

High resolution solar infrared Fourier transform spectrometry: application to the study and long-term monitoring of the Earth's atmosphere

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1. Project description

The Liège team has a long tradition in the monitoring of the Earth's atmosphere. Indeed, the first observations and investigations were carried out by Pr Marcel Migeotte and collaborators in the late 1940s, using a grating infrared spectrometer. This instrument was then installed at the Jungfraujoch station and infrared spectra were systematically recorded in 1950-1951 such as to cover the 2.8 to 23.7 micrometer (μm) spectral range (Migeotte et al., 1956). The next period was dedicated to the study of the sun and to the production of photometric solar atlases, using a 7 m grating spectrometer, in single then double pass mode. In the mid-1970s, the team resumed its atmospheric monitoring activities which are still ongoing nowadays. Since the mid-1980s, Fourier Transform InfraRed (FTIR) instruments are used, allowing to record very high resolution and signal-to-noise wide-band solar infrared spectra. This sustained effort has led to an unrivalled collection of infrared spectra which is unique worldwide in terms of length, measurement density and quality. At the end of 2019, we reached 36 years of continuous FTIR measurements at the Jungfraujoch station!

The main objectives of the team are essentially twofold: (i) maintain the instrumentation operational while also improving its performance, (ii) analyse the spectra in order to produce high-level geophysical parameters and valorise them.

In 2019, observations have been performed on site or recorded through a proprietary remote-control internet interface. Altogether, about 1200 high resolution infrared solar spectra have been collected on 72 days over the first eleven months of the year (the statistics for December are not yet available at the time of writing). A failure of the instrumentation (sun-tracker protection control) prevented the recording of spectra in August.

The analysis of our spectra allows us to determine the abundance of an increasing number of key constituents of the Earth

atmosphere (currently more than 30, see Table 1), playing a role in ozone depletion, climate change, or affecting air quality. Numerous target species are therefore relevant to the Montreal Protocol on substances that deplete stratospheric ozone (e.g. CFCs, HCFCs, HCl) and/or to the Paris Agreement (COP21) to mitigate climate change (e.g. CO₂, CH₄, N₂O).

Table 1. List of atmospheric species (>30) currently retrieved from the Jungfraujoch observational database.

Greenhouse gases; support to the Paris Agreement	H ₂ O, CO ₂ , CH ₄ , N ₂ O, CF ₄ , SF ₆
Ozone-related; support to the Montreal Protocol	O ₃ , NO, NO ₂ , HNO ₃ , ClONO ₂ , HCl, HF, COF ₂ , CFC-11, CFC-12, HCFC-22, HCFC-142b, CCl ₄ , CH ₃ Cl
Air quality; support to the EU-Copernicus programme	CO, CH ₃ OH, C ₂ H ₆ , C ₂ H ₂ , C ₂ H ₄ , HCN, HCHO, HCOOH, NH ₃ , PAN
Other	OCS, N ₂ , various isotopologues ¹

¹) an isotopologue is a molecular twin that differs from the reference molecule in the isotopic composition; for example, ¹³C¹⁶O and ¹²C¹⁸O are the isotopologues of the most abundant ¹²C¹⁶O.

2. Update of the long-lived halocarbon time series

As obvious from Table 1, several halogenated source and reservoir species are systematically retrieved from the Jungfraujoch FTIR spectra. The corresponding time series are regularly included in the Scientific Assessment of Ozone Depletion reports, supporting the evaluation of the success of the Montreal Protocol. This was again the case in the last edition of that report (WMO2018, see Figures 1-2, 1-13, 1-20 and Table 1-2 in its chapter 1).

Figure 1 provides the updated time series of the most abundant (hydro)chlorofluorocarbons ((H)CFCs). The HCFC-22 time series has

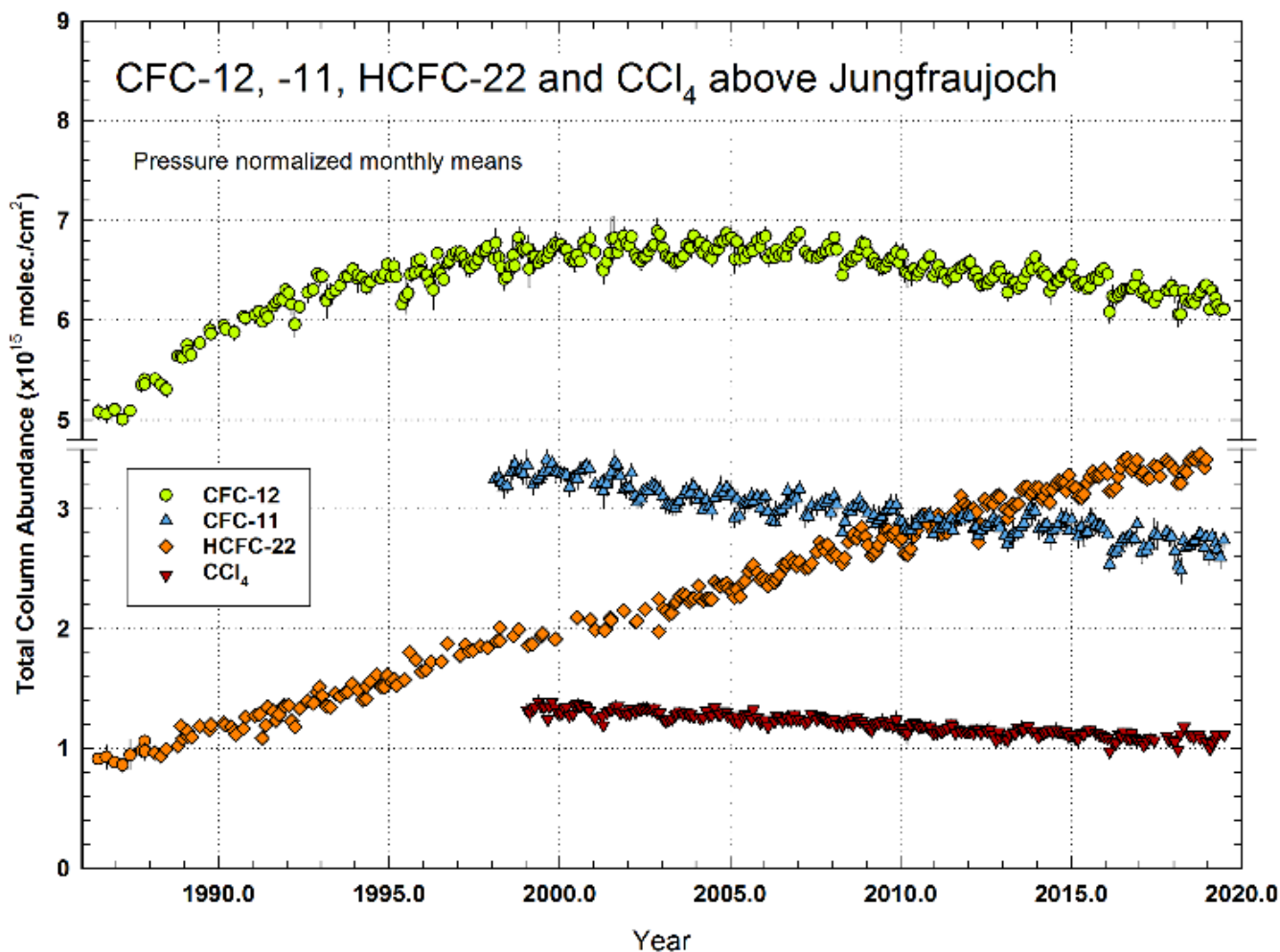


Figure 1. Monthly mean total vertical column abundances (in molecules per square centimeter) of CFC-12 (CCl_2F_2), CFC-11 (CCl_3F), CCl_4 and HCFC-22 (CHClF_2) as derived from the long-term monitoring program conducted by ULiège at the Jungfraujoch station. The CFC-11 and CFC-12 time series have been consistently updated from Zander et al. (2008), the CCl_4 time series from Rinsland et al. (2012). The HCFC-22 data set is described in Prignon et al (2019).

been recently updated and improved (Prignon et al., 2019) while the three other data sets have been extended until mid-2019. Current trends have been evaluated considering all available daily means for the 2013-2019 time period, and accounting for the auto-correlation present in the data sets following Santer et al. (1999). When compared with the rates of change reported in Table 1-2 of WMO2018 (using data from 2010-2016), we don't see significant differences for the CFC-11, CFC-12 and CCl_4 trends. For HCFC-22, the absolute yearly increase ($\text{E13 molec.cm}^{-2}.\text{yr}^{-1}$) drops from 7.58 over 2010-2016 to 5.65 over 2013-2019, i.e., lower by 25%. This corresponds to relative rates of increase of 2.54(20) and 1.77(12) $\%.\text{yr}^{-1}$, when using the year 2013 and 2016 as reference, respectively.

References

Migeotte, M., Neven, L. and Swensson, J.: The solar spectrum from 2.8 to 23.7 microns—part I: Photometric atlas, *Mém. Soc. Roy. Sci. Liège, Special vol 1*, 1956.

Prignon, M., Chabrilat, S., Minganti, D., O'Connell, Doherty, S., Servais, C., Stiller, G., Toon, G. C., Vollmer, M. K. and Mahieu, E.: Improved FTIR retrieval strategy for HCFC-22 (CHClF_2), comparisons with in situ and

satellite datasets with the support of models, and determination of its long-term trend above Jungfraujoch, *Atmos. Chem. Phys.*, 19(19), 12309–12324, doi:10.5194/acp-19-12309-2019, 2019.

Rinsland, C. P., Mahieu, E., Demoulin, P., Zander, R., Servais, C. and Hartmann, J.-M.: Decrease of the carbon tetrachloride (CCl_4) loading above Jungfraujoch, based on high resolution infrared solar spectra recorded between 1999 and 2011, *J. Quant. Spectrosc. Radiat. Transf.*, 113(11), 1322–1329, doi: 10.1016/j.jqsrt.2012.02.016, 2012.

Santer, B. D., Wigley, T. M. L., Boyle, J. S., Gaffen, D. J., Hnilo, J. J., Nychka, D., Parker, D. E. and Taylor, K. E.: Statistical significance of trends and trend differences in layer-average atmospheric temperature time series, *J. Geophys. Res.*, 105(D6), 7337–7356, doi:10.1029/1999JD901105, 2000.

Zander, R., Mahieu, E., Demoulin, P., Duchatelet, P., Roland, G., Servais, C., De Mazière, M., Reimann, S. and Rinsland, C. P.: Our changing atmosphere: Evidence based on long-term infrared solar observations at the Jungfraujoch since 1950, *Sci. Total Environ.*, 391(2–3), 184–195, doi: 10.1016/j.scitotenv.2007.10.018, 2008.

WMO (World Meteorological Organization), *Scientific Assessment of Ozone Depletion: 2018*, Global Ozone Research and Monitoring Project-Report No. 58, 588 pp., Geneva, Switzerland, 2018.

Internet data bases

<http://labos.ulg.ac.be/girpas/en/publications>
<http://labos.ulg.ac.be/girpas/en/>
<ftp://ftp.cpc.ncep.noaa.gov/ndacc/station/jungfrau/hdf/ftir/>
<ftp://ftp.cpc.ncep.noaa.gov/ndacc/RD/jungfrau/hdf/ftir/>

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Scientific publications and public outreach 2019**Refereed journal articles and their internet access**

Mahieu, E., O. Flock, J. Notholt, M. Palm, I. Pardo Cantos, M. Prignon, G. Roland, C. Servais, and A. Verma, Surveillance à long terme de l'atmosphère terrestre à la station du Jungfraujoch, *Bull. Soc. R. Sci. Liège*, **88**, 31–41, doi: 10.25518/0037-9565.9136, 2019.
<https://popups.uliege.be/0037-9565/index.php?id=9136>

Prignon, M., S. Chabrilat, D. Minganti, S. O'Doherty, C. Servais, G. Stiller, G.C. Toon, M.K. Vollmer, and E. Mahieu, Improved FTIR retrieval strategy for HCFC-22 (CHClF₂), comparisons with in situ and satellite datasets with the support of models, and determination of its long-term trend above Jungfraujoch, *Atmos. Chem. Phys.*, **19**, 19, 12309–12324, doi: 10.5194/acp-19-12309-2019, 2019.
<https://orbi.uliege.be/handle/2268/233951>

Stinecipher, J.R., P.J. Cameron-Smith, N.J. Blake, L. Kuai, B. Lejeune, E. Mahieu, I.J. Simpson, and J.E. Campbell, Biomass burning unlikely to account for missing source of carbonyl sulfide, *Geophys. Res. Lett.*, **46**, 24, 14912–14920 doi: 10.1029/2019GL085567, 2019.
<https://doi.org/10.1029/2019GL085567>

Tzompa-Sosa, Z.A., B.H. Henderson, C.A. Keller, K. Travis, E. Mahieu, B. Franco, M. Estes, D. Helmig, A. Fried, D. Richter, P. Weibring, J. Walega, D.R. Blake, J.W. Hannigan, I. Ortega, S. Conway, K. Strong, and E.V. Fischer, Atmospheric Implications of Large C₂-C₅ Alkane Emissions From the U.S. Oil and Gas Industry, *J. Geophys. Res. Atmos.*, 2013, 1–22, doi: 10.1029/2018JD028955, 2019.
<https://orbi.uliege.be/handle/2268/230483>

Conference Papers

Mahieu, E., M. Prignon, C. Servais, S. Chabrilat, Q. Errera, M. Friedrich, S. Smeeke, L. Froidevaux, R.J. Salawitch, P. Wales, J. Notholt, and M.P. Chipperfield, Post-peak trend of upper stratospheric hydrogen chloride derived from ground-based FTIR solar spectra and model simulations, Paper presented at European Geosciences Union General Assembly, Vienna, Austria, April 12, 2019. <http://hdl.handle.net/2268/237056>

Minganti, D., S. Chabrilat, Y. Christophe, Q. Errera, M. Prignon, E. Mahieu, and M. Abalos, Investigation of stratospheric circulation using long-lived tracers with WACCM, BASCOE CTM and a reanalysis of MLS observations, Poster presented at European Geosciences Union General Assembly, Vienna, Austria, April 12, 2019. <http://hdl.handle.net/2268/236874>

Prignon, M., P.F. Bernath, S. Chabrilat, M.P. Chipperfield, S. Dhomse, W. Feng, D. Minganti, C. Servais, D. Smale, and E. Mahieu, Impact of lower stratospheric dynamical variability on total inorganic fluorine derived from ground-based FTIR, satellite and model data, Poster presented at European Geosciences Union General Assembly, Vienna, Austria, April 12, 2019. <http://hdl.handle.net/2268/234238>

Data books and reports

Update on Ozone-Depleting Substances (ODSs) and Other Gases of Interest to the Montreal Protocol, Chapter 1 in *Scientific Assessment of Ozone Depletion: 2018*, Global Ozone Research and Monitoring Project-Report No. 58, World Meteorological Organization, Geneva, Switzerland, 2019.

Magazine and Newspaper articles

“Jungfraujoch, lanceur d'alerte pour le climat”, *L'Alpe*, n°84, Printemps 2019. <https://www.lalpe.com/lalpe-84-observatoires-un-oeil-sur-le-cosmos/>

“Climate scientists gather for conference”, *Otago Daily Times*, May 25-26, 2019. <https://www.odt.co.nz/regions/central-otago/climate-scientists-gather-conference>

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