

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

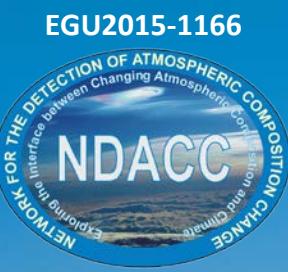
E. Mahieu¹ (emmanuel.mahieu@ulg.ac.be), W. Bader¹, B. Bovy¹, B. Franco¹,
B. Lejeune¹, C. Servais¹, J. Notholt², M. Palm², G.C. Toon³

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TARGET GASES: CFC-11, CFC-12, HCFC-22, HCFC-142b, CCl_4 , CH_3Cl , CF_4 , SF_6

SUBJECTS of the PRESENTATION:

- NEW or UPDATED TIME SERIES & TRENDS over 2000-2014
- ORGANIC CHLORINE AND FLUORINE BUDGETS (CCl_y^* and CF_y^*)
- IMPACT OF CIRCULATION CHANGES in the lower stratosphere



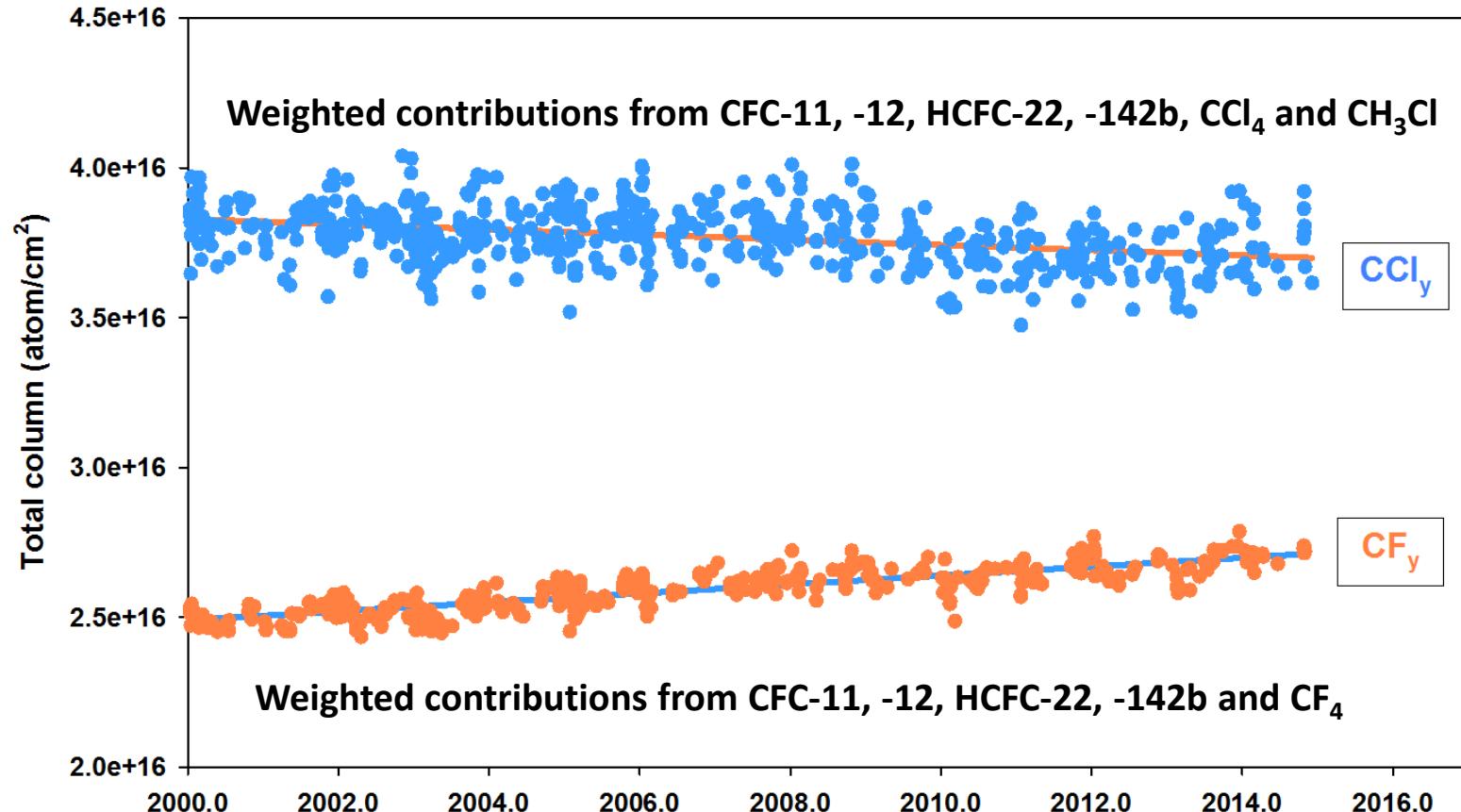
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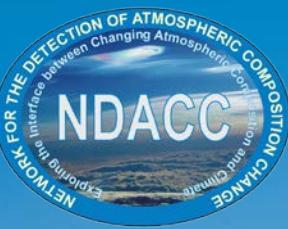


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Evolution of the CCl_y^* and CF_y^* loadings above Jungfraujoch

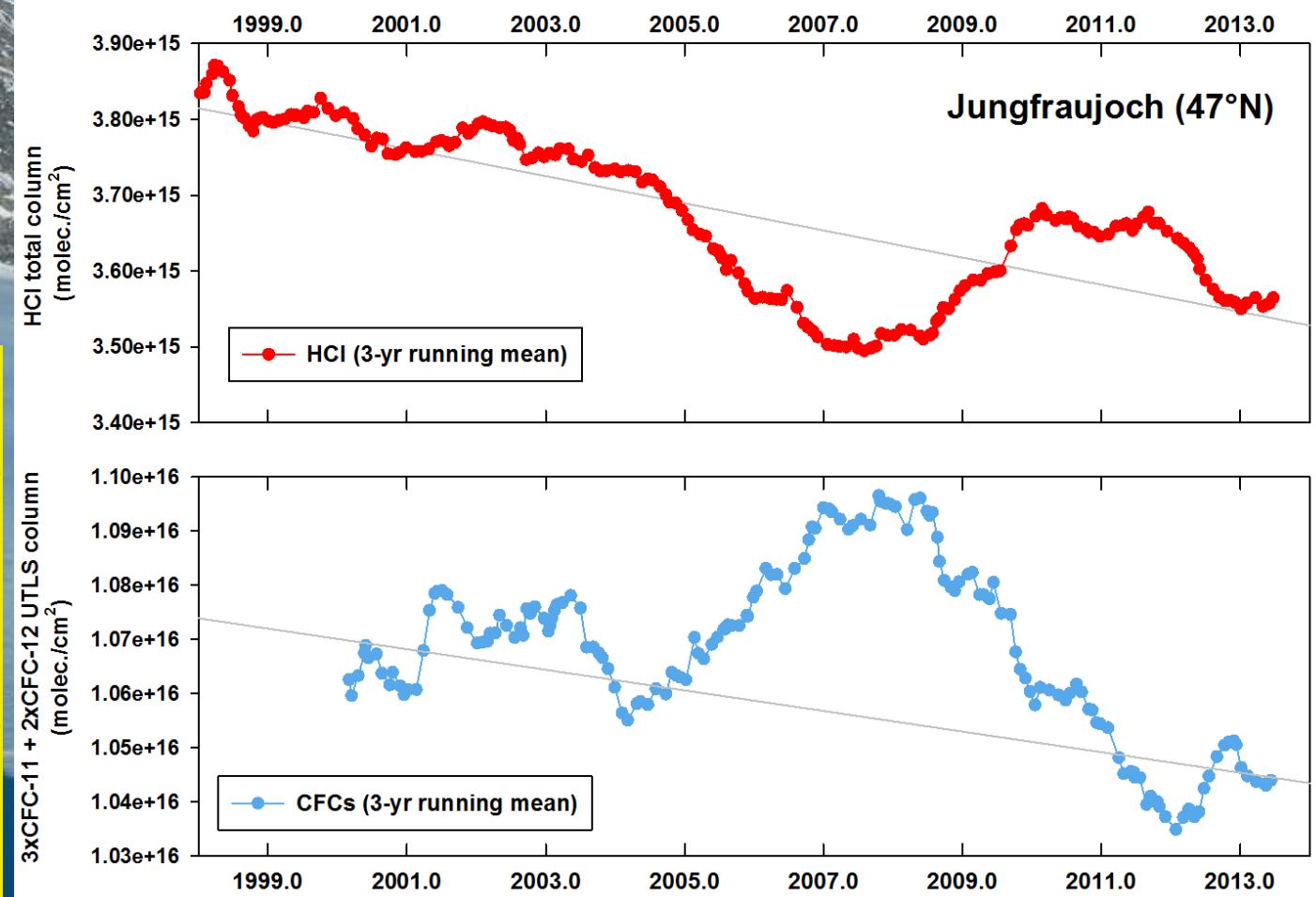




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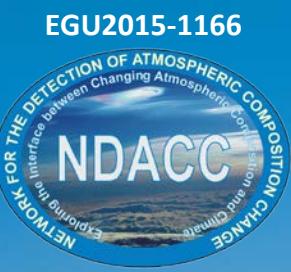
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HCl time series,
updated from
Mahieu et al.
(*nature*13857, 2014)



In the UTLS, CFC-11 and -12 exhibit an opposite signal when compared to HCl over 2004–2011, in contrast with their monotonic decrease observed in the troposphere.



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SITE and INSTRUMENTATION

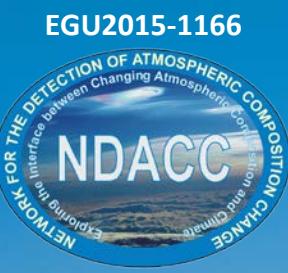
TIME SERIES

UPDATED TRENDS

TYPICAL INFORMATION CONTENT (CFCs)

CIRCULATION CHANGES

Acknowledgments



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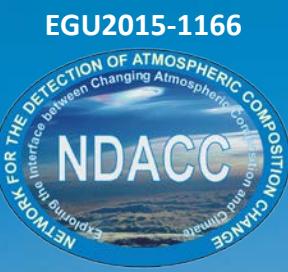
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SITE and INSTRUMENTATION:

- All spectra have been recorded with a high-resolution Fourier Transform InfraRed (FTIR) spectrometer (Bruker 120HR), under clear-sky conditions at the high-altitude International Scientific Station of the Jungfraujoch (ISSJ, Swiss Alps, 46.5°N, 8.0°E, 3580m a.s.l.). This site is located on the saddle between the Jungfrau (4158m) and the Mönch (4107m) summits. FTIR monitoring activities are conducted at that site within the framework of the Network for the Detection of Atmospheric Composition Change (NDACC, see <http://www.ndacc.org>)
- The Bruker spectrometer is equipped with cooled HgCdTe and InSb detectors, allowing covering the 650 to 4500 cm⁻¹ region of the electromagnetic spectrum. For the present investigations, we use high-resolution (0.006 cm⁻¹) IR solar absorption spectra spanning the 700-1400 cm⁻¹ interval. Typical signal-to-noise ratios amount to 800.

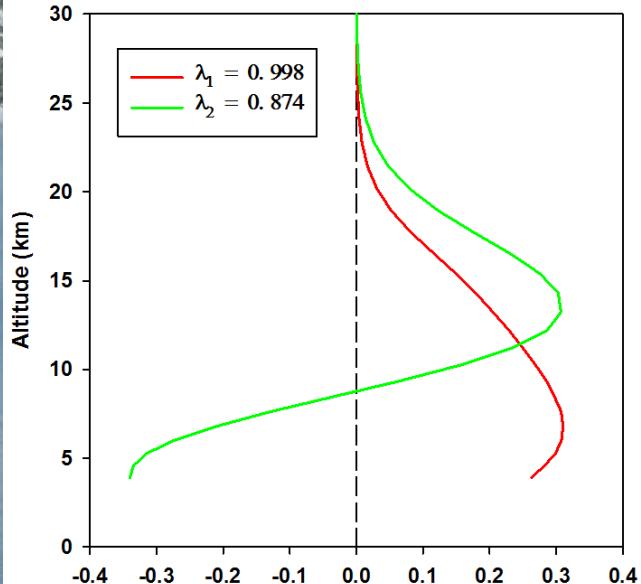


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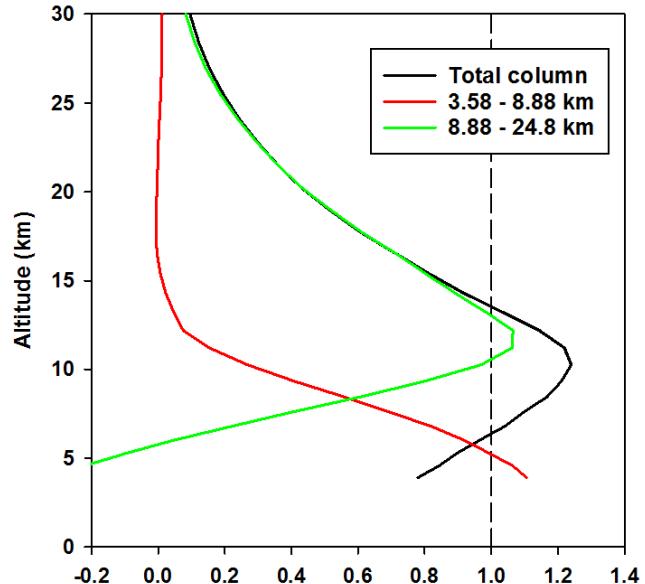
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First Eigen vectors
(CFC-12)

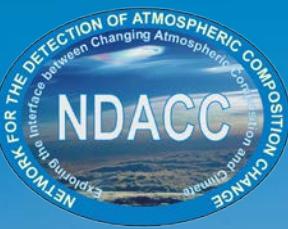


Averaging kernels for merged layers
(CFC-12)



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.

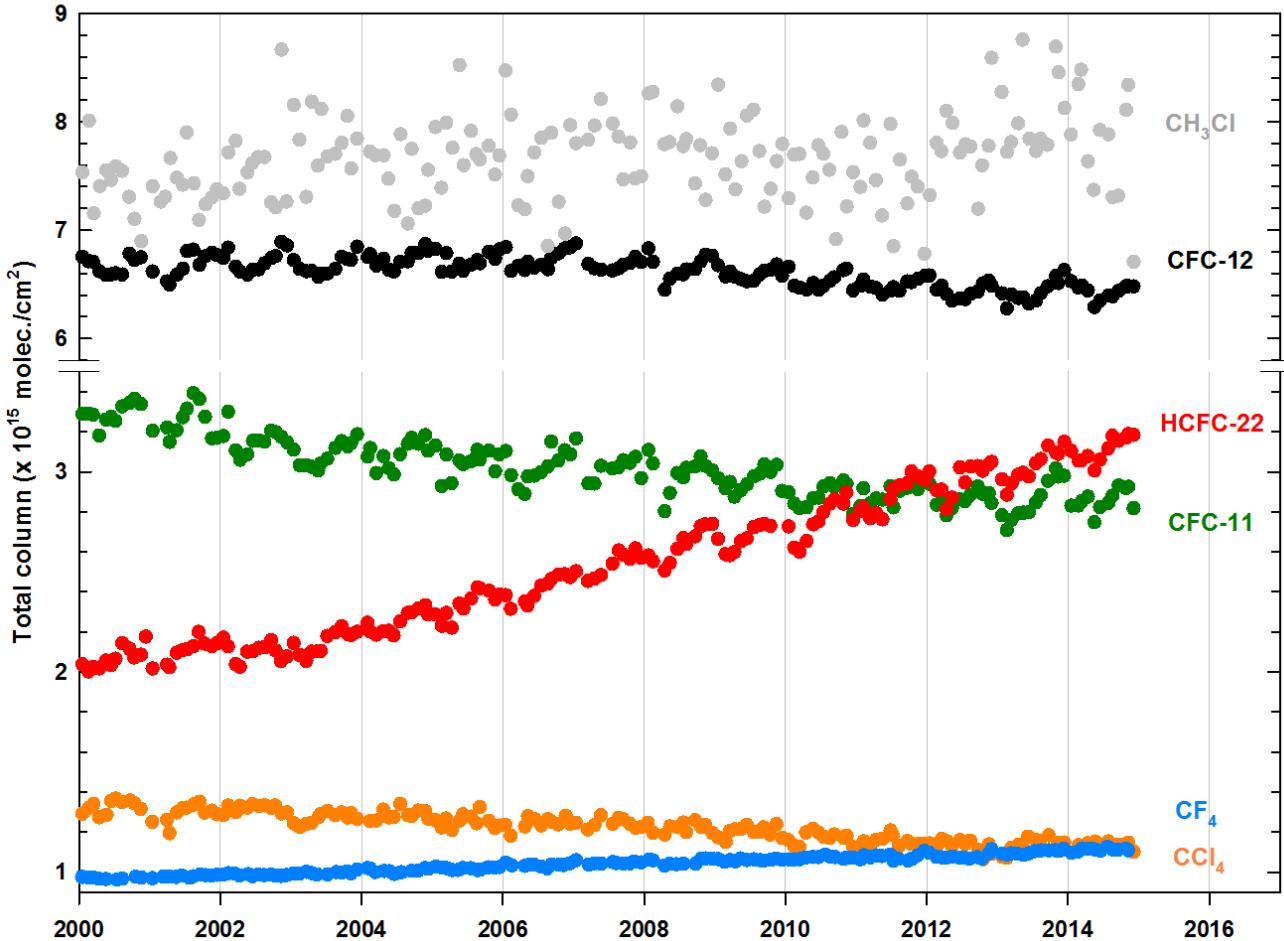


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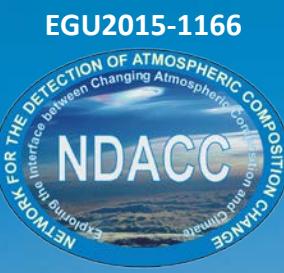
Evolution of a suite of halogenated source gases above Jungfraujoch



TIME SERIES:

All retrievals have been performed with the SFIT-2 algorithm (v3.91) which is based on the semi-empirical implementation of the Optimal Estimation Method of C.D. Rodgers. This code allows in most cases to retrieve information on the vertical mixing ratio profile of the species accessible to the ground-based FTIR technique

All spectra recorded over the last fifteen years have been analyzed, the corresponding time series for CH₃Cl, CFC-11, -12, HCFC-22, CCl₄, and CF₄ are shown here



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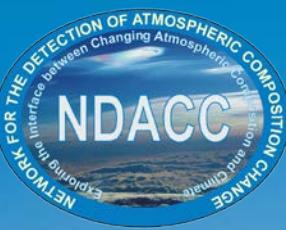
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UPDATED TRENDS:

Species	Time period	Annual absolute and relative trends $\pm 2\sigma$ (FTIR)	in situ 2004-2010
CH_3Cl	[2000-2014]	$+(2.32 \pm 0.80) \times 10^{13} \text{ molec./cm}^2$	$+(0.31 \pm 0.11) \%$
CCl_3F (CFC-11)	[2000-2014]	$-(3.04 \pm 0.11) \times 10^{13} \text{ molec./cm}^2$	$-(0.94 \pm 0.03) \%$
CCl_2F_2 (CFC-12)	[2004-2014]	$-(3.67 \pm 0.18) \times 10^{13} \text{ molec./cm}^2$	$-(0.54 \pm 0.03) \%$
CHClF_2 (HCFC-22)	[2000-2014]	$+(8.04 \pm 0.10) \times 10^{13} \text{ molec./cm}^2$	$+(4.16 \pm 0.05) \%$
CH_3CClF_2 (HCFC-142b)	[2000-2014]	$+(1.01 \pm 0.06) \times 10^{13} \text{ molec./cm}^2$	$+(5.98 \pm 0.32) \%$
CCl_4	[1999-2014]	$-(1.49 \pm 0.06) \times 10^{13} \text{ molec./cm}^2$	$-(1.10 \pm 0.04) \%$
CF_4	[1998-2014]	$+(9.91 \pm 0.07) \times 10^{12} \text{ molec./cm}^2$	$+(0.88 \pm 0.01) \%$
CCl_y^*	[2000-2014]	$-(8.84 \pm 1.79) \times 10^{13} \text{ molec./cm}^2$	$-(0.23 \pm 0.05) \%$
CF_y^*	[2000-2014]	$+(1.48 \pm 0.06) \times 10^{14} \text{ molec./cm}^2$	$+(0.60 \pm 0.03) \%$
SF_6	[2000-2014]	$+(4.00 \pm 0.12) \times 10^{12} \text{ molec./cm}^2$	$+(5.96 \pm 0.17) \%$

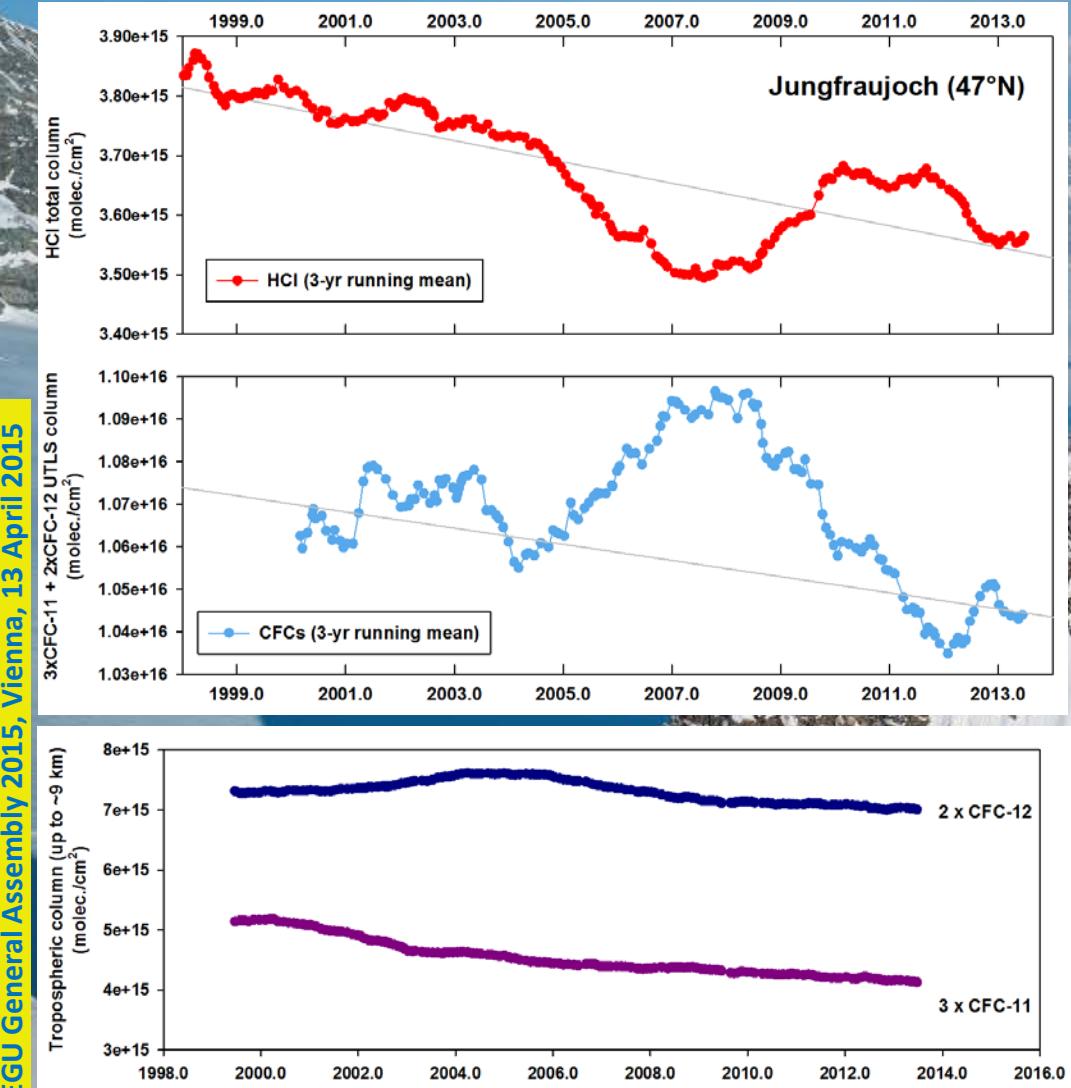
Note: for all relative trends, the abundance of the species for the first year of the time period under consideration is taken as reference
in situ trends from WMO-2014, for 30°N-90°N (adapted from Table 1-2 such as to use 2004 levels as reference)



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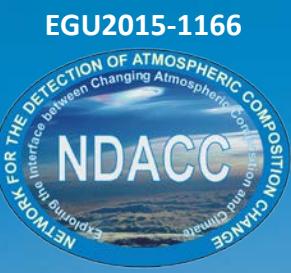
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CIRCULATION CHANGES:

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., *nature*13857, 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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Acknowledgments:

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Permanent link: <http://hdl.handle.net/2268/180469>