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

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TARGET GASES: CFC-11, CFC-12, HCFC-22, HCFC-142b, CCl₄, CH₃Cl, CF₄, SF₆

SUBJECTS of the PRESENTATION:
• NEW or UPDATED TIME SERIES & TRENDS over 2000-2014
• ORGANIC CHLORINE AND FLUORINE BUDGETS (CClₓ* and CFₓ*)
• IMPACT OF CIRCULATION CHANGES in the lower stratosphere
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Weighted contributions from CFC-11, -12, HCFC-22, -142b, CCl₄ and CH₃Cl

-0.23%/yr

Weighted contributions from CFC-11, -12, HCFC-22, -142b, CCl₄ and CH₃Cl

+0.60%/yr

Evolution of the CClₓ* and CFₓ* loadings above Jungfraujoch
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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.

HCl time series, updated from Mahieu et al. (nature13857, 2014)
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\textbf{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 \url{http://www.ndacc.org}).

- The Bruker spectrometer is equipped with cooled HgCdTe and InSb detectors, allowing covering the 650 to 4500 cm\textsuperscript{-1} region of the electromagnetic spectrum. For the present investigations, we use high-resolution (0.006 cm\textsuperscript{-1}) IR solar absorption spectra spanning the 700-1400 cm\textsuperscript{-1} interval. Typical signal-to-noise ratios amount to 800.
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**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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\textbf{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 CH\textsubscript{3}Cl, CFC-11, -12, HCFC-22, CCl\textsubscript{4}, and CF\textsubscript{4} are shown here.
### Updated Trends:

<table>
<thead>
<tr>
<th>Species</th>
<th>Time period</th>
<th>Annual absolute and relative trends ± 2σ (FTIR)</th>
<th>in situ 2004-2010</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH₃Cl</td>
<td>[2000-2014]</td>
<td>+(2.32 ± 0.80) x10¹³ molec./cm²</td>
<td>+(0.31 ± 0.11) %</td>
</tr>
<tr>
<td>CCl₃F (CFC-11)</td>
<td>[2000-2014]</td>
<td>-(3.04 ± 0.11) x10¹³ molec./cm²</td>
<td>-(0.94 ± 0.03) %</td>
</tr>
<tr>
<td>CCl₂F₂ (CFC-12)</td>
<td>[2004-2014]</td>
<td>-(3.67 ± 0.18) x10¹³ molec./cm²</td>
<td>-(0.54 ± 0.03) %</td>
</tr>
<tr>
<td>CHClF₂ (HCFC-22)</td>
<td>[2000-2014]</td>
<td>+(8.04 ± 0.10) x10¹³ molec./cm²</td>
<td>+(4.16 ± 0.05) %</td>
</tr>
<tr>
<td>CH₃CClF₂ (HCFC-142b)</td>
<td>[2000-2014]</td>
<td>+(1.01 ± 0.06) x10¹³ molec./cm²</td>
<td>+(5.98 ± 0.32) %</td>
</tr>
<tr>
<td>CCl₄</td>
<td>[1999-2014]</td>
<td>-(1.49 ± 0.06) x10¹³ molec./cm²</td>
<td>-(1.10 ± 0.04) %</td>
</tr>
<tr>
<td>CF₄</td>
<td>[1998-2014]</td>
<td>+(9.91 ± 0.07) x10¹² molec./cm²</td>
<td>+(1.05 ± 0.01) %</td>
</tr>
<tr>
<td>CCl₄*</td>
<td>[2000-2014]</td>
<td>-(8.84 ± 1.79) x10¹³ molec./cm²</td>
<td>-(0.23 ± 0.05) %</td>
</tr>
<tr>
<td>CF₄*</td>
<td>[2000-2014]</td>
<td>+(1.48 ± 0.06) x10¹⁴ molec./cm²</td>
<td>+(0.60 ± 0.03) %</td>
</tr>
<tr>
<td>SF₆</td>
<td>[2000-2014]</td>
<td>+(4.00 ± 0.12) x10¹² molec./cm²</td>
<td>+(5.96 ± 0.17) %</td>
</tr>
</tbody>
</table>

**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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**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\(_4\) 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:
The University of Liège contribution to the present work has primarily been supported by the AGACC-II project of the SSD program funded by the Belgian Federal Science Policy Office (BELSPO), Brussels. E. Mahieu is Research Associate with the F.R.S. – FNRS. Laboratory developments and mission expenses at the Jungfraujoch station were funded by the F.R.S. – FNRS and the Fédération Wallonie-Bruxelles, respectively. We thank the International Foundation High Altitude Research Stations Jungfraujoch and Gornergrat (HFSJG, Bern) for supporting the facilities needed to perform the observations. We further acknowledge the vital contribution from all the Belgian colleagues in performing the Jungfraujoch observations used here.

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