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 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. At the end of 2021, we reached 38 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 to produce high-level geophysical parameters and valorise them.

In 2021 and given the extension of the covid-19 pandemic and related cross-border travel restrictions, it was only possible to collect remote observations. Altogether and before any averaging, only about 1500 high resolution infrared solar spectra were collected on 53 days. Indeed, a new failure of our KVM adapter prevented the recording of spectra during the first half of the year. The shortage of electronic components substantially delayed the production and delivery of a new device which entered operation before the end of June thanks to the vital local support of the custodians.

The systematic 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). It is also worth noting that long-lived tracers can also be used to provide useful insights regarding global atmospheric circulation.

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.

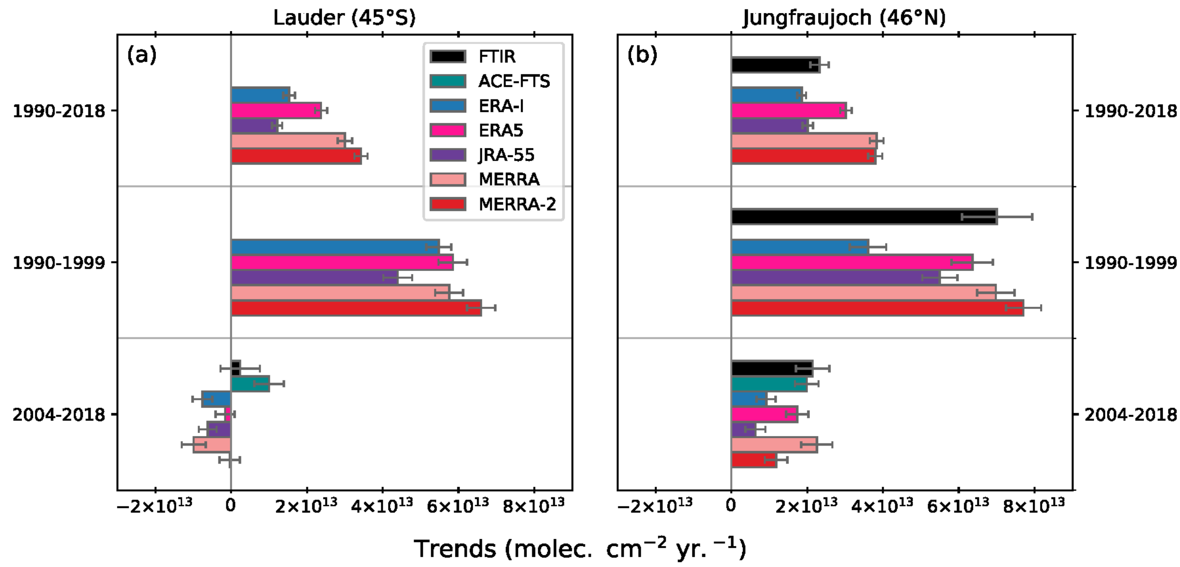

Figure 1. Inorganic fluorine trends above Lauder (a) and Jungfraujoch (b) computed for ground-based FTIR and ACE-FTS observations as well as for BASCOE CTM simulations driven by five meteorological reanalyses. See the colour key for their identification. Horizontal thin bars corresponds to the 95% uncertainty range around the estimated trend values (see Prignon et al., 2021).

2. Research highlights

Buckinx (2021) investigated the impact of the covid-19 lockdown on air quality at the Jungfraujoch station. To that effect, the time series of a suite of eleven trace constituents were analysed to determine whether their abundances were significantly altered during 2020 in comparison with the previous 10 or 20 years. A first step involved the detrending of the preceding years, assuming a function combining a linear trend and a third order Fourier series, using a bootstrap resampling tool (Gardiner et al., 2008). The comparison between the 2020 data and the detrended multiyear times series showed evidence for a significant reduction in C_2H_2, C_2H_4, C_2H_6, CH_3OH, H_2CO, HCOOH and tropospheric ozone, the latter results being in agreement with the study of Steinbrecht et al. (2021). In contrast, no clear signal was found for the remaining targets of this study, namely CH_4, CO, HCN and NH_3. Taken together, this leads to the preliminary conclusion that the drop in tropospheric ozone concentrations was driven by a decrease of the volatile organic compounds (VOCs) rather than by CH_4 and CO, two of its important precursors. Still, it remains to be determined to what extent the meteorological conditions also affected the air pollutants amounts.

Using FTIR time series of inorganic fluorine above Lauder (45°S) and Jungfraujoch (46.5°N), supplemented with satellite data collected by the ACE-FTS instrument, and model simulations performed with the BASCOE chemistry transport model (CTM) implementing all major meteorological reanalyses, Prignon et al. (2021) investigated stratospheric circulation change over the last two decades. This study notably concluded that inorganic fluorine (F_y) has been accumulating less rapidly in the Southern Hemisphere than in the Northern Hemisphere over the last 15 years, as obvious from Figure 1. Furthermore, the 5-7 year interhemispheric variability depicted in Strahan et al. (2020) using observations and simulations of the HCl and HNO_3 stratospheric tracers is confirmed here by all the F_y datasets involved. Last but not least, it is worth mentioning that long-term F_y time series are of primary importance for the evaluation of the fulfilment of the Montreal Protocol on substances that deplete the ozone layer, not only as a proxy of inorganic chlorine, but also to track the build-up of stratospheric fluorine resulting from the photolysis and oxidation of the hydrofluorocarbons (HFCs), these manmade halocarbons being now targeted by this international treaty following the Kigali Amendment of 2016.

Peroxyacetyl nitrate ($CH_3COO_2NO_2$, or PAN) is the main atmospheric reservoir of NO_x ($NO+NO_2$). PAN is characterized by a lifetime that can reach several months in the cold upper troposphere, allowing the long-range transport of NO_x. The subsequent thermal decomposition of PAN can release NO_x, potentially leading to the efficient formation of tropospheric ozone far from the polluted regions of emission. The satellite community has progressively developed nearly global PAN products, but these suffer from the poor availability of concurrent observations, preventing their proper validation. This was one of the motivations that led us to develop a retrieval strategy for PAN from ground-based FTIR solar spectra. First multi-year FTIR datasets have been derived for PAN from the Jungfraujoch and a few other NDACC remote stations. They were successfully compared with GEOS-Chem model simulations as well as with IASI satellite measurements performed from aboard the Metop platforms, revealing significant seasonal modulations and a possible underestimation of the accumulation of PAN in the Arctic atmosphere. More information on these first comparisons and on the perspective they open can be found in Mahieu et al. (2021).

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