

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:

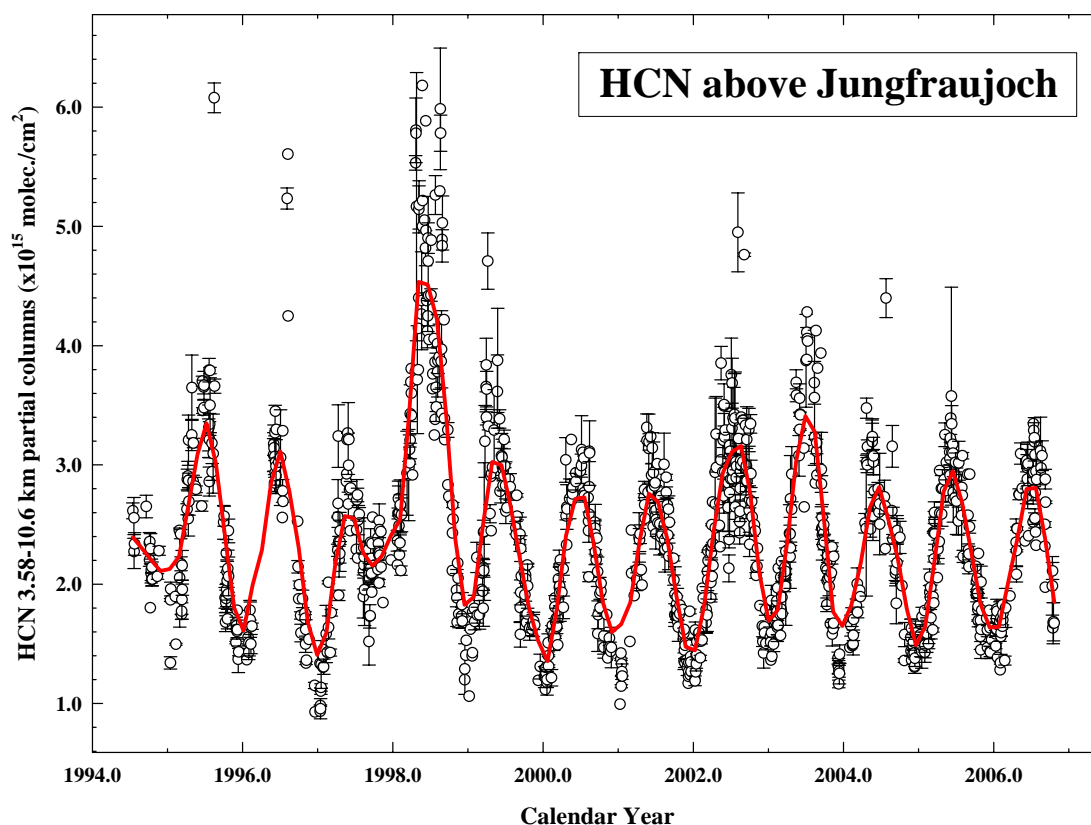
The main activity of the Liège group at the Jungfraujoch was the continuation of the long-term monitoring of the Earth atmosphere. The observations achieved by the two high-performance Fourier-transform infrared (FTIR) spectrometers allow to routinely derive abundances of more than 20 constituents related to the erosion of the ozone layer in the stratosphere (HCl, ClONO<sub>2</sub>, HNO<sub>3</sub>, NO, NO<sub>2</sub>, HF, COF<sub>2</sub>, O<sub>3</sub>, CCl<sub>2</sub>F<sub>2</sub>, CHClF<sub>2</sub>, CCl<sub>3</sub>F...), affecting our climate and monitored in the frame of the Kyoto protocol (N<sub>2</sub>O, CH<sub>4</sub>, CO<sub>2</sub>, SF<sub>6</sub>...) or altering the oxidization processes in the troposphere (CO, C<sub>2</sub>H<sub>2</sub>, C<sub>2</sub>H<sub>6</sub>, OCS, HCN, H<sub>2</sub>CO...). The resulting databases allow the determination of the short-term variability, seasonal modulations, as well as long-term changes affecting most of these species.

During 2006, observers spent 225 days at the Jungfraujoch. Good weather conditions enabled solar observations on 102 days. Regular measurements with a sealed cell containing HBr gas have also been realized, intended to characterize the instrumental line shape and check the good quality of the spectra.

For a number of the species listed above, a complete re-analysis of the archived spectra is currently under way with SFIT-2, a modern retrieval algorithm that provides in most cases information on the distribution of the molecules versus altitude. This algorithm allows determining partial columns (e.g. to distinguish between tropospheric and stratospheric contents) as well as more accurate total columns.

For example, a retrieval strategy for hydrogen cyanide (HCN), using simultaneously five HCN lines and specific microwindows for the proper simulation of water vapour absorptions (the major interference in the fitted windows), has been developed, validated and applied to Jungfraujoch FTIR observations. Figure 1 shows the results of this re-analysis in terms of tropospheric partial columns.

In addition to the constituents routinely retrieved, emphasis was placed in 2006 on some gases related to the tropospheric processes, for example HCN (already mentioned above), carbonyl fluoride COF<sub>2</sub>, formaldehyde H<sub>2</sub>CO, ethylene C<sub>2</sub>H<sub>4</sub> and methane isotopologues.

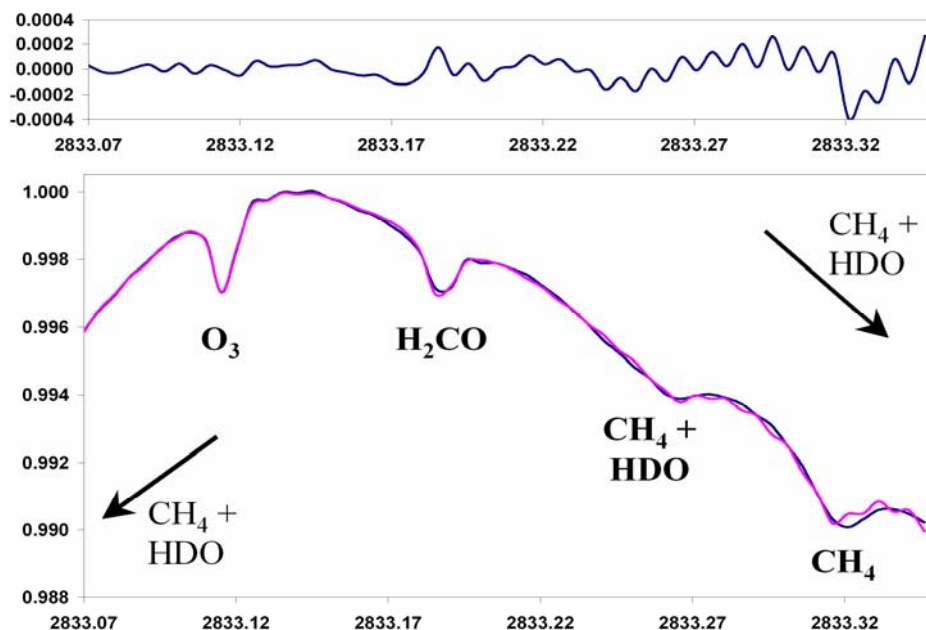


**Figure 1:** Daily-mean partial columns of HCN above the Jungfraujoch, from the ground upwards to 10.6 km and from 1994 onwards, derived from FTIR spectra. One notices very high tropospheric columns in 1998 (and to a lesser extent in 2003) correlated with documented high values of carbon monoxide resulting from important biomass burning. Also obvious is the strong seasonal variation, with maximum tropospheric columns generally observed in July.

Formaldehyde results from the incomplete combustion of carbon-containing materials. It is a prominent product of biomass burning and an important element in air quality monitoring. However, even the most favourable infrared absorptions of formaldehyde are extremely weak.

Up to now, reliable H<sub>2</sub>CO retrievals from Jungfraujoch FTIR spectra were only possible with weekly- or monthly-averaged spectra, preventing proper characterization of its variability and the production of data relevant for satellite validation. Efforts have therefore been undertaken to improve the signal-to-noise ratio of the solar observations, with the hope to be able to retrieve H<sub>2</sub>CO abundances from individual spectra, with sufficient precision. For this purpose, a tuneable optical filter covering the 2810 to 2850 cm<sup>-1</sup> spectral range has been installed in December 2005 in the Bruker FTS. The combination of this narrower interval with a larger aperture has resulted in H<sub>2</sub>CO spectra with S/N between 7000 and 15000 – an improvement by more than a factor 4 – with the resolution unchanged. This specific experimental setup has been regularly used since December 2005, allowing to record nearly 400 atmospheric spectra in 2006. Preliminary results are displayed in Figure 2 and 3.

In the Earth's atmosphere, the cycle of methane – one of the main greenhouse gases – is complex and its understanding requires a complete study of its sources and sinks.



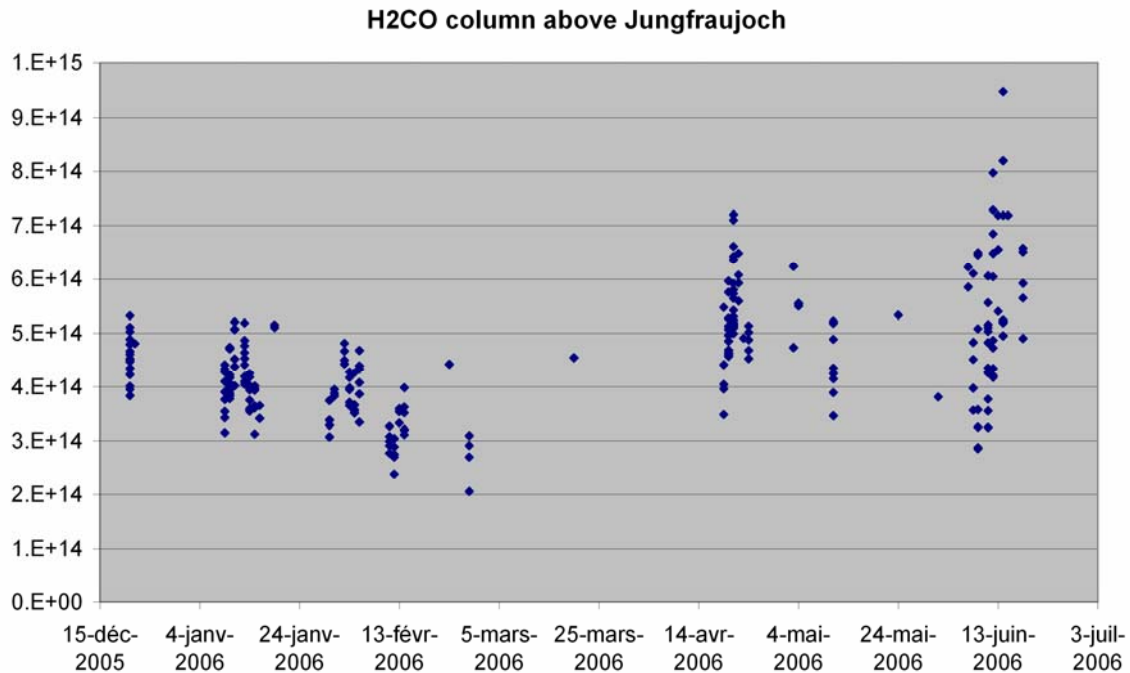
**Figure 2:** Spectral domain used for the retrieval of formaldehyde  $H_2CO$  from Jungfraujoch spectra. In the lower frame, the blue trace corresponds to a mean of 74 spectra and the magenta trace to a synthetic spectrum fitted to the observation. The upper frame shows the differences between computed and observed spectra. Main interferences are also displayed on the figure. Notice the high quality of the spectrum and the extremely faint  $H_2CO$  absorption (less than 0.2 %, for a solar elevation of  $8.2^\circ$ ).

Different processes are known to fractionate the common isotopologues of methane, and hence measuring their isotopic ratios may yield constraints on the nature of the methane sources. Preliminary retrieval strategies have been established to retrieve the isotopologues of methane  $^{13}CH_4$  and  $CH_3D$  from the Jungfraujoch FTIR spectra and the first results seem promising.

In support of the validation of the Canadian ACE-FTS spectrometer flying onboard the SCISAT-1 satellite, specific observational campaigns were organized at the Jungfraujoch in 2006, to record as many coincident measurements as possible. Amongst about 40 occultations of ACE-FTS that occurred in 2006 in the vicinity of Jungfraujoch, we succeeded in obtaining coincident measurements for half of them.

Our group participated to the international conference “Research at Jungfraujoch – Top of Science”, held at Interlaken on September 11-14, 2006, in celebration of the 75<sup>th</sup> anniversary of the High Altitude Research Station Jungfraujoch. Four posters were presented (“The evolution of inorganic chlorine above the Jungfraujoch station: an update”. “Recent evolution of atmospheric OCS above the Jungfraujoch station: Implications for the stratospheric aerosol layer”. “Solar spectroscopy 1950 - ...” “M. Migeotte observations in 1950-1951.”) as well as an oral presentation by R. Zander and S. Reimann (“Long-term monitoring of greenhouse and ozone-depleting gases at Jungfraujoch”).

In May 2006, authorities from the Liège University and from the Belgian Federal Science Policy showed their interest and recognized the quality of our work, when they visited our laboratory at the Jungfraujoch.



**Figure 3:** Preliminary H<sub>2</sub>CO total columns for the December 2005 to June 2006 period. These first results indicate that: (i) minimum values of about  $3 \times 10^{14}$  molec./cm<sup>2</sup> are found in February-March; (ii) higher columns ranging up to  $8 \times 10^{14}$  molec./cm<sup>2</sup> are observed during spring; (iii) spring is characterized by higher variability.

In February 2006, Luc Delbouille, who has been during many years the leader of the group, received an award "in recognition of 50 years of outstanding achievements in solar spectroscopy and atmospheric research at the High Altitude Research Station Jungfrauoch and in grateful acknowledgement of his personal endeavours for the benefit of the Research Station and the Foundation".



Active in electronics, optics and computers, he developed the solar spectroscopy laboratory at the Jungfrauoch since 1956, designing the huge grating spectrometer that remained in operation until 1989 and the first Fourier transform spectrometer that is still in use now. Very sadly, Luc Delbouille suddenly died on September 7<sup>th</sup>, 2006 at the age of 77. His death came as a terrible shock for the members of the group and for the scientific community. He was still very active, spending every year more than 80 days at the Jungfrauoch.

Key words:

Earth atmosphere, ozone layer, greenhouse gases, climate change, 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/>) / partners of the EC-project HYMN (<http://www.knmi.nl/samenw/hymn/>) / NASA Langley Research Center / ACE science team / NASA JPL / University of Oslo / EMPA / University of Leeds / IMK (Forschungszentrum Karlsruhe) / satellite experiments: ACE-FTS, ENVISAT, MOPITT / ...

Scientific publications and public outreach 2006:

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Hase, F., P. Demoulin, A. J. Sauval, G. C. Toon, P. Bernath, A. Goldman, J. W. Hannigan, C. Rinsland, An empirical line-by-line model for the infrared solar transmittance spectrum from 700 to 5000 cm<sup>-1</sup>, *J. Quant. Spectrosc. Radiat. Transfer*, **102**, 450-463, 2006.

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