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

Emmanuel Mahieu[1], Christian Servais[1], Whitney Bader[1], Olivier Flock[1], Maxime Prignon[1], and Ginette Roland[1]

[1] Institute of Astrophysics and Geophysics – University of Liège, Liège, Belgium

emmanuel.mahieu@uliege.be

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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 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 a collection of infrared spectra which is unique worldwide, in terms of length, measurement density and quality. A meaningful statistic reveals that over the last twenty-five years, the longest time period without observation never exceeded 53 days, underlining the long-term commitment of the team members and their capacity to quickly fix any instrumental failure. We would like to mention that the BOLO museum came once more to collect old HP equipment and our Benson graph plotter. It is also worth noting that in 2018, Dr Ginette Roland celebrated the 60th anniversary of her first stay at the Jungfraujoch. Since then, she has been up there every year and has spent altogether nearly 15 years at the Sphinx!

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 2018, observations have been performed on site or recorded through a proprietary remote-control internet interface. Altogether, about 1500 high resolution infrared solar spectra have been collected on 87 days over the first eleven months of the year (the statistics for December are not yet available at the time of writing).

The team has recently finalised the implementation of the SFIT-4 retrieval algorithm (version 0.9.4.4). This new tool represents a significant improvement with respect to SFIT-2. A very useful feature of this code is the computation "on the fly" of full per-spectrum uncertainty budgets for the various systematic and random components, still increasing the objective characterisation of our data sets.

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

Greenhouse gases; support to the Paris Agreement	H_2O, CO_2, CH_4, N_2O, 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_3Cl
Air quality; support to the EU-Copernicus programme	CO, CH_3OH, C_2H_6, C_2H_2, C_2H_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, $^{13}C^{16}O$ and $^{12}C^{18}O$ are the isotopologues of the most abundant $^{12}C^{16}O$

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. CO_2, CH_4, N_2O).

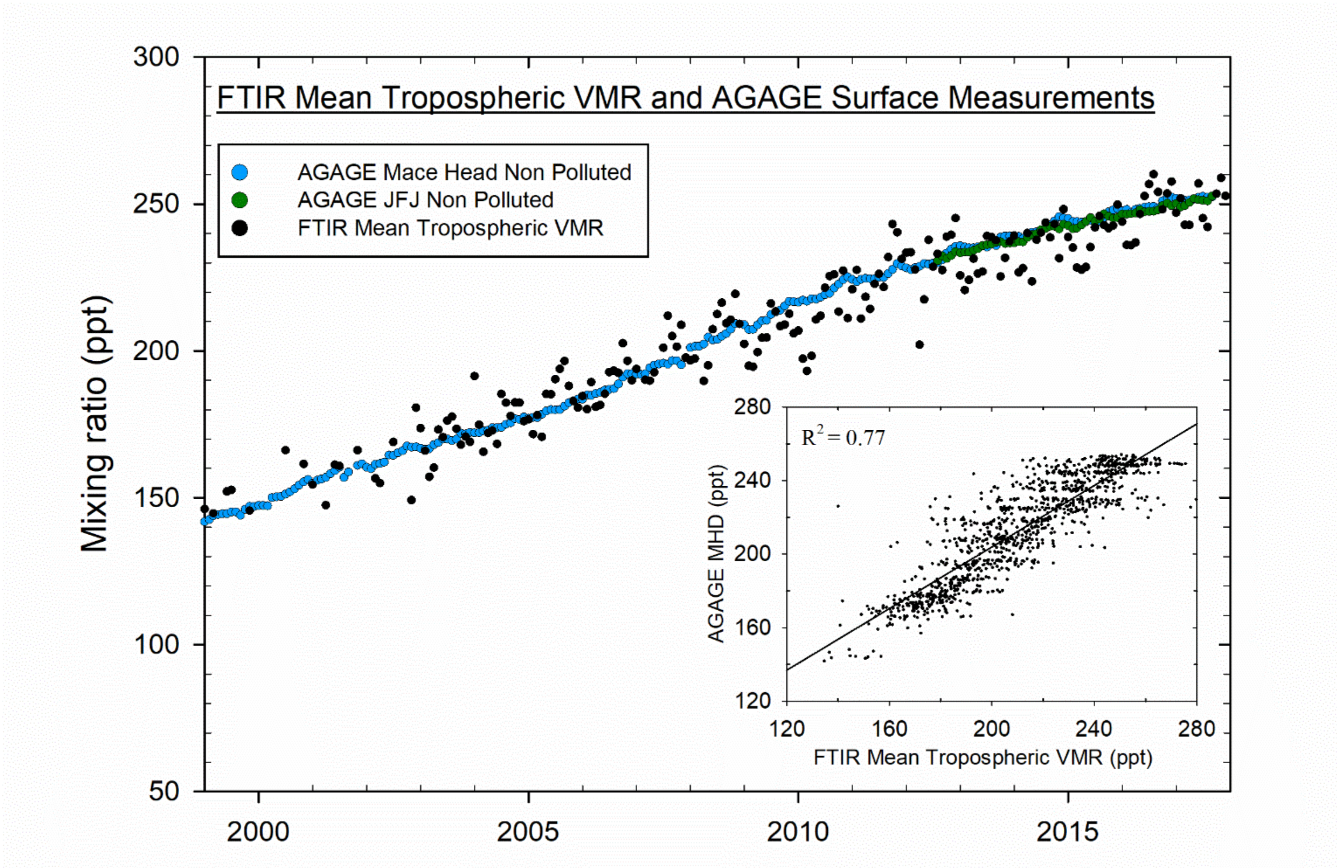

Figure 1 Tropospheric monthly mean time series of HCFC-22 at Jungfraujoch. FTIR series (black) is derived by taking the average of all the layers below 11.2 km, the altitude limit objectively defined by the retrieval information content analysis. AGAGE in situ series from Mace Head (light blue) and Jungfraujoch (green) do not include polluted (flagged by AGAGE teams) measurements. Daily coincidences between the Mace Head and FTIR data are depicted in the lower right scatter plot.

In 2018, we further settled a new service aiming at the rapid delivery of FTIR data in support to the Copernicus Atmospheric Monitoring Programme of the European Union. Currently, height-resolved FTIR data (augmented with information content and uncertainty budgets) are provided for carbon monoxide, methane and ozone, as soon as possible after spectra acquisition, and in all cases within a delay remaining below a month. These data are needed for the verification of the air quality model forecasts delivered by Copernicus at the global, regional and local scales. On the scientific side, the team has developed or improved retrieval strategies for heavy isotopologues of methane (CH_3D and $^{13}CH_4$), for HCFC-22 and PAN (peroxyacetyl nitrate, a stable reservoir for the NO_x pollutants). A study dealing with the new multi-decadal time series of HCFC-22 is in preparation by Prignon et al., we present some results in the next section. For a complete view of the team's scientific output in 2018, the reader is invited to consult the literature (see the "Refereed journal articles" section) and the team's web site under the publications tab.

2. Improved 30-year time series of HCFC-22 (CHClF2)

Although HCFC-22 ($CHClF_2$, among the first substitutes to the manmade chlorofluorocarbons, or CFCs) is a traditional target of the FTIR monitoring programme at Jungfraujoch, the corresponding product has thus far been essentially limited to total column time series, with at most limited information content and hence, vertical resolution. Prignon et al. have thoroughly revisited the retrieval strategy, re-evaluating all available micro-windows and regularisation methods. They eventually selected a single microwindow approach (centered on the $2\nu_6$ Q-branch at 829 cm^{-1}) and a Tikhonov L1 regularisation. For the Bruker instrument, this improved strategy enables the determination of two independent pieces of information with only a limited impact of the a priori state. Indeed, the mean degree of freedom (DOFS) amounts to 1.97, and the second eigenvector provides partial column time series below and above the tropopause, with most of the information coming from the retrieval (eigenvalue of 0.85). A complete uncertainty budget has been carefully established, indicating random and systematic errors of 2.7 and 5.5 % on the individual total columns, respectively.

In order to check the validity of the tropospheric and stratospheric FTIR time series, in-depth comparisons have been conducted, involving independent datasets available in the literature. MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) data spanning the 2005-2012 time period were shown to be in excellent agreement with the stratospheric FTIR time series, with a non-significant FTIR-MIPAS bias of (-4.64 ± 6.1) % (1σ). The lower stratospheric trends from both experiments are also in excellent agreement, close to 3.3%/yr for the coincident time period 2005-2012.

The mean tropospheric mixing ratio time series has been compared with unpolluted surface Gas Chromatography-Mass Spectrometry

(GC-MS) measurements by the AGAGE (Advanced Global Atmospheric Gases Experiment) network, considering baseline data from Mace Head (MHD, 55°N, Ireland) and Jungfraujoch (Empa). Figure 1 shows the AGAGE and FTIR time series for the common measurement time period. Here again, there is no significant bias between the ensembles. When comparing the MHD and FTIR series, we derive a men bias of (-1.1 ± 6.6 %; 1σ). The insert in Fig. 1 shows the good correlation between the individual daily means, with an R-correlation factor of 0.87. The trend investigations highlighted a slowing down of the HCFC-22 accumulation in the troposphere, with successive relative decadal trends of 3.7 and 2.3%/yr, for 1999-2008 and 2008-2017, respectively.

This updated data set therefore represents a significant step forward; it is indeed now possible to monitor, from the ground, the long-term evolution of HCFC-22 in the troposphere as well as in the stratosphere. More information will soon be available in Prignon *et al.*

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

Collaborating partners / networks

Dr. S. Reimann, Dr. M. Steinbacher, and Dr M.K. Vollmer, Empa, Dübendorf
Dr. F. Conen, Institut für Umweltgeowissenschaften, Universität Basel
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Dr M.P. Chipperfield and team, University of Leeds, Leeds, U.K.
Dr J. Notholt and M. Palm, University of Bremen, Bremen, Germany
Dr G.C. Toon, California Institute of Technology, NASA-JPL, Pasadena, CA
Mr J. Hannigan, National Center for Atmsopheric Resaerch, Boulder, CO
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Address

GIRPAS (Groupe Infra-Rouge de Physique Atmosphérique et Solaire)
Institut d'Astrophysique et de Géophysique – Université de Liège
Quartier Agora, Allée du 6 Août, 19, Building B5A
B-4000 Liège (Sart Tilman)
Belgium

Contacts

Christian Servais – Project leader
Tel.: +32 4 366 9784
e-mail: christian.servais@uliege.be
Whitney Bader
Tel.: +32 4 366 9789
e-mail: whitney.bader@uliege.be
Olivier Flock
Tel.: +32 4 366 9790
e-mail: o.flock@uliege.be
Emmanuel Mahieu
Tel.: +32 4 366 9786
e-mail: emmanuel.mahieu@uliege.be
Maxime Prignon
Tel.: +32 4 366 9785
e-mail: maxime.prignon@uliege.be
Ginette Roland
+32 4 342 2594
e-mail: g.roland@uliege.be
Vincent Van De Weerdt
+32 4 366 9767
Vincent.vandeweerdt@uliege.be
Diane Zander
e-mail: diane.zander@uliege.be
+32 4 366 9770