Post-peak trend in inorganic chlorine (Cl_y) from Jungfraujoch, ACE-FTS and HALOE infrared solar observations

Emmanuel.Mahieu (@ulg.ac.be)
with the GIRPAS-team, University of Liège, BE
and the ACE-team, University of Waterloo, CA

emmanuel.mahieu@ulg.ac.be

The Jungfraujoch station & its multi-decadal Infrared dataset

- **FTIR** measurements are performed year-round at the Jungfraujoch station [Swiss Alps, **46.5ºN**, 8.0ºE, **3580m a.s.l.**, a site of the NDACC(*) network, see www.ndacc.org]
- Two high-resolution Fourier Transform Infrared (FTIR) spectrometers (one homemade and one Bruker 120HR) are operated under clear-sky conditions at the Jungfraujoch
- FTIR regular observations are available since 1984, with high density of measurements (~120 days/yr on average) since the mid-1990s, when the second spectrometer entered into regular operation
- Remote operation of the Bruker instrument is operational since late 2008 [design and implementation by *Ch. Servais*, ULg]
- In addition, grating spectra recorded essentially from 1976 to 1989 are also available, they cover narrow IR intervals (HCl, HF, CH_4, N_2O…). We are in the process of valorizing them (analysis with modern retrieval tools)
- Altogether : more than 35 years of uninterrupted IR monitoring in the Alps
- Total and partial column abundances of more than two dozen stratospheric and/or tropospheric species are systematically retrieved from our spectra.

(*) NDACC: Network for the Detection of Atmospheric Composition Change

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 specific applications, e.g. for water vapor retrievals on log scales
- Both codes implement 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-2008 or HITRAN-2004 including the 2006 updates are assumed in most 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 laboratory spectra are used for numerous target or interfering species (e.g. CFCs, HCFCs, CCl_4, $ClONO_2$, C_2H_6)

emmanuel.mahieu@ulg.ac.be

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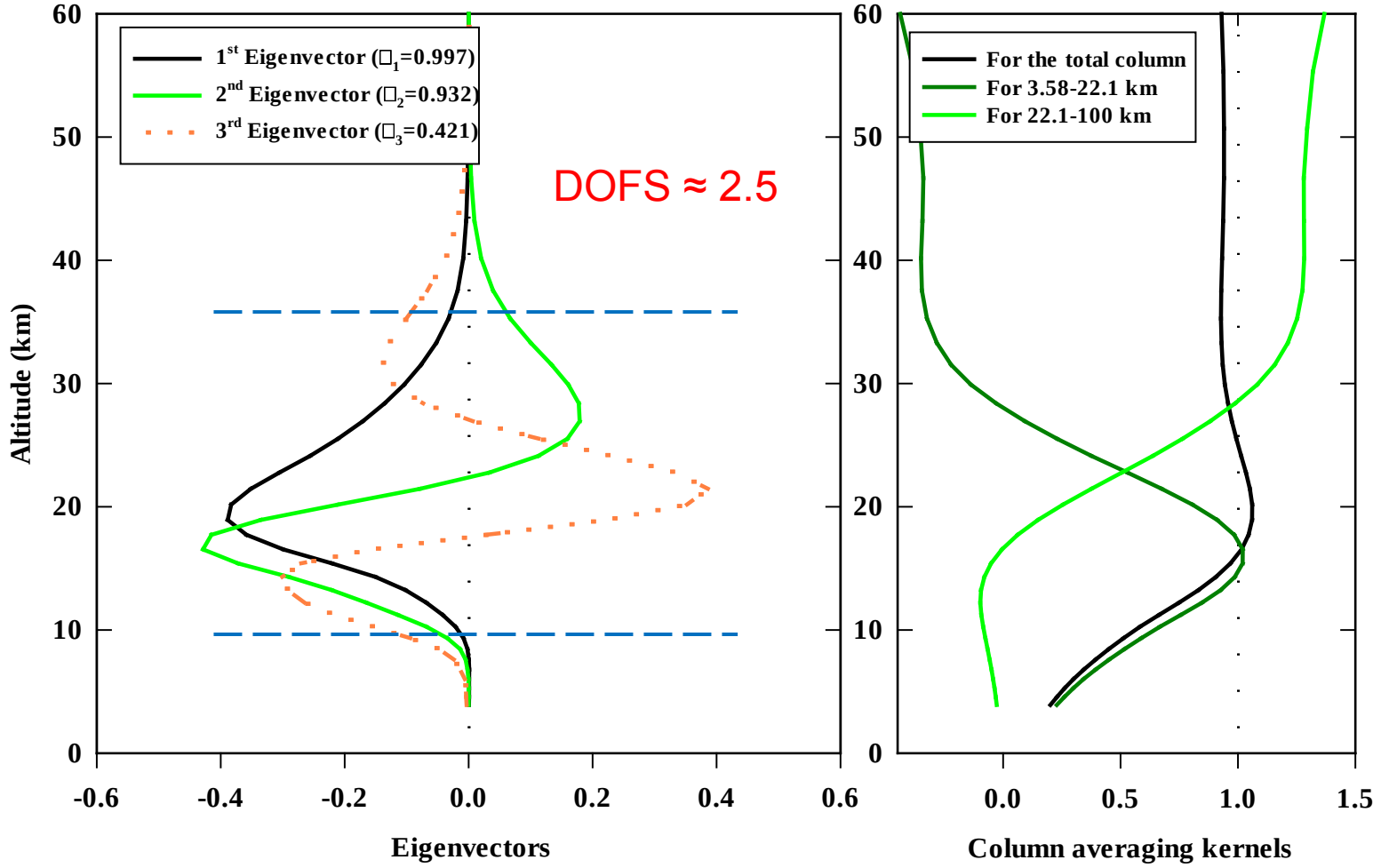
Typical information content for HCl



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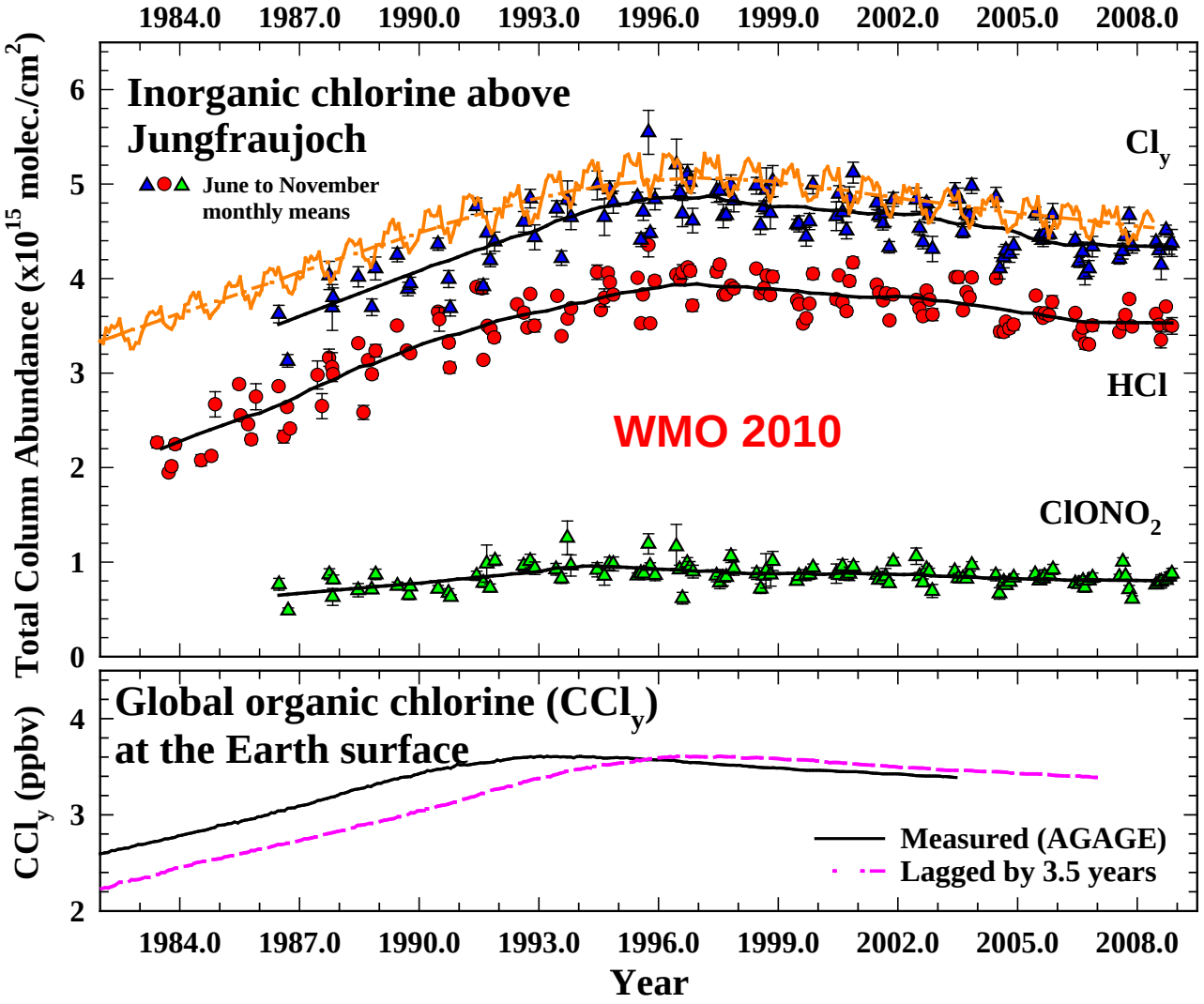
Long-term trend for chlorine (46.5°N)



HCl and $ClONO_2$, the two most abundant chlorine reservoirs in the stratopshere are accessible to the gb-FTIR technique, the long-term Jungfraujoch Cl_y time series shows:

-increase at ~4%/yr

-maximum load in 1996

-decrease at ~ -1%/yr

Good agreement with models and CCl_y build up in the troposphere

emmanuel.mahieu@ulg.ac.be

Kohlhepp et al., ACP, 12, 2012

- NDACC-wide effort, involving **17 FTIR sites** from 80°N to 78°S, allowing a “near-global” coverage
- **2000-2009** time series of HCl, $ClONO_2$ (and HF) were compared to simulations performed by **5 models** (2 CTMs, 2 CCMs and a 2-D model)
- The decrease of inorganic chlorine is confirmed, at rates close to **-1%/yr**, in overall agreement with the model calculations
- There is however a larger spread in the measured trends than in the modeled ones

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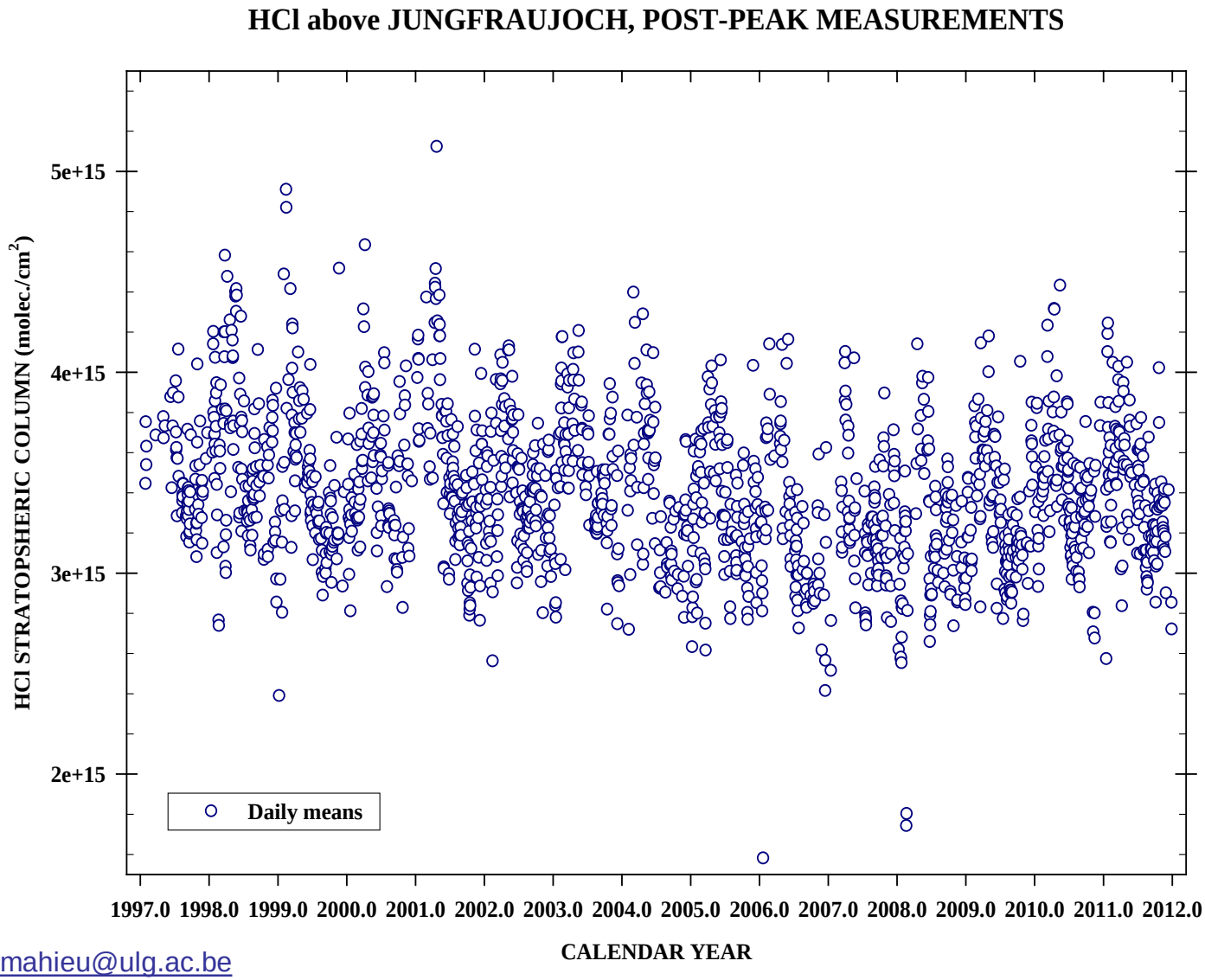
When extending to 2011…



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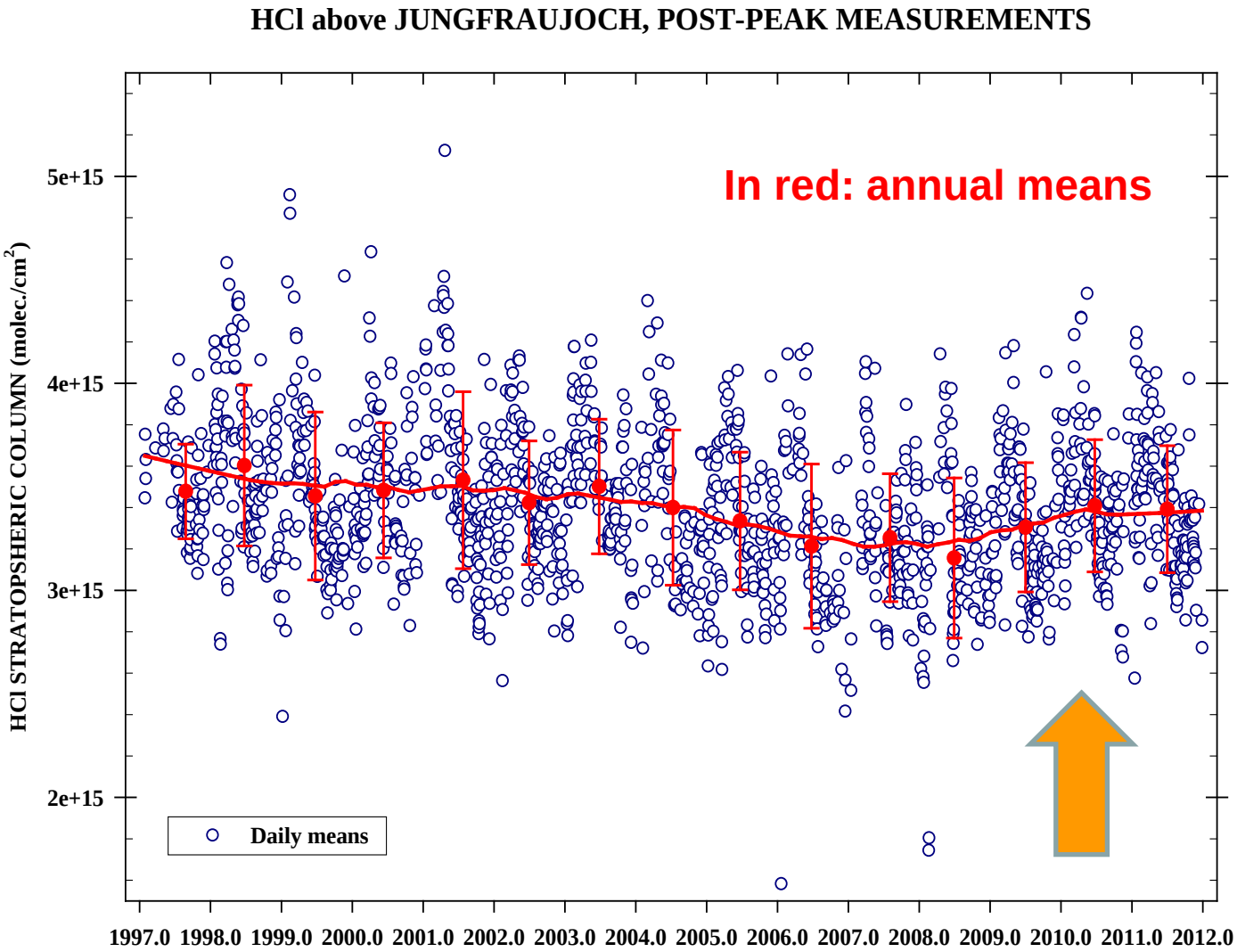
1997-2011 data set for HCl



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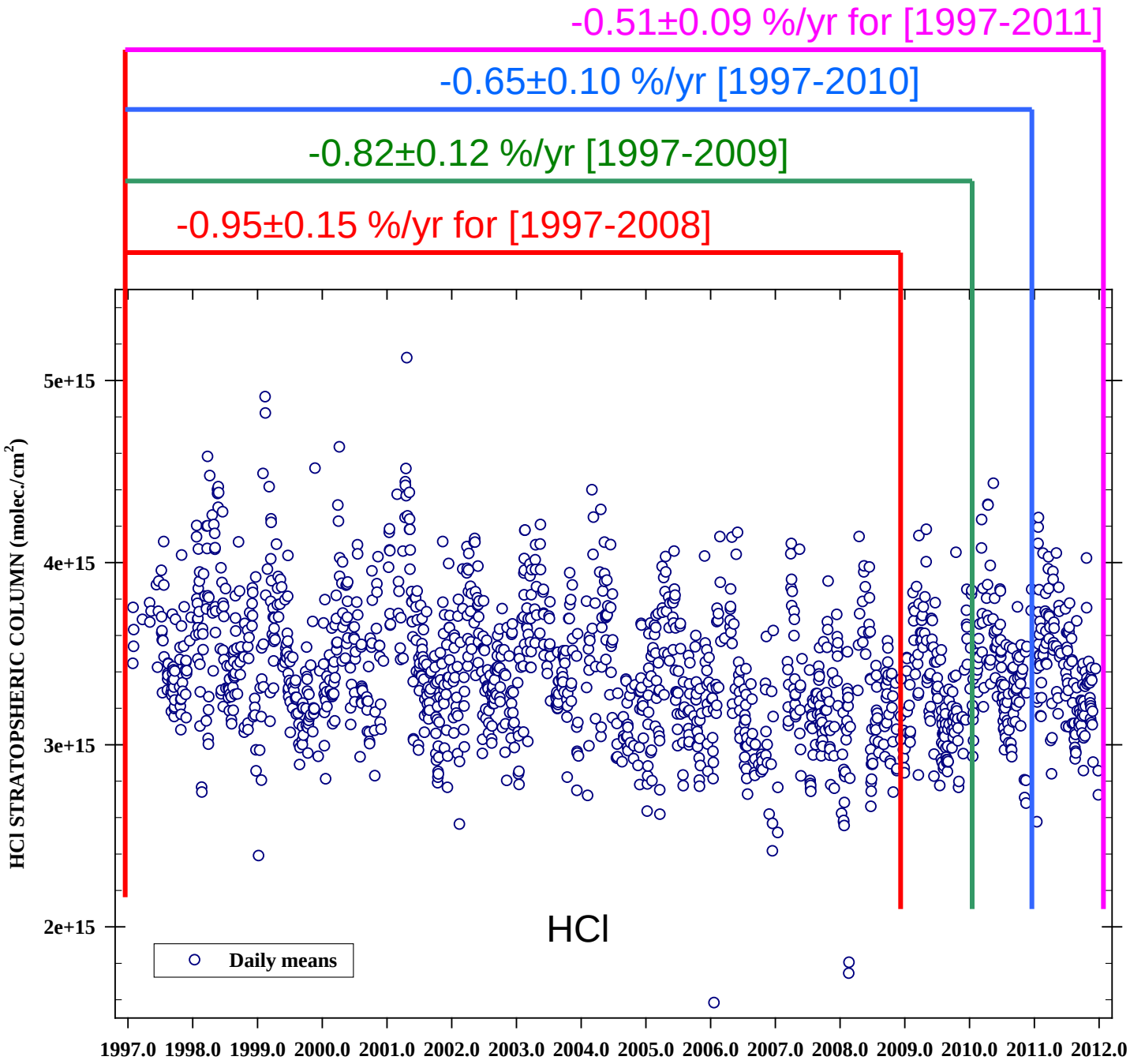

- Trend analyses performed with the bootstrap resampling tool developed by Gardiner et al. (ACP, 8, 2008)
- 2-sigma confidence levels are provided
- Although still significantly negative, the **HCl** trend is nearly divided by 2 when adding the last three years…
- Consistent findings when considering the HCl + $ClONO_2$ data set

emmanuel.mahieu@ulg.ac.be

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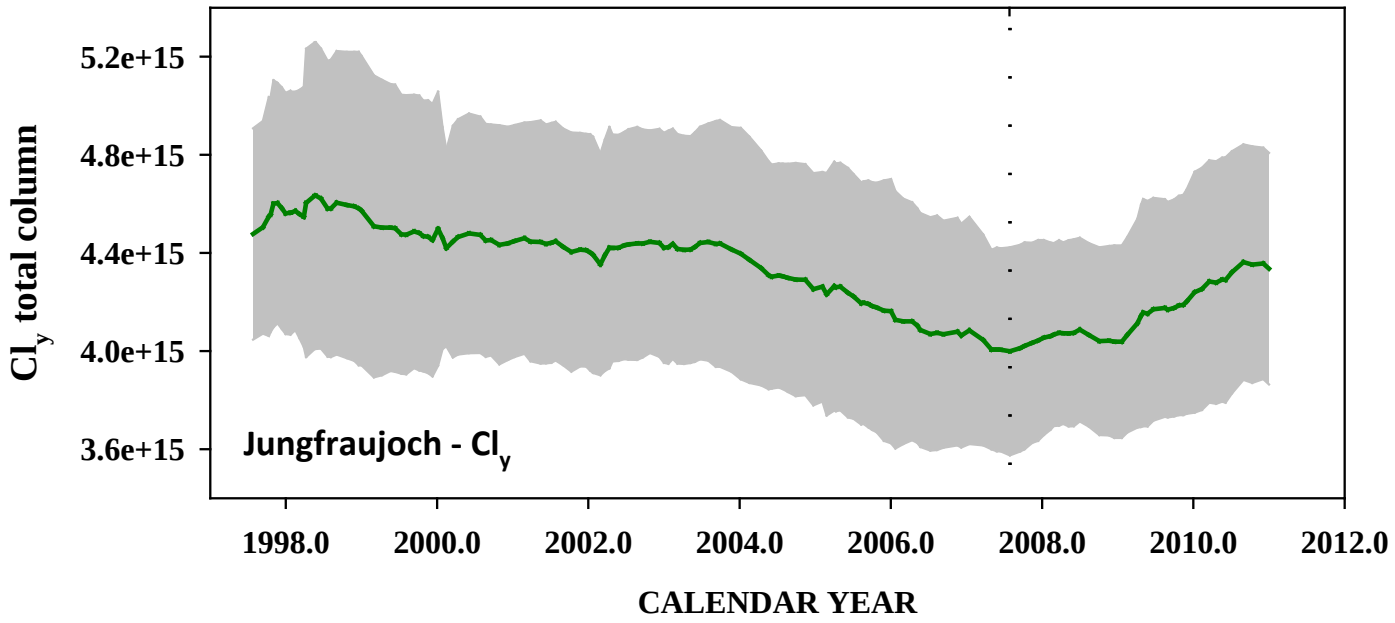
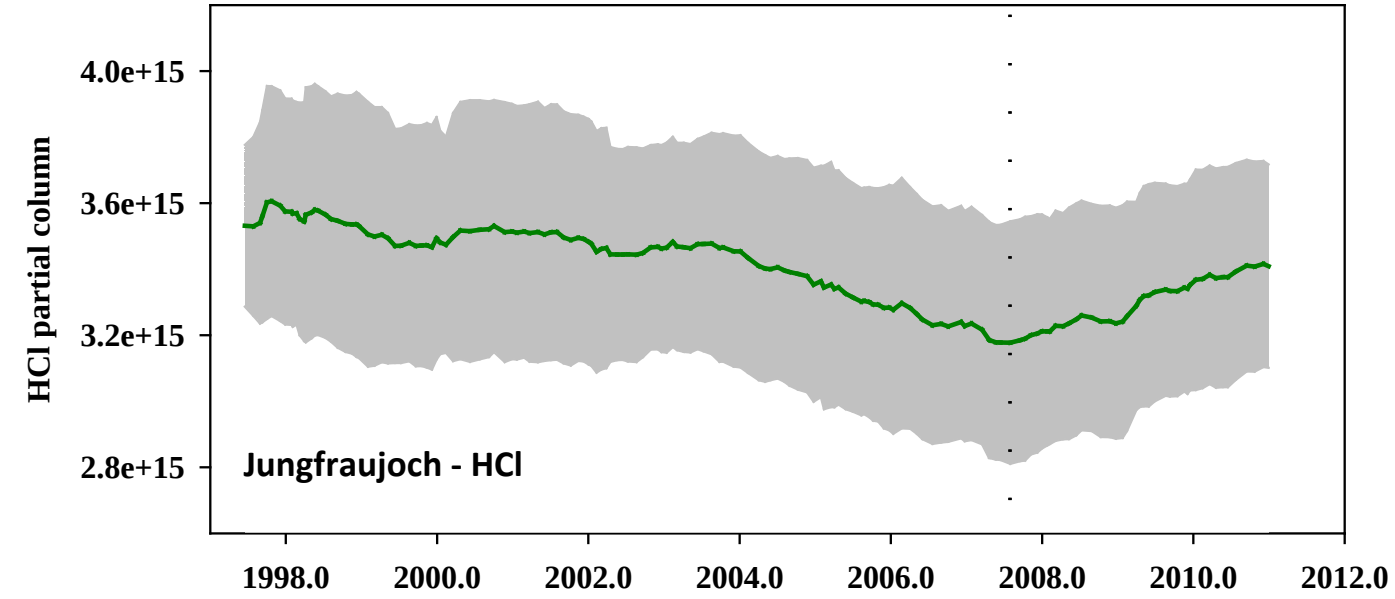


- **Running mean analyses** on the Cl_y and HCl time series indicate minimum columns around July 2007
- Since then, both sets show steady increase of the local means



- Local averages are computed for each month over 2-year intervals
- The grey area correspond to the standard deviations around the means

emmanuel.mahieu@ulg.ac.be

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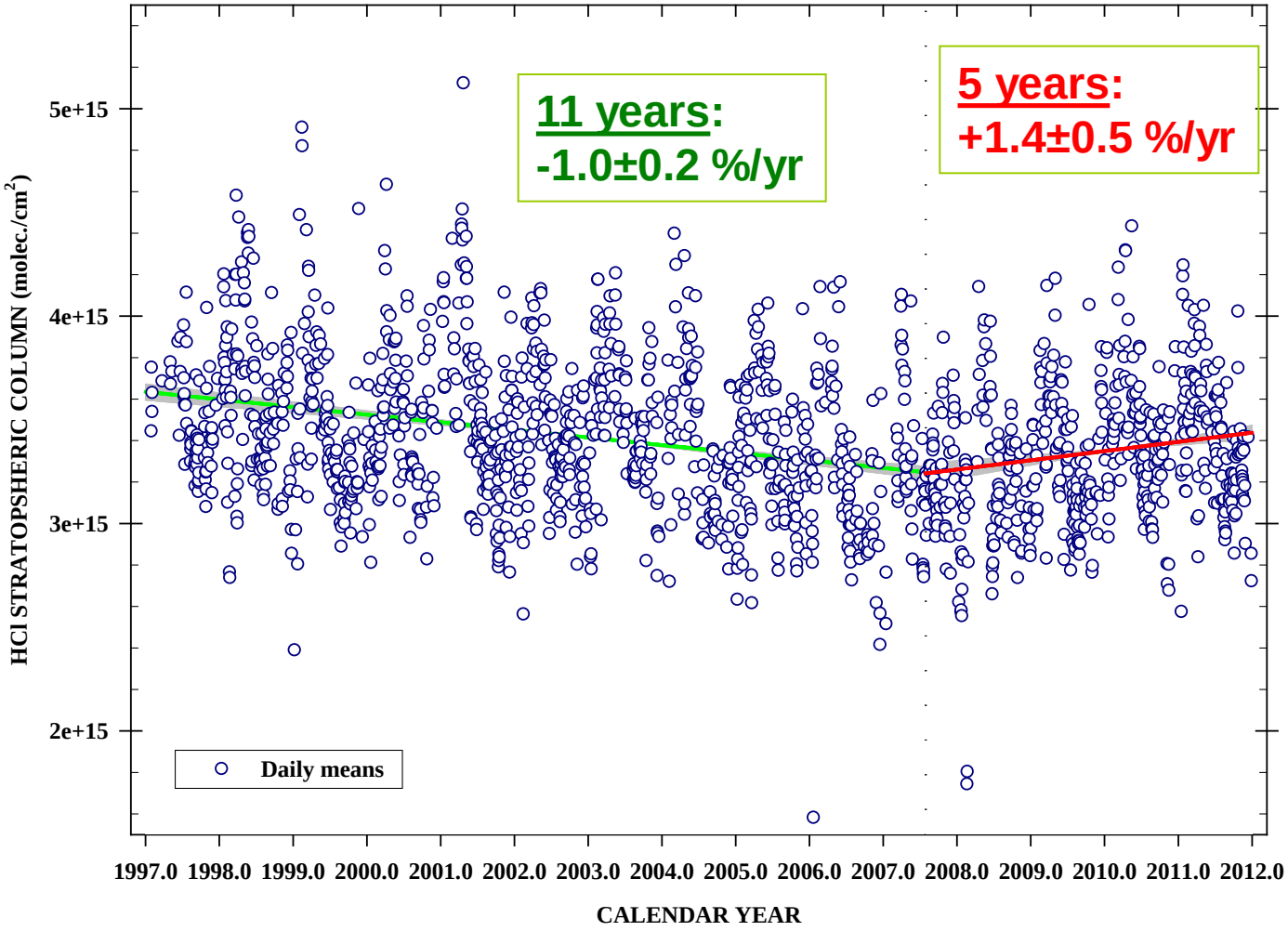
When considering the two regimes separately…



- Decrease over 1997-2007, with a rate of -3.7x10^{13} molec./cm^2/yr
- Followed by a significant increase of 4.4x10^{13} molec./cm^2/yr
- The upward trend is almost three times larger in magnitude than its 2-sigma uncertainty
- Mean levels at the end of 2011 = those of early 2002
- But short time period of only 5 years

emmanuel.mahieu@ulg.ac.be

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What about satellite observations?

- **HALOE** (1991-2005), version 19 (netcdf files)
- **ACE-FTS**, version 3 (from 2004 onwards)
- *Also available: AURA/MLS data (Froidevaux et al., GRL, 33, 2006), not considered thus far*
- But these time series cannot be combined directly to form a consistent set due to a known bias between them, as noticed in ACE validation studies (McHugh et al., GRL, 32, 2005; Mahieu et al., ACP, 8, 2008) as well as in Lary et al., GRL, 34, 2007)
- This bias is obvious when displaying both time series, HALOE being low

emmanuel.mahieu@ulg.ac.be

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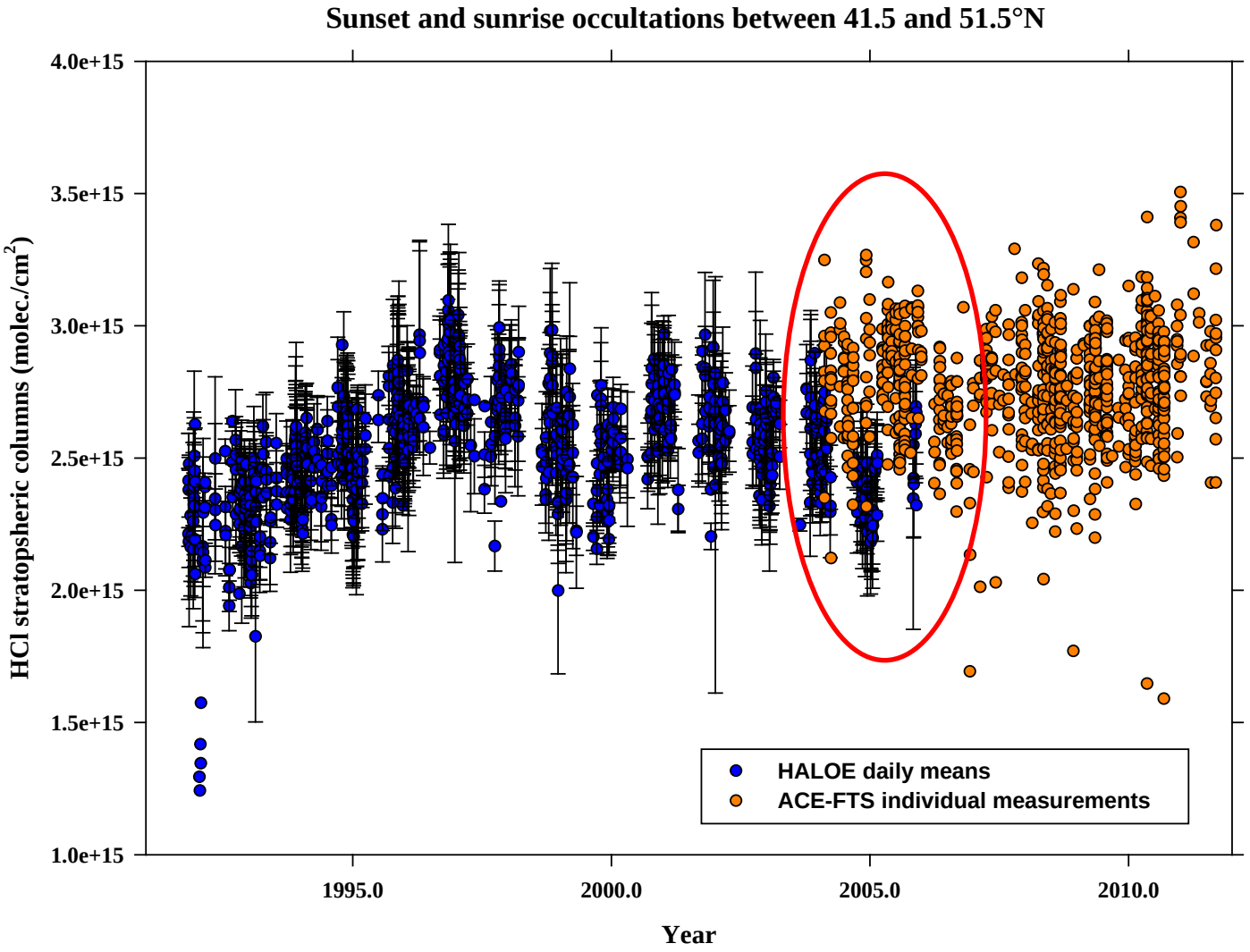
HALOE V19 and ACE-FTS V3



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Accounting for the bias

- The relative bias has been re-evaluated, considering all available coincidences (within 2h and 500 km) as well as the mean difference between both sets in the 41.5-51.5°N zone
- Both approaches led to the determination of a relative bias of 14 %, with ACE higher than HALOE, in agreement with previous studies (bias of 10-20% quoted by McHugh et al., 2005; 10-15% by Mahieu et al., 2008).
- HALOE was therefore scaled high by 14% to match ACE-FTS

emmanuel.mahieu@ulg.ac.be

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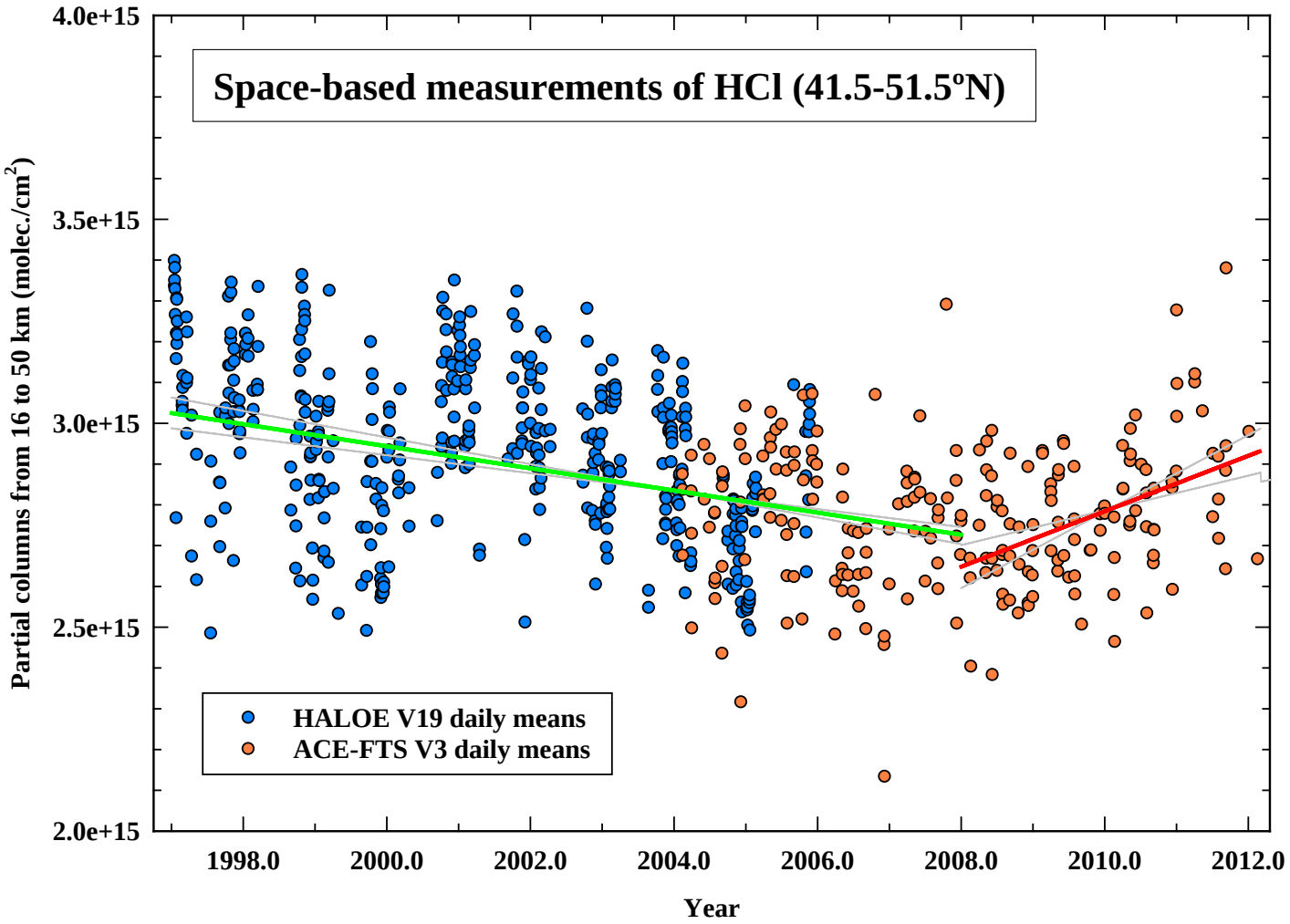
Harmonized satellite time series



All trends in 10^{13} molec./cm^2 or in % per year

[1997-2007] trend:
-3.4±2.5 (-1.1%/yr)

From the ground:
-3.7±0.6 (-1.0%/yr)

[2008-2011] trend:
+6.8±2.6 (+2.6%/yr)

From the ground:
+4.4±1.8 (+1.4%/yr)

Uncertainty:
2-sigma level

emmanuel.mahieu@ulg.ac.be

Current conclusions

- Good agreement between trends derived from the Jungfraujoch and composite satellite time series, suggesting at least a recent and significant slowing down of HCl (and of Cl_y) at Northern mid-latitudes, *or even a positive trend over the last 5 years*
- Persistence of this upward trend needs to be verified by extending the time series with the newer Jungfraujoch and ACE-FTS data
- This new feature in the HCl and Cl_y time series is unexpected: chlorine has steadily decreased in the troposphere over the 1993-2008 time frame, as shown by the AGAGE & NOAA ins situ measurements, at rates between about -0.4 and -1%/yr

emmanuel.mahieu@ulg.ac.be

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Questions

- Do we observe the consequences of a –temporary– change in global circulation, affecting the representativity of the site? (ACE-FTS reveals a meridional gradient in the HCl columns of ~1%/deg. lat.)
- Is this verified in other latitude bins and in the Southern hemisphere?
- Possible impact on (the onset of) ozone recovery?
- What about other tracers (e.g. HF, HNO_3, COF_2)?

emmanuel.mahieu@ulg.ac.be

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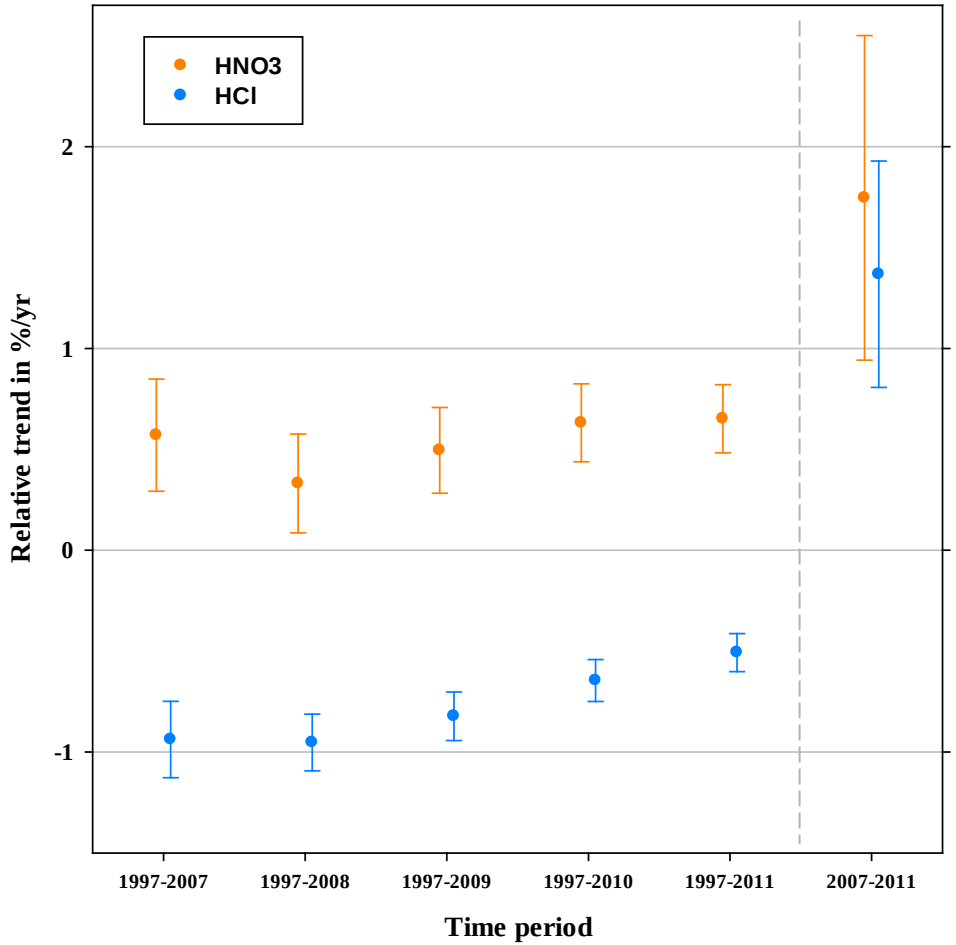
Trends comparison: HNO_3 vs HCl



- 2-sigma uncertainty levels
- Steady build up of the main HNO_3 source (N_2O) at about 0.25/%yr

Preliminary results for HNO_3!!

emmanuel.mahieu@ulg.ac.be

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Thank you!

Jungfraujoch FTIR data are available (in hdf)

from the NDACC-DHF:

ftp://ftp.cpc.ncep.noaa.gov/ndacc/station/jungfrau/hdf/ftir/

or upon request by email:

emmanuel.mahieu@ulg.ac.be

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