Post-peak **trend of upper stratospheric HCl** derived from ground-based FTIR solar spectra and model simulations

E. Mahieu, M. Prignon, C. Servais, S. Chabrillat, Q. Errera, M. Friedrich, S. Smeekes, L. Froidevaux, R.J. Salavitch, P. Wales, J. Notholt, and M.P. Chipperfield

emmanuel.mahieu@uliege.be

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Primary motivation of this 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)**

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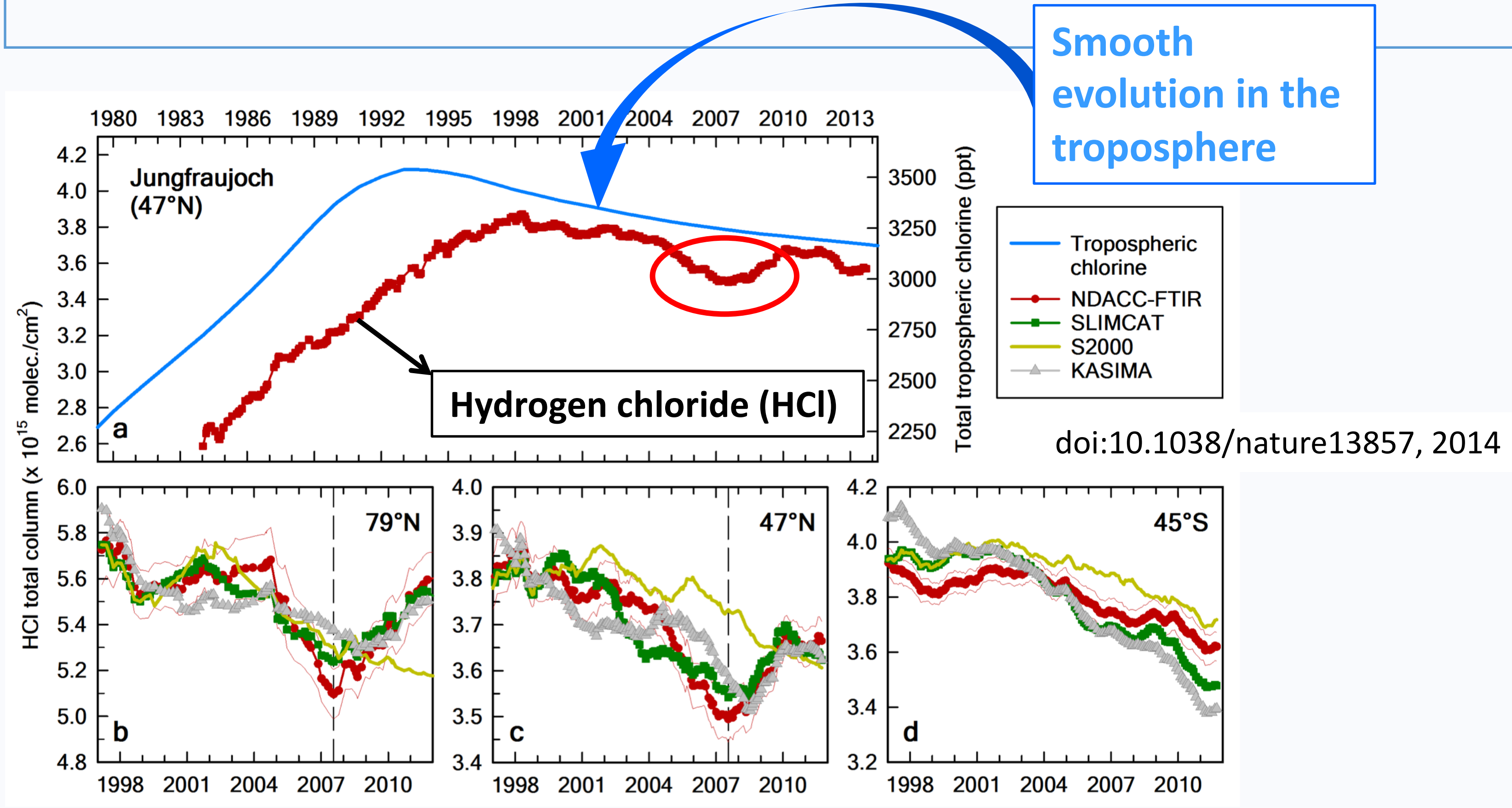
Atmospheric Chlorine time series



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

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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

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How to get around this issue? [1]

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

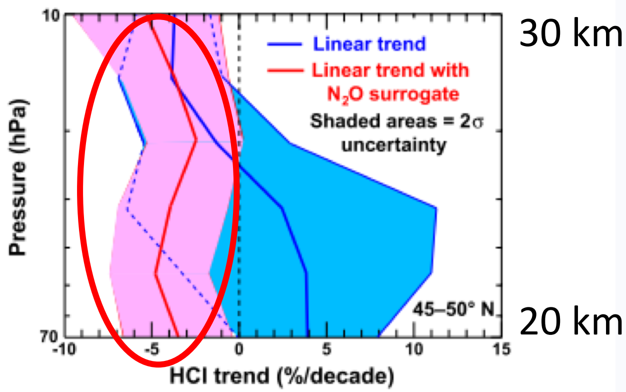


Figure 5. Linear trend in HCl concentrations determined from MLS measurements between 70 and 10 hPa (approximately 20 to 30 km altitude) for the latitude band of 45–50° N. The blue line is the trend determined from the raw deseasonalized data. The red curve is the trend determined while including the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

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

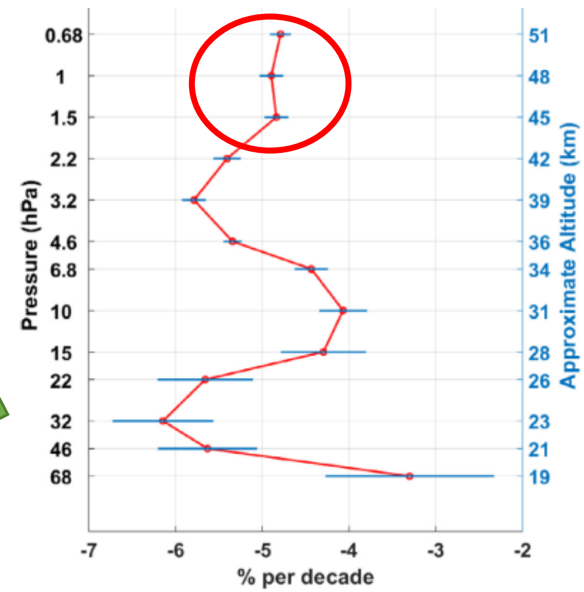


Fig. 2. Linear HCl trends as a function of pressure (approximate altitudes are on the right) for 2004 to 2017 for 60°S–60°N with one standard deviation error bars (see text).

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**

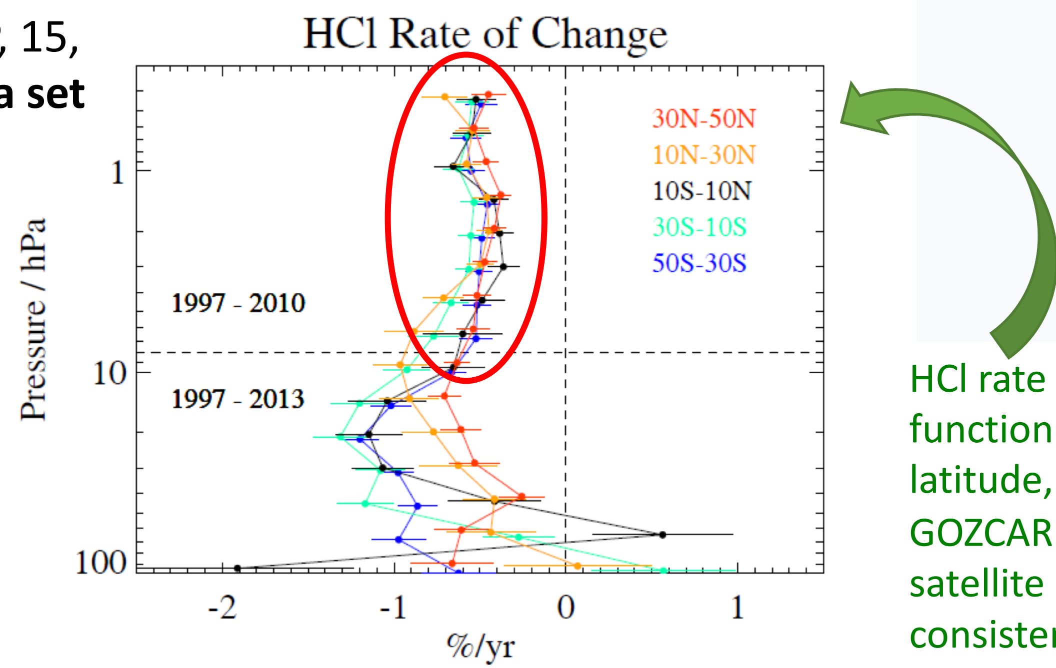
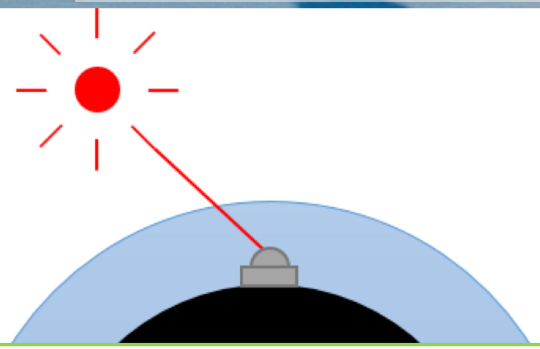


Figure 8. The average rate of change (percent per year) for HCl as a function of pressure for different latitude bin averages (see legend) for time periods corresponding to the appropriate GOZCARDS HCl values (see text) in the upper stratosphere (January 1997–September 2010) and lower stratosphere (January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

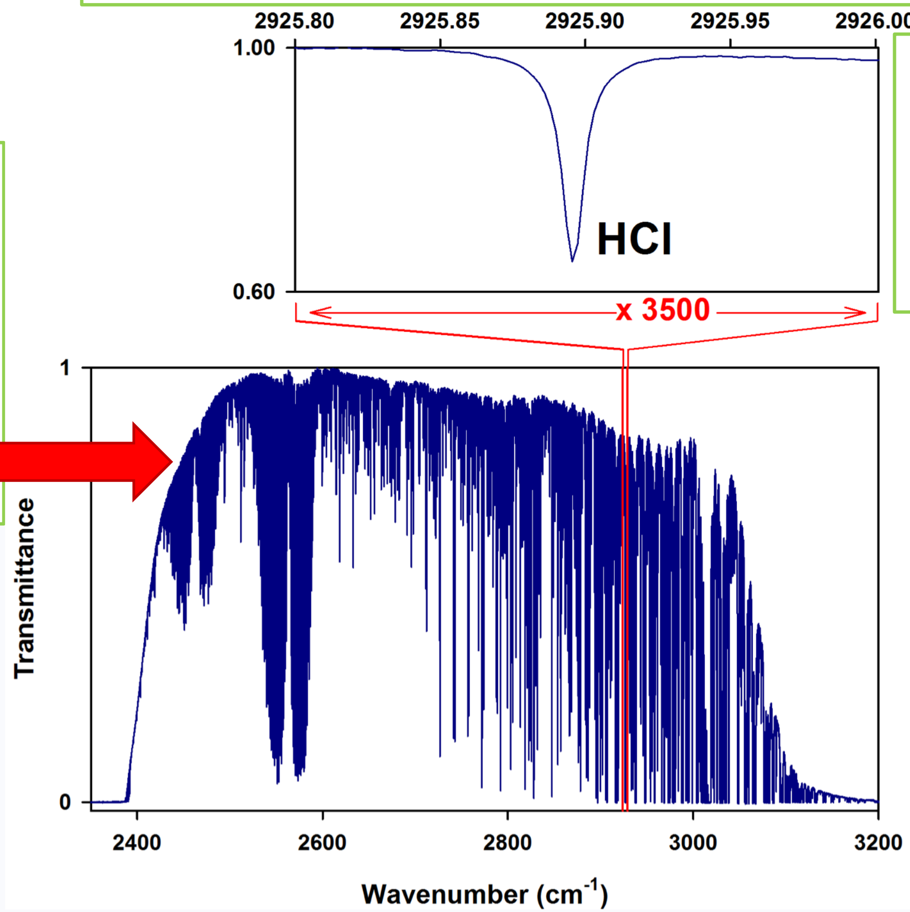
HCl rate of change as a function of altitude and latitude, from the GOZCARDS merged satellite data set: more consistent and robust trends are derived in the upper stratosphere, for all latitude bins

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***

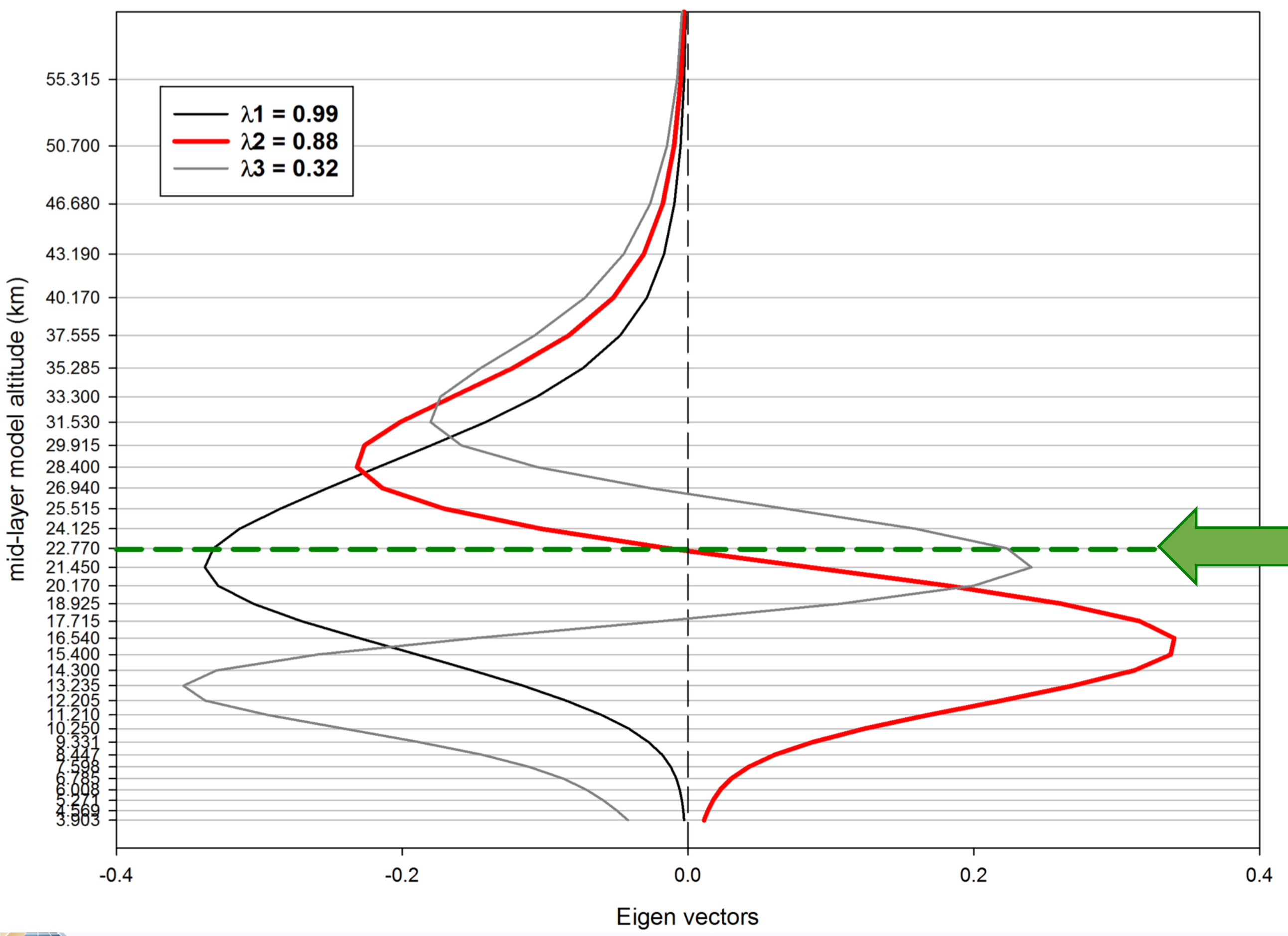
Narrow spectral ranges, or micro-windows are analyzed with the SFIT-4 retrieval software, implementing the OEM method (Rodgers, 2000)



- **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")

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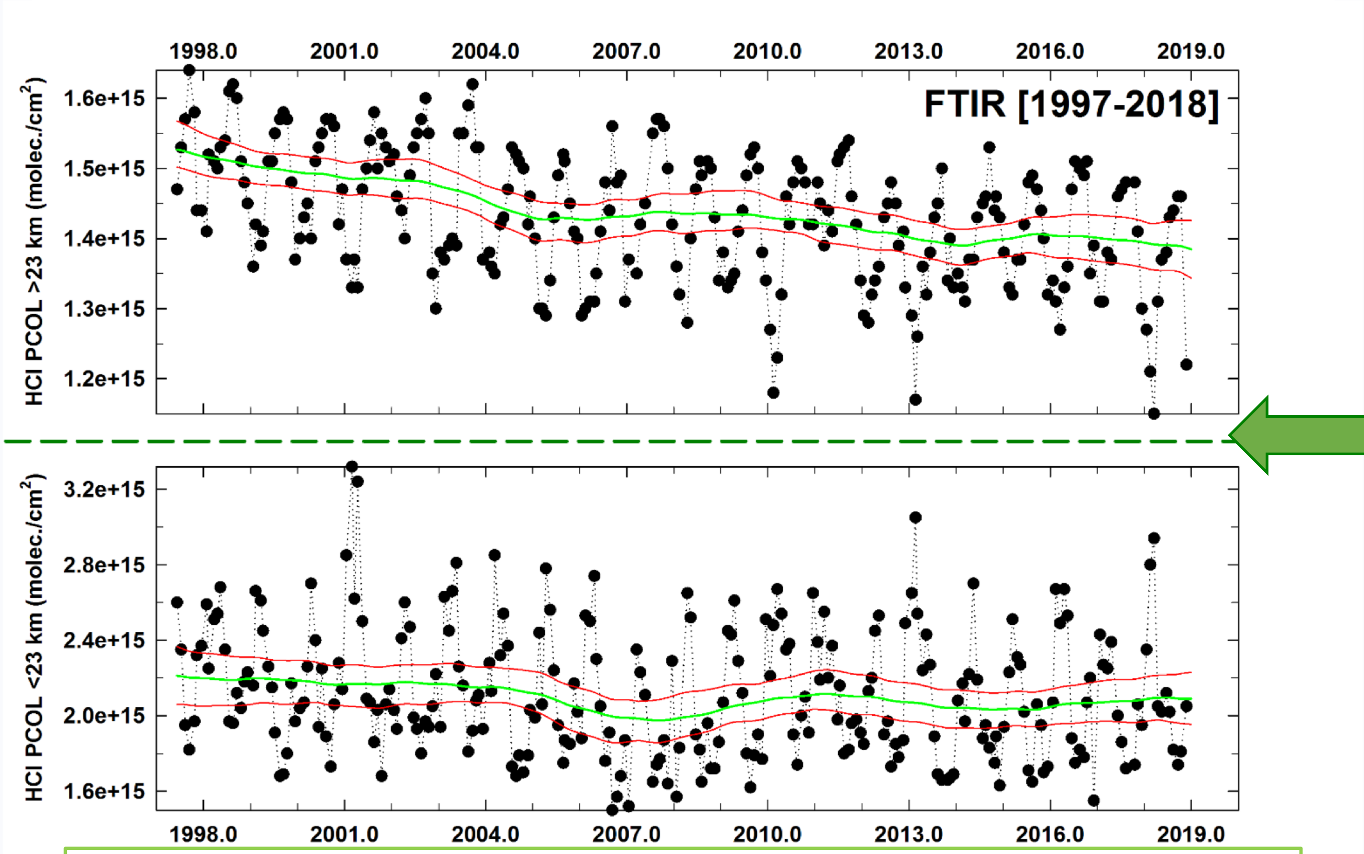
Post-peak FTIR time series

Partial column time series of HCl above and below 23 km

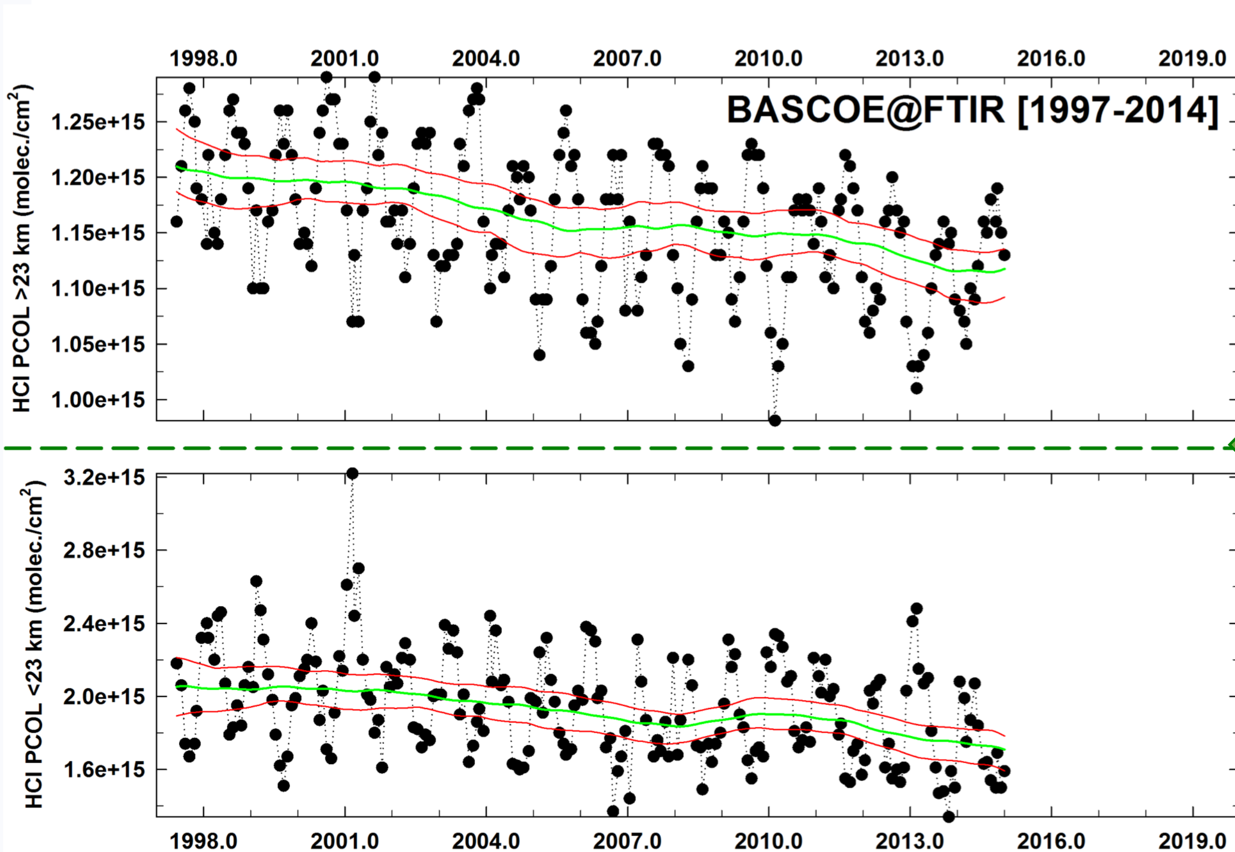


The 2007 minimum is absent from the "upper" time series; overall, the year-to-year variability is lower in mid/upper stratosphere and we observe a more monotonous decrease of HCl

Non parametric estimation of the HCl trends (green) and their corresponding confidence bands (95%; red)

Post-peak BASCOE model time series

Partial column time series of HCl above and below 23 km

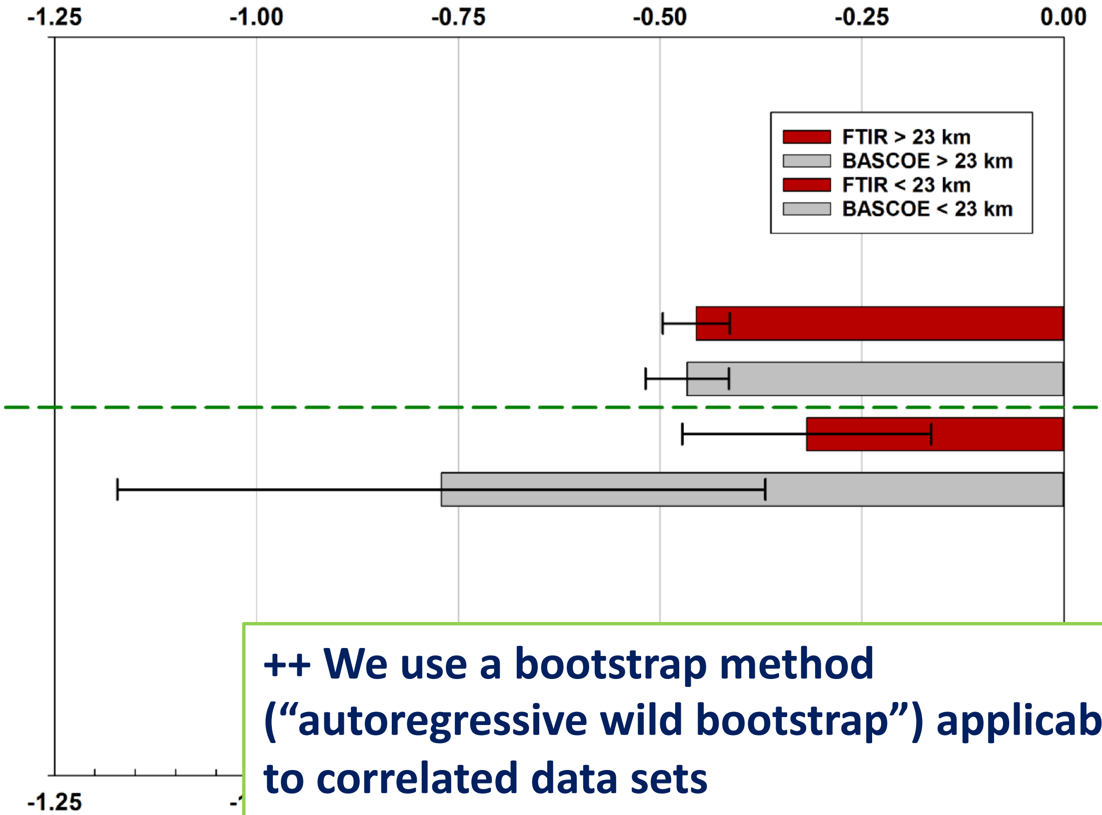


A 2°x2.5° simulation was performed with the 3-D CTM BASCOE model, driven by ERA-Interim, with lower boundary conditions of Meinshausen et al. (GMD, 10, 2017). Agreement btw the FTIR and BASCOE time series is good, albeit a systematic bias of 20% for the mid-upper stratospheric columns

Non parametric estimation of the HCl trends (green) and their corresponding confidence bands (95%; red):

Evaluation of the post-peak trend of HCl

Relative rates of change (%/yr) for the HCl partial column time series above and below 23 km

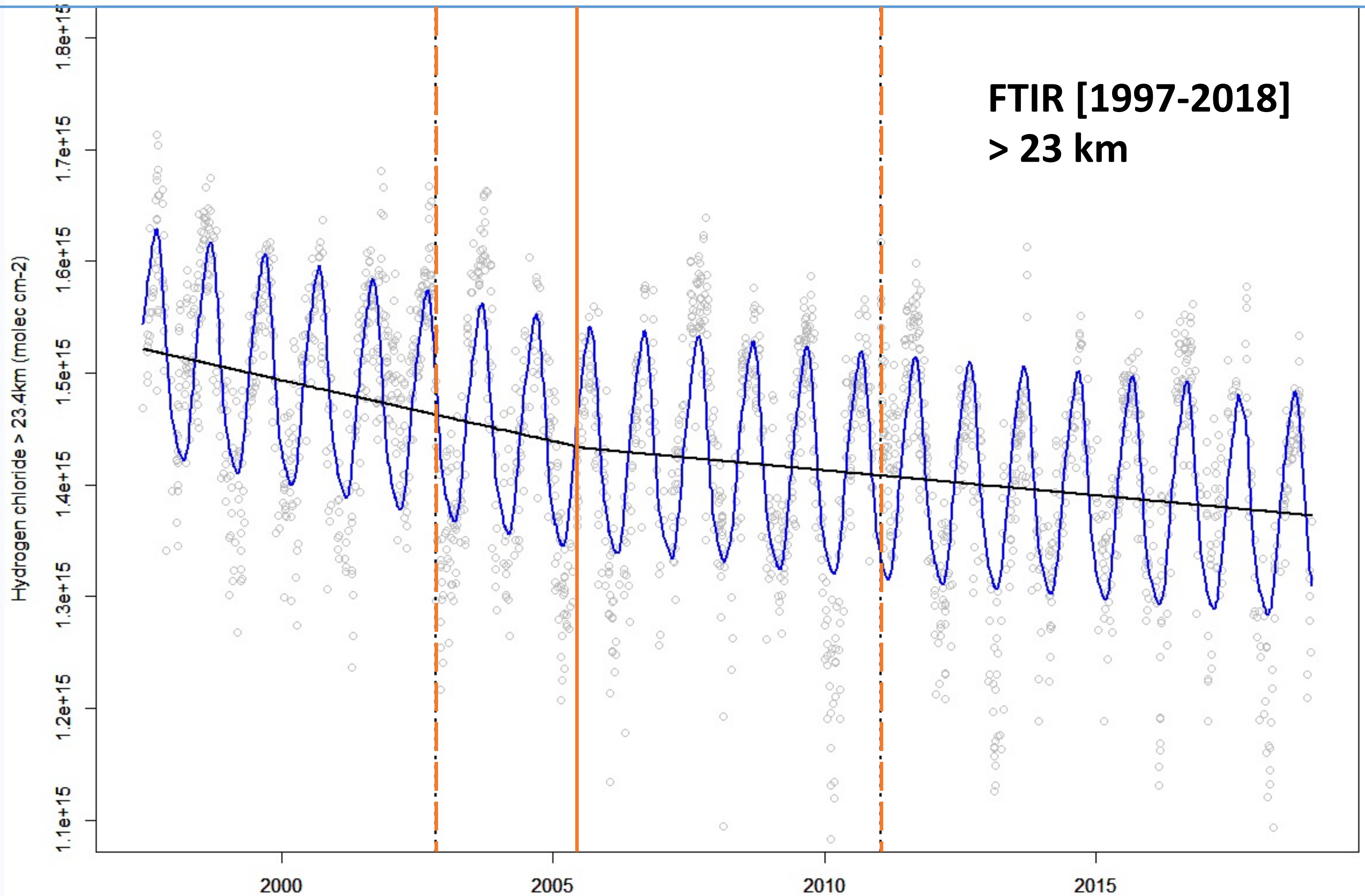


++ We use a bootstrap method ("autoregressive wild bootstrap") applicable to correlated data sets

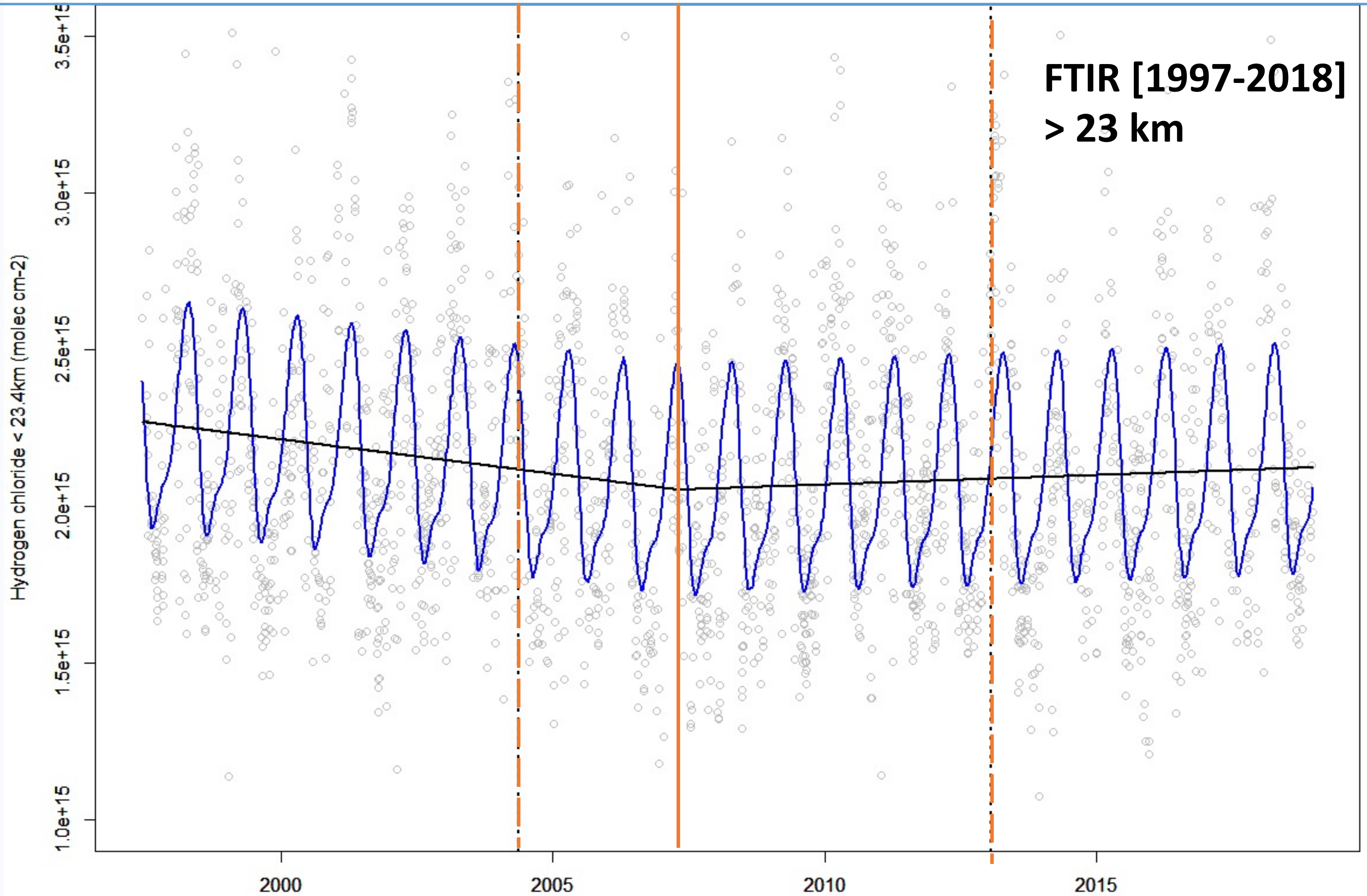
++ A linear function combined with a 3-term Fourier series is used

- We note a very good agreement for the FTIR and BASCOE trends > 23 km
- The rates of change are consistent with ACE-FTS, MLS and *in situ* surface findings (-0.5%/yr)
- The trends for the lower stratosphere are much more noisy and less consistent/robust

The broken trend approach



The broken trend approach



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 is more appropriate for this purpose, also for our FTIR data
- Trends of -5%/decade are derived from the *in situ* networks, from ACE-FTS, MLS and Jungfraujoch FTIR, demonstrating with great precision and consistency the success and effectiveness of the Montreal Protocol

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

emmanuel.mahieu@uliege.be

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Contact information

emmanuel.mahieu@uliege.be

http://labos.ulg.ac.be/girpas/en

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