

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 Prof. 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). It is important to mention that all these spectra, available on paper-rolls, have now been digitized and calibrated in terms of wavenumber and signal strength (Makkor et al. 2025).

The next two decades were 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 have been 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.

The current main objectives of the team are essentially twofold: (i) maintain the instrumentation at the state-of-the art, (ii) analyse the spectra to produce high-level geophysical parameters and valorise them.

In 2024, a major change occurred, with the replacement of our 120HR FTIR spectrometer by a 125HR instrument, also from Bruker Optics. This only briefly interrupted our monitoring program. Indeed, solar observations have been collected remotely, but also on-site, during all months but August. 982 spectra were obtained on 34 days with the 120HR. And 1185

spectra were collected with the 125HR spectrometer on 31 days.

Table 1: List of atmospheric species 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	ClONO ₂ , HCl, HF, COF ₂ , CFC-11, CFC-12, HCFC-22, HCFC-142b, CCl ₄ , CH ₃ Cl, HFC-134a, HFC-23
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.

The systematic analysis of our solar FTIR spectra allows us to determine the abundance of an increasing number of key constituents of the Earth atmosphere (currently more than 35, 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, HFCs, HCl) and/or to the Paris Agreement (COP21) to mitigate climate change (e.g., CO₂, CH₄, N₂O). It is also worth noting that long-lived tracers such as, e.g., HCl, HF, N₂O, can also be used to provide useful insights regarding global atmospheric circulation (e.g., Minganti et al. 2022; Prignon et al. 2021).



Figure 1: Removal on July 22, 2024, of the 120HR instrument via the balcony of the FTIR laboratory, using the hoist located on the upper Sphinx terrace. Picture E. Mahieu

2 2024 highlights

This is after 35 years of uninterrupted service that the 120HR instrument ceased operation, after the recording of the last atmospheric spectrum on July 19, and of the last laboratory spectrum on July 21. This instrument has then been carefully disassembled in three segments to allow its transport to its next destination, the UNESCO World Heritage Swiss Alps Jungfrau-Aletsch Museum in Naters (visit wnf.ch/ftir), to be used in a permanent exhibition designed to raise young people's awareness of the accumulation of greenhouse gases in the Earth's atmosphere and of climate change. Figure 1 illustrates the evacuation of one of the three sections.

After its entire emptying, the FTIR laboratory underwent a complete renovation in August, including the replacement of the electrical installation. The deployment of the new FTIR spectrometer took place the first week of September, including its optical alignment and a suite of tests. First atmospheric spectra were recorded on September 6, 2024. Routine operation of the 125HR instrument started in October, and it is continuing nominally.

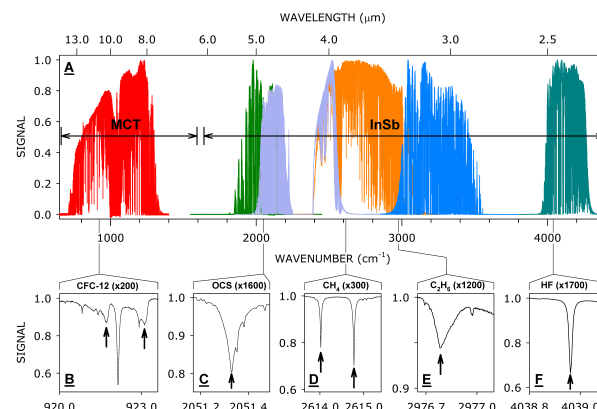


Figure 2: The spectral ranges covered by the six optical filters and MCT or InSb detectors used at the Jungfraujoch station for the new Bruker 125HR spectrometer are shown in Box A, after normalization of the vertical scale to 1. These bandwidths are also influenced by the absorption of the main greenhouse gases, enabling “atmospheric windows” to be defined. Frames B to F show a selection of spectral micro-windows, including absorptions of a few target molecules, after magnifying the horizontal scale by 200 to 1700. Mind the vertical scales for the different panels.

The 125HR instrument has a maximum optical path difference of 258 cm, optimum for atmospheric monitoring. Its configuration was selected such as to avoid any extra chamber or mirror. It has three detector positions, allowing to easily perform pairwise intercomparison of Mercury-Cadmium-Telluride (HgCdTe or MCT) or Indium-Antimonide (InSb) detectors, or to later add an Indium-Gallium-Arsenide (InGaAs) detector such as to also cover the near infrared range of the electromagnetic spectrum. The detectors in operation in the 120HR proved superior with respect to new or other devices available to us, and they were selected for routine operation. In addition, we also kept almost all optical filters that were implemented in the 120HR, maximizing the instrumental continuity and consistency of our FTIR monitoring program. The spectral range covered by the 125HR instrument is depicted in Figure 2.

The motivations for renewing the FTIR spectrometer were multiple, including reliability issues with the 120HR, aggravated by the lack of spare parts (electronic boards, mechanical items) allowing to fix major failures and the discontinued support from its manufacturer. Regarding the new instrument, we can point out one of its major advantages: its operation speed, with observations now recorded three times quicker, with identical spectral performance, thanks to the new electronics and computer. This is a critical advantage when tracking diurnal variations, or to cover all optical filters on a short sunny day.

As it was not possible to operate the 120HR and 125HR simultaneously, their intercalibration will be performed through other ways. This will be ensured using laboratory cell spectra (HBr and N₂O cells), atmospheric features for gases with well-known abundances (N₂ and CO₂), and by exploiting parallel observations performed by a Vertex 80V mid-resolution

spectrometer, thanks to a close collaboration with UBremen. This instrument is operated on the second floor of the Sphinx observatory since May 2024. It has recorded spectra in parallel with the 120HR, then the 125HR, further bridging the gap between the 120 and 125HR. Analysis of these spectra is ongoing for a suite of selected targets.

Another step consisted in the replacement of our in-house liquid nitrogen (LN2) system needed to cool the detectors with a commercial microdosing equipment from Norhof. This equipment has proved highly satisfactory, enabling two detectors to be cooled remotely with drastically reduced LN2 consumption.

The update of the solar tracker is planned for spring 2025. At that time, all the FTIR instrumentation will have been renewed, giving a new momentum and opening perspectives for the continuation of our very long-term monitoring effort.

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References

- Makkor, J. et al. “Digitization and calibration of historical solar absorption infrared spectra from the Jungfraujoch site”. *Atmos. Meas. Tech. Discuss. [preprint]* (2025). In press, 2025. DOI: 10.5194/amt-2024-93.
- Migeotte, M., L. Neven, and J. Swensson. “The solar spectrum from 2.8 to 23.7 microns—part I: Photometric atlas”. *Mém. Soc. Roy. Sci. Liège Special vol 1* (1956).
- Minganti, D. et al. “Evaluation of the N₂O Rate of Change to Understand the Stratospheric Brewer-Dobson Circulation in a Chemistry-Climate Model”. *J. Geophys. Res. Atmos.* **127** (2022), 1–22. DOI: 10.1029/2021JD036390.
- Prignon, M. et al. “Stratospheric fluorine as a tracer of circulation changes: comparison between infrared remote-sensing observations and simulations with five modern re-analyses”. *J. Geophys. Res. Atmos.* (2021). DOI: 10.1029/2021JD034995.

Internet data bases

<http://girpas.uliege.be/>
www-air.larc.nasa.gov/pub/NDACC/PUBLIC/stations/jungfraujoch/hdf/ftir/
www-air.larc.nasa.gov/pub/NDACC/PUBLIC/RD/jungfraujoch/hdf/ftir/

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Refereed journal articles

- Müller, J.-F. et al. “Bias correction of OMI HCHO columns based on FTIR and aircraft measurements and impact on top-down emission estimates”. *Atmos. Chem. Phys.* **24**, 4 (2024), 2207–2237. DOI: 10.5194/acp-24-2207-2024.
- Pardo Cantos, I., E. Mahieu, et al. “First HFC-134a retrievals from ground-based FTIR solar absorption spectra, comparison with TOMCAT model simulations, in-situ AGAGE observations, and ACE-FTS satellite data for the Jungfraujoch station”. *J. Quant. Spectrosc. Radiat. Transf.* **318** (2024), 108938. DOI: 10.1016/j.jqsrt.2024.108938.
- Wizenberg, T. et al. “Measured and Modeled Trends of Seven Tropospheric Pollutants in the High Arctic From 1999 to 2022”. *J. Geophys. Res. Atmos.* **129**, 12 (2024). DOI: 10.1029/2023JD040544.
- Zhou, M. et al. “Recent Decreases in the Growth Rate of Atmospheric HCFC-22 Column Derived From the Ground-Based FTIR Harmonized Retrievals at 16 NDACC Sites”. *Geophys. Res. Lett.* **51**, 22 (2024), 1–10. DOI: 10.1029/2024GL112470.

Conference papers

- Makkor, J. et al. “Digitization and use of historical spectra from 1950/51 for the retrieval of various trace gases from the Jungfraujoch site (46.55°N, 7.98°E, 3580m)”. *EGU General Assembly 2024*. Vienna, Austria, 2024, 14–19 April 2024. DOI: 10.5194/egusphere-egu24-11539.

Pardo Cantos, I. and E. Mahieu. “Determination of time series of halogenated gases relevant to the Montreal Protocol using ground-based, satellite and model data”. *European Research Course on Atmospheres*. Grenoble, France, 2024, 14 January–3 February. URL: <https://hdl.handle.net/2268/310904>.

Theses

Callewaert, S. “Application of the WRF-GHG model for the interpretation of ground-based observations of atmospheric greenhouse gas concentrations”. PhD thesis. Université de Liège, 2024, pp. 1–167.

Yombo Phaka, R. “First MAX-DOAS observations of tropospheric NO₂ and H₂CO in Central Africa: impact on air quality and validation of the TROPOMI satellite instrument”. PhD thesis. Université de Liège, 2024, pp. 1–180.

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