

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 performed at the Jungfraujoch by 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 oxidation 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.

During 2007, observers spent 230 days at the Jungfraujoch. Good weather conditions enabled solar observations on 106 days. 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, emphasis was placed in 2007 on some gases related to the tropospheric processes, for example hydrogen cyanide HCN , formaldehyde H_2CO , formic acid H_2CO_2 , ethylene C_2H_4 and methane isotopologues. Furthermore, systematic investigations aiming at the retrieval of information on the vertical distribution of water vapour from infrared spectra have been initiated this year.

Methane isotopologues

Due to its high warming potential and its relatively long chemical lifetime (~9 years), atmospheric methane CH_4 plays a major role in the radiative forcing responsible of the greenhouse effect. The cycle of methane is complex and his characterization requires a thoroughly study of the sources and sinks of its main isotopologue as well as the other isotopic species. Different processes are known to fractionate these isotopologues, and hence isotopic abundances are particularly useful to differentiate between various sources of atmospheric methane.

Systematic investigations have been performed to determine the best set of microwindows for the retrieval of $^{13}CH_4$ and CH_3D from the Jungfraujoch FTIR spectra.

The selected $^{13}\text{CH}_4$ absorption features are located in two different spectral regions: around 1234 cm^{-1} and from 2817 to 2926 cm^{-1} . They allow to derive total columns of $^{13}\text{CH}_4$, as well as partial columns between 3.58 and 14 km .

For CH_3D retrievals, four microwindows, ranging from 2950 to 3090 cm^{-1} , have been selected. They are fitted together to increase the collected information. This multi-windows approach has been compared with a multi-spectra approach, i.e. using only one micro-window, but fitting simultaneously several spectra recorded on the same day and at the same spectral resolution, also to increase the information content. The multi-microwindows strategy provides slightly more vertical information.

Preliminary results of the retrievals of these methane isotopologues are displayed in Figure 1 and 2.

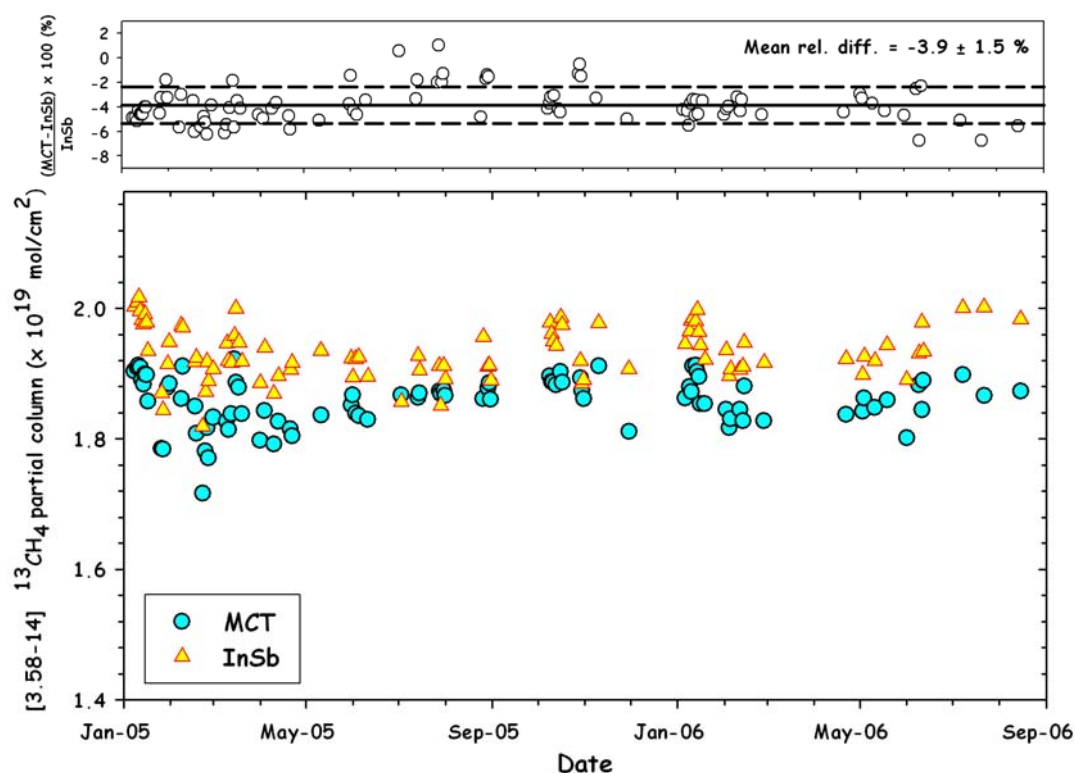


Figure 1. Comparison of $^{13}\text{CH}_4$ partial columns between 3.58 and 14 km , derived for the period January 2005 to August 2006 from two different spectral regions: one line around 1234.2 cm^{-1} (“MCT”, light blue dots) and 7 lines from 2817 to 2926 cm^{-1} (“InSb”, yellow triangles). The relative differences (top panel, $100 \times (\text{MCT} - \text{InSb}) / \text{InSb}$) reveal a significant mean bias close to 4% between both $^{13}\text{CH}_4$ partial columns time series, probably arising from inconsistencies in spectroscopic parameters.

Ethylene (C_2H_4)

Ethylene originates from a variety of anthropogenic (e.g. cars in urban areas) and natural (e.g. plants, volcanoes, forest fires) sources. Given its very short chemical lifetime combined with weak infrared absorption, C_2H_4 detection is difficult from FTIR spectra, in particular at a remote high-altitude site like the Jungfrauoch. However, during special events, e.g. under enhanced biomass burning, it is possible to clearly see ethylene absorptions in our spectra. Figure 3 presents such an example of ethylene enhancement above the Jungfrauoch, with an atmospheric contents more than 4 times the normal value.

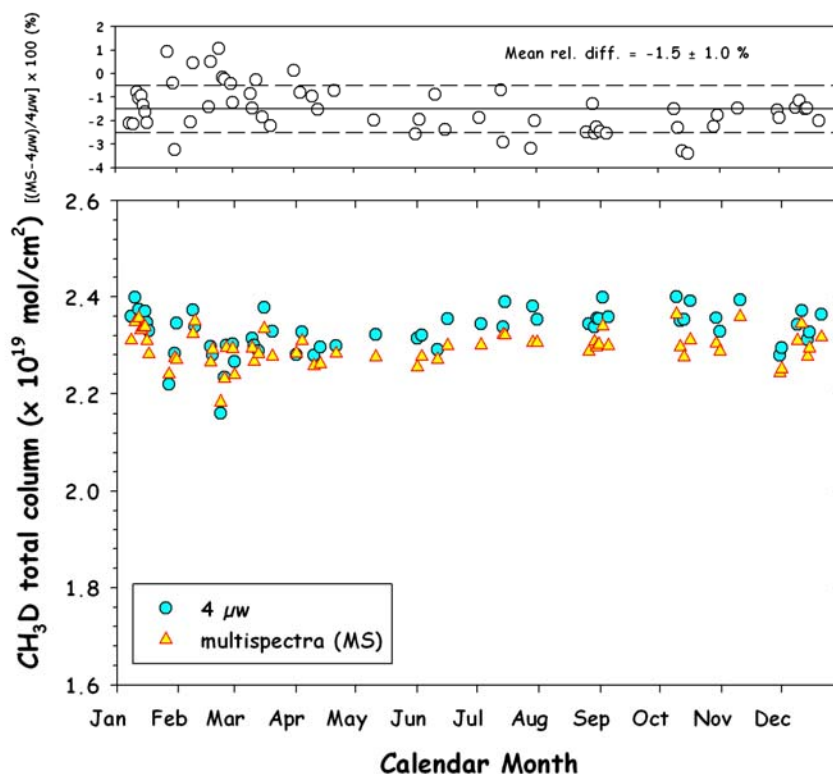


Figure 2. Daily means CH₃D total columns above Jungfraujoch for the year 2005, derived from the multi-microwindows approach [four microwindows, ranging from 2950 to 3090 cm⁻¹] (light blue dots) and from the multi-spectra approach [one microwindow around 3070.9 cm⁻¹; simultaneous fitting of several spectra recorded on the same day and at the same spectral resolution] (yellow triangles). The multi-microwindows strategy provides slightly more vertical information. Only FTIR spectra recorded at solar zenith angles between 70 and 80° have been analyzed here. Relative differences between the two retrieval strategies are plotted in the upper panel. A slight significant relative difference of 1.5 ± 1.0 % can be noticed, with the multi-microwindows approach giving higher abundances.

Water vapour

In the frame of GAW-CH¹ and AGACC² projects, preliminary investigations have been performed to derive total and partial vertical abundances of water vapour from Jungfraujoch infrared solar observations, including historical atmospheric spectra recorded with a grating spectrometer from the 1968 to the late 80s. To retrieve water vapour from the FTIR spectra, more than 60 microwindows, encompassing one or several H₂O lines and located in spectral regions ranging from 700 to 4300 cm⁻¹, have been selected and tested to develop a robust fitting strategy. At this stage, averaging kernels analysis indicates a quite good sensitivity from the altitude site (3.58 km) up to 11 km. We hope to derive 3 to 4 independent pieces of information on the water vertical profile. In the most favorable cases, the sensitivity range may extend up to 15 km.

Investigation of micro-windows containing water vapour isotopologues (H₂¹⁷O, H₂¹⁸O and HDO) has also started. The analysis of the atmospheric isotopologic water vapour composition provides valuable information on many climate, chemical and atmospheric circulation processes.

¹ Swiss Global Atmosphere Watch

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

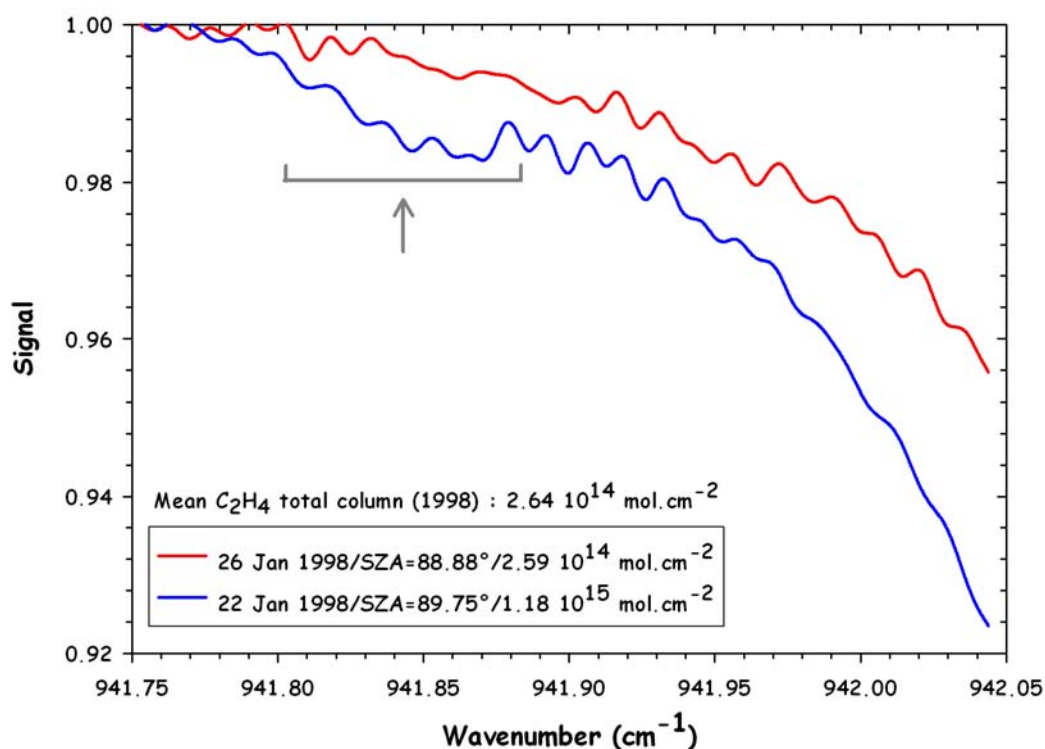


Figure 3. Example of detection of ethylene (C₂H₄) enhancement above the Jungfraujoch. The blue curve shows a spectrum recorded in January 1998 at a zenith angle of 89.8°, where the ethylene absorption is clearly visible (grey arrow). Four days later, another FTIR spectrum (red curve), obtained under similar geometric conditions, corresponds to background atmospheric ethylene contents. A total column of 1.18 10¹⁵ molec./cm² was retrieved in the first case, i.e. about four times larger than the mean total column value of 2.64 10¹⁴ molec./cm² derived for 1998.

Comparison with ACE-FTS measurements

In support of the validation of the Canadian ACE-FTS spectrometer, flying onboard the SCISAT-1 satellite, specific observational campaigns have again been organized at the Jungfraujoch in 2007, to record as many coincident measurements as possible. Amongst the 38 occultations of ACE-FTS that occurred in 2007 in the vicinity of Jungfraujoch (closer than 1000 km), we succeeded in obtaining coincident measurements for 13 of them.

Besides specific ongoing validation efforts, we have compared the abundance of different gases retrieved from the Jungfraujoch spectra with ACE-FTS data. One of these gases is hydrogen cyanide (HCN), for which we recently developed a new retrieval strategy, using simultaneously five HCN lines and specific microwindows for the proper simulation of water vapour absorptions (the major interference in the fitted microwindows, even at the high-altitude site of the Jungfraujoch). Our HCN retrievals provide good sensitivity up to 20 km, thus permitting comparisons with the ACE-FTS measurements in the upper troposphere and lower stratosphere. Figure 4 shows such a comparison, for 117 occultations recorded between March 2004 and May 2007. The two data sets are in excellent agreement in terms of absolute value, amplitude and phase of the seasonal modulation.

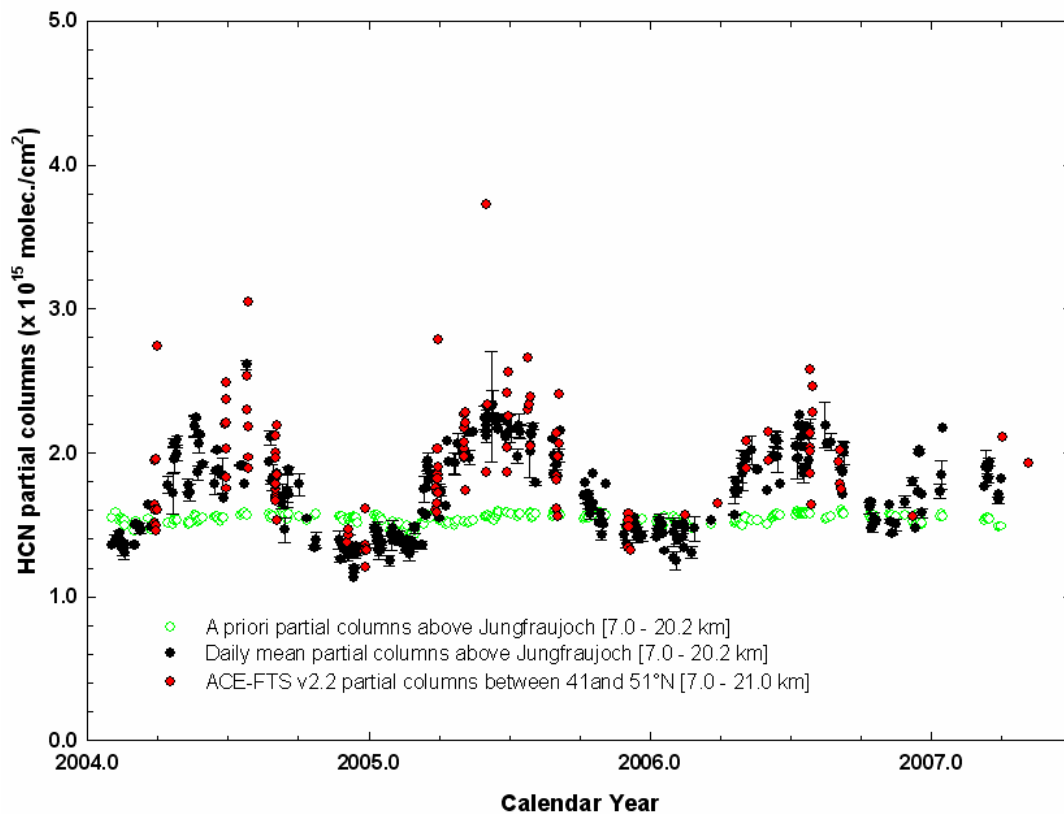


Figure 4. Comparison of HCN partial columns retrieved from Jungfrauoch spectra (black dots) or by ACE-FTS satellite instrument (red dots), for a similar altitude range (7-20 km for Jungfrauoch and 7-21 km for ACE). The 117 occultations from the ACE instrument were recorded between March 2004 and May 2007 in the 41-51°N latitudinal belt. Green circles show the Jungfrauoch a priori partial columns (i.e. the starting point for the retrieval procedure).

OMI validation

We are also participating to the validation of the OMI (Ozone Monitoring Instrument) experiment, flying on board the NASA Aura satellite. For that reason, we regularly measure formaldehyde (H_2CO) – one of the OMI products – at the Jungfrauoch, thanks to a dedicated optical filter. During 2007, 349 formaldehyde spectra were recorded. Although the infrared H_2CO absorption is extremely weak, the quality of the FTIR spectra is sufficient to deduce the formaldehyde column with a single measurement precision estimated to about 10 % and H_2CO is now becoming a standard product provided by our group.

Aura / IRWG campaign

In support of the validation of the 4 remote-sounding instruments aboard the NASA Aura satellite (MLS, TES, HIRDLS and OMI, <http://aura.gsfc.nasa.gov/instruments/>), we participated in the 2007 NDACC/IRWG³ - Aura intercomparison campaign, which took place from 15 August to 15 September 2007. During this period, we obtained 383 spectra at the Jungfrauoch, recorded over 19 days. Vertical volume mixing ratio profiles of O_3 , HNO_3 and CO and the total column of O_3 will be compared with Aura instruments measurements.

³ InfraRed Working Group of the Network for the Detection of Atmospheric Composition Change

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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