

Name of research institute or organization:

**Institut d'Astrophysique et de Géophysique, Université de Liège**

Title of project:

High resolution, solar infrared Fourier Transform spectrometry. Application to the study of the Earth atmosphere

Project leader and team:

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Project description:

Contribution to the long-term monitoring of the Earth's atmosphere has remained the central activity of the Liège group. Regular observations carried out at the Jungfraujoch with our two high-performance Fourier-transform infrared (FTIR) spectrometers allow to derive abundances of more than 25 constituents affecting our climate and monitored in the frame of the Kyoto protocol ( $N_2O$ ,  $CH_4$ ,  $CO_2$ ,  $SF_6$ ...), related to the erosion of the ozone layer in the stratosphere ( $HCl$ ,  $ClONO_2$ ,  $HNO_3$ ,  $NO$ ,  $NO_2$ ,  $HF$ ,  $COF_2$ ,  $O_3$ ,  $CCl_2F_2$ ,  $CHClF_2$ ,  $CCl_3F$ ...), or altering the oxidization processes in the troposphere ( $CO$ ,  $C_2H_2$ ,  $C_2H_6$ ,  $OCS$ ,  $HCN$ ,  $H_2CO$ ...). The resulting databases allow the determination of the short-term variability, seasonal modulations, as well as long-term changes affecting most of these species.

In the frame of a permanent development of the instruments, numerous improvements to the remote operation of the Bruker FTIR spectrometer have been installed, including remotely operated mirrors protection flaps, cameras and a new liquid nitrogen detector cooling system. These hardware oriented tasks have been complemented by the development of the corresponding application-specific control software, enabling remote access to all the necessary parameters of the spectrometer, sun tracker and protections. In mid-October 2008, for the first time, we have been able to remotely observe with the Bruker FTIR spectrometer at the Jungfraujoch and to successfully record solar spectra from Liège.

During 2008, observers spent 248 days at the Jungfraujoch. Good weather conditions enabled solar observations on 108 days, including 12 days with spectra remotely recorded from Liège. Regular measurements with a sealed cell containing  $HBr$  gas have also been realized, in order to characterize the instrumental line shape. This objectively warrants that the observations are performed consistently at the highest level of quality/performance.

In addition to the constituents routinely retrieved, here are a few examples where emphasis was placed in 2008:

### **Water vapour**

In the frame of GAW-CH<sup>1</sup> and AGACC<sup>2</sup> projects, preliminary investigations have been performed to derive total and partial vertical abundances of water vapour from

<sup>1</sup> Swiss Global Atmosphere Watch

<sup>2</sup> Belgian Advanced exploitation of Ground-based measurements for Atmospheric Chemistry and Climate Applications, <http://www.oma.be/AGACC/Home.html>

Jungfraujoch solar observations, including historical atmospheric spectra recorded with a grating spectrometer from 1968 to the late 80s.

Numerous water vapour lines are present in the spectral range accessible by the Jungfraujoch FTIR spectrometers. One part of the work consists in finding appropriate H<sub>2</sub>O lines, temperature-insensitive, free of interferences... in different spectral regions. A selection of 63 micro-windows, distributed between 700 and 4300 cm<sup>-1</sup>, has been retained. Combination of several micro-windows with strong and weak lines should allow to increase the quantity of information retrieved from the FTIR spectra, in particular the information characterizing the vertical distribution of water vapour.

In the case of the grating spectra, the narrow spectral domains recorded for regular atmospheric trend studies contain very few H<sub>2</sub>O lines and do not provide many possibilities in the lines selection. Figure 1 shows an example of the HF windows recorded in 1976 with the Jungfraujoch grating spectrometer.

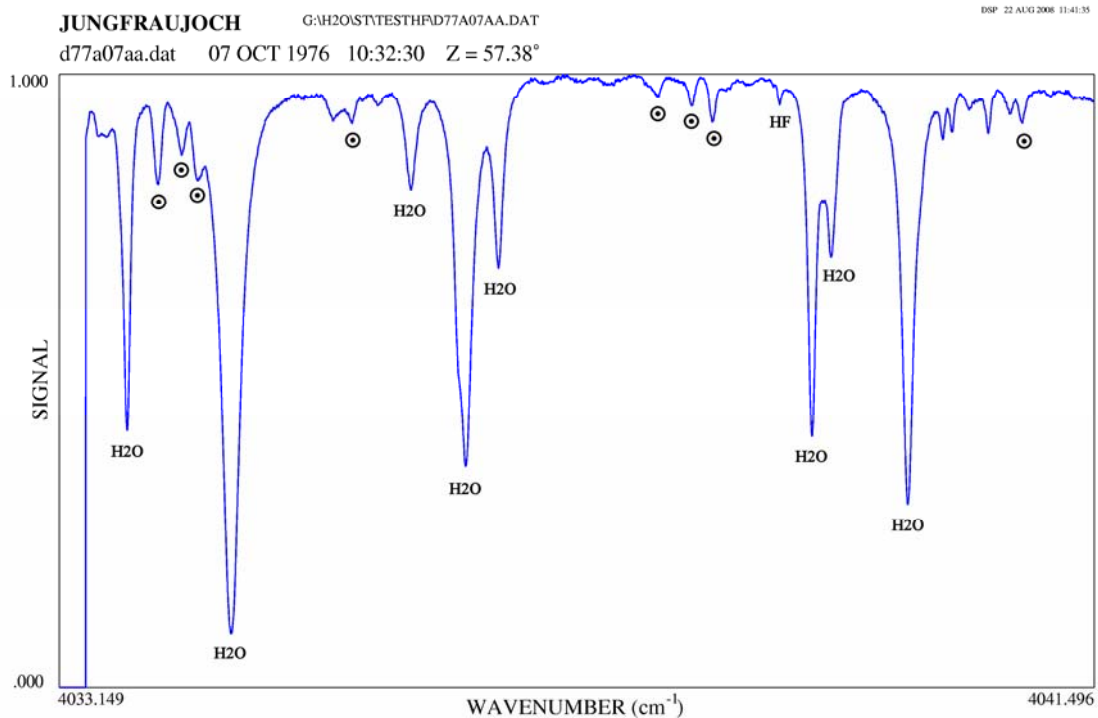


Figure 1. Example of a spectrum recorded on 7 October 1976 at the Jungfraujoch with the Liège grating spectrometer, in the region of the R1 line of HF. Several H<sub>2</sub>O lines recorded simultaneously with HF allow to derive water vapour contents during that day.

One major difficulty associated to this work will be to carefully intercalibrate the various H<sub>2</sub>O lines selected for retrieval in different spectral regions, because the spectroscopic parameters of these lines are not always known with high precision and biases regularly exist between intensities of lines from different bands. Fortunately, the successive Jungfraujoch spectrometers were always operated with some overlap and it should therefore be possible to intercalibrate the lines in different spectral domains by using observations simultaneously performed by two instruments.

A new retrieval algorithm, PROFFIT, developed at IMK (Karlsruhe) has also been implemented. This program can perform spectra inversion on a linear or on a

logarithmic VMR scale, this later being more adapted for H<sub>2</sub>O retrievals, because of the huge vertical gradient of water vapour in the atmosphere. Figure 2 illustrates an example of a 7-years H<sub>2</sub>O total columns time series derived from FTIR spectra with the PROFFIT code.

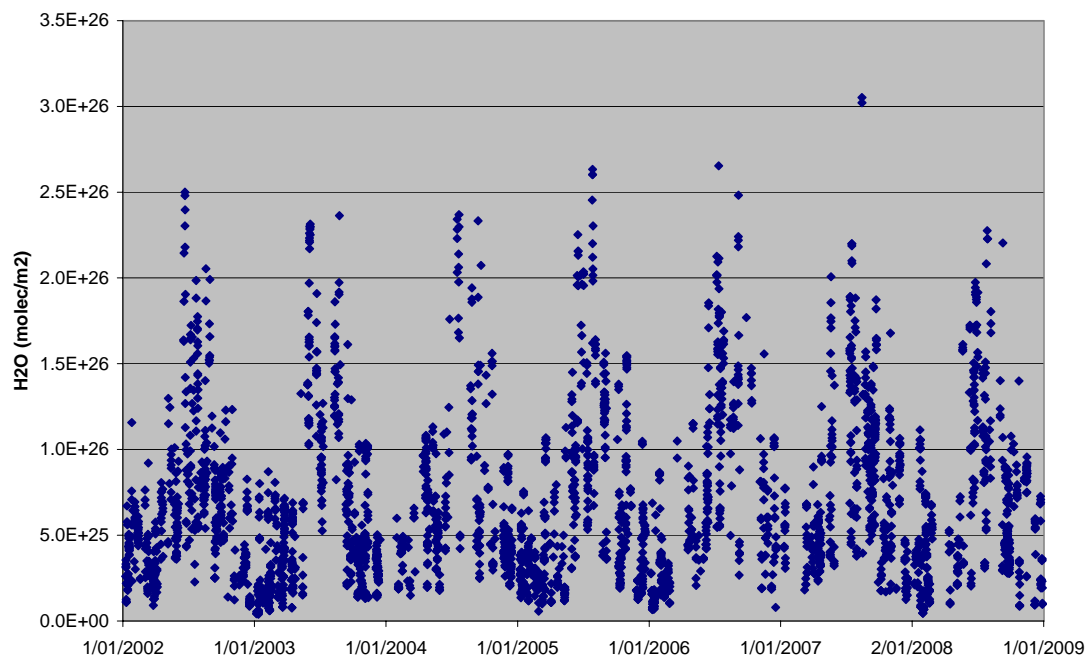


Figure 2. Water vapour total column above Jungfraujoch, from 2002 to 2008, derived with the PROFFIT retrieval tool, from a 6 micro-windows combination (1110.00-1113.00, 1117.30-1117.90, 1120.10-1122.00, 1196.00-1200.40, 1220.50-1221.50 and 1251.75-1253.00 cm<sup>-1</sup>). Note the huge difference between winter and summer water vapour contents (up to a factor of 50 !)

### Carbon monoxide isotopologues

A new approach has been developed to retrieve carbon monoxide isotopologues <sup>12</sup>CO and <sup>13</sup>CO from FTIR spectra. Six unsaturated lines from the (2-0) band of <sup>12</sup>CO have been selected in the 4200-4300 cm<sup>-1</sup> spectral interval, essentially on the basis of minimum interference by CH<sub>4</sub>, HDO and the solar spectrum. For <sup>13</sup>CO, four lines from the 2055-2155 cm<sup>-1</sup> range have been kept, with interferences by O<sub>3</sub> and the solar spectrum only. Information content analysis of this micro-windows setting indicates that, typically, 2.7 and 2.1 pieces of information may be derived, with errors on tropospheric columns of 7 and 6 %, respectively for <sup>12</sup>CO and <sup>13</sup>CO.

Since the 2 isotopologues are recorded with different optical filters, simultaneous measurements are not available and only data taken within maximum 3.5 hours interval have been considered. Mean tropospheric VMRs have been compared, monthly mean VMR relative differences (defined as 200 x (<sup>13</sup>CO-<sup>12</sup>CO)/(<sup>13</sup>CO+<sup>12</sup>CO), in %) are plotted versus the year fraction in Figure 3, for all available data since 1997. Among striking features, we notice the significant seasonal modulation, with a maximum in the middle of the year; this probably reflects significant changes in specific sources and/or sinks of the isotopologues. We also see interannual change, with possible impact of strong biomass burning events, with e.g. 1998 showing relative differences above the mean signal. However, these data are still preliminary, further investigations will be performed in 2009, including comparison with numerical models and results available from the scientific literature.

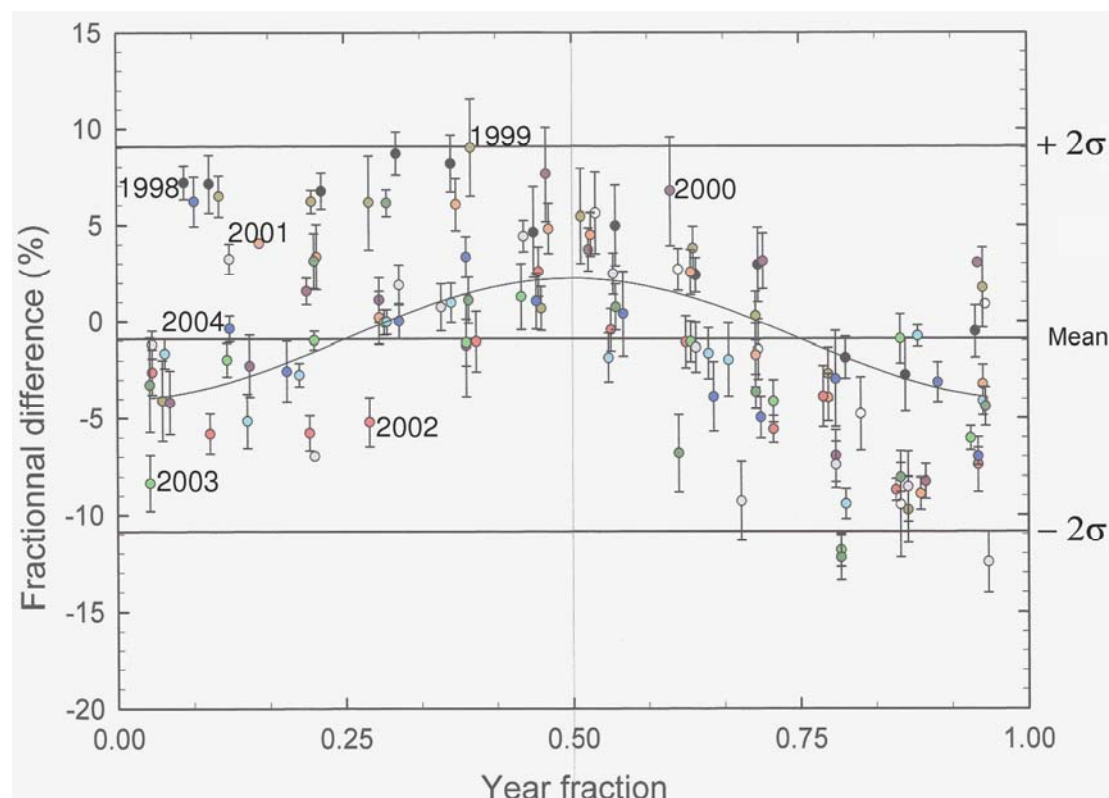


Figure 3. Seasonal modulation of the monthly mean relative differences between  $^{13}\text{CO}$  and  $^{12}\text{CO}$  isotopologues, in the troposphere above the Jungfraujoch station.

### HCFC-142b

Among the replacement products for the CFCs, one of the most commonly used is the HCFC-142b ( $\text{CH}_3\text{CClF}_2$ ), with applications in refrigeration and foam blowing. This has resulted in significant release to the atmosphere, and hence to large growth rates, e.g. 4.2 %/yr for the period 2003-2004 (WMO, 2006). At present times, it is the third most abundant HCFC in the atmosphere, after HCFC-22 and HCFC-141b.

HCFCs, which are ozone depleting substances, since they are relatively long-lived chlorine-bearing source gases, have recently started to be regulated under the Montreal Protocol, with 100 % phase-out production actually settled for 2029. It is therefore important to monitor their accumulation in the atmosphere, in order to make sure that the observed growth rates are consistent with the reported emissions. In addition, these species are also potent greenhouse gases, with several absorption features in the infrared.

Relatively broad HCFC-142b absorption features have been identified in the Jungfraujoch infrared observations. In all cases, the absorptions are very weak, even in the most recent observations. Three microwindows have been selected to retrieve HCFC-142b in our spectra: around 904, 967 and 1192  $\text{cm}^{-1}$ . Synthetic spectra of the corresponding intervals are reproduced in Figure 4. Although the calculations have been performed for low sun conditions (zenith angle of 85°), the total absorption for the target gas remains weak, with values of 0.36, 0.12 and 0.47 %, to be compared to total absorptions of 7.1, 11.1 and 26.6 % in the 904, 967 and 1192  $\text{cm}^{-1}$  intervals, respectively.

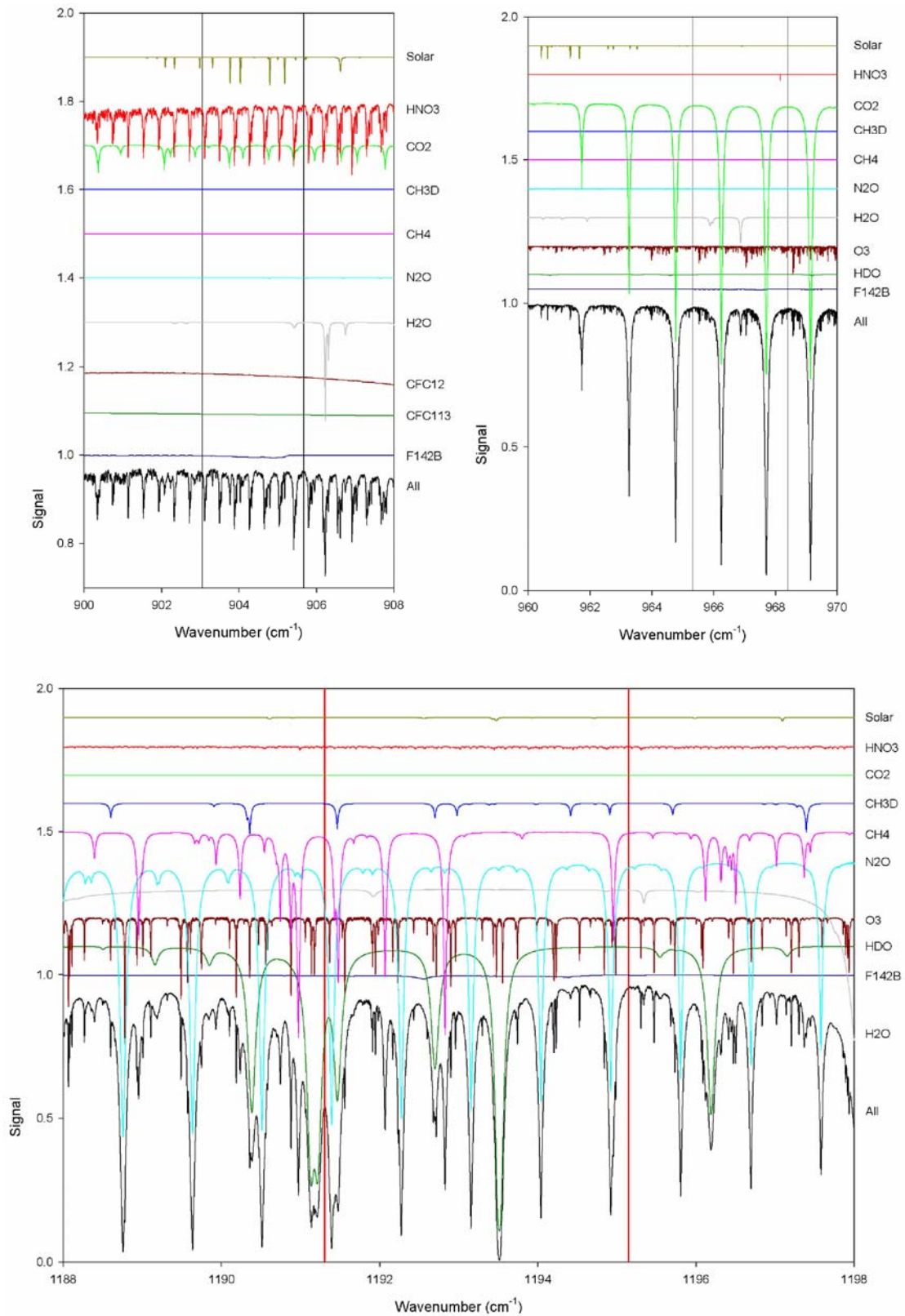


Figure 4. Synthetic spectrum calculations performed for a zenith angle of  $85^\circ$  and for the micro-windows used for the retrieval of HCFC-142b, further extended to the left and the right to allow the identification of adjacent absorptions that could influence the local continuum. Absorptions by individual gases and total spectra are reproduced and identified on the right of each frame. Traces have been offset for clarity.

In order to reach sufficient information content despite these unfavourable and challenging conditions, we have performed the retrievals using simultaneous fits of the three microwindows, further combining several consecutive observations of the same day.

Jungfraujoch observations collected over the 2002-2007 time period have been systematically fitted to produce a consistent time series. Monthly mean total columns have been computed, they are ranging from about  $1.0E14$  to  $2.5E14$  molec./cm<sup>2</sup>. A linear fit to the whole time series indicated a mean trend slightly lower than 8 %/yr. This value is more than two times larger than expected, and we have tried to identify possible causes for this bias. The most likely one is the interference by HFC-134a, which also absorbs in the 1192 cm<sup>-1</sup> micro-window. This species is currently accumulating at a very high rate in the atmosphere (10 %/yr in over 2004-2006). Unfortunately, pseudolines for HFC-134a are actually unavailable, preventing us to perform retrievals accounting for this interference. It is therefore not possible to validate our approach – and hence to perform reliable HCFC-142b retrievals – until these pseudolines are available.

Key words:

Earth atmosphere, climate change, greenhouse gases, ozone layer, long-term monitoring, infrared spectroscopy

Internet data bases:

<ftp://ftp.cpc.ncep.noaa.gov/ndacc/>, <http://www.nilu.no/nadir/>

Collaborating partners/networks:

Main collaborations: IASB (Institut d'Aéronomie Spatiale de Belgique) / NDACC (Network for the Detection of Atmospheric Composition Change, previously NDSC; <http://www.ndacc.org/>) / GAW-CH / partners of the EC-project HYMN (<http://www.knmi.nl/samenw/hymn/>) and GEOMON (<http://geomon.ipsl.jussieu.fr/>) / NASA Langley Research Center / ACE science team / NASA JPL / University of Oslo / EMPA / University of Leeds / IMK (Forschungszentrum Karlsruhe) / satellite experiments: IASI, AURA, OMI, ACE, ENVISAT / ...

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