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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Jacqueline Bosseloirs, Olivier Flock, Vincent Van De Weerdt, Diane Zander

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, H_2CO_2…). The resulting databases allow the determination of the short-term variability, seasonal modulations, as well as long-term changes affecting most of these species.

Remote operation of the Bruker FTIR spectrometer has been further improved in 2009, including installation of additional cameras, protections and detectors. 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 as well as enabling total control, including UDP camera data streams, from firewall protected internet addresses. Network infrastructure of the Jungfraujoch has been completely upgraded for the coming acquisition system and its foreseen large and fast data exchange.

During 2009, Liège observers spent 221 days at the Jungfraujoch and recorded about 2500 high-resolution FTIR solar spectra on 139 different days, including 51 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 routinely retrieved constituents, here are a few examples where emphasis was placed in 2009:

Methane isotopologues

Atmospheric methane (CH_4), the second most important anthropogenic greenhouse gas, has significant chemical impacts on both the troposphere and the stratosphere. Because the different sources of methane (natural and anthropogenic like wetlands, rice paddies, termites, natural gas escape, biomass burning, etc) have distinct $^{13}C/^{12}C$ ratios, measurements of atmospheric $^{13}CH_4$ content, in addition to those of the main

isotopologue $^{12}CH_4$, can be used to investigate individual source strengths as well as their spatial and temporal distributions. Characterization of the isotopic fractionation of methane is therefore important, for example, to help models constrain estimates of the global methane budget. However, experimental data for the $^{13}C/^{12}C$ isotope ratio are sparse. In that context, a new retrieval strategy has been developed in order to retrieve $^{13}CH_4$ amounts from Jungfraujoch FTIR spectra. Corresponding information content analysis indicates that, in addition to its total column, $^{13}CH_4$ partial columns in the layer [3.58-18] km can be obtained from our measurements.
Comparisons with satellite ACE-FTS measurements have been performed and show a good agreement (see Figure 1).
Analysis of the results reveals a seasonal behaviour of the $^{13}C/^{12}C$ isotope ratio, reaching its maximum in summer (July/August) and its minimum during winter (December/January), which is consistent with previous in situ studies.
Comparisons with numerical model data are still under evaluation. Available results were presented at the EGU2009 General Assembly (Duchatelet et al., 2009) and at the ACE science team meeting (Mahieu et al., 2009).

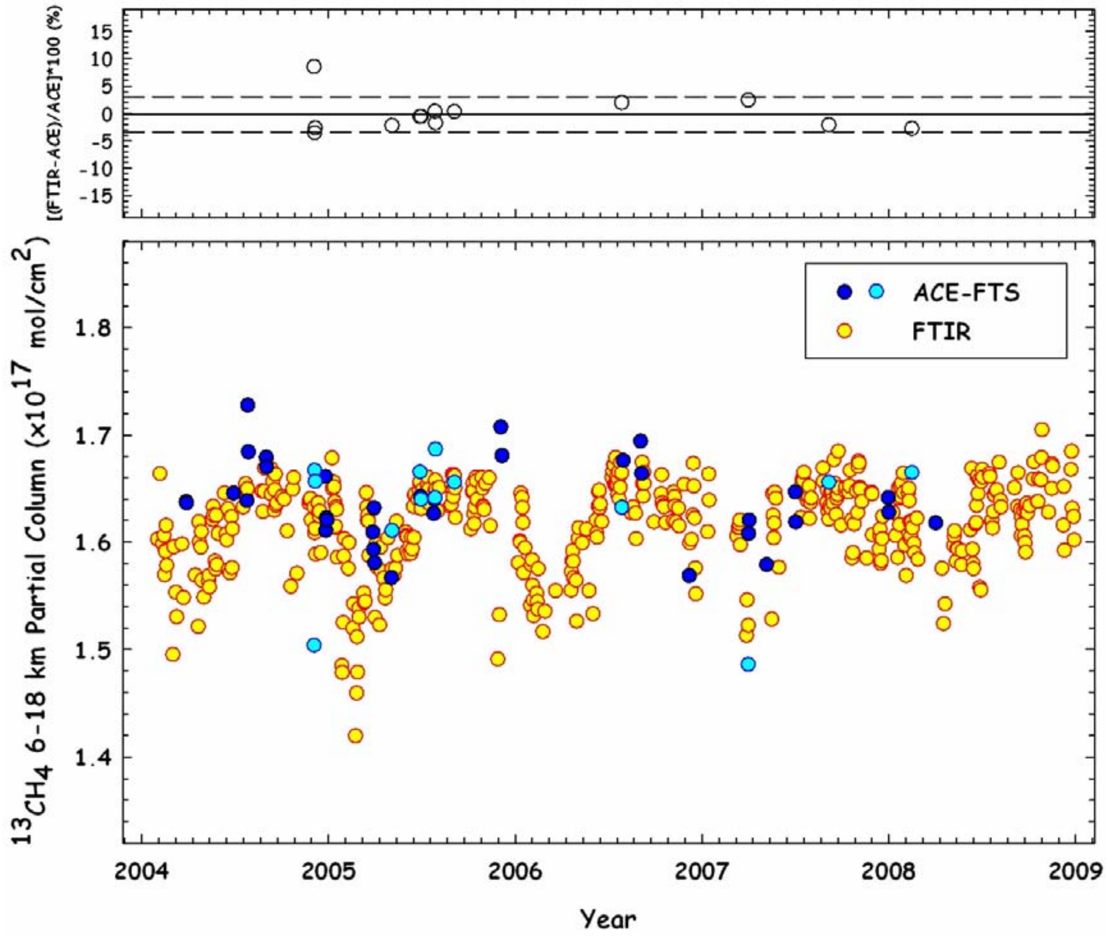

Figure 1 - Daily mean $^{13}CH_4$ 6-18 km partial columns time series as observed by the ACE-FTS space instrument (blue and light blue circles) and by FTIR spectrometry at Jungfraujoch (yellow circles). Light blue circles correspond to coincident days between satellite and ground-based data. Top panel reproduces relative differences between FTIR and ACE-FTS data (solid line: mean relative difference; dotted lines: 1-σ standard deviation on the mean).

Historical observations analysis

Work on water vapour retrievals from old grating spectra continued to progress: we are now able to analyze these spectra with the same software as we use for the modern FTIR spectra. In the last months, we concentrated our investigations on the spectral region recorded around the HF absorption at 4038.96 cm^{-1}, which also contains a few H_2O absorption lines. To test the validity of our approach, we first re-analysed the HF line, which has been measured in the 80s with a planimeter instrument. The agreement between both methods is rather good for the years 1983 to 1989, but is poorer from 1977 to 1980, suggesting that the equivalent width measurements were not consistently performed over the years. The HF total column time series for this period has therefore been significantly improved (see Figure 2).
Water vapour total columns have also been derived from the same spectra. The next step we will be to retrieve H_2O columns from the other spectral domains recorded with the old grating spectrometer.

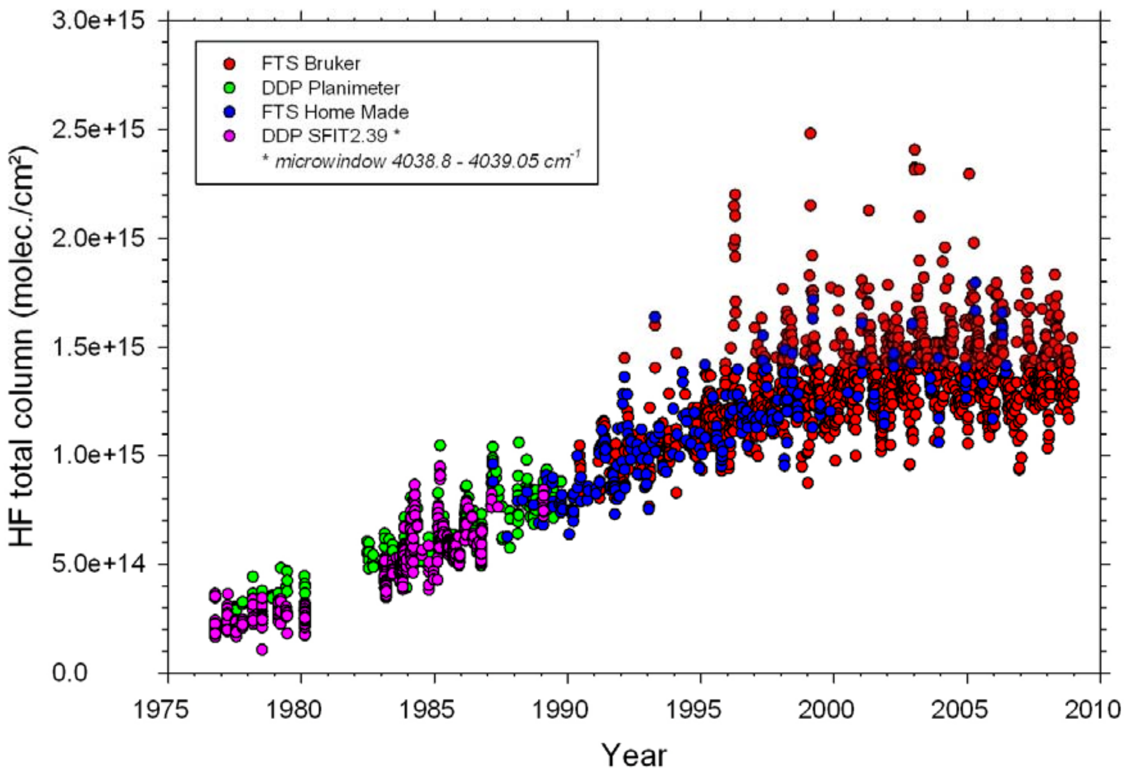

Figure 2 – Total column of fluorhydric acid HF above Jungfraujoch, derived from spectra acquired by 3 different instruments: the 7-m grating spectrometer from 1976 to 1989 ("DDP"), the home-made Fourier transform spectrometer (FTS) from 1985 to 2006, and the Bruker FTS from 1990 to now. Grating spectra have been analysed by two different methods: in the 80s with a planimeter instrument (green dots), and recently with the SFIT2 software (pink dots).

Water vapour from FTIR spectra

We have made significant progress in 2009 with respect to water vapour retrievals. In collaboration with IMK-IFU (Garmisch-Partenkirchen, Germany), a strategy to retrieve integrated water vapour (IWV) from FTIR spectra has been set up and optimized so that IWV retrievals match radiosonde characteristics. After implementation of this new strategy, IWV time series from both sites (Jungfraujoch and Zugspitze) have been consistently retrieved.
Using timely coincident spectra from the two Jungfraujoch FTIR spectrometers, we have estimated the precision on IVW retrievals to better than 0.05 mm (i.e. ~3 % of the mean IWV). Given the water vapour atmospheric variability, the differences of IWV retrieved from the two instruments show an exponential increase as a function of the temporal mismatch, already starting at $\Delta t \approx 1$ minute. This extremely high variability of water vapour with space and time puts strong constraints when it comes to define collocation criteria for intercomparisons between instruments.
Trend analysis of the two FTIR long-term IWV records shows, for the [1996 - 2008] time interval, a positive trend of 0.8 ± 0.2 mm/decade for Zugspitze, whereas Jungfraujoch data exhibit a trend of only 0.1 ± 0.1 mm/decade. The reasons for these dissimilarity still need to be clarified; part of it may be due to the horizontal distance (≈ 250 km) and/or the altitude difference (3.58 versus 2.96 km) of the two stations. It has been shown that the IWV trend patterns above continents are heterogeneous, contrarily to what is observed above the oceans, where a good correlation is found with changes in sea-surface temperatures. More details are available in Sussmann et al. (2009).
Also, we have started to use the PROFFIT algorithm developed at IMK (Karlsruhe, Germany) to perform water vapour retrievals. Its ability to perform spectra inversion on a logarithmic VMR scale is particularly well suited for H_2O and HDO retrievals, because of the huge vertical gradient of water vapour in the atmosphere.

Ozone

Ozone trends above Jungfraujoch have been updated, using a retrieval strategy that optimizes the vertical information content (Vigouroux et al., Atmos. Chem. Phys., 8, 6865-6886, 2008). Trends have been evaluated by a bootstrap resampling method, for total columns and for partial columns in 4 independent layers of the atmosphere. The following values of the ozone trends and uncertainties (95% confidence limits), in %/year, have been obtained for the period 1995-2008: total column: 0.00 ± 0.13, ground-10 km: -0.30 ± 0.29, 10-18 km: -0.15 ± 0.42, 18-27 km: 0.03 ± 0.10, 27-42 km: 0.12 ± 0.10. These results have been submitted for inclusion in the next WMO/UNEP Ozone Assessment 2010. Similar partial column products were also used for the validation of ACE-FTS and MAESTRO level 2 products (Dupuy et al., 2009).

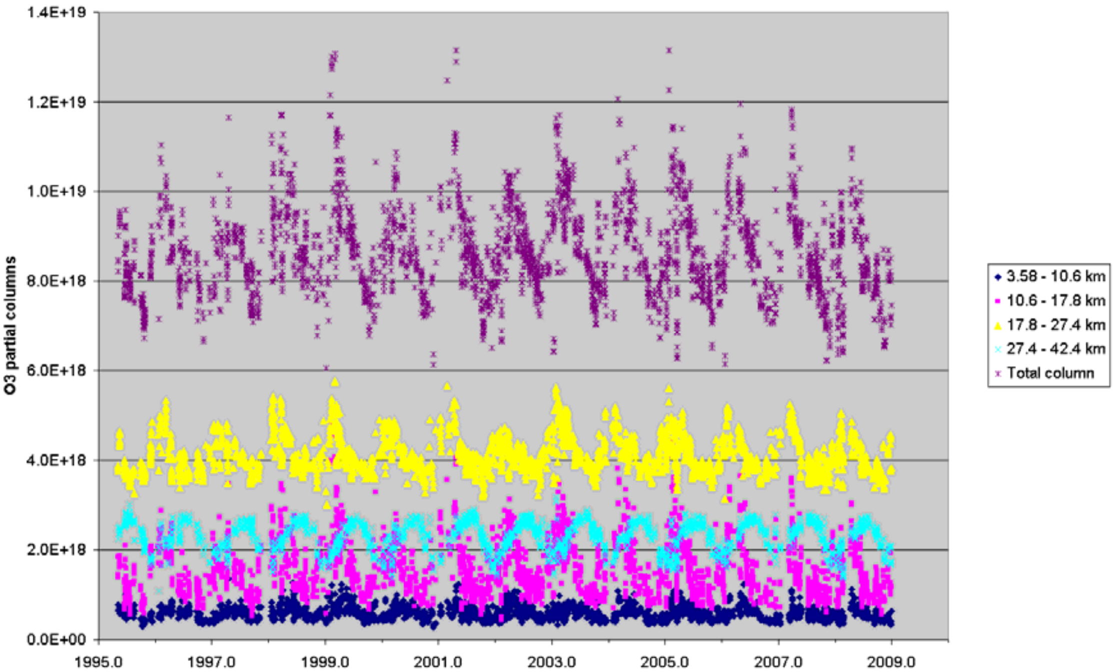

Figure 3 – Partial columns of ozone in 4 four independent layers of the atmosphere (four lower traces), and ozone total columns (upper trace).

Formic acid H_2CO_2

Total vertical column abundances of formic acid above the Jungfraujoch station have been derived for the September 1985 - September 2007 time period. The investigation was based on the spectrometric fitting of five spectral intervals, one encompassing the H_2CO_2 ν_6 band Q-branch at 1105 cm^{-1}, and four additional ones allowing to account for major temperature-sensitive or timely changing interferences by other atmospheric gases, in particular HDO, O_3, CCl_2F_2 and $CHClF_2$. The a priori H_2CO_2 vertical profile used in the retrievals presents a slope which is consistent with the very short lifetime (from a few hours to a few days) of this reactive species and commensurate with numerical values deduced from earlier studies for northern mid-latitudes. Validity of the H_2CO_2 adopted profile in the upper troposphere and up through the lower stratosphere was further assessed by comparison with ACE-FTS solar occultation measurements performed in the geographical region of interest. A major improvement in the absolute determination of the atmospheric H_2CO_2 columns has resulted from the adoption of new spectral line intensities for the ν_6 band of trans-formic acid, resulting in retrieved free tropospheric loadings being about a factor of two smaller than those derived with previous spectroscopic parameters. The main other results derived from this formic acid database indicate that the free tropospheric burden of H_2CO_2 above the Jungfraujoch undergoes important short-term daytime variability, seasonal and inter-annual modulations. For example, Figure 4 reveals an overall seasonal variation, with a broad summer maximum peaking in July, and a November-December-January minimum. This result constitutes an important finding, establishing, for the first time, the existence of a seasonal variation of the free tropospheric loading of H_2CO_2 above a continental site at northern mid-latitudes.

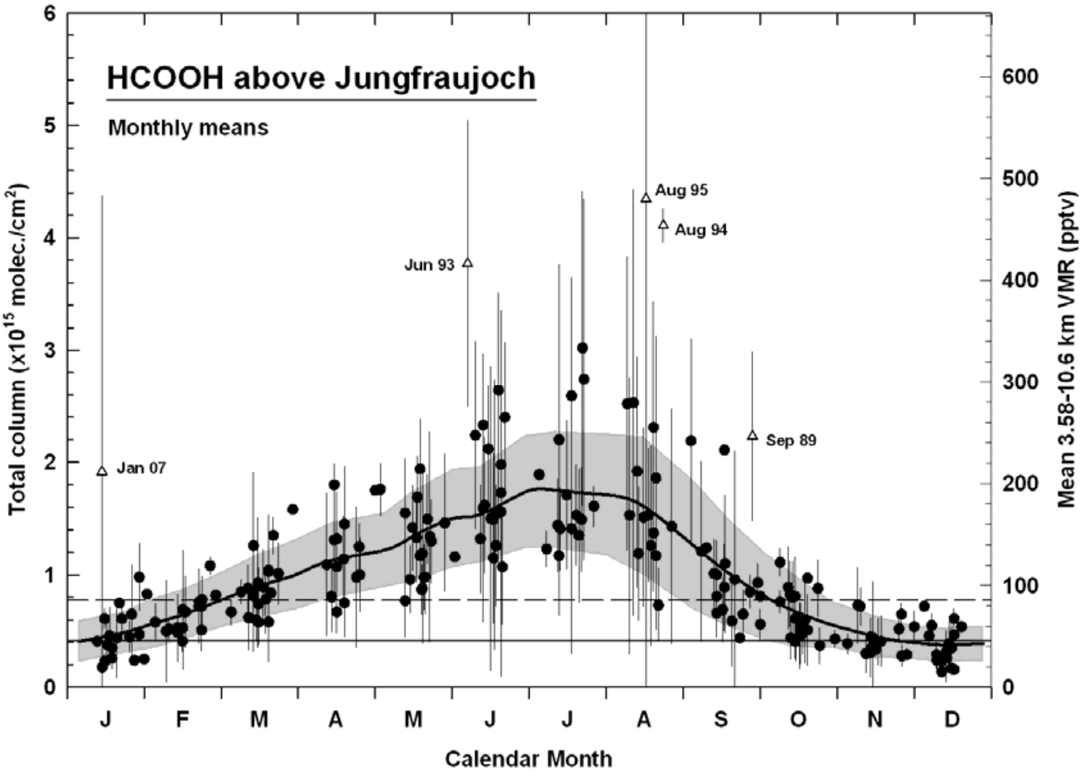

Figure 4 - Monthly mean column abundances and associated standard deviation bars, displayed on a one-year time base, with 5 obvious monthly mean outliers identified by triangular symbols. The thick curve corresponds to a running mean fit to the black filled symbols only, with a 15-day step and a 2-month wide integration time. The shaded area visualizes the 1-σ standard deviation associated to the running mean curve and reflects primarily the observed inter-annual variability of H_2CO_2. The right side scale allows converting an observed total H_2CO_2 column abundance read off of the left side scale into a mean VMR concentration (expressed in parts per trillion by volume, i.e., pptv) over the 3.58 to 10.6 km altitude range.

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/station/jungfrau/, 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 / …

Scientific publications and public outreach 2009:

The complete list of GIRPAS publications can be found at
http://girpas.astro.ulg.ac.be/girpas/publi03e.htm and
http://girpas.astro.ulg.ac.be/girpas/Communic.htm.

Refereed journal articles and their internet access

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Radio and television

Visit at the Jungfraujoch of Sabine Laruelle, Belgian Minister of Science Policy, on November 17, 2009: interview of Christian Servais and Olivier Flock, RTL-TVi television, November 18, 2009.

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