Monitoring the Earth's atmosphere at Jungfraujoch: the challenges and benefits of a long-term commitment

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Monitoring the composition of the Earth's atmosphere

Science… and… Tourism!
(more than 1 million visitors per year since 2015)

The Jungfraujoch high-altitude station (46.5° N, 8.0° E, 3580 m a.s.l.)

=> 1991. *Network for the Detection of Atmospheric Composition Change* (NDACC)

Map of the talk…

- **A bit of history: it all started more than 70 years ago (early 1950s)!**

- **Why the Jungfraujoch for a Belgian team?**

- **A few words about our remote-sensing technique (Fourier Transform Infrared – FTIR): how and what do we measure?**

- **Applications and key results:**

  - **Air quality surveillance/tropospheric ozone precursors:**
    - **Ethane and the oil & gas sector**
    - **First retrievals of PAN from FTIR spectra**

  - **Support to the Montreal Protocol:**
    - **CFC-11 and its undeclared production and renewed emissions**
    - **F-bearing gases in the stratosphere**
    - **Robust trends for hydrogen chloride**

A BIT OF HISTORY
AND WHY THE JUNGFRAUJOCH?

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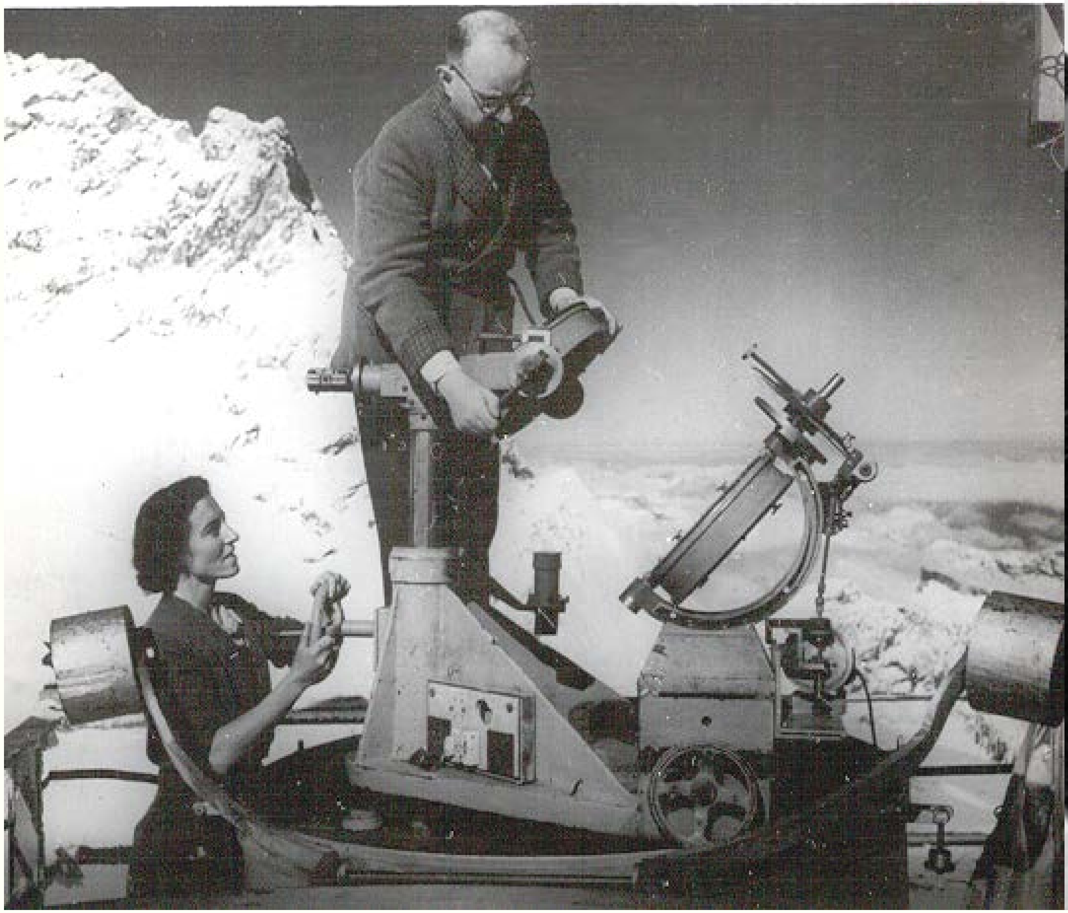
1950: the first measurements!



Pr Marcel Migeotte on the Sphinx terrace

Pr. Migeotte's vision...

" It will be very interesting to systematically record telluric bands due to CH_4, N_2O and CO in view to study or detect intensity variations with time".

Marcel Migeotte, 1951

In: "Zwanzig Jahre Hochalpine Forschungsstation Jungfraujoch"
Editor : A. von Muralt
Verlag Stämpfli & Cie, Bern, 1951

... is now - and more than ever - our mission!

Why/When/How? Some milestones…

- **<u>1945</u>:** M. Migeotte thesis on high-resolution spectroscopy
- **<u>End of the 1940s</u>:** detection of CH_4, N_2O, CO (Columbus, Ohio) => need an **unpolluted** site to confirm their ubiquity
- **<u>1950</u>:** First measurements at Jungfraujoch, extended in 1951 => atlas (2.8 – 23.7 µm) + line identifications (1958)
- **<u>1958-1974</u>:** from IGY onwards, "focus" on the sun and successive improvement of the instrumentation (7-m spectrometer) (prism/grating/double-pass)
- **<u>1974</u>:** Molina & Rowland hypothesis (CFC ↓O_3) receives a large echo in the scientific community

Some more milestones…

- **The same year (1974)**: R. Zander detects HF (*) in the upper stratosphere in IR spectra recorded from Palestine (TX) with the Liège gondola $\Rightarrow$ the team resumes its atmospheric observational program (HCl, HF, N_2O, CH_4…) in **1976-1977**
- **1984**: beginning of routine operation for the homemade **FTS**
- **1990**: installation of a Bruker 120HR FTS, after dismantling the 7-m grating spectrometer, this FTS is still running today…
- **2008**: implem. of remote operation of instrum. (↑ statistics)
- **Today**: the program continues to extend our database, unique worldwide (45+ years; 60000+ FTIR spectra!!)

(*) Hydrogen fluoride

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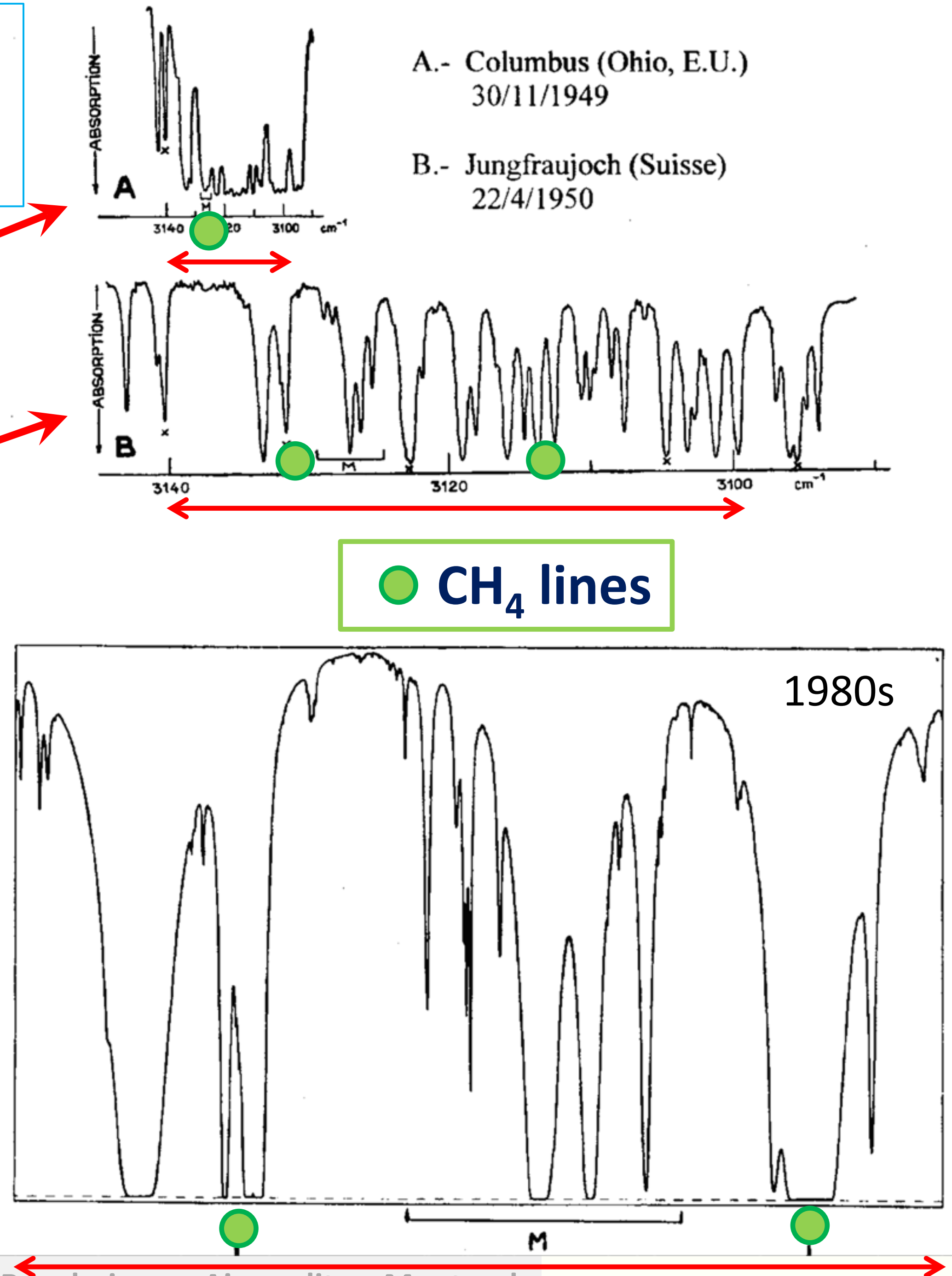
Pioneering observations of atmospheric methane

Columbus – grating

Jungfraujoch – grating

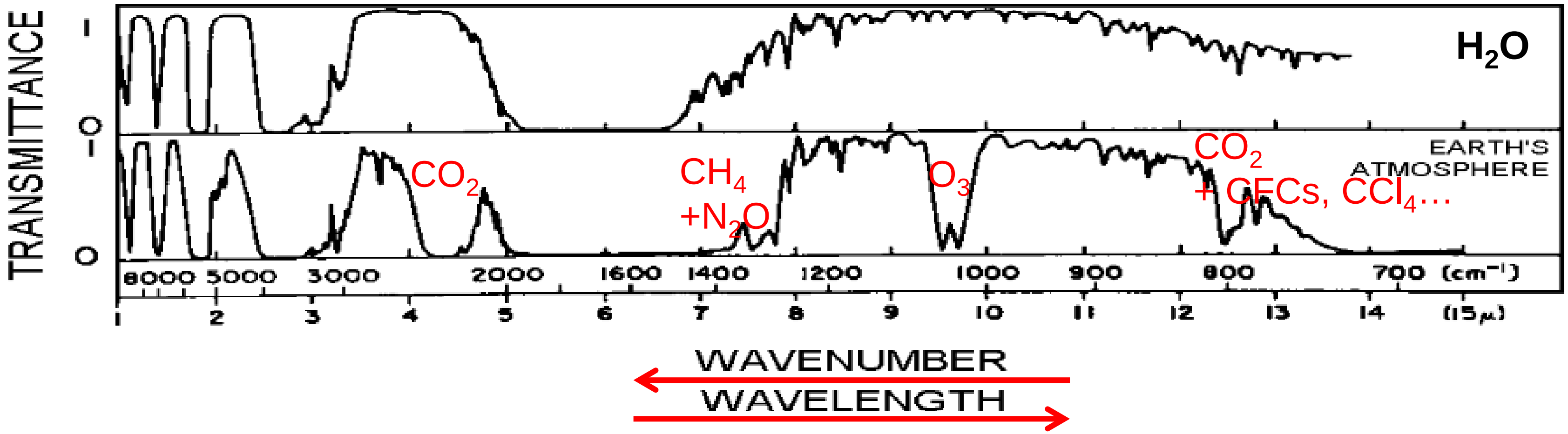
Jungfraujoch – FTIR



Signif. improvement of the instrumental performances

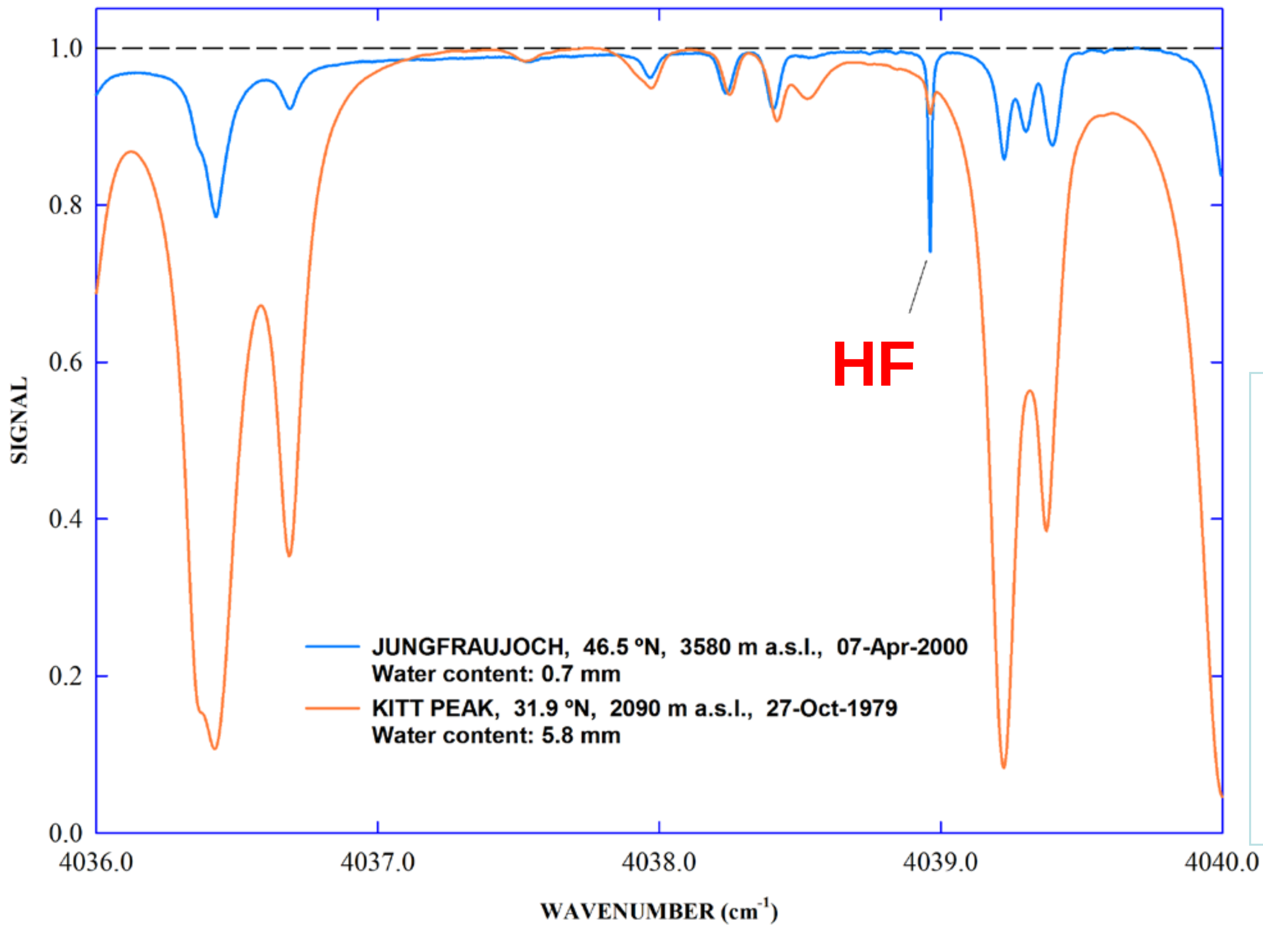
The Jungfraujoch site: why?

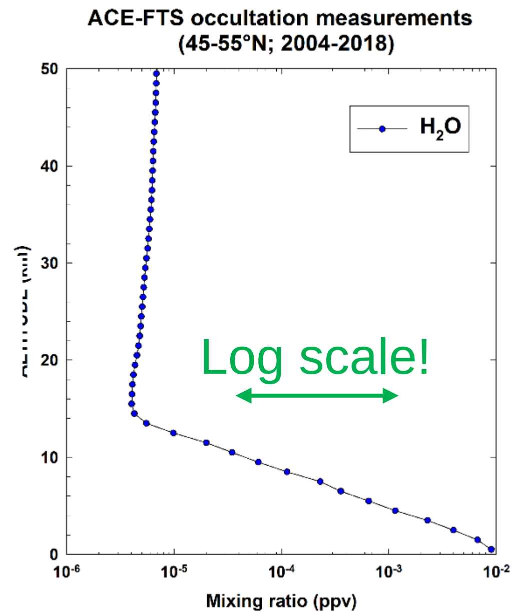
- **Unpolluted… but not only!**
- **Water vapor absorptions between 1 and 14 µm**



The infrared absorption spectrum of water vapor (top) and of the entire atmosphere (bottom) (Stephens 1994).

Infrared spectra at 2 high-altitude sites

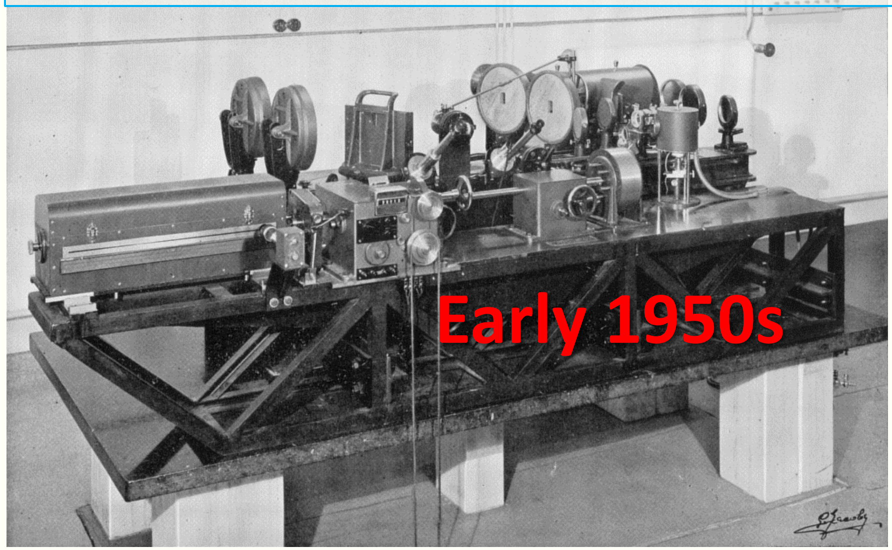


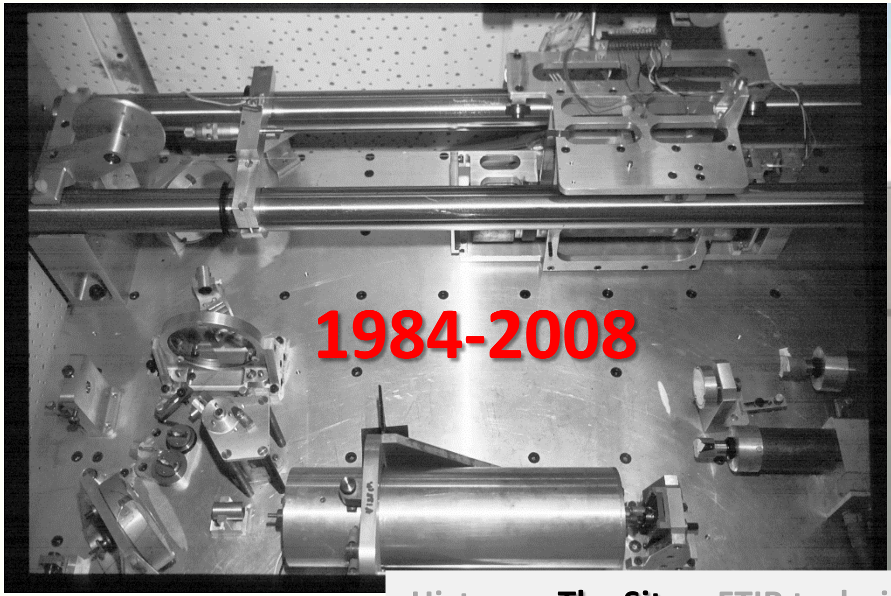


- Kitt Peak: site close to the Arizona desert, at 2 km altitude
- Both spectra were recorded under very dry conditions

≥ 2/3 of the water vapor column found below 3.6 km !!, half of it below 1.5 km => interference of H_2O strongly reduced

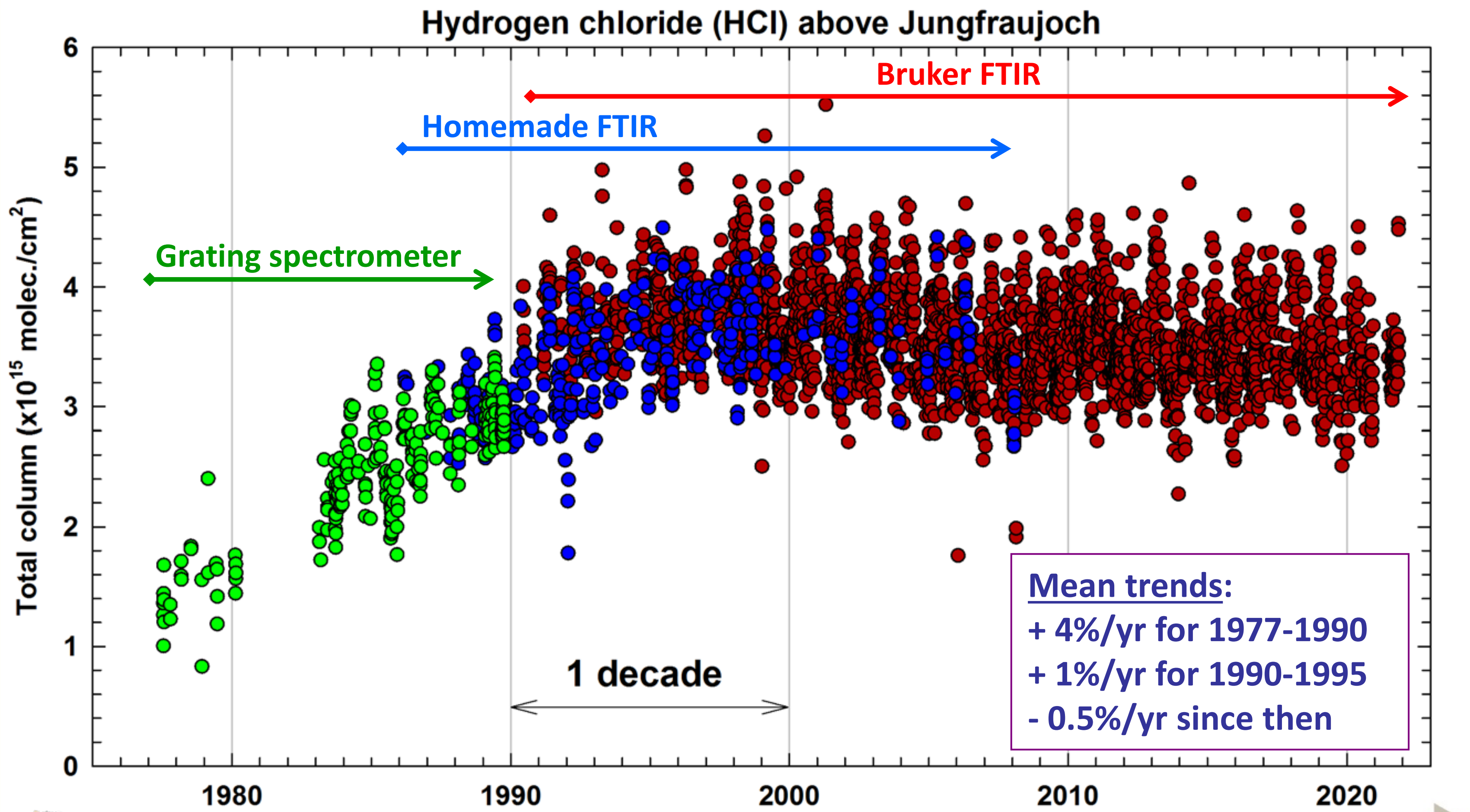
Team's instrumentation at Jungfraujoch over time…

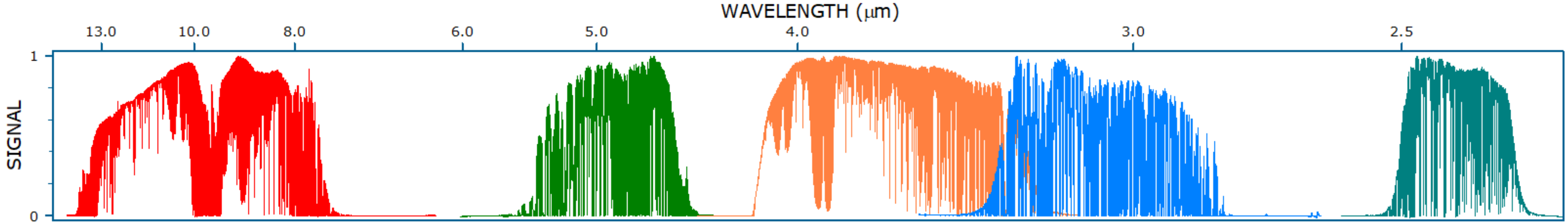




Overlaps help ensuring… consistency!

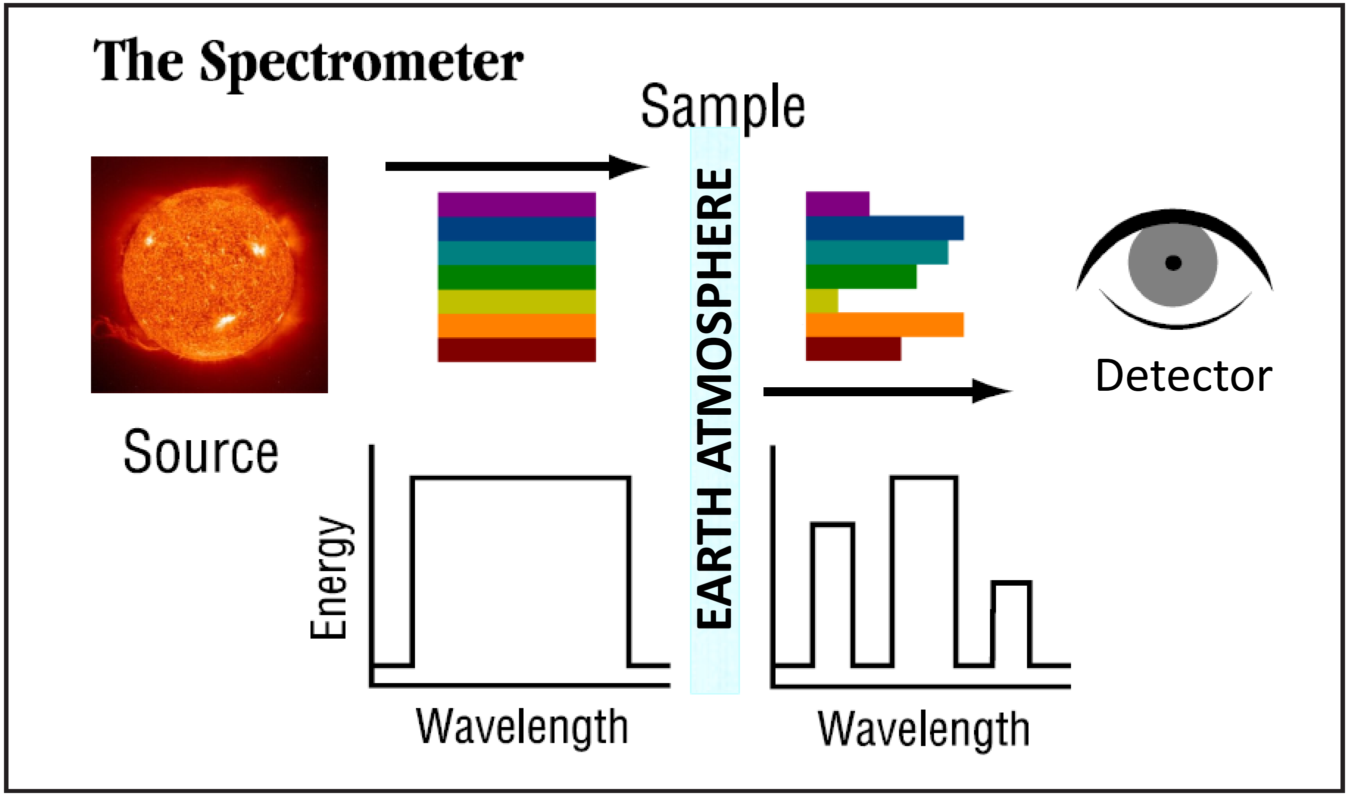
Now 45 years of continuous measurements, with three instruments!



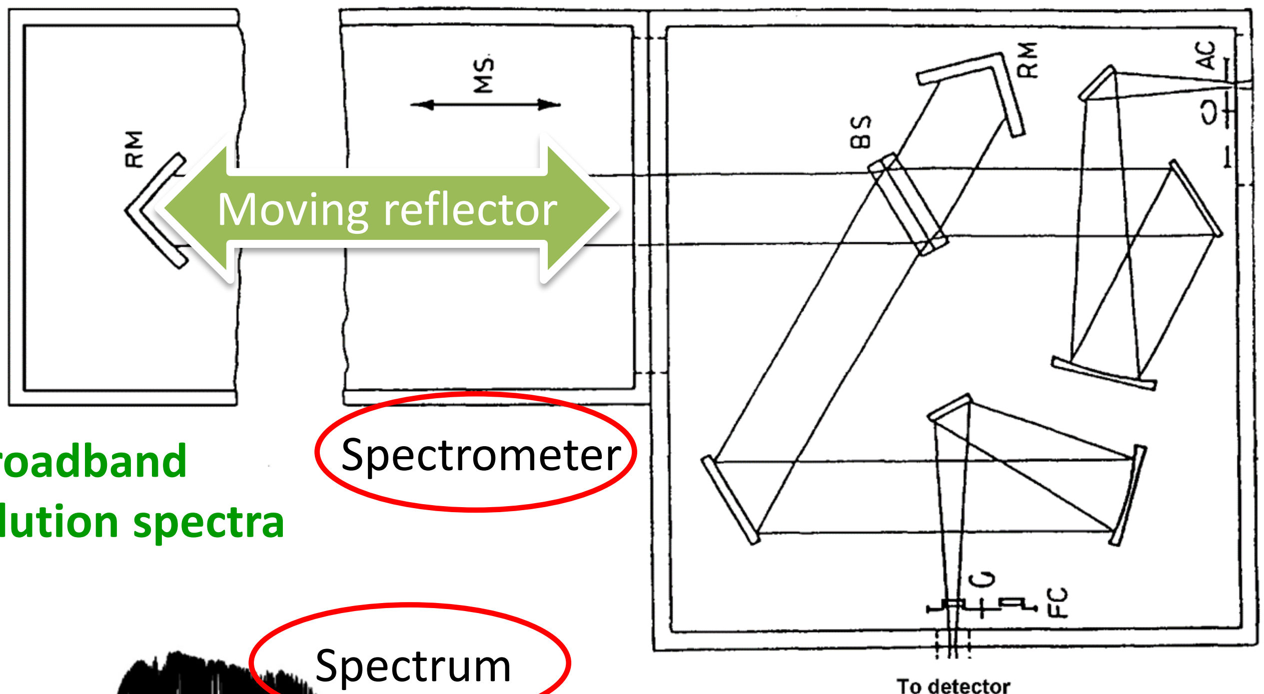


INFRARED REMOTE-SENSING

Remote-sensing of the atmospheric composition: the basic principle



Remote-sensing of the atmospheric composition with FTIR instruments

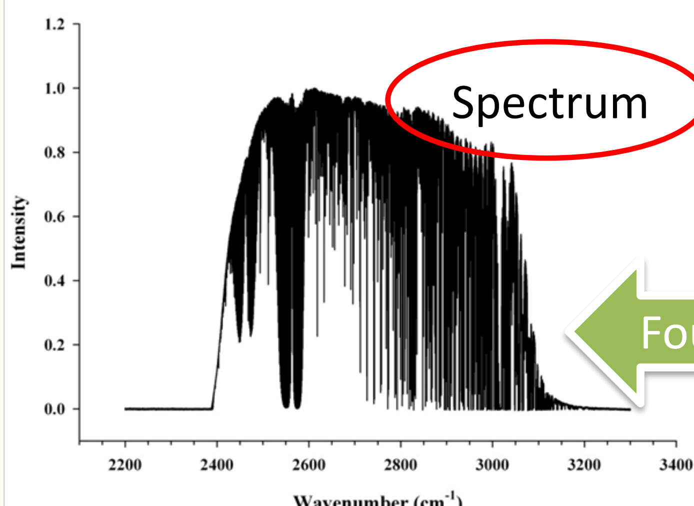


Moving reflector

Spectrometer

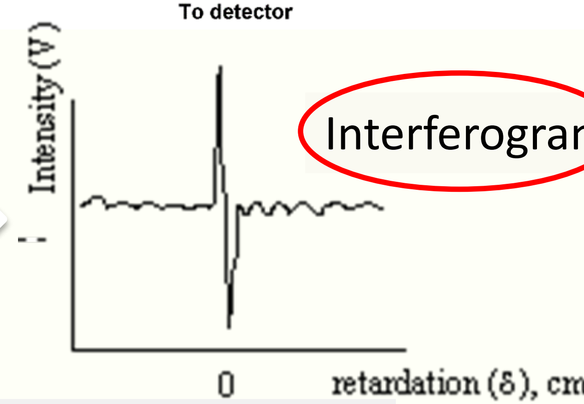
Clear-sky mandatory!

⇒ broadband high-resolution spectra



Spectrum

Fourier



Interferogram

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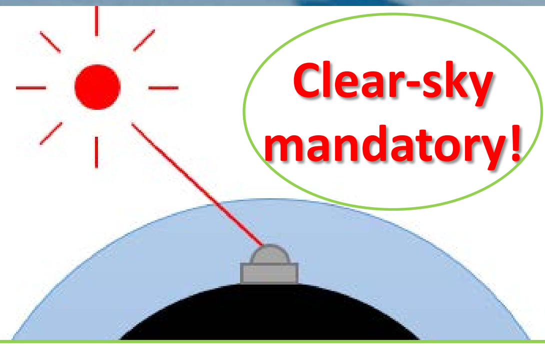
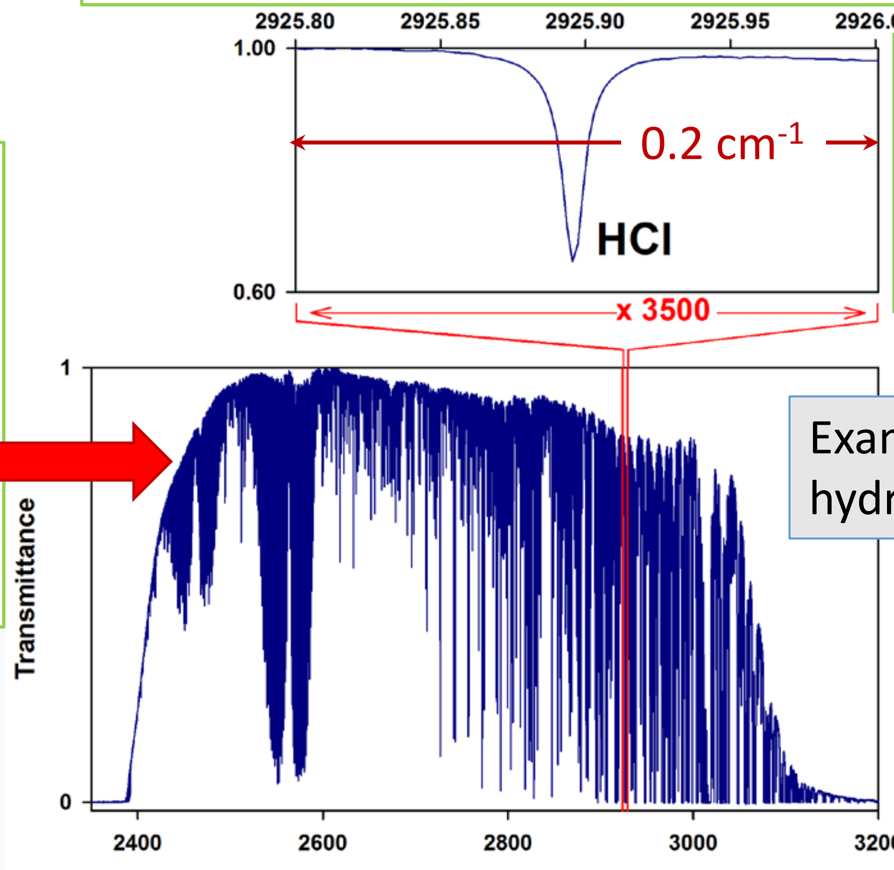
Ground-based remote sensing



- Fourier Transform Infrared (FTIR) instruments
- operated year-round under clear-sky conditions
- interferograms ⇔ (FT) *broadband* *and high-resolution* *and high S/N ratio* *IR solar absorption spectra*

Narrow spectral ranges, or micro-windows are analyzed with the SFIT-4 retrieval software, implementing the OEM method (Rodgers, 2000)



- **Area** of the lines => *total columns*
- **Shape** of the lines => *concentration profiles*

Example for hydrogen chloride (HCl)

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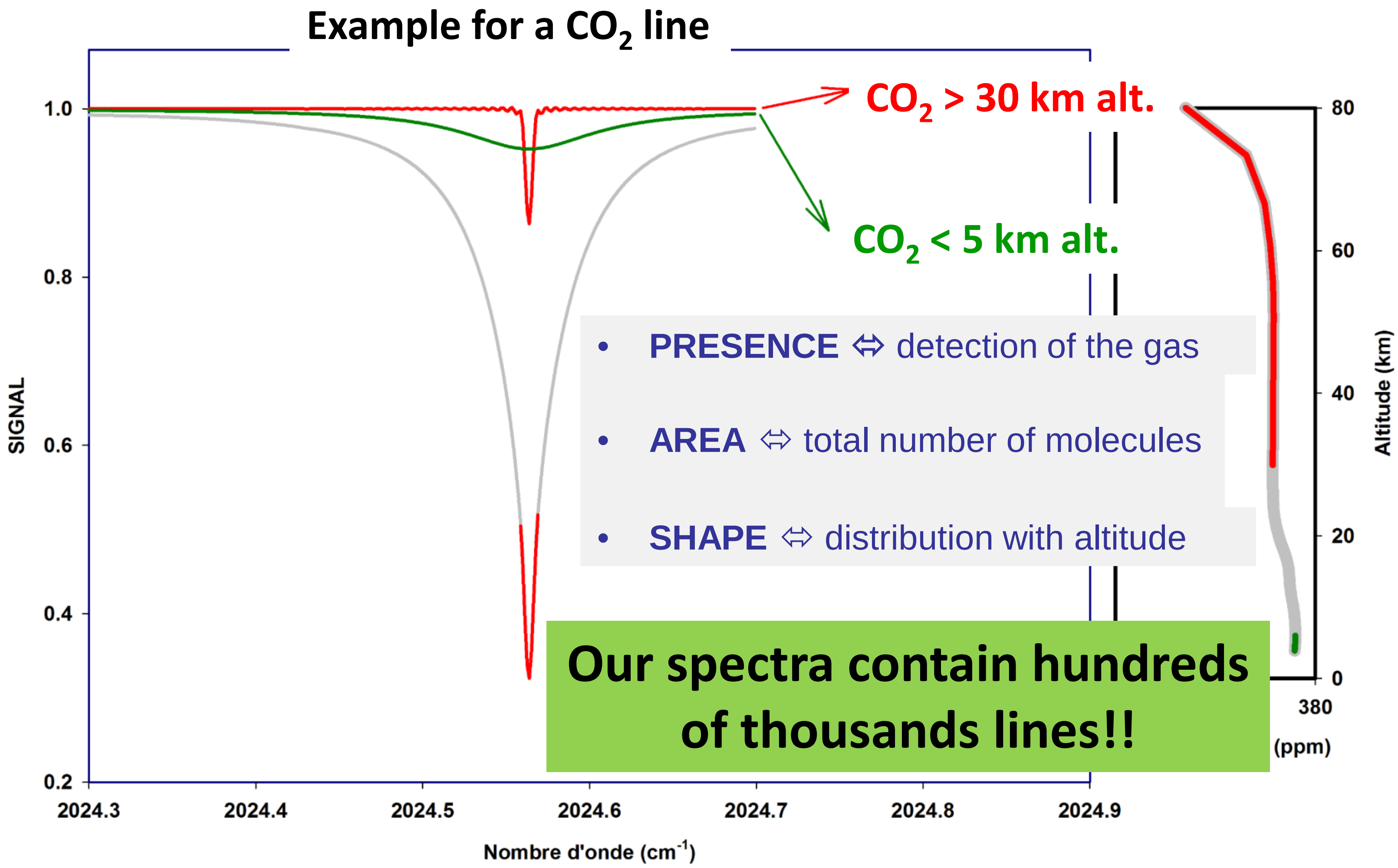
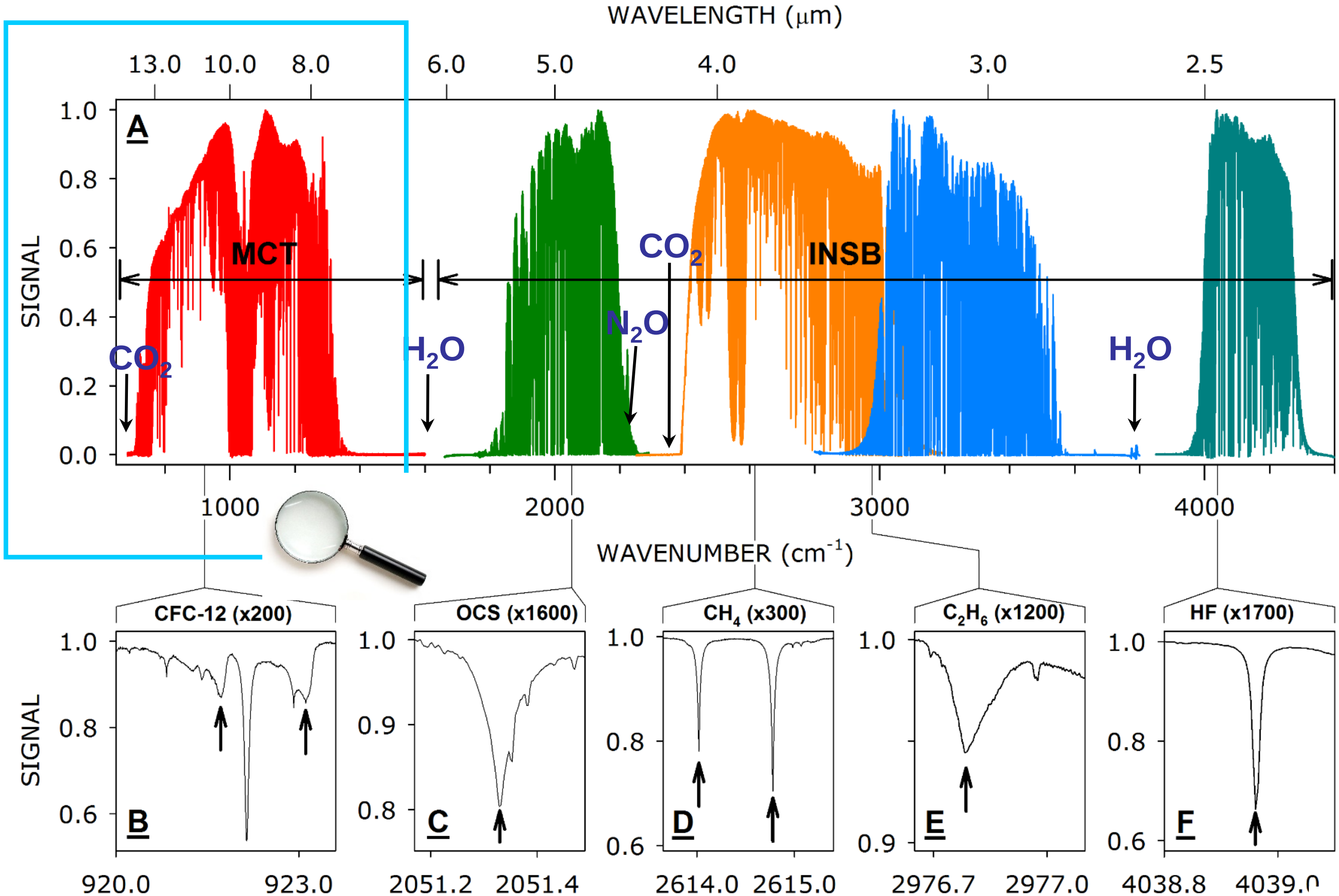
What do we learn from a single line?

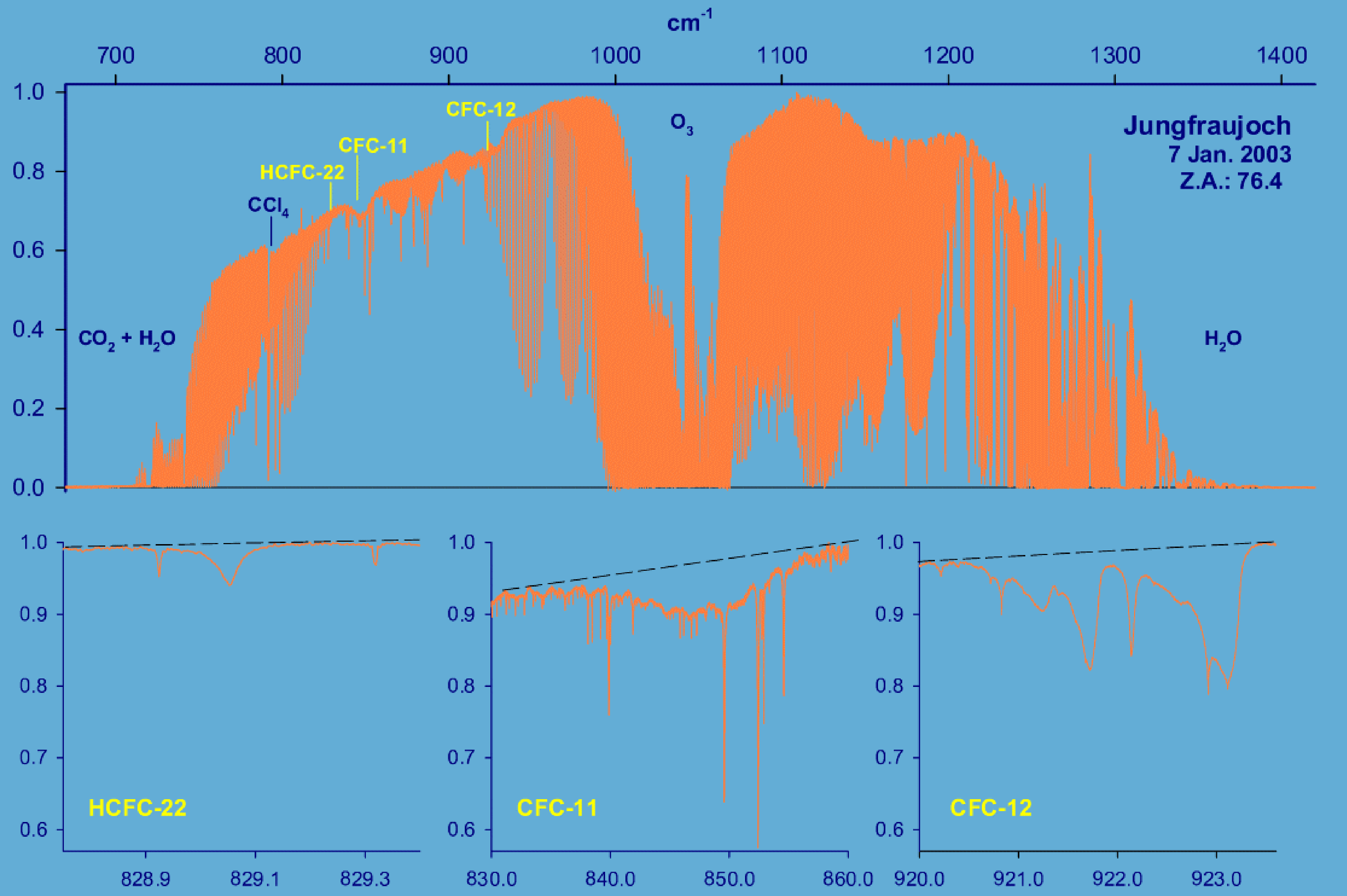


- **PRESENCE** ⇔ detection of the gas
- **AREA** ⇔ total number of molecules
- **SHAPE** ⇔ distribution with altitude

Our spectra contain hundreds of thousands lines!!

Bruker FTIR setup@Jungfraujoch: two detectors and 5 optical filters




cm-1
700
800
900
1000
1100
1200
1300
1400
1.0
0.8
0.6
0.4
0.2
0.0
CFC-12
CFC-11
HCFC-22
CCl_4
O_3
Jungfraujoch
7 Jan. 2003
Z.A.: 76.4
$CO_2 + H_2O$
H_2O
1.0
0.9
0.8
0.7
0.6
HCFC-22
828.9
829.1
829.3
CFC-11
830.0
840.0
850.0
860.0
CFC-12
920.0
921.0
922.0
923.0

Current list of FTIR target gases at Jungfraujoch (35+, still expanding!)

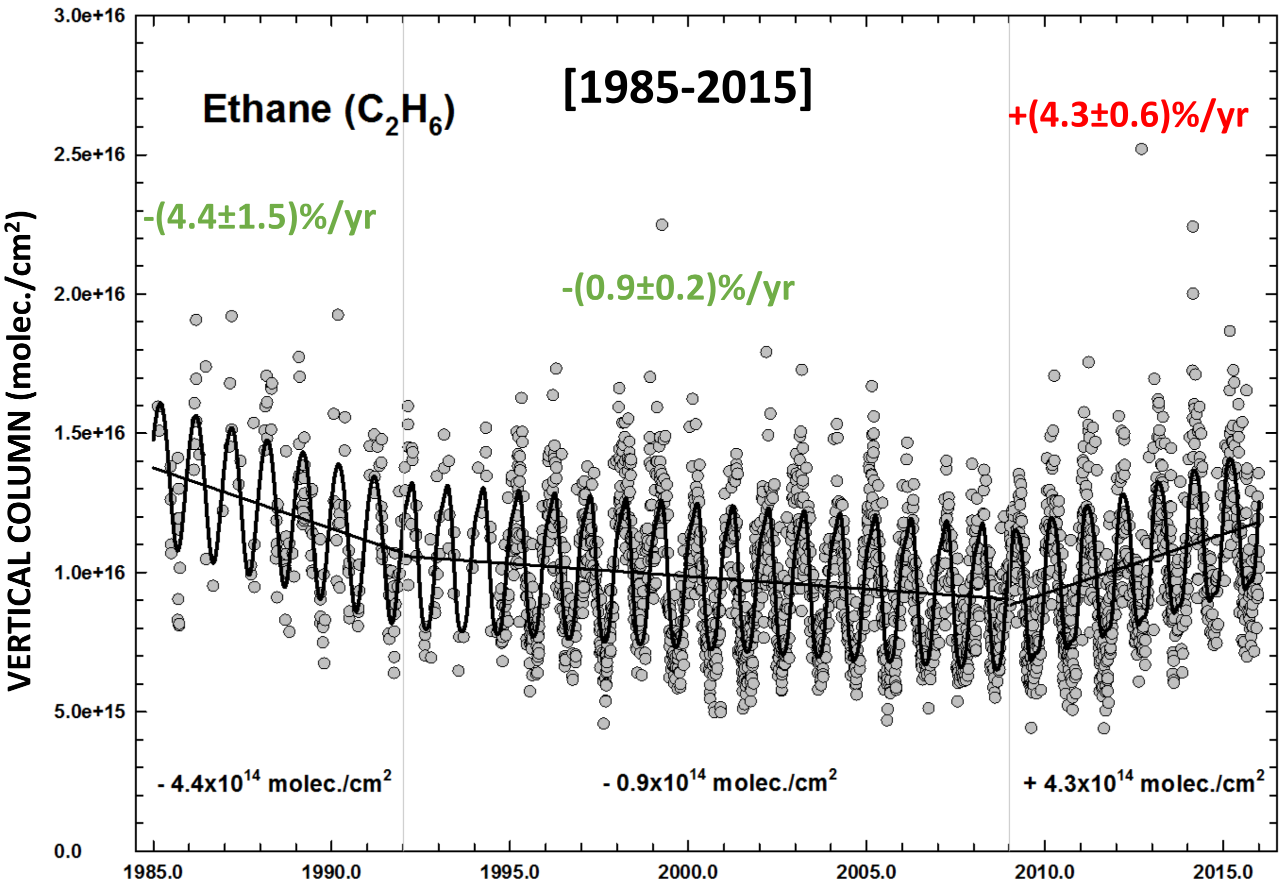
Currently, more than 35 gases are routinely retrieved from our spectra		
Major greenhouse gases	H_2O, CO_2, CH_4, N_2O, CF_4, SF_6	Support to the **Kyoto Protocol** and the **Paris Agreement** (COP21)
Related to stratospheric ozone depletion	O_3, NO, NO_2, HNO_3, $ClONO_2$, HCl, HF, COF_2, CFC-11, CFC-12, HCFC-22, HCFC-142b, CCl_4	Support to the **Montreal Protocol** on substances that deplete ozone
Air quality, biomass burning, oil production and transport	CO, CH_3OH, C_2H_6, C_2H_2, C_2H_4, HCN, HCHO, HCOOH, PAN, NH_3	**Europe's eyes on Earth**
Other	OCS, N_2, numerous isotopologues (HDO, CH_3D, $^{13}CH_4$, ^{13}CO…)	Various applications; e.g., source apportionment/attribution

ETHANE (C_2H_6) AND THE OIL & GAS SECTOR

Why ethane?

- Most abundant non-methane hydrocarbon (NMHC) in the Earth's atmosphere, **lifetime** of about 2-6 months
- Main **sink**: removal through oxidation by OH; source/precursor of tropospheric ozone ($HO_x/NO_x/O_x$)
- Main **sources**: of anthropogenic origin, with typically 62% from leakage during **production *and* transport** of natural gas, 20% from **biofuel combustion**, 18% from **biomass burning** (Xiao et al., JGR, 2008)
- Until about 2009, a **prolonged decrease** (-1 to -2.7%/yr) has been documented, with **global emissions dropping from 14 to 11 Tg/yr over 1984-2010**: attributed to successful measures aiming at reducing fugitive emissions from its fossil fuel sources

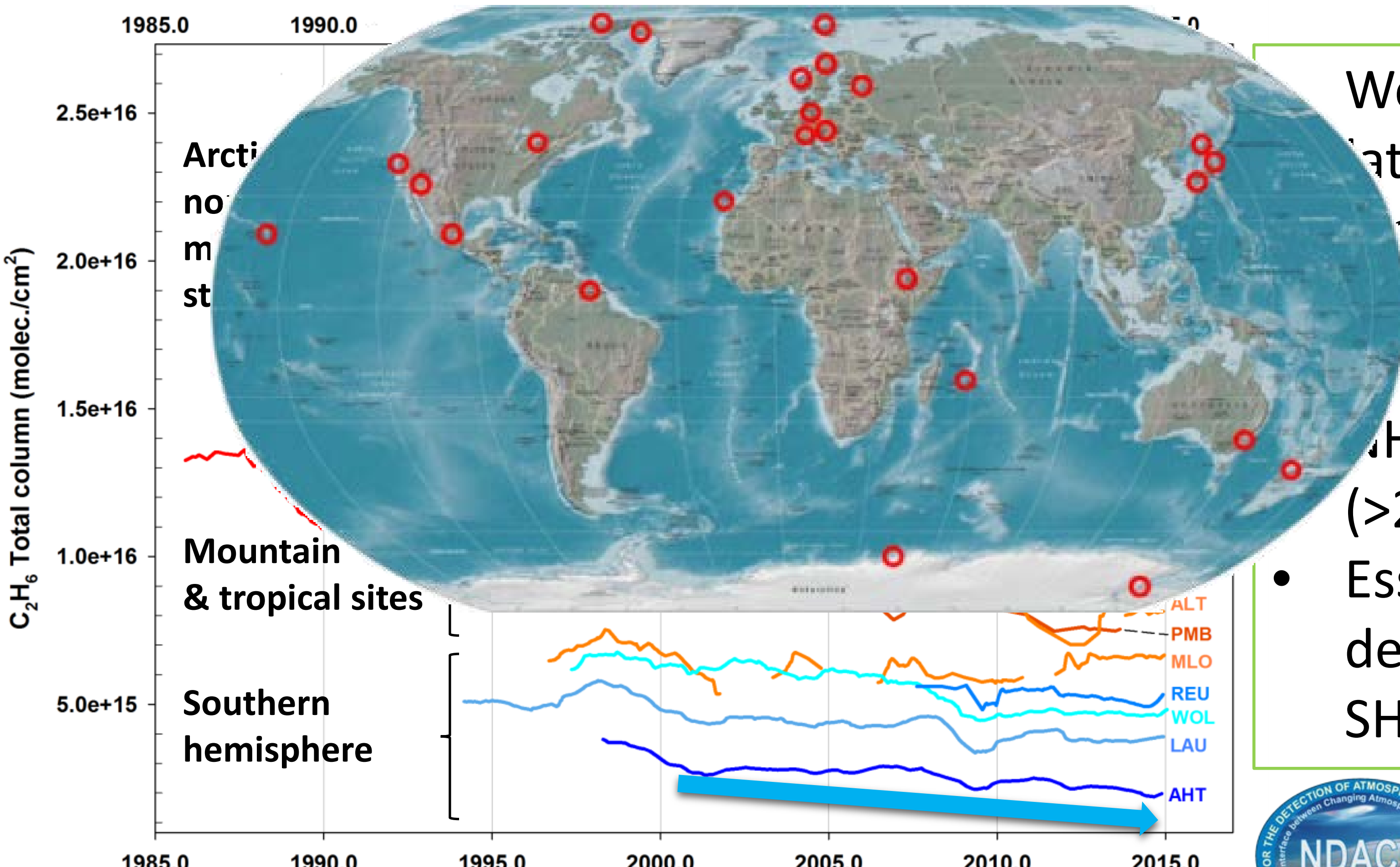
Multi-decadal time series of the Jungfraujoch: detection of an upturn



Updated from Franco et al., JQSRT, 2015

- Sharp increase took place from ~2009 onwards, after 20+ years of decrease
- Observed at a remote location (Swiss Alps, 3.6 km a.s.l.)
- Increase throughout the troposphere and lower stratosphere (partial column time series)

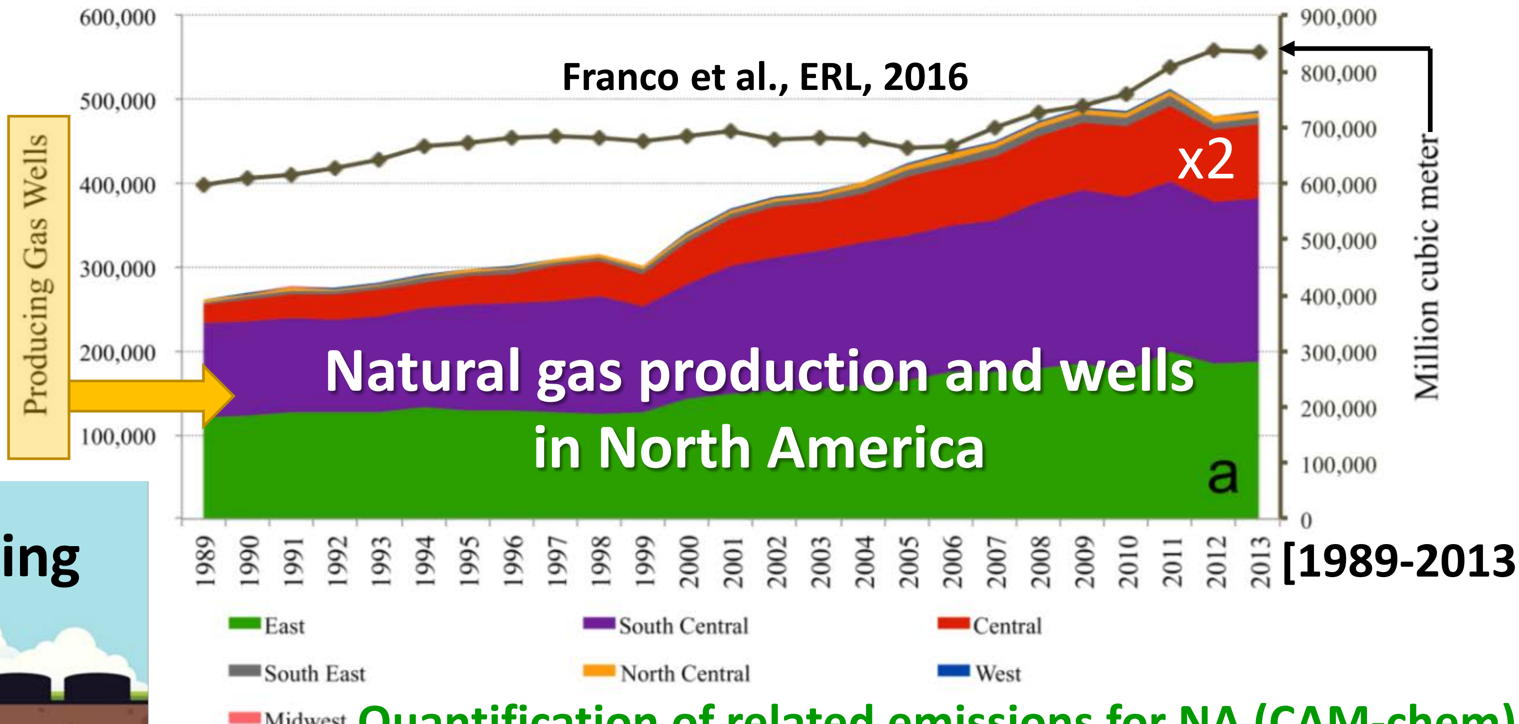
This was a semi-global feature

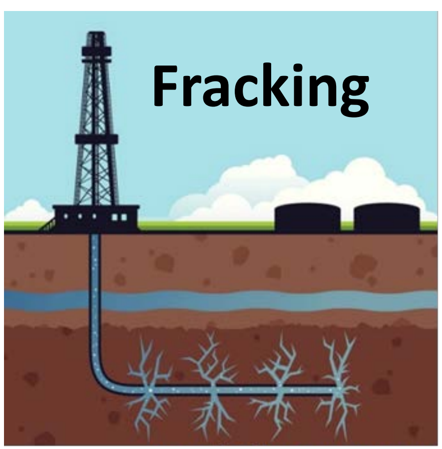


Well-known latitudinal gradient
significant increase at all NH sites (>2006-2009)

- Essentially decreasing for SH (2015…)

Resulting from the oil & natural gas boom in North America





www.huffingtonpost.com

Quantification of related emissions for NA (CAM-chem)

Increase of the NA anthropogenic emissions by 75%,
from 1.6 Tg/yr in 2008 to 2.8 Tg/yr in 2014
Using a single C_2H_6/CH_4 emission ratio per source category, we estimated that the concurrent CH_4 O&G emissions have increased from 20 Tg/yr in 2008 to 35 Tg/yr in 2014

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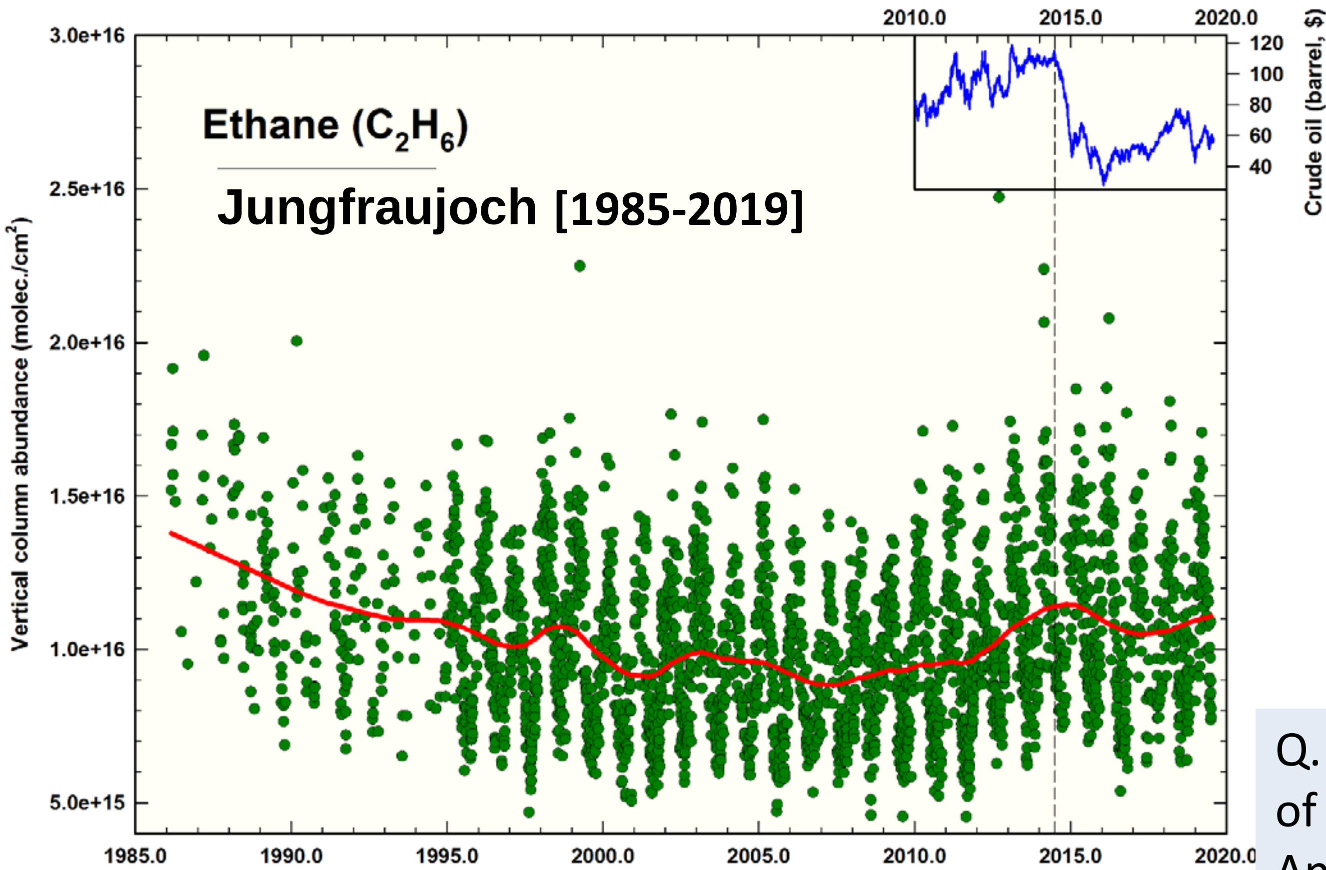
Ethane time series update

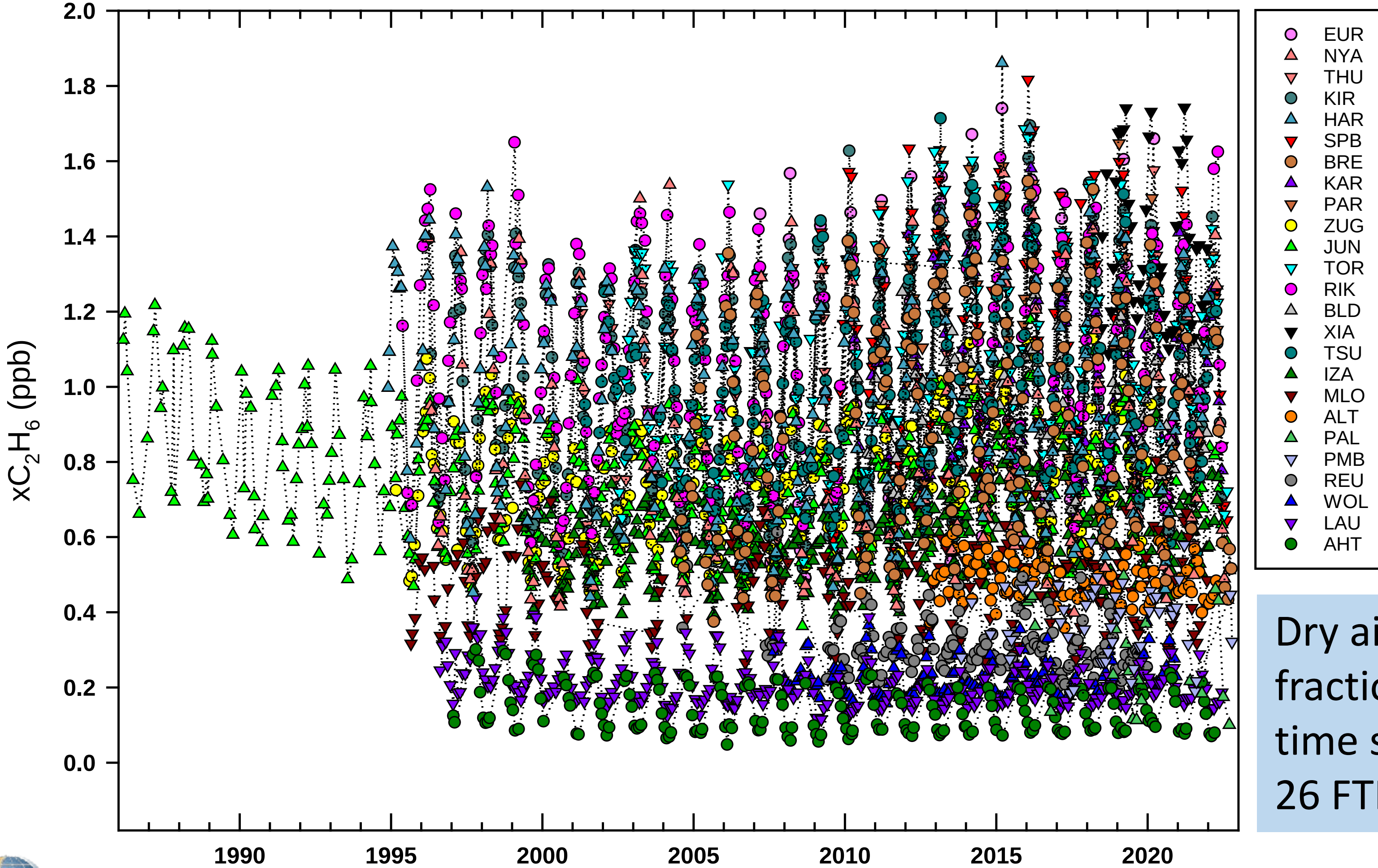


- The post-2008 rise lasts until ~2015
- Then we see a drop
- There is a parallel with the evolution of the crude oil price (see top panel)
- Fracking is not profitable under low oil prices

Q.: what will be the impact of the war in Ukraine?
And of LNG production and transport?

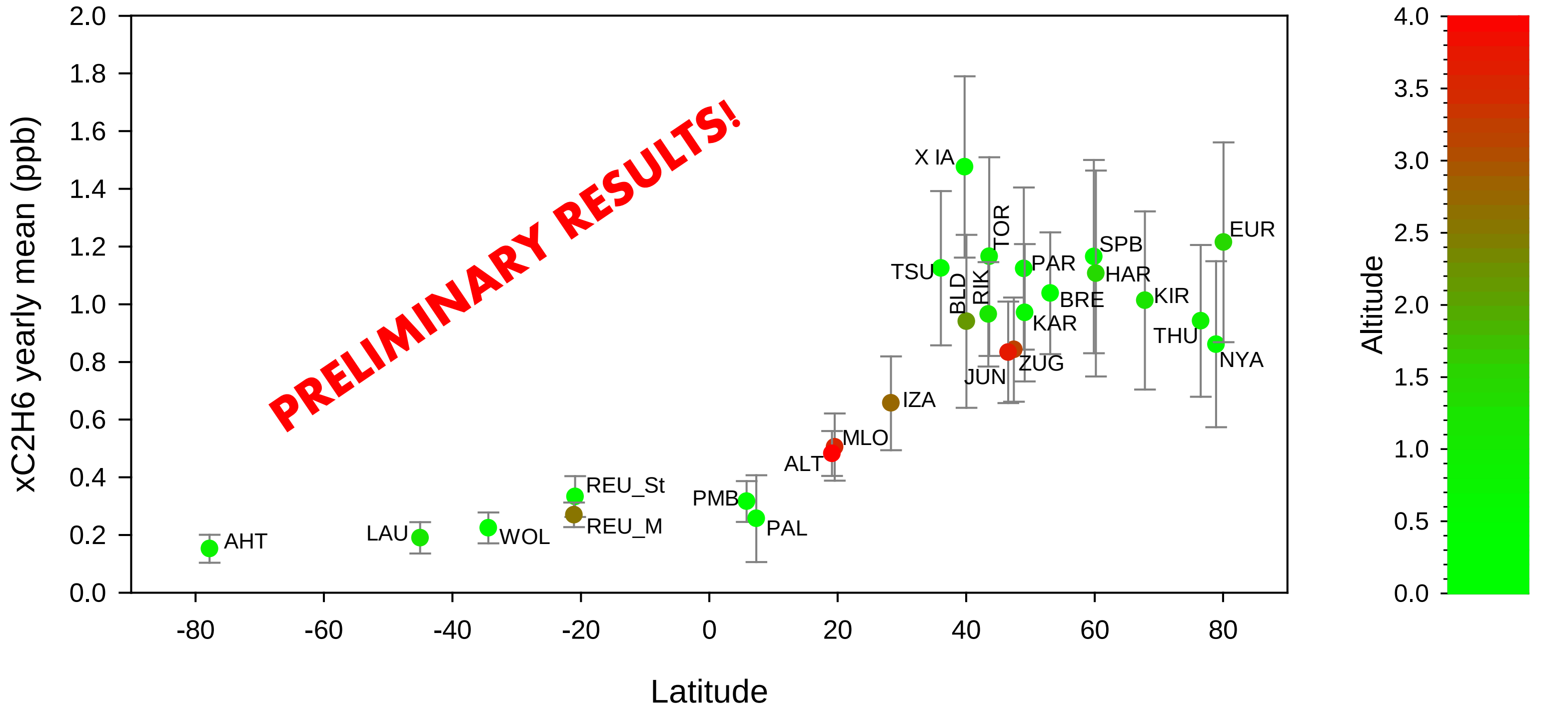
At the global scale

Atmospheric ethane from FTIR remote-sensing measurements



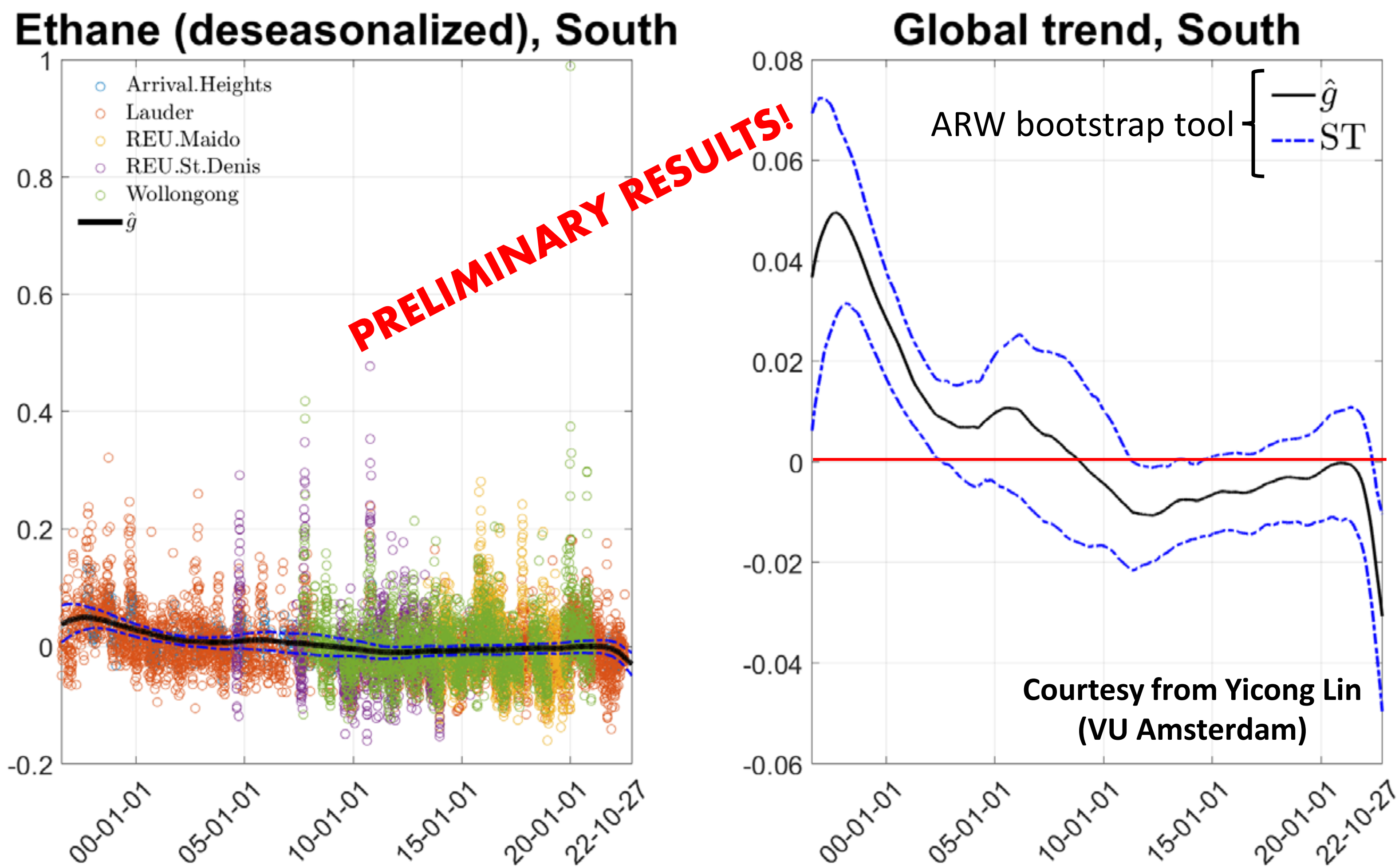
Dry air mole fraction (xC_2H_6) time series at 26 FTIR sites

At the global scale

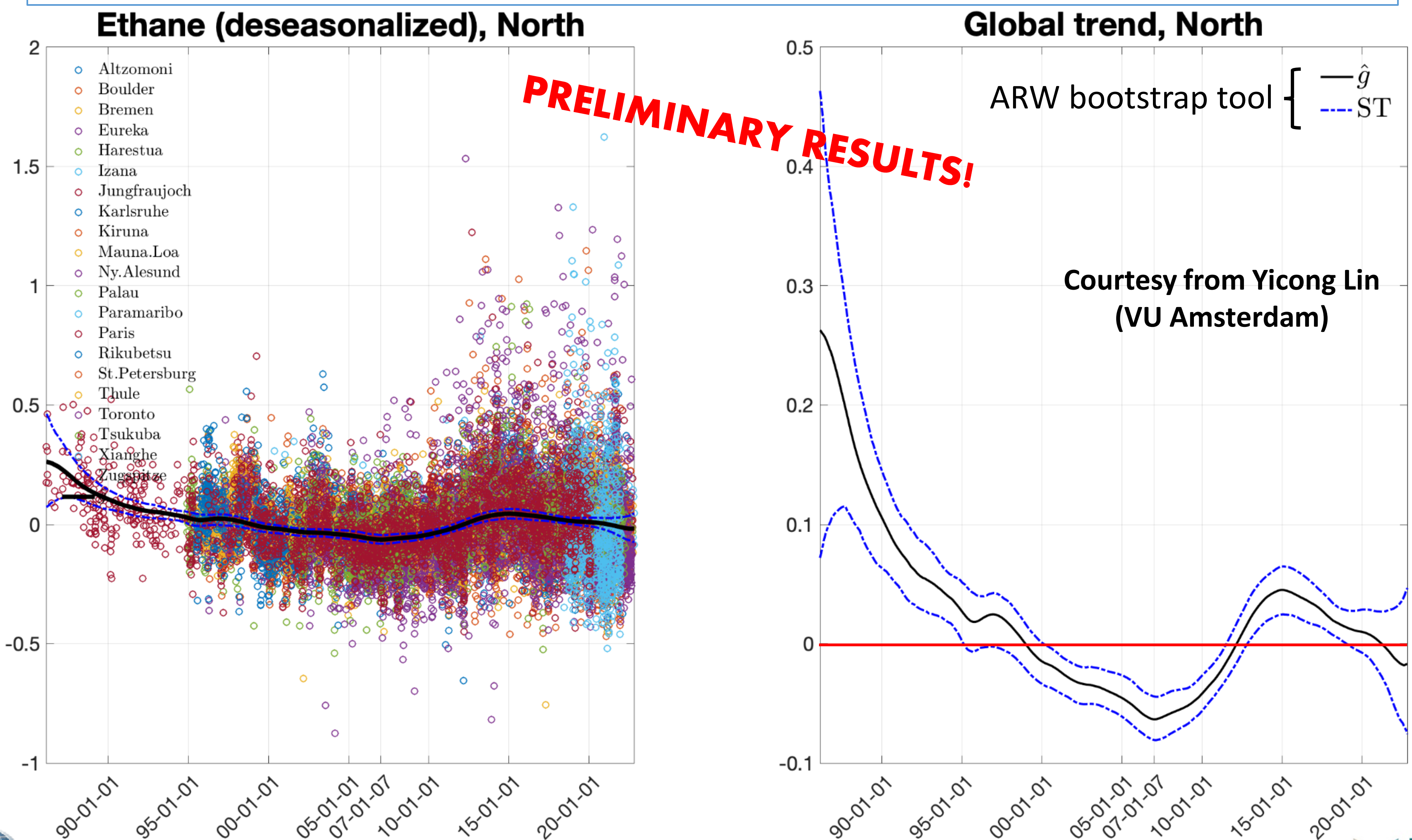


Yearly averages for 2019 as a function of latitude and altitude

Interhemispheric trends: SH



Interhemispheric trends: NH



$CH_3-C(O)-O_2-NO_2$

FIRST GROUND-BASED FTIR RETRIEVALS OF PEROXYACETYL NITRATE (PAN)

Background information on PAN(*)

- PAN is a reservoir of NO_x (=NO+NO_2) ("reversible storage" of NO_x)
- PAN is formed when non-methane volatile organic compounds (NMVOCs) oxidation products react with NO_x, meaning that it has anthropogenic and natural sources (fossil fuel combustion, biomass burning, lightning, and processes responsible for NMVOC emissions)
- Since PAN lifetime can reach several months in the cold upper troposphere, NO_x can be transported in the form of PAN far away from the region of primary emission and formation
- NO_x is released by thermal decomposition
- Its thermal decomposition in remote areas leads to the efficient formation and redistribution of tropospheric ozone (O_3), with important implications for both tropospheric oxidative capacity and air quality

(*) Tereszchuk et al., 2013; Fischer et al., 2014

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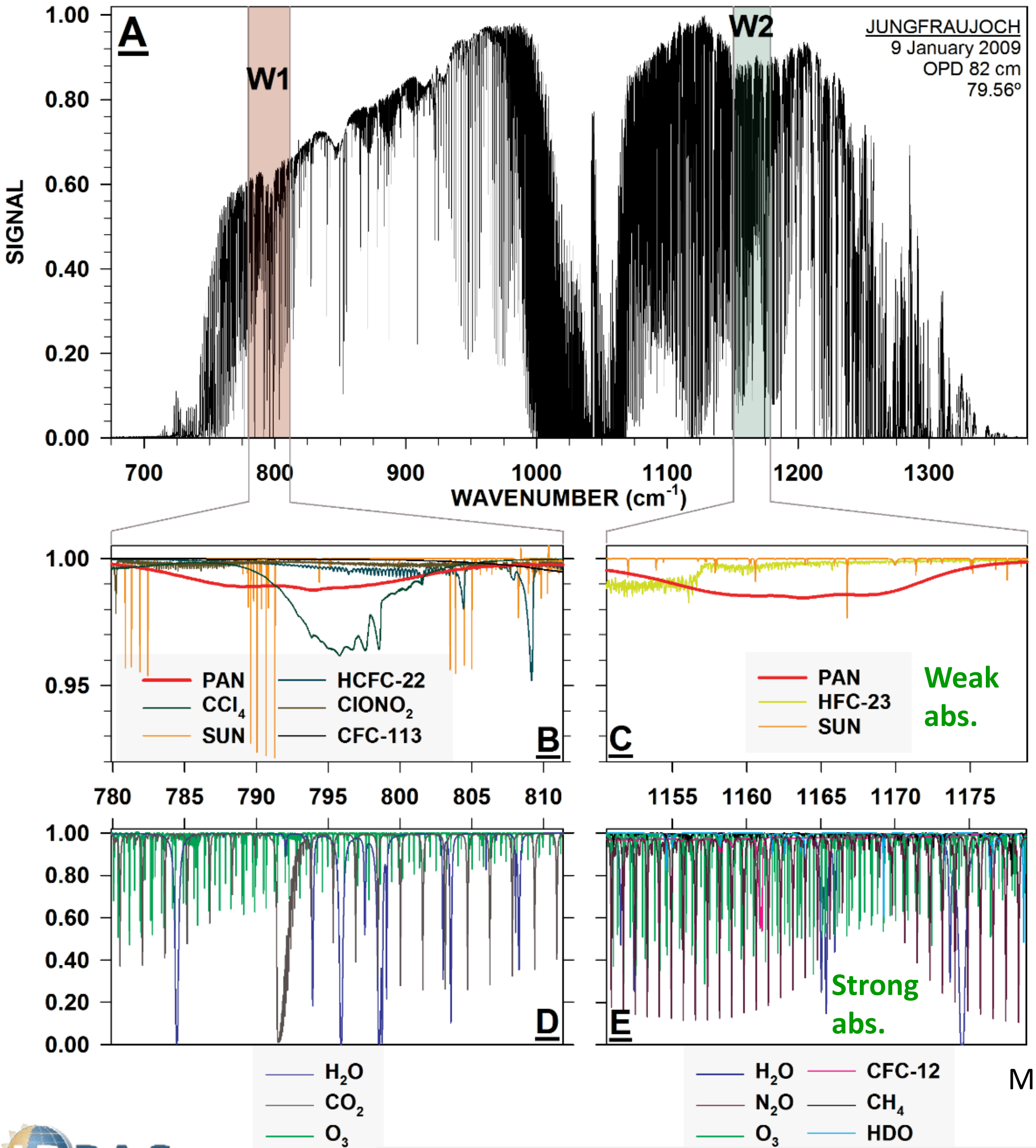


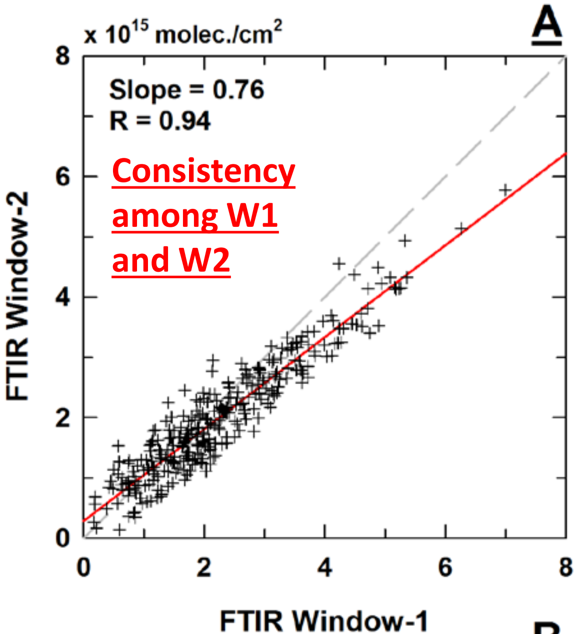
Retrieval windows

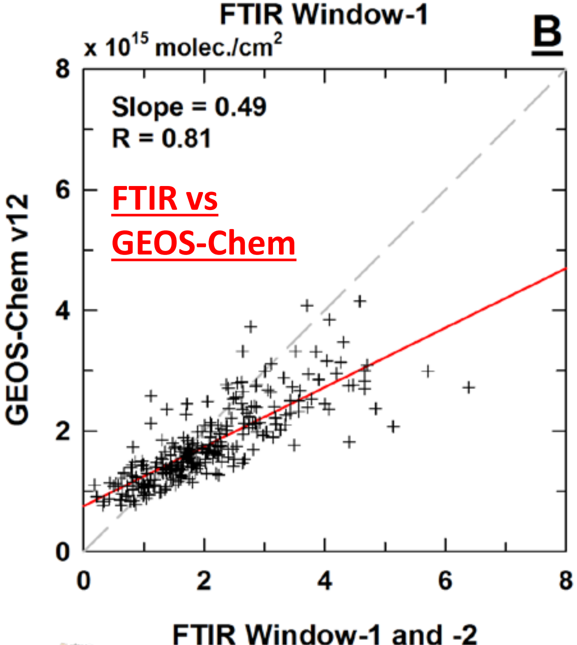
- From the ground, there are two unblinded PAN features
- These are broad, unstructured and quite weak (few %)
- The retrieval is challenging, with broad windows spanning many interferences

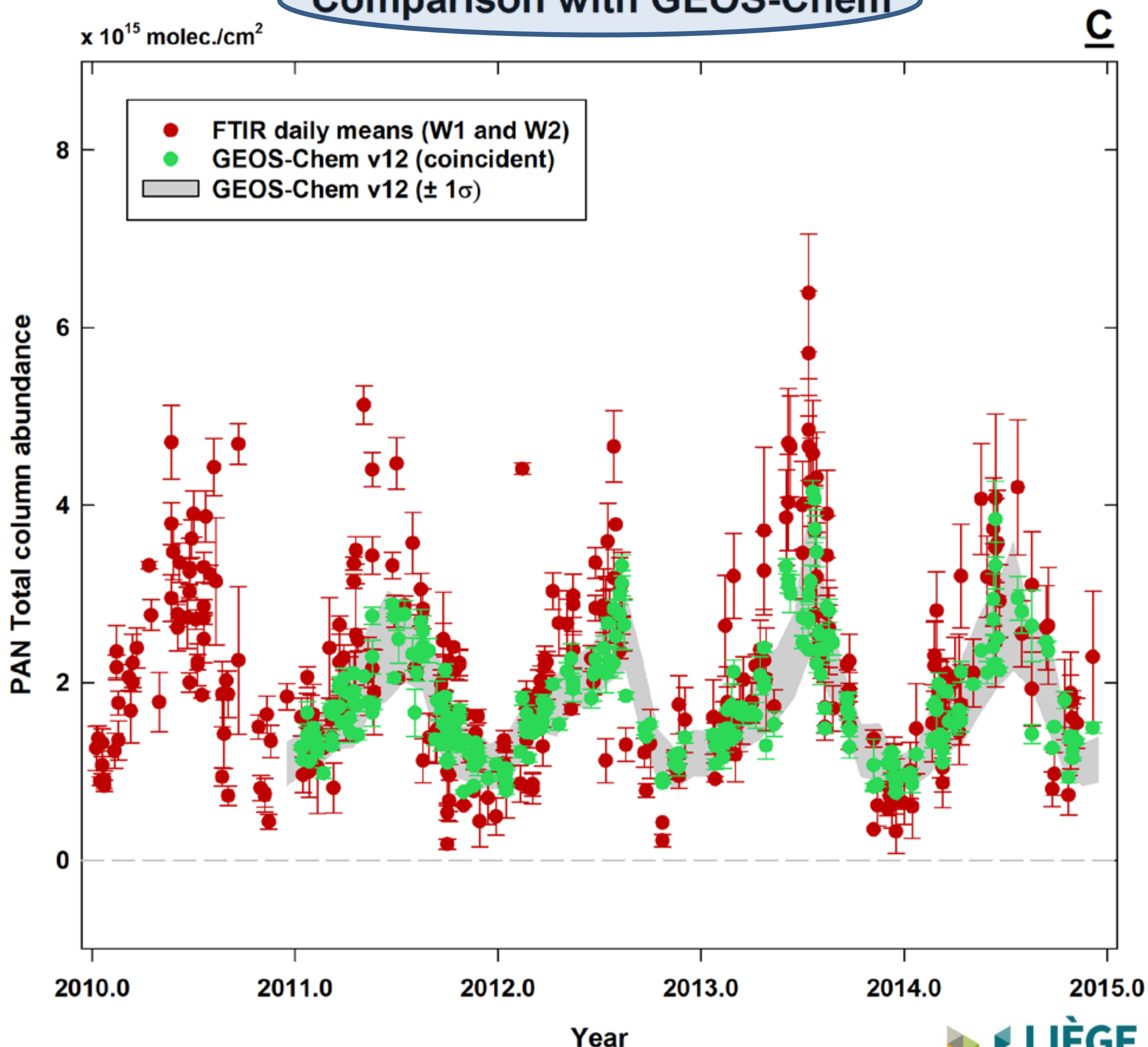
Mahieu et al., 2021

PAN above Jungfraujoch

Comparison with GEOS-Chem

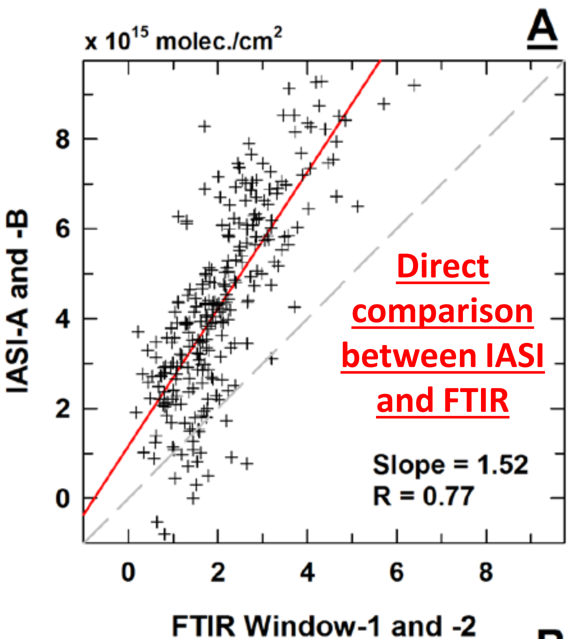


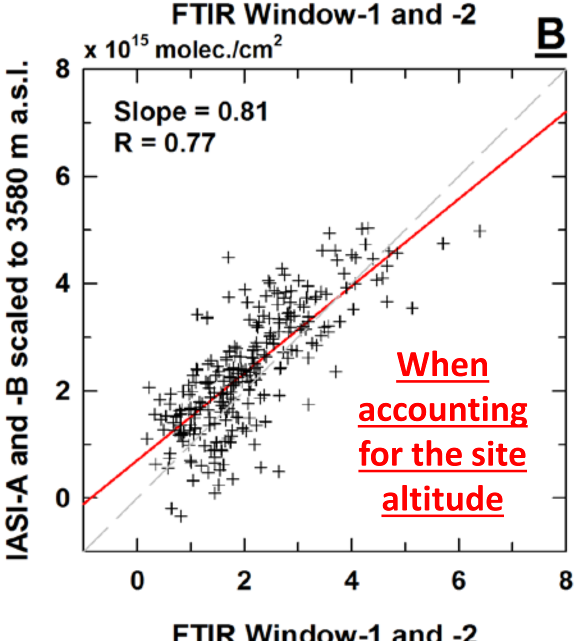


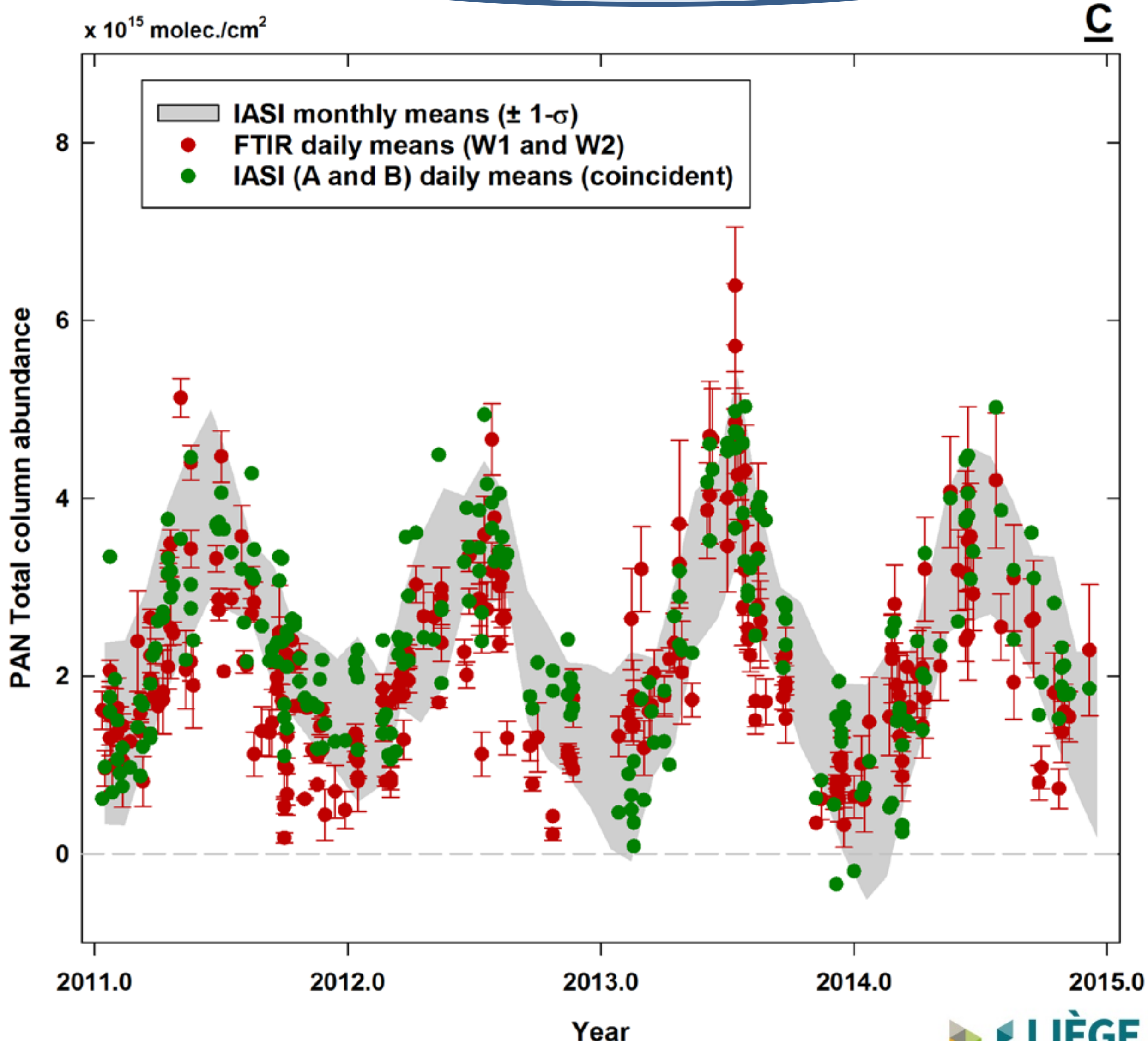


PAN above Jungfraujoch

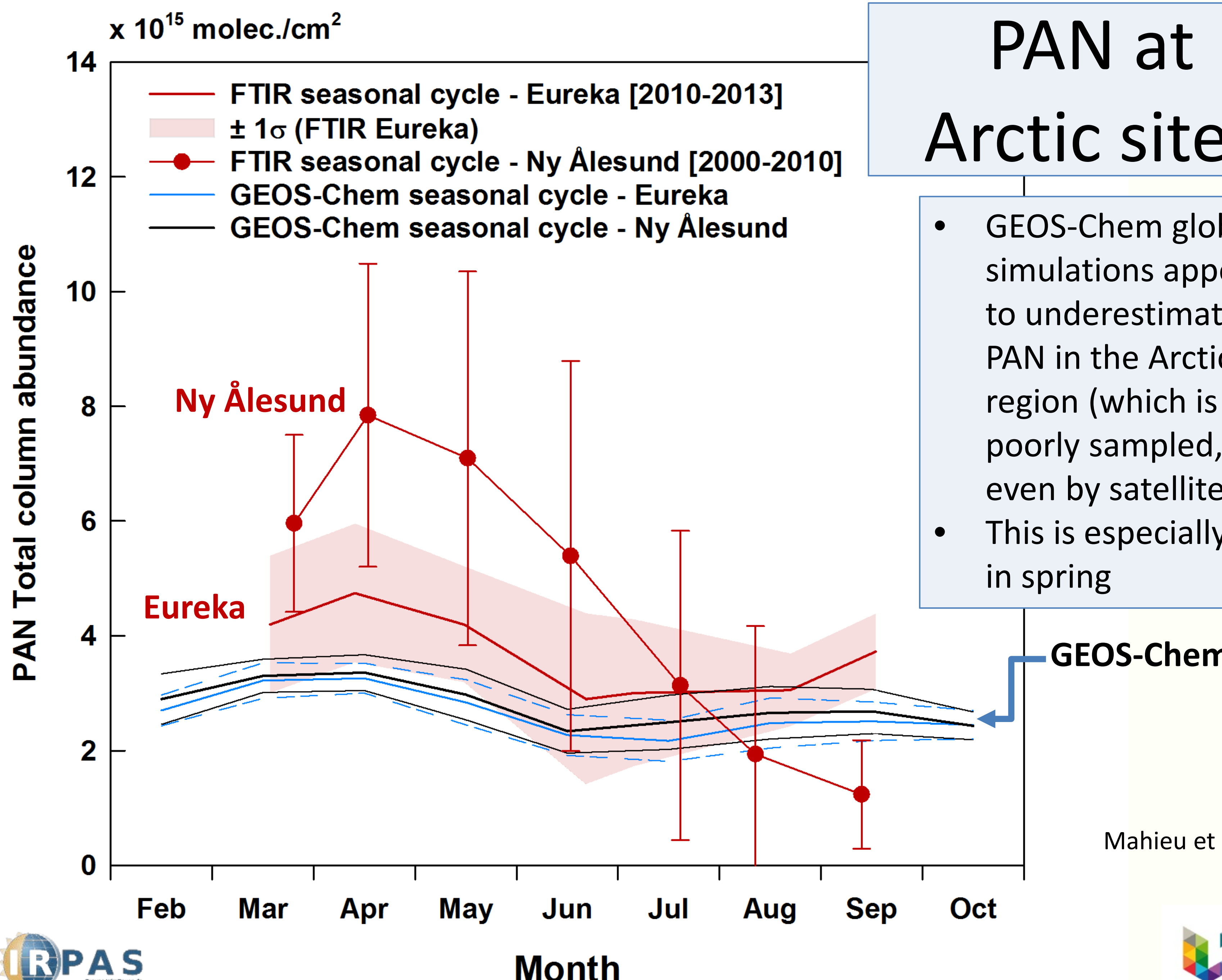
Comparison with IASI-A and -B







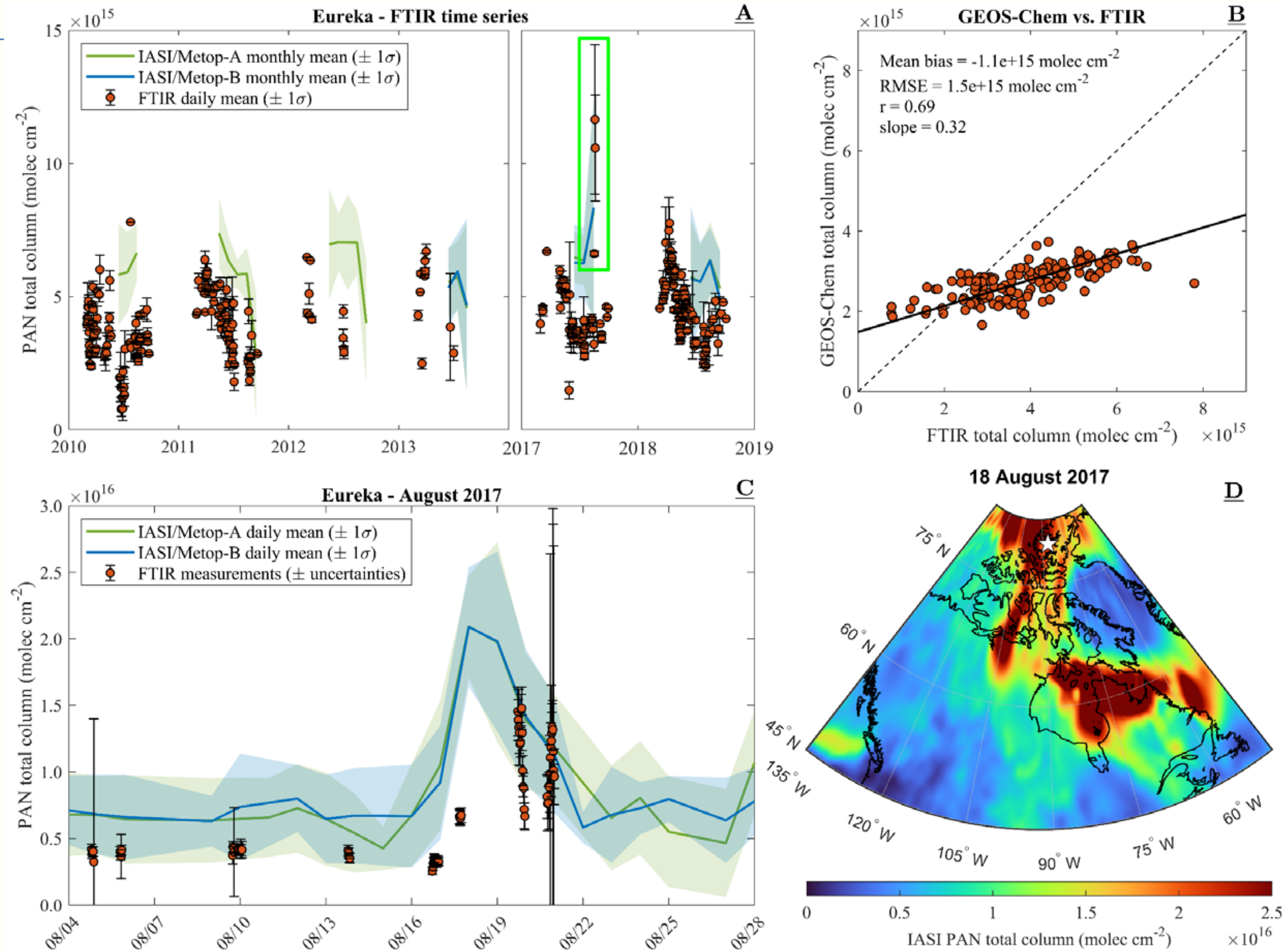
PAN at Arctic sites



- GEOS-Chem global simulations appear to underestimate PAN in the Arctic region (which is poorly sampled, even by satellites!)
- This is especially true in spring

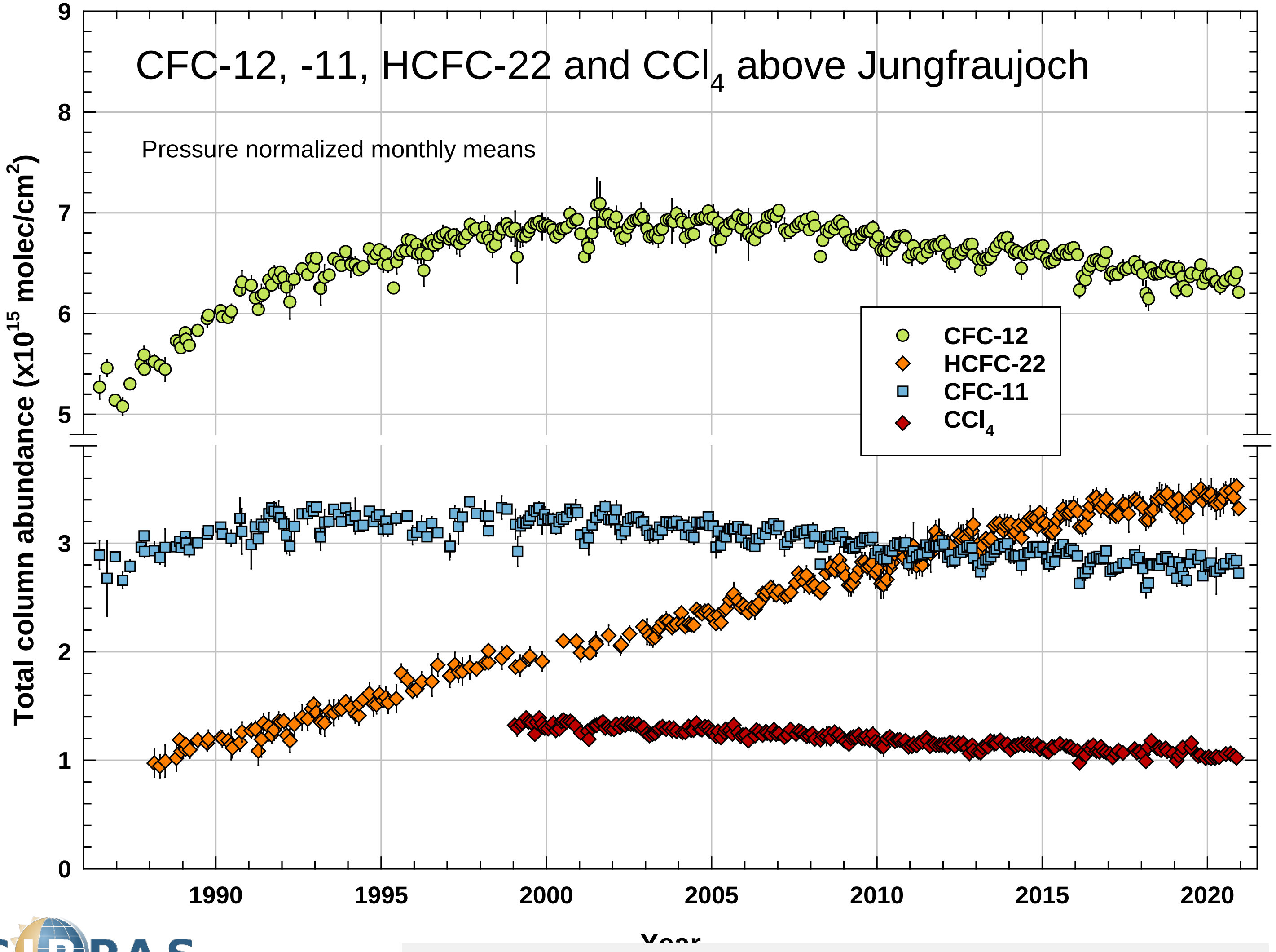
Mahieu et al., 2021

PAN in fire plumes



SUPPORT TO THE MONTREAL PROTOCOL ON SUBSTANCES THAT DEPLETE THE OZONE LAYER

Monitoring of source gases



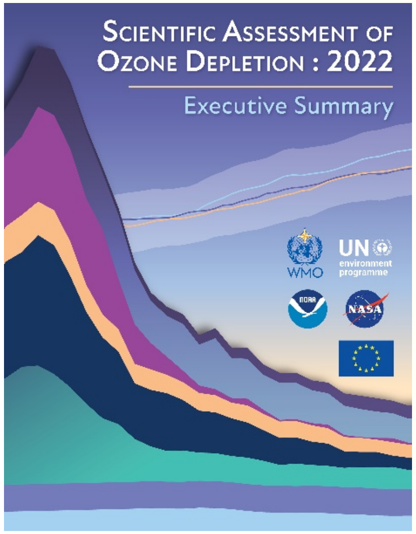


Figure to appear in next edition of the Scientific Assessment of Ozone Depletion (2022)

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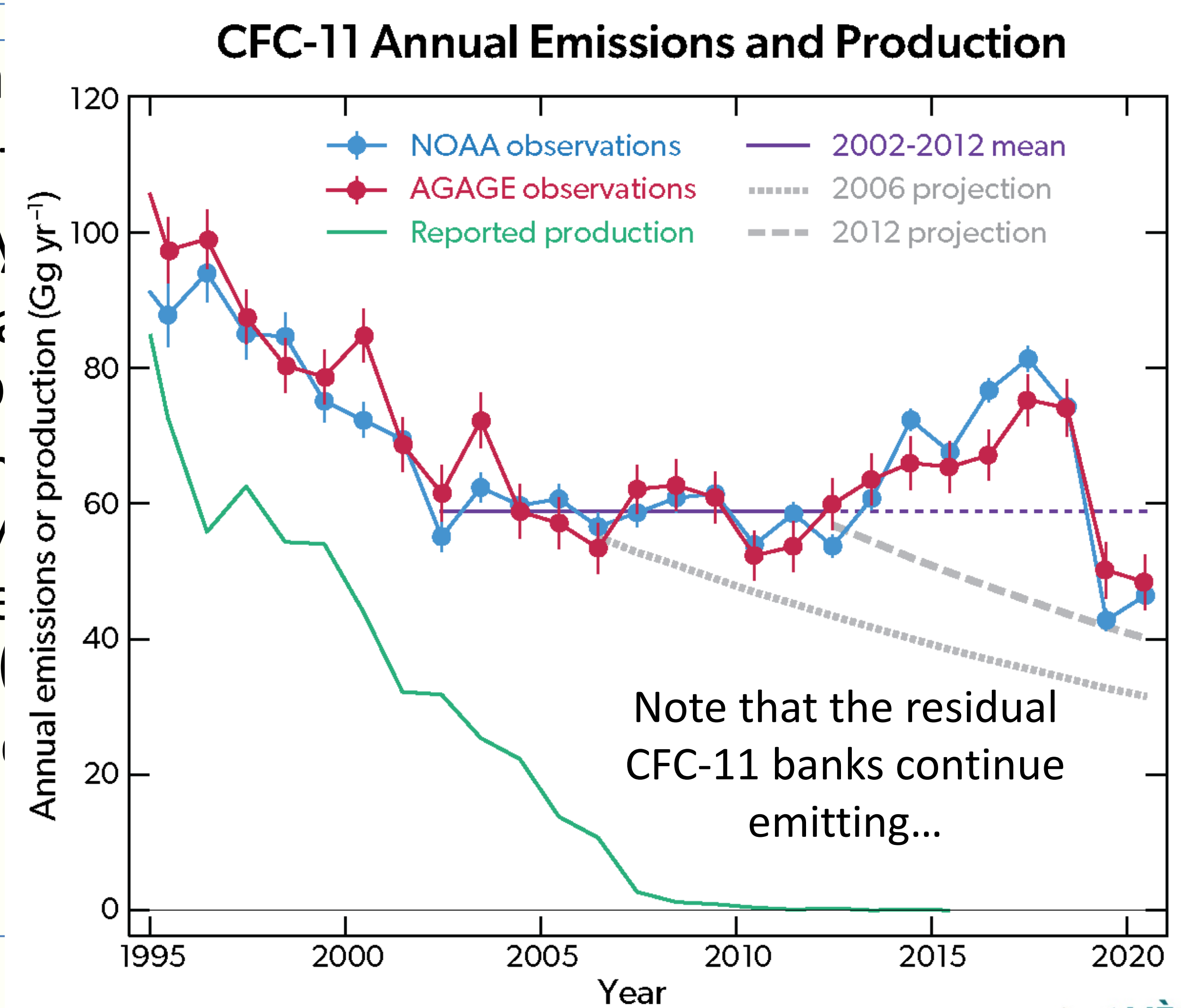
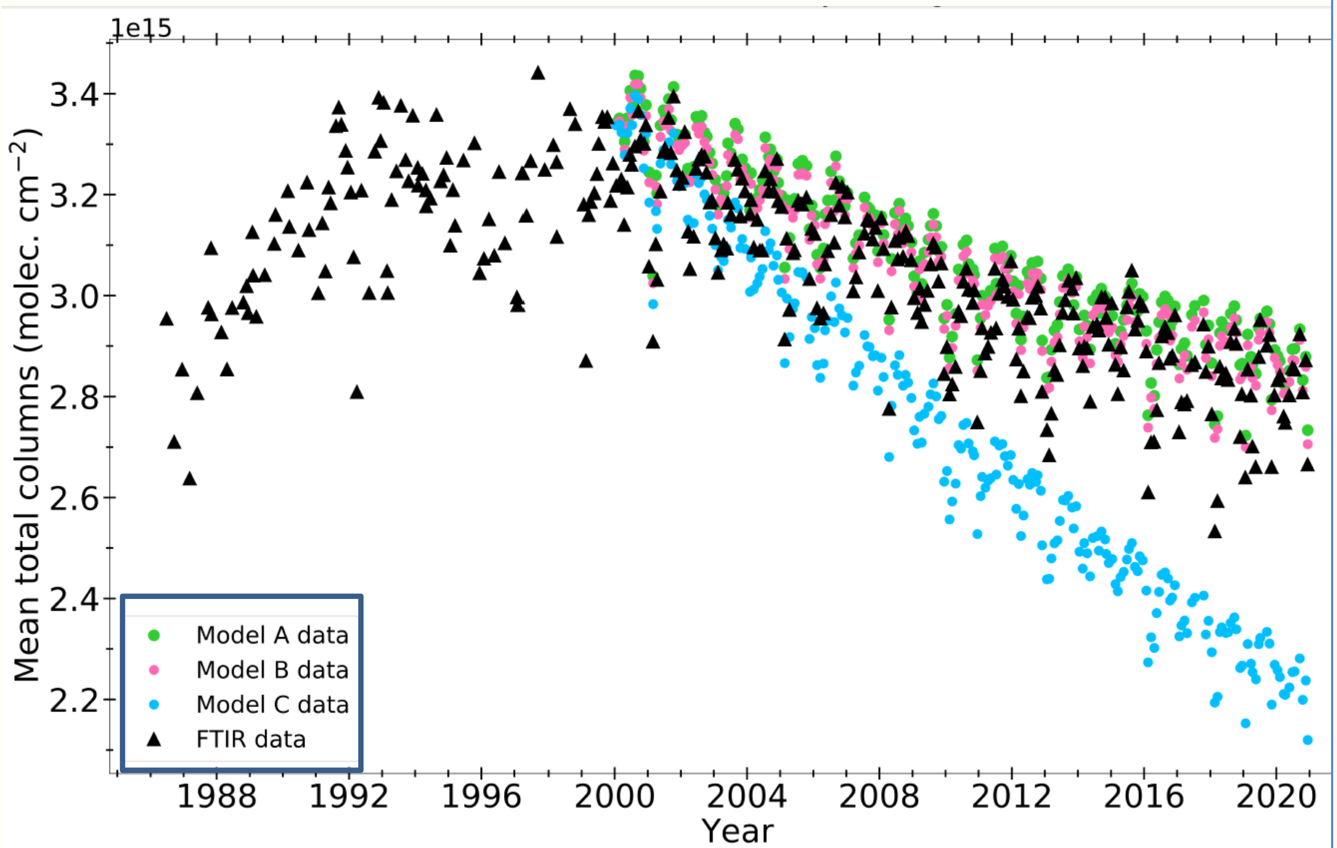
CFC-11: undeclared production and emissions

- Second most a
- Solely from an
- Long-lived (52y
- Production pha tropospheric b
- **BUT** this declin identified by M mostly to unde eastern China (
- The correspon (13±5)Gg



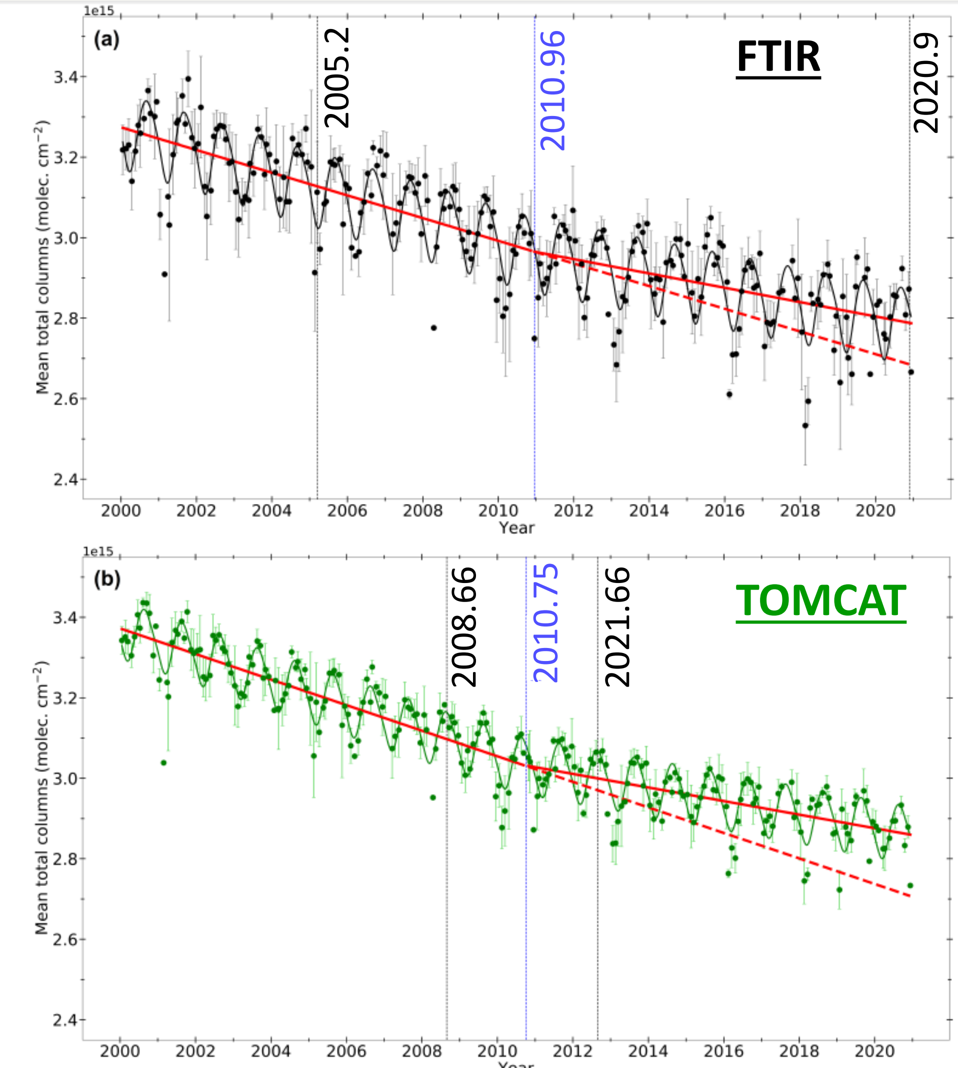
The Jungfraujoch time series of CFC-11



Pardo Cantos et al., 2022

=> Comparison with TOMCAT runs (ULeeds)
=> The model A and B runs implement best emission scenarii for CFC-11, including the undeclared emissions as derived from AGAGE
=> The model C run corresponds to the simple decay of CFC-11 (zero emissions)
=> Very good agreement is observed between FTIR and TOMCAT columns

The CFC-11 trends for Jungfraujoch



Pardo Cantos et al., 2022

=> Using the broken trend approach (Friedrich et al., 2020)
=> The statistical tool (auto-regressive wild bootstrap/ARW) is searching for a break point in the dataset and then determines robust uncertainties for its location as well as the pre- and post-linear functions
=> The dashed lines denote the expected evolution of CFC-11 without the renewed production and emissions

	Break	Rate1	Rate2
FTIR	2010.96	-0.95±0.13	-0.61±0.15
TOMCAT	2010.75	-1.03±0.08	-0.55±0.09

2σ level of uncertainty

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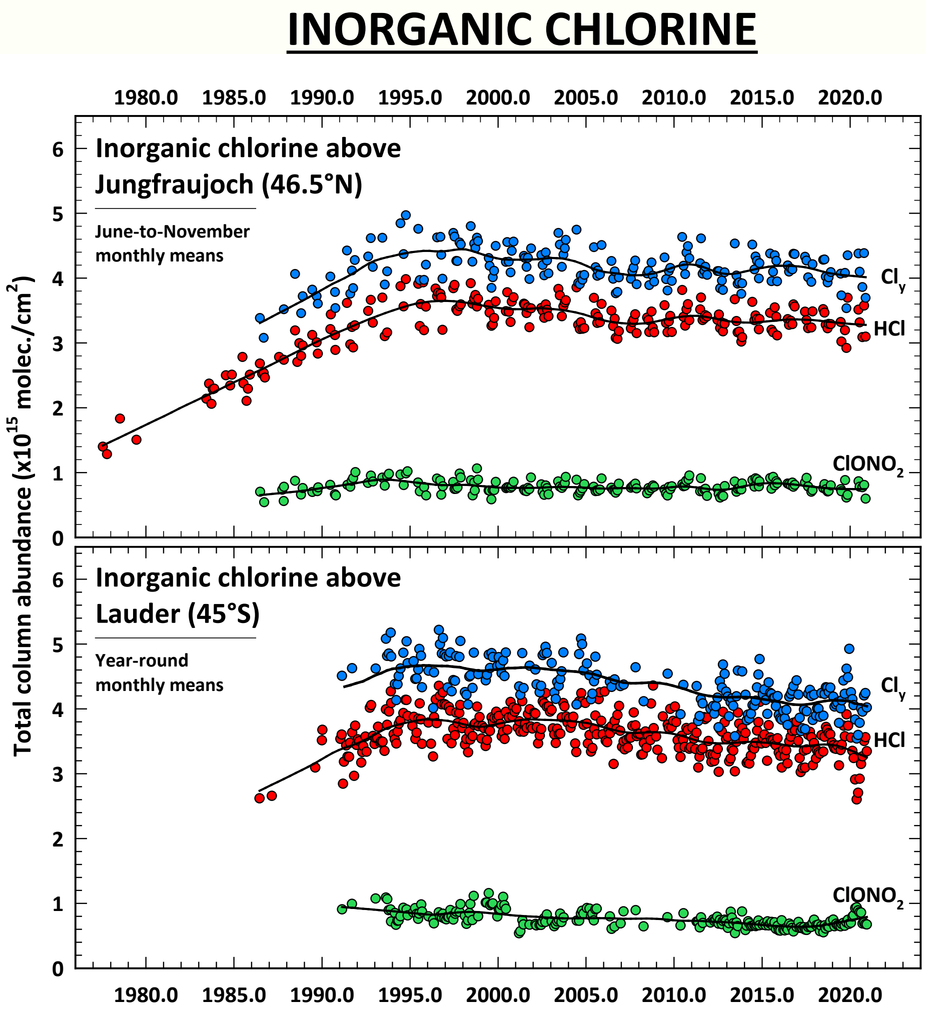
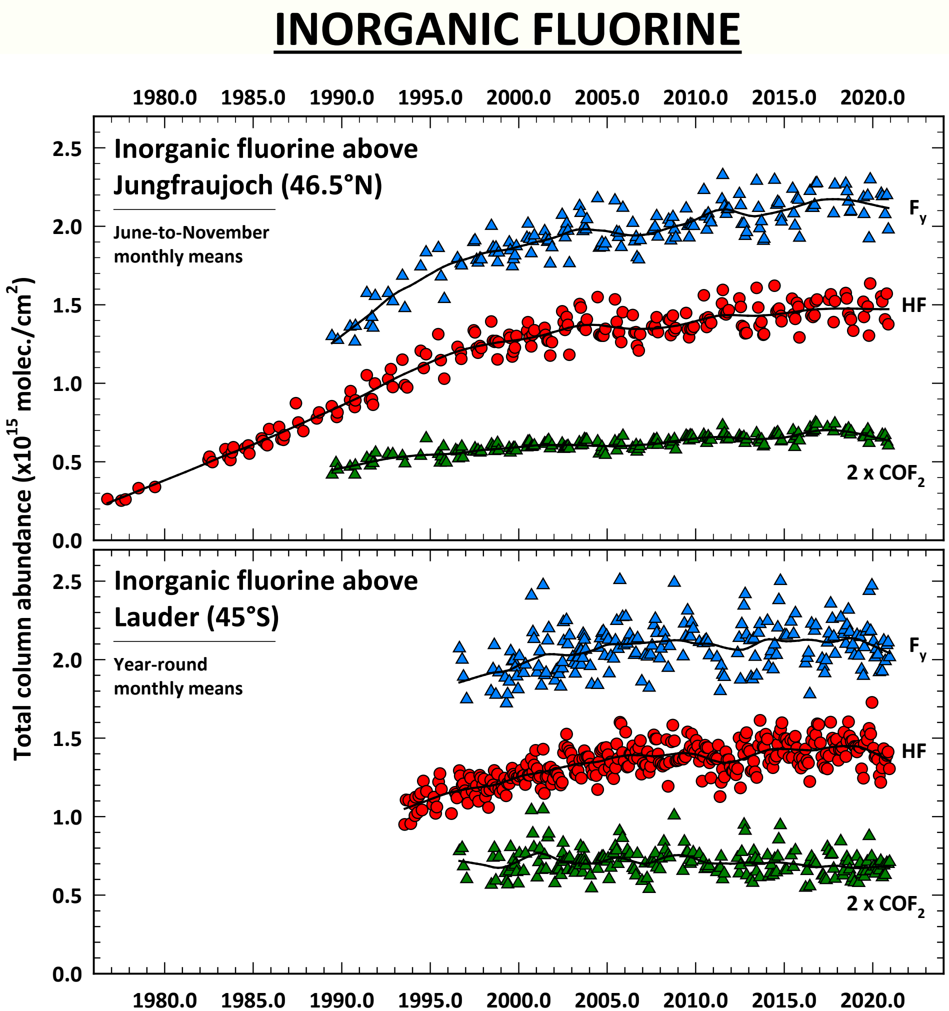
Monitoring of reservoir species





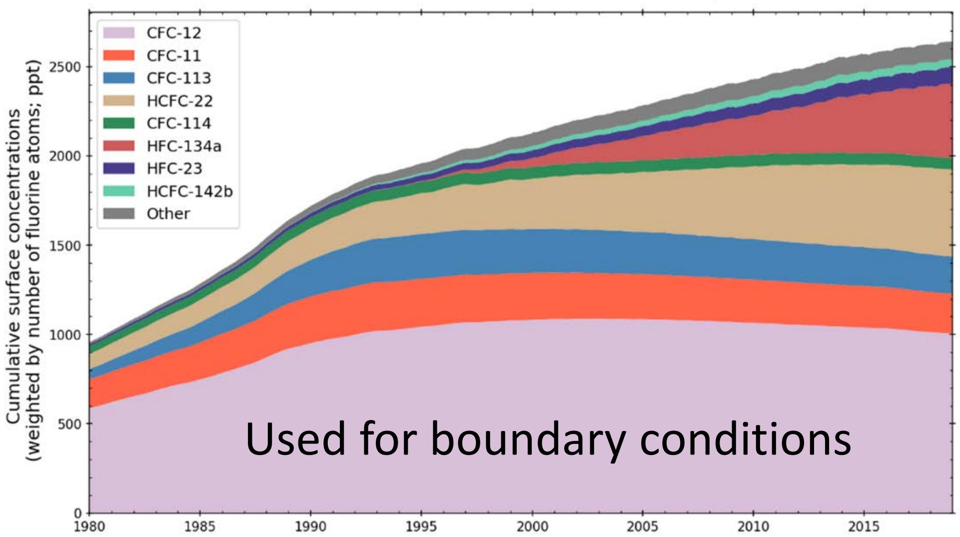
EVOLUTION OF INORGANIC FLUORINE

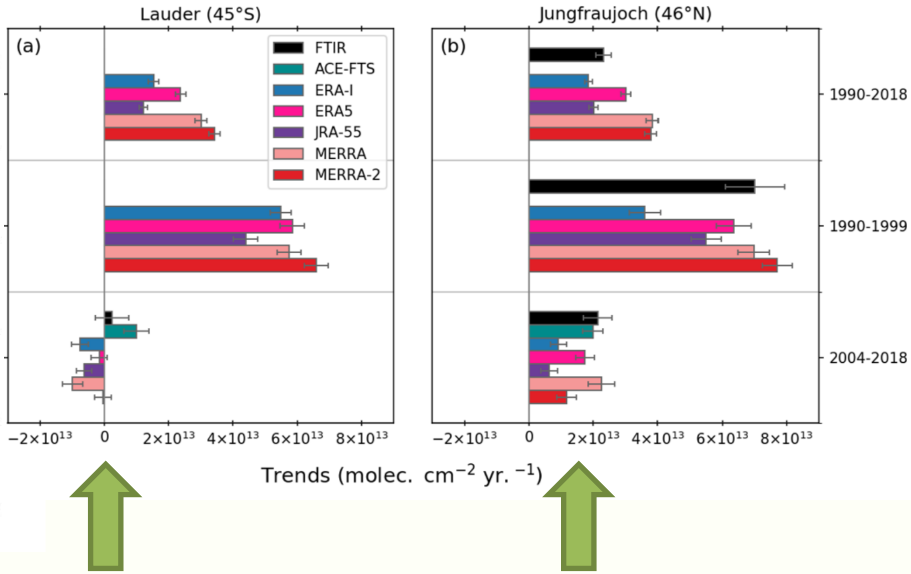
Monitoring of reservoir species

ORGANIC FLUORINE

INORGANIC FLUORINE

Global surface concentrations of main fluorinated species



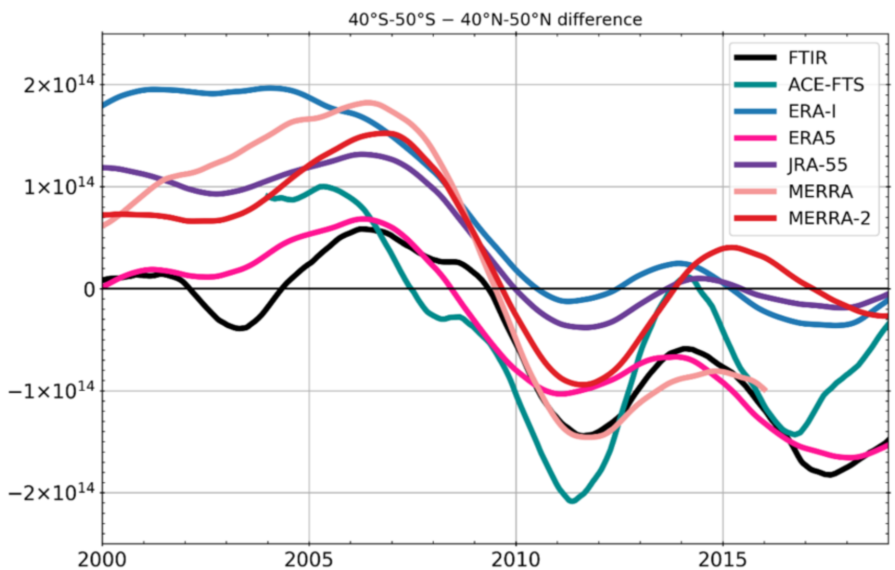


[F-gases] from Meinshausen et al. (2020)
Regular buildup of tropospheric fluorine
HCFC-22 and HFC-134a are now prominent contributors

Dissimilar trends for SH (Lauder) and NH (Jungfraujoch) for 2004-2018; supported by the BASCOE model runs

Prignon et al., 2021

Inorganic fluorine: hemispheric contrast



SH – NH midlatitude differences of F_y

- Asymmetrical change of the BDC* over the past 20 years from observations <u>and</u> CTM simulations with 5 modern reanalyses
- Speeding up of the BDC in the Southern Hemisphere, the SH branch gets stronger relative to the NH one
- Opposes to CCMs in charge of O_3 recovery projections
- 5-7 year variability (fluctuations) superimposed on the overall trend

* Brewer-Dobson circulation

Prignon et al., 2021

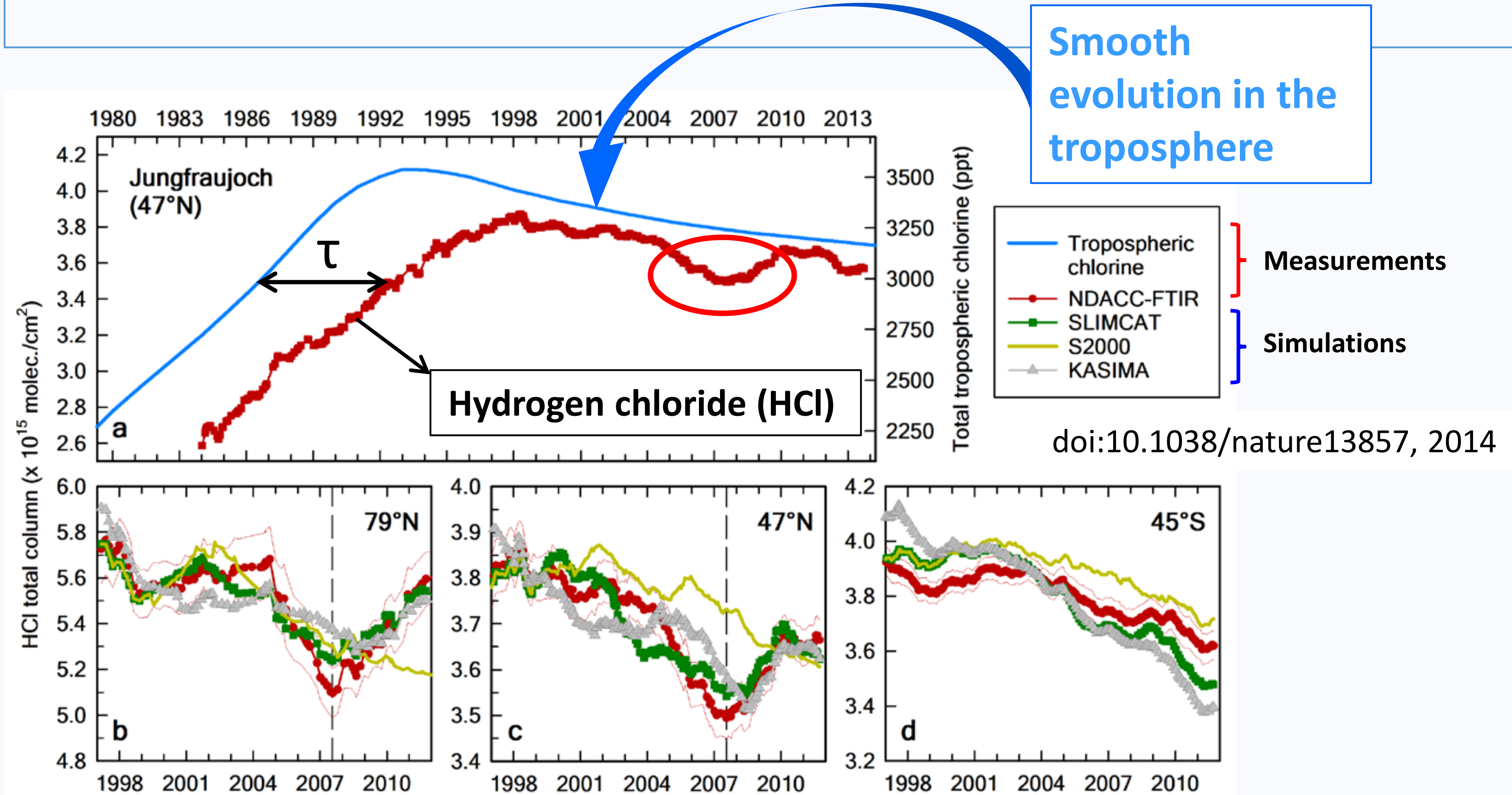
ROBUST TRENDS
FOR HYDROGEN CHLORINE (HCL)?

Primary motivation of this (ongoing!) work

- Hydrogen chloride (HCl), is the end product of the photolysis of the chlorine-bearing source gases (CFCs, HCFCs, CCl_4, CH_3Cl…) which are monitored by the *in situ* surface networks (AGAGE, NOAA-ESRL…), HCl is the main reservoir for chlorine in the stratosphere
- HCl is therefore the most relevant indicator of the stratospheric chlorine loading, and the target of the present investigations

➢ **Derive robust/valid trends enabling the precise characterization of the evolution of stratospheric chlorine, to measure the success of the Montreal Protocol on substances that deplete stratospheric ozone (ODSs)**

Atmospheric Chlorine time series



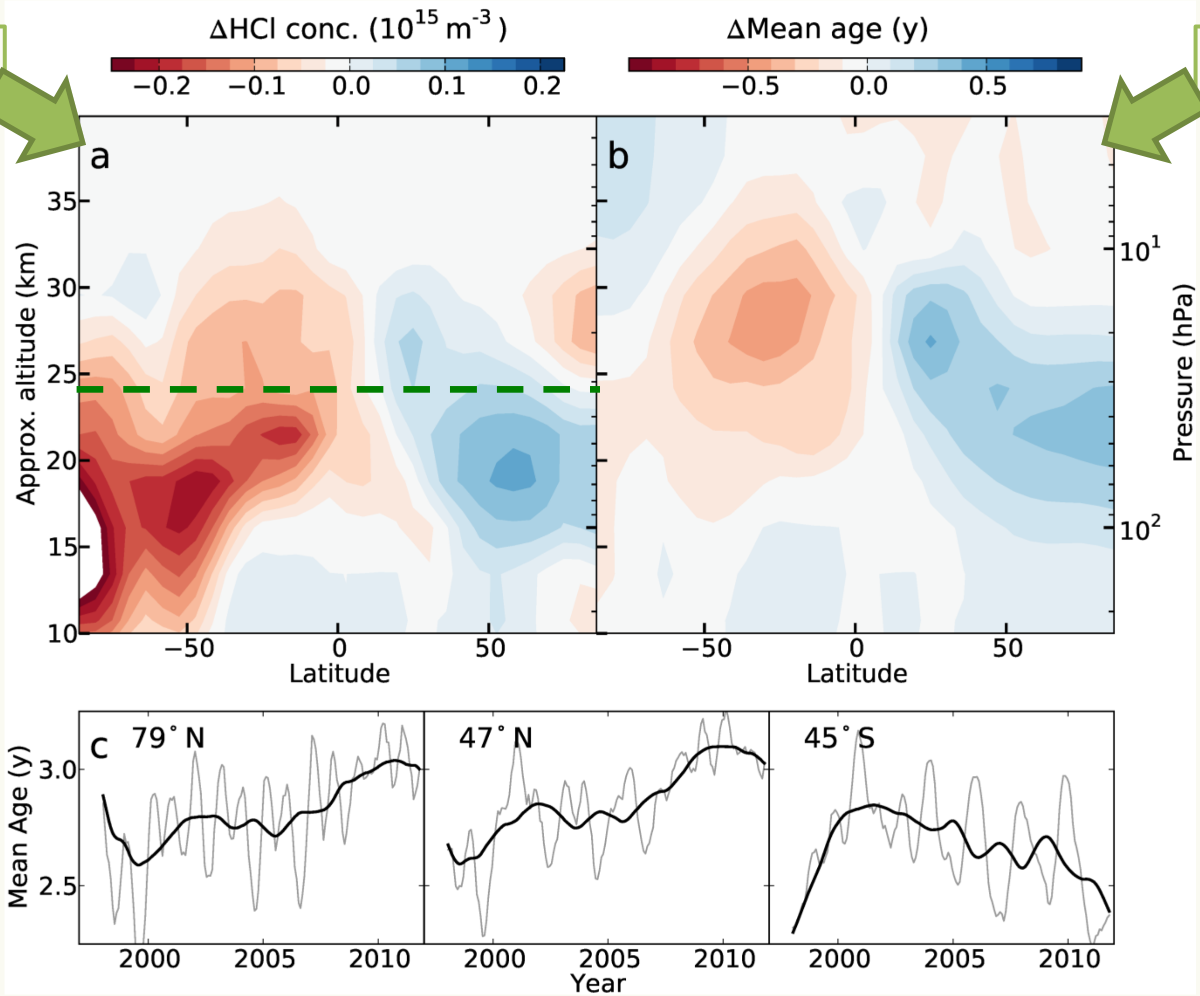
doi:10.1038/nature13857, 2014

The Northern hemisphere anomaly [2007-2011] is explained by a slowing down of the atmospheric circulation in the lower stratosphere, ultimately leading to a change in the balance between the chlorine sources and reservoirs

Spatial changes for HCl and age of air between 2005 and 2010

ΔHCl

ΔAoA



Mahieu et al., 2014

Challenge

- HCl has exhibited significant **multi-year variability** in the last decade, driven by changes in the Brewer Dobson Circulation
- Several studies showed that the perturbations were mainly located **in the lower stratosphere**
- These "anomalies" **complicate the determination of precise long-term trends** for HCl, for direct comparison with the tropospheric evolution of the source gases (AGAGE network…) and for measuring the success of the Montreal Protocol in the stratosphere

How to get around this issue? [1]

Stolarski et al., ACP, 18, 2018;
MLS data

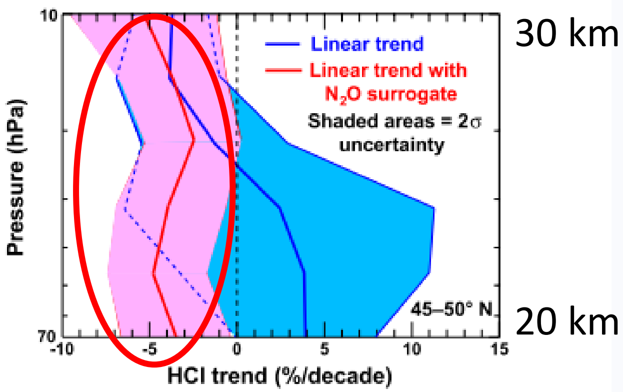


Figure 5. Linear trend in HCl concentrations determined from MLS measurements between 70 and 10 hPa (approximately 20 to 30 km altitude) for the latitude band of 45–50° N. The blue line is the trend determined from the raw deseasonalized data. The red curve is the trend determined while including the N_2O time series as an explanatory variable. The shaded areas represent 2σ uncertainties for each.

Bernath & Fernando, JQSRT, 217, 2018; **ACE-FTS data**

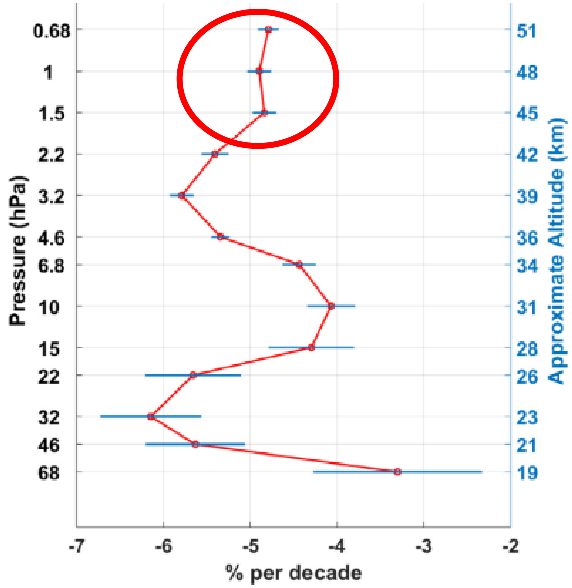


Fig. 2. Linear HCl trends as a function of pressure (approximate altitudes are on the right) for 2004 to 2017 for 60°S–60°N with one standard deviation error bars (see text).

Information from another long-lived stratospheric tracer is used to remove dynamical effects: the trend is then found close to the expected value of -0.5%/yr in the whole range (red vs blue curves), statistical uncertainty is also reduced.

Alternatively, and when sufficient vertical information is available, one can use measurements in the mid- and/or upper stratosphere; here again, the trend is closer to the expected value and the uncertainty significantly lower (error bars).

How to get around this issue? [2]

Froidevaux et al., ACP, 15, 2015; **GOZCARDS data set**

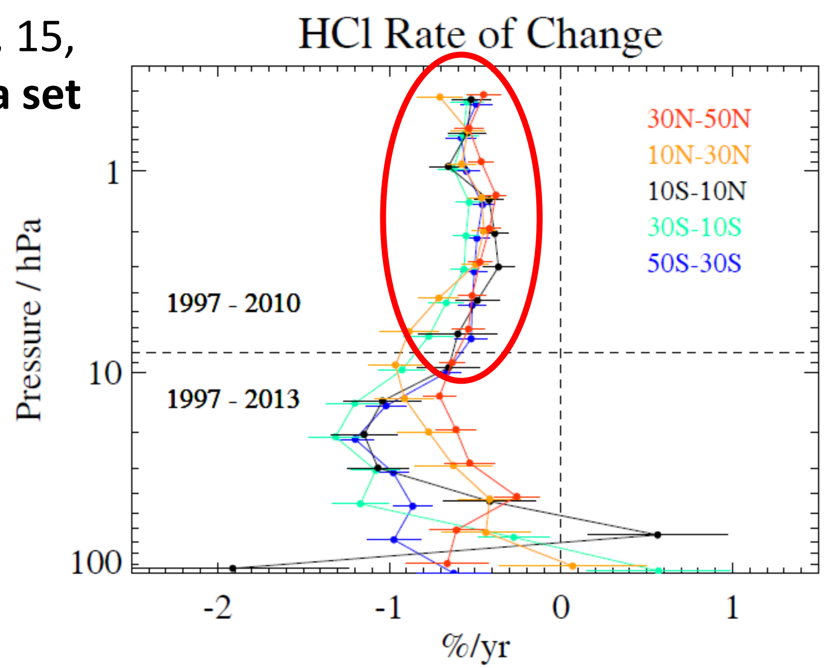
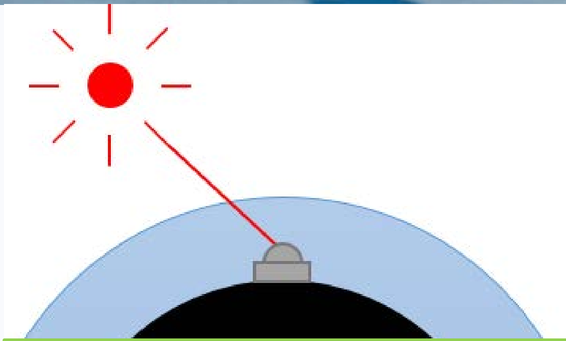


Figure 8. The average rate of change (percent per year) for HCl as a function of pressure for different latitude bin averages (see legend) for time periods corresponding to the appropriate GOZCARDS HCl values (see text) in the upper stratosphere (January 1997–September 2010) and lower stratosphere (January 1997–December 2012). Deseasonalized monthly data were used to obtain a long-term trend for these time periods; 2σ error bars are shown.

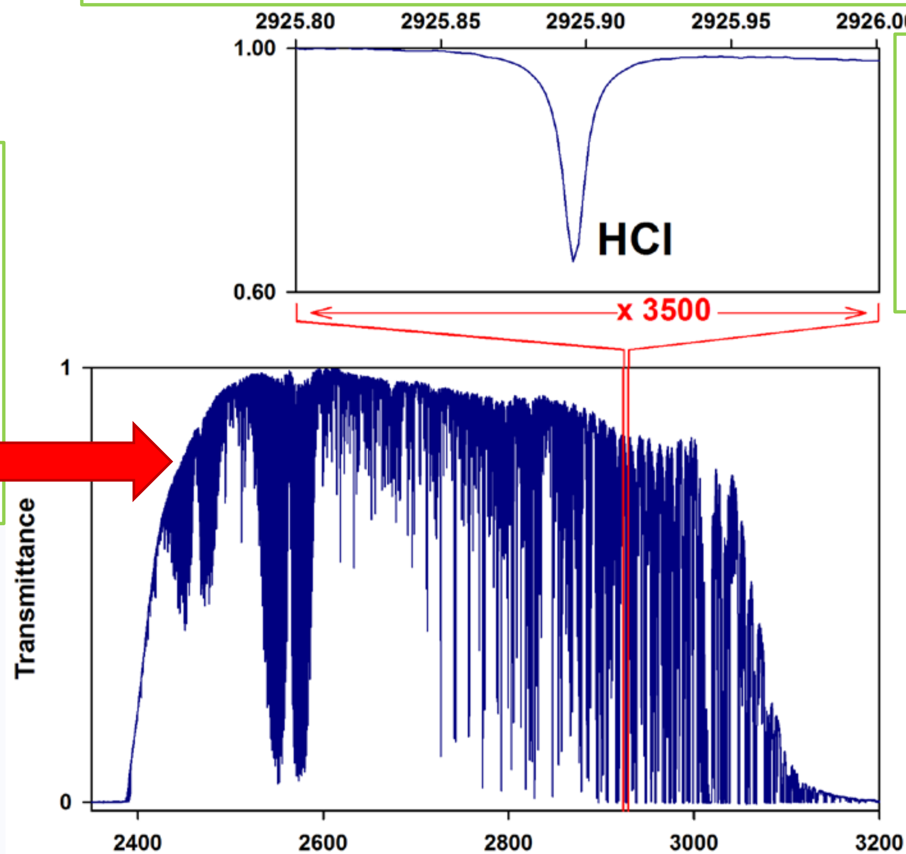
HCl rate of change as a function of altitude and latitude, from the GOZCARDS merged satellite data set: more consistent and robust trends are derived in the upper stratosphere, for all latitude bins

Ground-based remote-sensing of HCl



- Fourier Transform Infrared (FTIR) instruments
- operated year-round under clear-sky conditions
- interferograms ⇔ (FT) *broadband high-resolution IR solar absorption spectra*

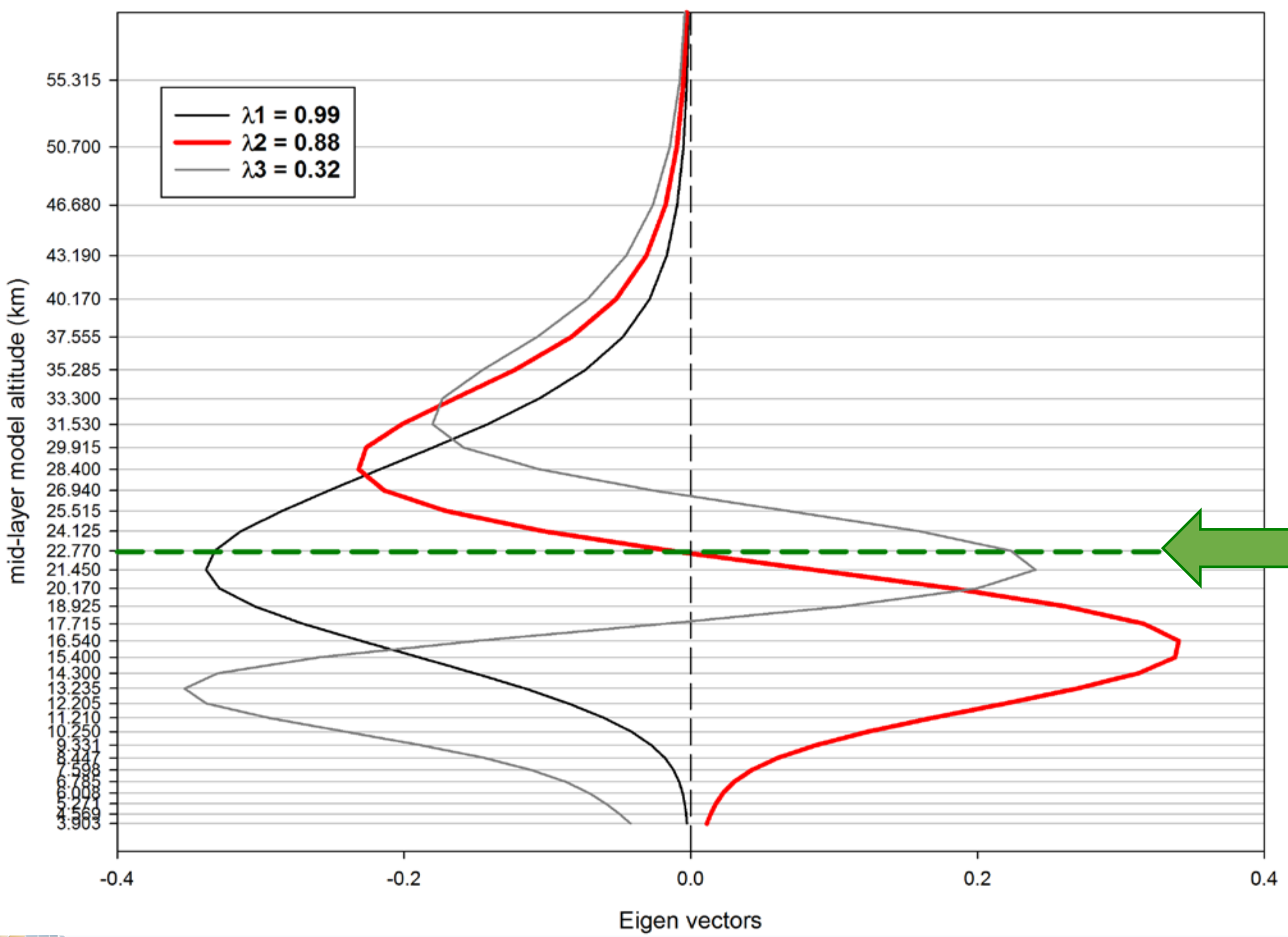
Narrow spectral ranges, or micro-windows are analyzed with the SFIT-4 retrieval software, implementing the OEM method (Rodgers, 2000)



- **Area** of the lines => *total columns*
- **Shape** of the lines => *concentration profiles*

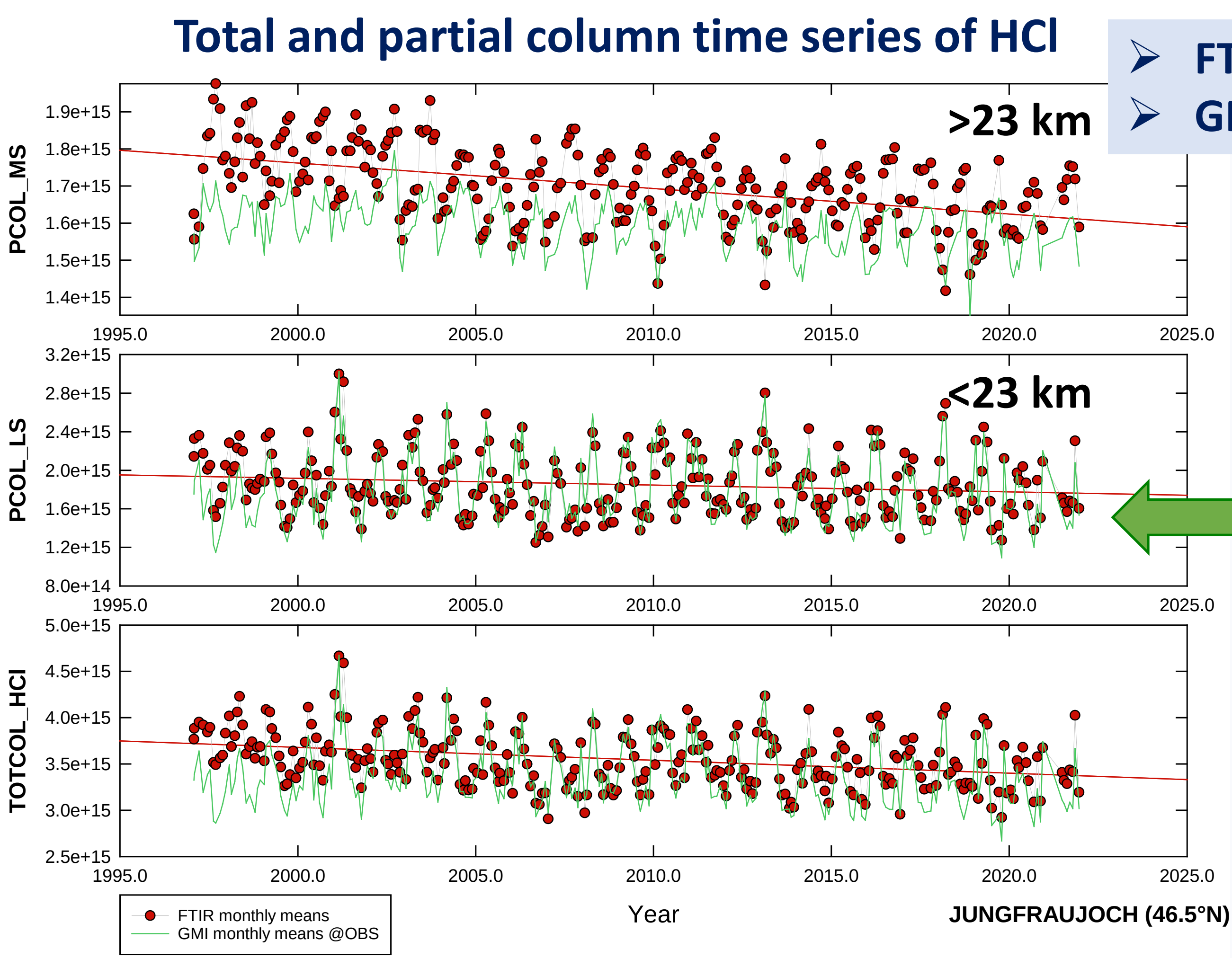
Available information content (optimal estimation method)

Leading Eigen Vectors of the averaging kernel matrix



The red Eigen vector indicates that we can distinguish between HCl partial columns below and above ~23 km (where it crosses the "zero line")

Post-peak HCl time series



- FTIR at Jungfraujoch
- GMI model run (MERRA2)

The 2007 minimum is absent from the "upper" time series; overall, the year-to-year variability is lower in mid/upper stratosphere, and we observe a more monotonous decrease of HCl

The GMI simulations were provided by Susan Strahan (GSFC)

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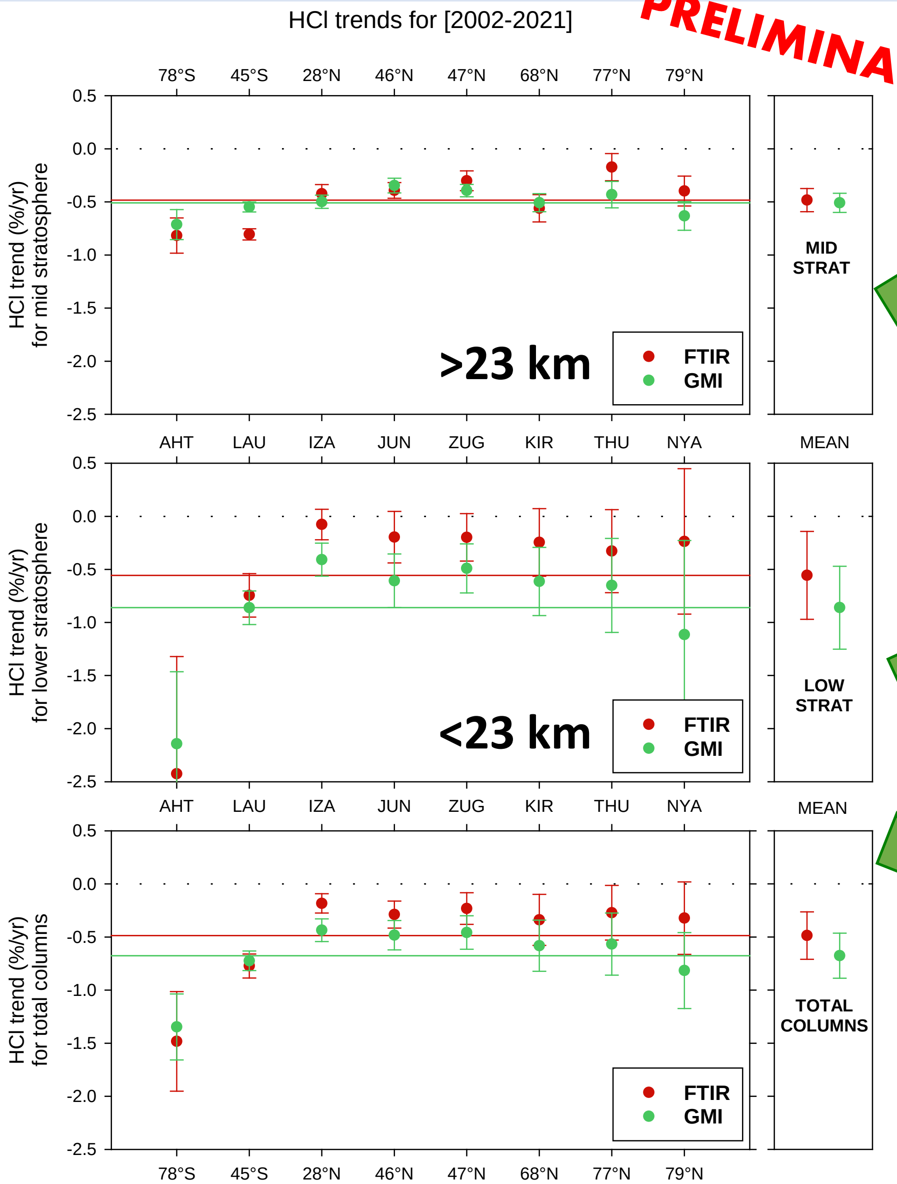
The post-peak trend of HCl at 8 FTIR sites

PRELIMINARY RESULTS!



- We note a very good agreement for the FTIR and GMI trends > 23 km at most sites
- The rates of change are consistent with ACE-FTS, MLS and *in situ* surface findings (-0.5%/yr)
- The trends for the lower stratosphere/TC are noisier

++ Trends and confidence ranges: we use a bootstrap method (autoregressive wild bootstrap) applicable to “autocorrelated” data sets
++ A linear function combined with a 3-term Fourier series is used to fit the data

Current conclusions

- We used FTIR and model data for HCl below and above 23 km as well as a bootstrap tool/method accounting for auto-correlation in the data sets
- We derive more robust/valid and defined trends in the mid/upper stratosphere, a region which seems more appropriate for this purpose, also for our FTIR data, and even at polar stations (less variability and signal still strong enough)
- Very consistent trends of -5%/decade are derived from the *in situ* networks, from ACE-FTS, MLS and Jungfraujoch FTIR, demonstrating the effectiveness of the Montreal Protocol

➢ Perfect alignment of the FTIR instrument is critical in order to derive sensible partial column time series

Thank you for your attention!
Questions are welcome

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