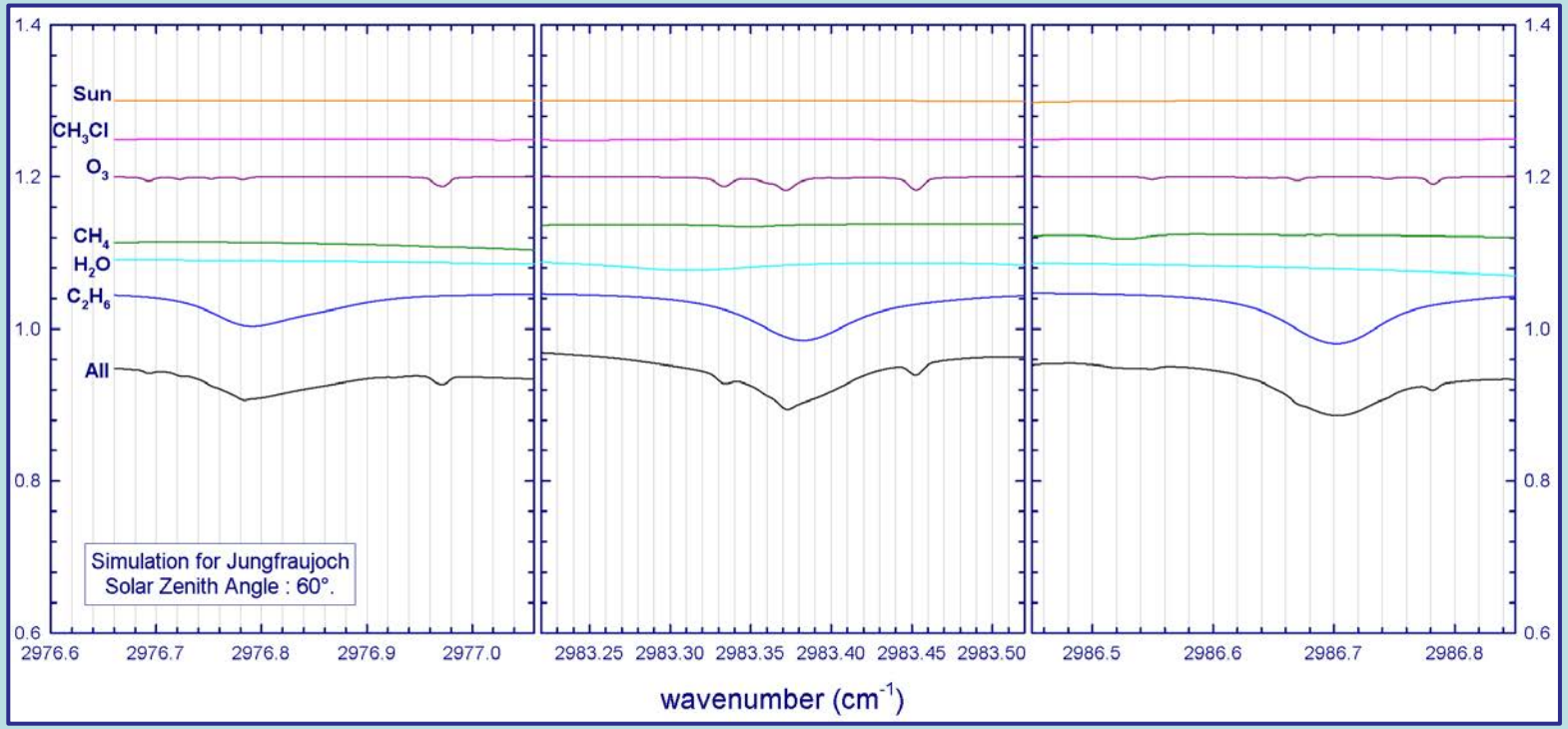
Bande d'élongation C-H

$_{P}Q^{3}$	$_{P}Q^{1}$	$_{R}Q^{0}$ Sites secs @ hautes latitudes



Fortes interférences de H_2O

Using C_2H_6 pseudolines based on Harrison et al. lab spectra (JQSRT, 2010): allowing combining the three windows (Franco et al., *JQSRT*, *160*, 2015)

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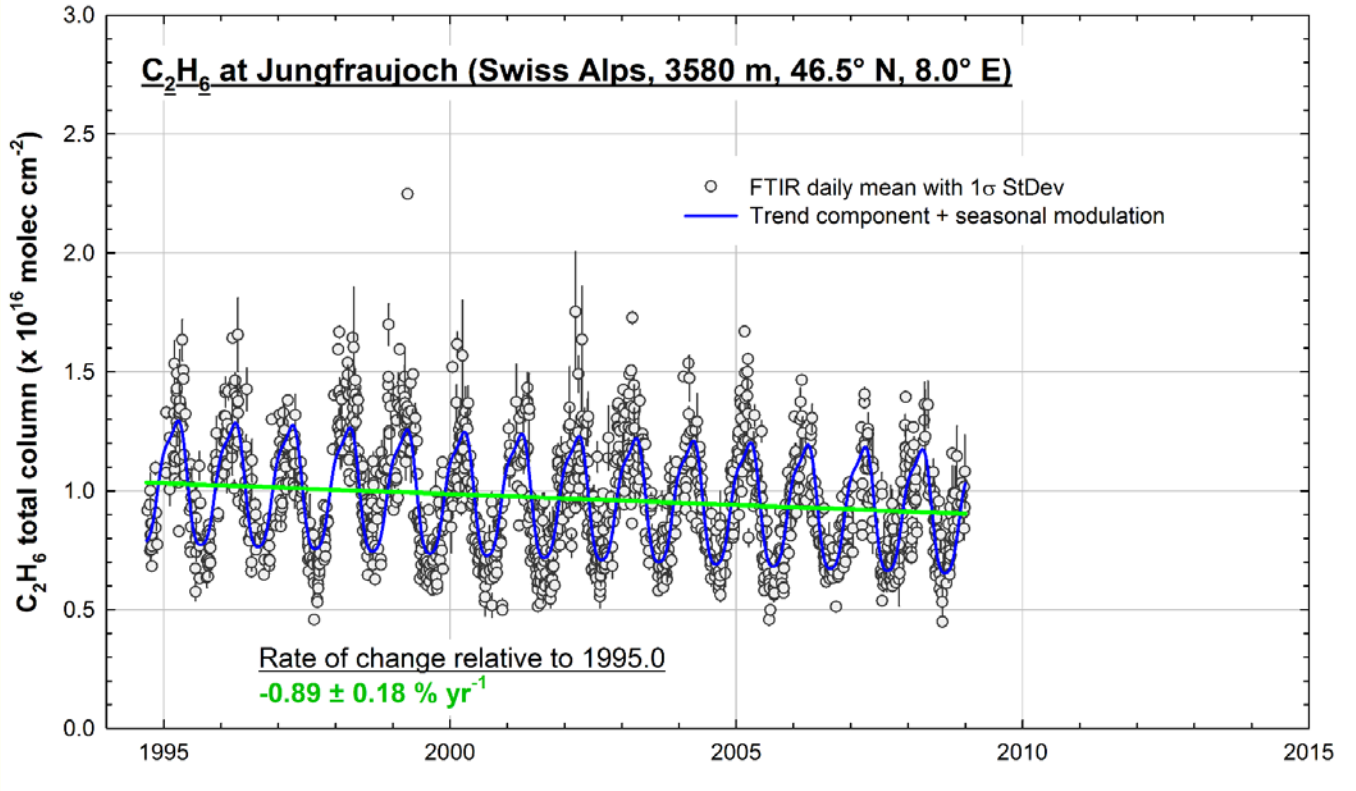
Recent increase of ethane detected in the remote atmosphere of the Northern Hemisphere

B. Franco[1], W. Bader[1], B. Bovy[1], E. Mahieu[1], E. V. Fischer[2], K. Strong[3], S. Conway[3], J. W. Hannigan[4], E. Nussbaumer[4], P. F. Bernath[5,6,7], C. D. Boone[7] & K. A. Walker[3,7]

[1]University of Liège (bruno.franco@ulg.ac.be), [2]Colorado State University, [3]University of Toronto, [4]NCAR Boulder, [5]Old Dominion University, [6]University of York, [7]University of Waterloo

Regular decrease of atmospheric C_2H_6 burden consistent with the global decline of fugitive emissions from fossil fuel sources from the mid-1980s (Aydin et al., 2011; Simpson et al., 2012)



EGU General Assembly 2015, April 13th, Vienna, Austria

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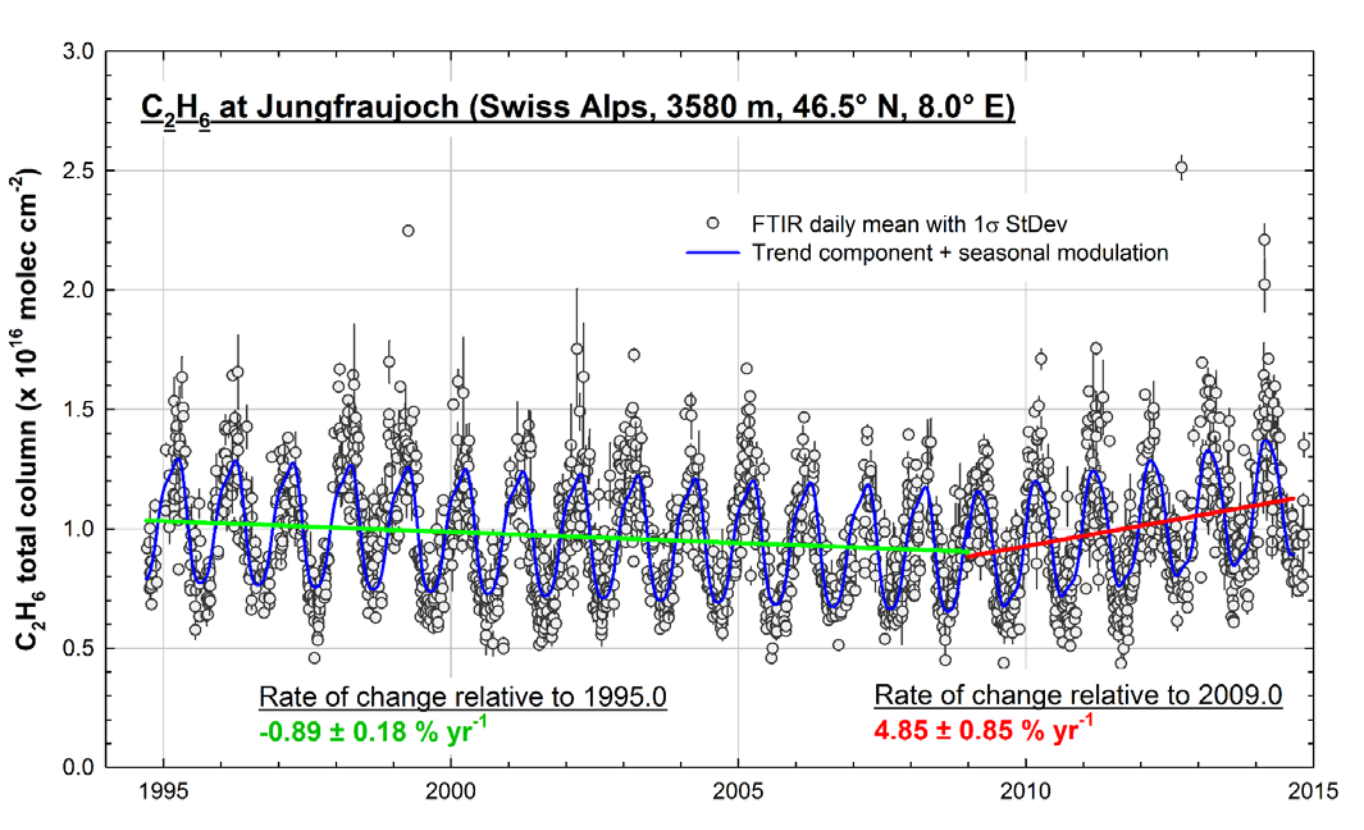
But since 2009, the atmospheric C_2H_6 burden is increasing at a rate of **5 % yr^{-1}**



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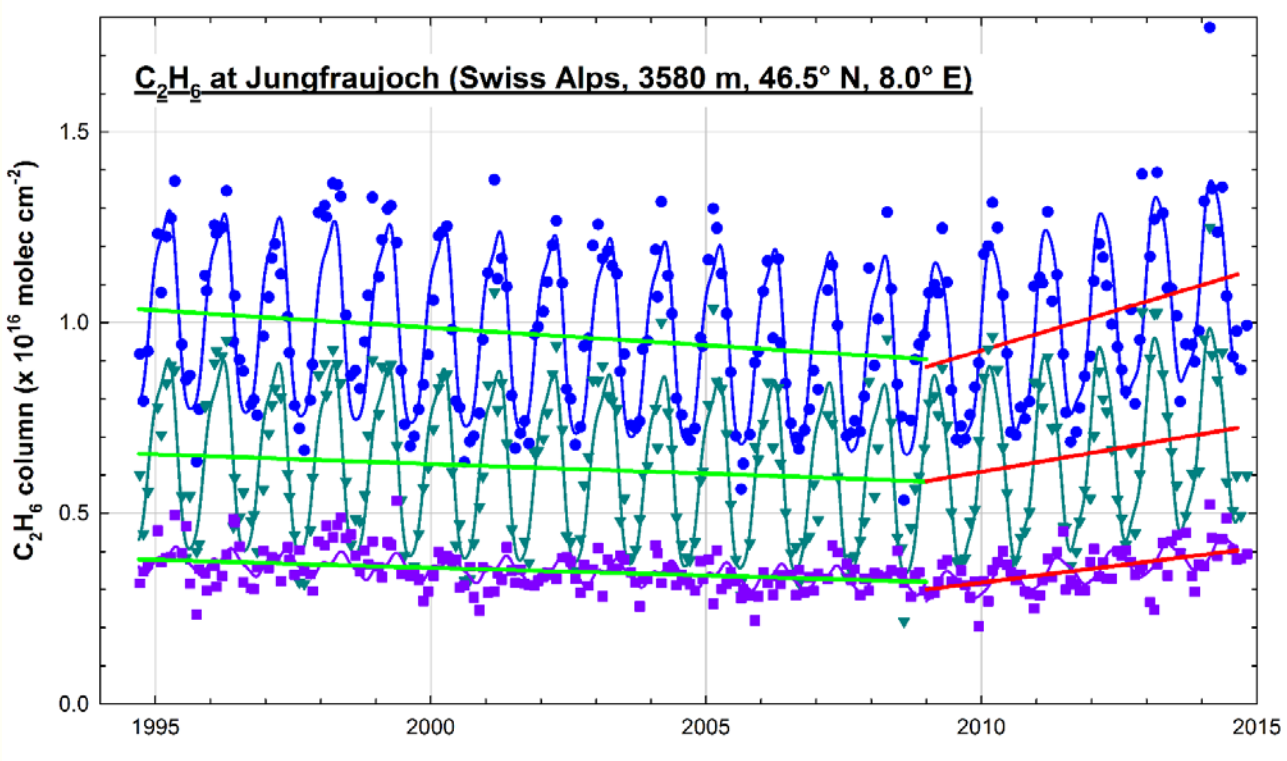
- Two independent partial columns may be deduced from the FTIR retrievals (DOFS ≈ 2.1)
- **Vertically-homogeneous** increase of C_2H_6 consistent throughout the troposphere and lower stratosphere



Rates of change relative to 2009.0

- Total column
4.85 ± 0.85 % yr^{-1}

- Lower troposphere (3.6-8 km)
4.23 ± 1.06 % yr^{-1}

- Upper trop. – lower strat. (8-21 km)
6.05 ± 1.09 % yr^{-1}

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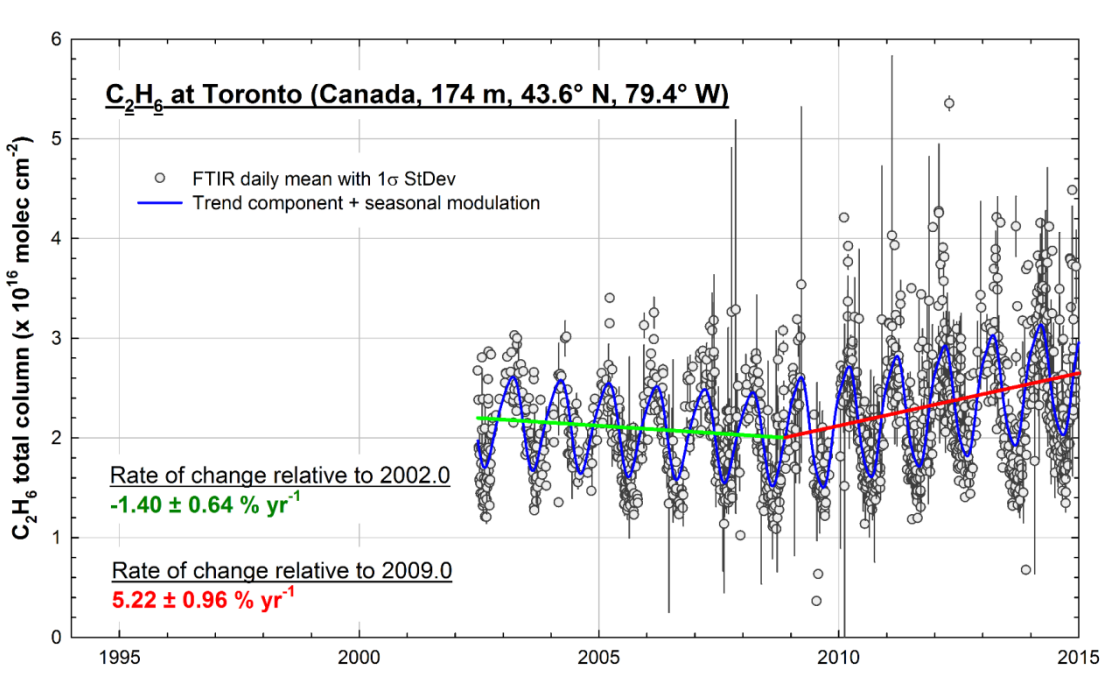
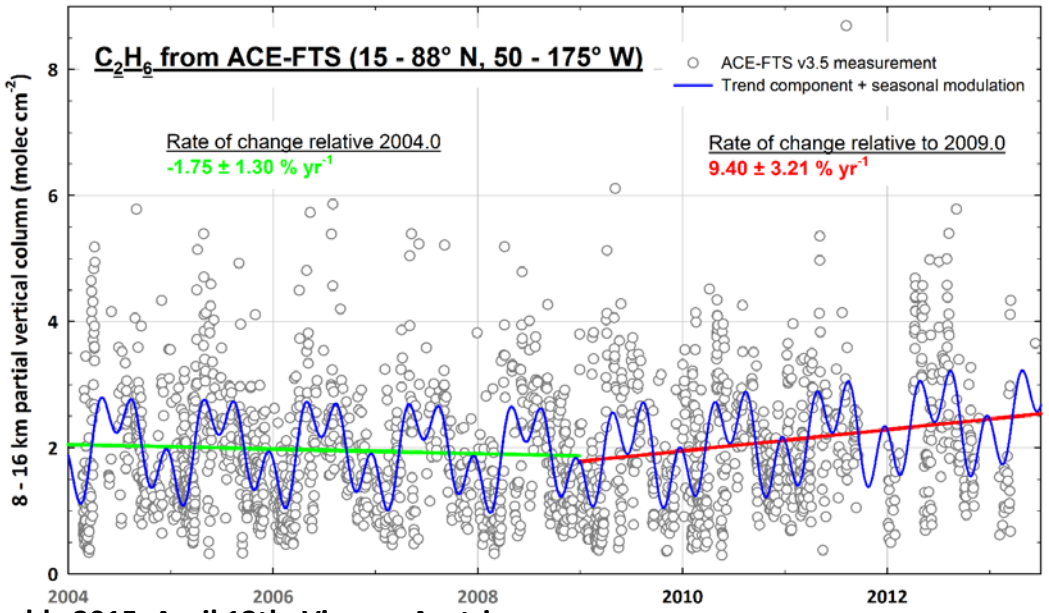


Recent upward trend confirmed at Toronto, Boulder, Eureka, Thule…

And by ACE-FTS occultation measurements above North America

No increase detected in the Southern Hemisphere (Lauder, ACE-FTS)

Massive exploitation of shale gas (fracking) in the US is the likely cause for the C_2H_6 trend upturn: important implications for CH_4 coincident emissions and air quality degradation!



CFC-11 and CFC-12

Changes in atmospheric circulation

emmanuel.mahieu@ulg.ac.be

Université de Liège

EGU2015-1166

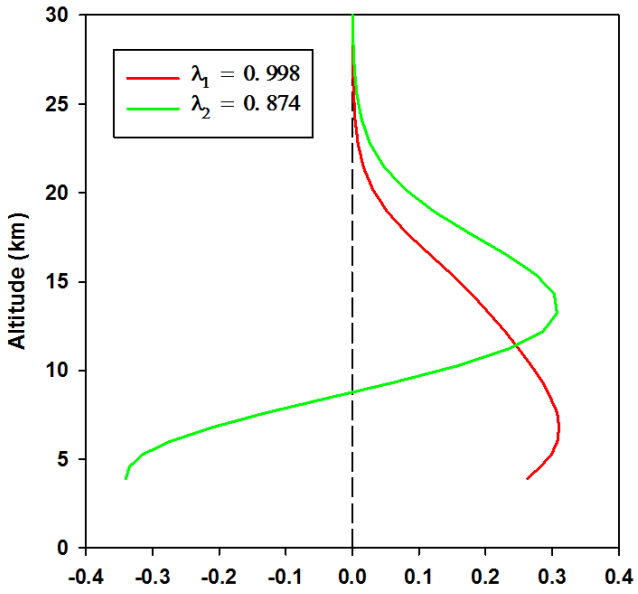
Halogenated source gases measured by FTIR at the Jungfraujoch station: updated trends and new target species

E. Mahieu[1] (emmanuel.mahieu@ulg.ac.be), W. Bader[1], B. Bovy[1], B. Franco[1], B. Lejeune[1], C. Servais[1], J. Notholt[2], M. Palm[2], G.C. Toon[3]

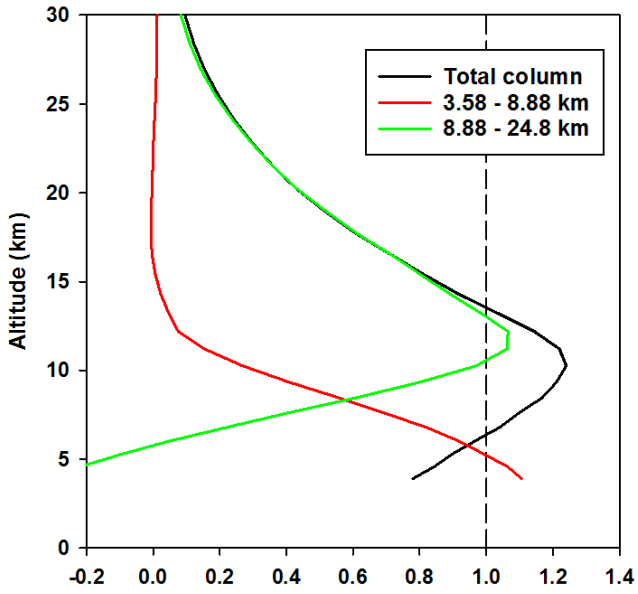
[1]University of Liège – [2]University of Bremen – [3]Jet Propulsion Laboratory - Caltech

EGU General Assembly 2015, Vienna, 13 April 2015

First Eigen vectors (CFC-12)



Averaging kernels for merged layers (CFC-12)



INFORMATION CONTENT:

The retrieval of CFC-12 provides sensitivity in the 3.58 – 20 km altitude range. Two pieces of information are available, with significant contribution from the retrieval (99 and 87% for the two first Eigen vectors), providing some vertical resolution. In addition to total columns, it is therefore possible to derive information on the evolution of CFC-12 in the low troposphere and in the upper troposphere-lower stratosphere. The situation is similar for CFC-11.

EGU2015-1166

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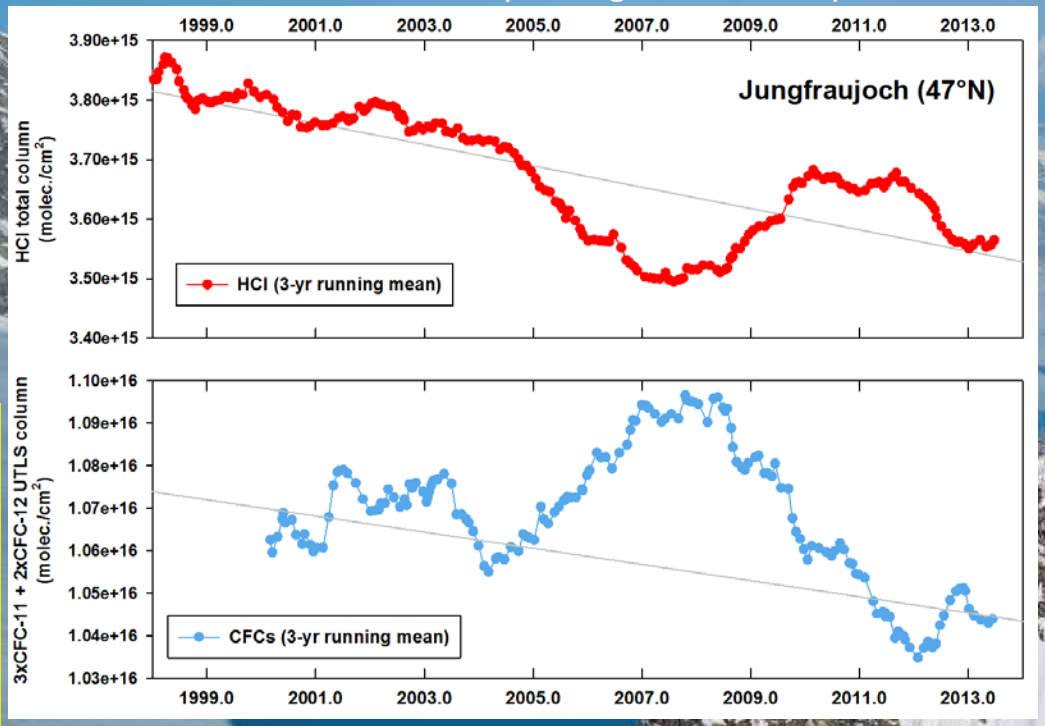

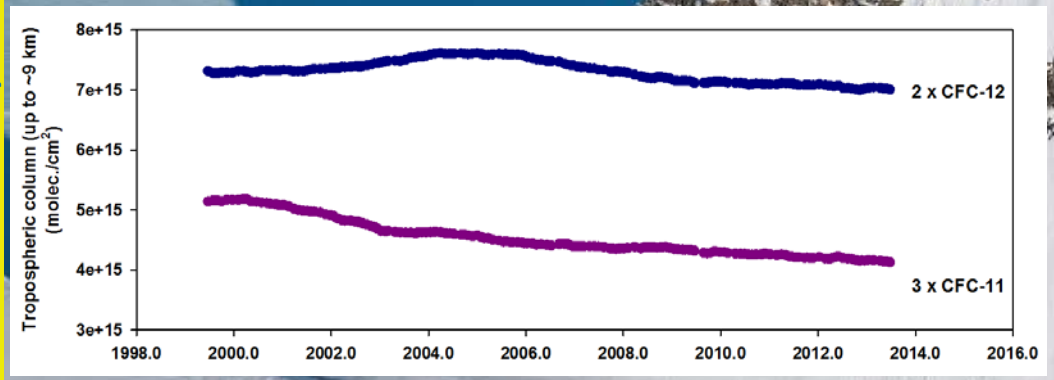

CIRCULATION CHANGES:

Due to a circulation slowdown (and hence an age-of-air increase) which occurred in the Northern Hemisphere lower stratosphere during 2007-2011 (see Mahieu et al., *nature13857*, 2014), the balance between the source and reservoir species of chlorine has been affected. More aged air was transported to the lower stratosphere, characterized by a larger relative conversion of source gases to HCl. This resulted in deviations from a smooth evolution for these species (upper frames) in this atmospheric region, in contrast with the situation prevailing in the troposphere (see the weighted contributions of the CFC-11 and -12 to the CCl_y budget; lower panel). Overall however, chlorine is decreasing in both the troposphere and stratosphere, demonstrating the effectiveness of the Montreal Protocol on substances that deplete the ozone layer.

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Selected publications (2014-2015)

- Bader et al., Long-term evolution and seasonal modulation of **methanol** above Jungfraujoch: Optimisation of the retrieval strategy, comparison with model simulations and independent observations, *AMT*, *7*, 2014
- Franco et al., Retrievals of **formaldehyde** from ground-based FTIR and MAX-DOAS observations at the Jungfraujoch station and comparisons with GEOS-Chem and IMAGES model simulations, *AMT*, 8, 2015
- Franco et al., Retrieval of **ethane** from ground-based FTIR solar spectra using improved spectroscopy: recent burden increase above Jungfraujoch, *JQSRT*, *160*, 2015
- Mahieu et al., Recent Northern Hemisphere stratospheric **HCl** increase due to atmospheric circulation changes, *Nature*, *515*, 2014

Contribution to the WMO 2014 Assessment of ozone depletion (Chapter 1: Update on Ozone-Depleting Substances and Other Gases of Interest to the Montreal Protocol)

- Bader et al., Recent **methane** increase derived from NDACC-FTIR, ACE-FTS and GEOS-Chem data sets, ***in preparation***, 2015
- Franco et al., Increase of **ethane** above North America: comparison between observations and simulations involving updated emission inventories, ***in preparation***, 2015

emmanuel.mahieu@ulg.ac.be

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Table 1-2. Comparison of annual trends of ODSs, HFC-23, CF_4, and SF_6 from in-situ measurements vs. remote sensing measurements. Relative trends in ODSs and halogenated greenhouse gases for the common 2004–2010 time period (except when specified) derived from in-situ surface measurements and remote sensing observations from the ground and from space. Surface trends were derived from monthly mean mole fractions, weighted by surface area in the region 30°N–90°N. Shown are the average and standard deviation of trends derived independently from NOAA and AGAGE data (% yr^{-1} relative to 2007 annual mean). For CF_4 and HFC-23, only AGAGE data were used, and the uncertainty was derived from uncertainties (one standard deviation) in the slope and 2007 annual mean. For HFC-23, global mean data were used from 2007 through 2010, supplemented with data from Miller et al. (2010) for 2004–2007. Ground-based remote sensing trends were derived from daily mean total column measurements performed at Jungfraujoch (46.5°N). The ACE-FTS trends were determined using tropical occultations (30°N–30°S), after averaging the mixing ratios in molecule-dependent altitude ranges (Brown et al., 2011). For HFC-23, the 40°N–40°S occultations were considered in the 10–25 km altitude range. For MIPAS CFC-11 and -12, mean rates of change for the 20°N–20°S and 10–15 km altitude range are provided, including observations between 2002 and 2011 (Kellmann et al., 2012). For SF_6, the trend characterizes the 2006–2009 time period between 17.5°N–17.5°S latitude and 9–15 km altitude (Stiller et al., 2012).

Substance	Annual Trend 2004–2010 (% yr^{-1} relative to 2007)			Data Sources
	In-situ 30°N–90°N	Remote sensing ground (total columns)	Remote sensing satellite	
CFC-11	−0.84 ± 0.09	−0.99 ± 0.10	MIPAS: −1.03±0.09 ACE-FTS: −0.9 ± 0.1	NOAA, AGAGE Zander et al., 2008 Kellmann et al., 2012 Brown et al., 2011
CFC-12	−0.39 ± 0.05	−0.38 ± 0.07	MIPAS: −0.51± 0.09 ACE-FTS: −0.4 ± 0.1	NOAA, AGAGE Zander et al., 2008 Kellmann et al., 2012 Brown et al., 2011
CFC-113	−0.93 ± 0.02		ACE-FTS: −1.2 ± 0.1	NOAA, AGAGE Brown et al., 2011
CCl_4	−1.35 ± 0.08	−1.31 ± 0.15	ACE-FTS −1.2 ± 0.1	NOAA, AGAGE Rinsland et al., 2012 Brown et al., 2011
HCFC-22	3.97 ± 0.06	3.52 ± 0.08	ACE-FTS 3.7 ± 0.1	NOAA, AGAGE Zander et al., 2008 Brown et al., 2011
HCFC-141b	2.57 ± 0.07		ACE-FTS 0.74 ± 0.5	NOAA, AGAGE Brown et al., 2011
HCFC-142b	5.44 ± 0.03		ACE-FTS 7.0 ± 0.4	NOAA, AGAGE Brown et al., 2011
HFC-23	4.2 ± 0.2		ACE-FTS 3.9 ± 1.2	AGAGE Harrison et al., 2012
CF_4	0.86 ± 0.01	1.02 ± 0.05	ACE-FTS 0.74 ± 0.04	AGAGE Mahieu et al., 2014 Brown et al., 2011
SF_6	4.27 ± 0.07	4.14 ± 0.32	MIPAS 4.3 ACE-FTS: 4.2 ± 0.1	NOAA, AGAGE Zander et al., 2008 Stiller et al., 2012 Brown et al., 2011

Last WMO: new table involving remote-sensing measurements!

Thank you!

NB: funding status is really poor, with e.g. support to our involvement in ACE ending in 2015…

emmanuel.mahieu@ulg.ac.be

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