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:

Christian Servais, project leader
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Project description:

The team's objectives are essentially twofold: (i) improve the performance of the instrumentation and perform the observations, (ii) analyse the spectra in order to produce high-level geophysical parameters and valorize them.

Over the last years, significant efforts have been invested in the development and implementation of a reliable system allowing to remotely and safely control the whole instrumentation, among which the spectrometer itself, the cooling of the detectors, the suntracker and its protective lid. Nevertheless, local support from the custodians remains critical, e.g. to remove heavy snow from the lid, to fill in the LN2 Dewar flask. The remote control system led to a significant improvement of the observation statistics, taking advantage of most days with clear-sky conditions. In 2013, Liège observers were at the Jungfraujoch on 212 days. Overall, more than 2200 high resolution infrared solar spectra have been recorded on 121 days, including 37 days from a remote location.

Analysis of our spectra allows us to determine the abundance of more than 25 constituents of the Earth atmosphere, playing a role in ozone depletion, climate change or affecting the air quality. Numerous target species are therefore relevant to the Montreal Protocol on substances that deplete stratospheric ozone (e.g. CFC-11, -12, HCFC-22, CCl_4, HCl, $ClONO_2$) and/or to the Kyoto Protocol (e.g. CO_2, CH_4, N_2O, CF_4, SF_6). We present hereafter a selection of key results derived in 2013 from the scientific exploitation of our observational database.

Methanol (CH_3OH)

For the first time, a strategy providing information on the vertical distribution of methanol (CH_3OH, the second most abundant organic compound in the Earth atmosphere) has been developed and optimized for ground-based FTIR spectra. The approach is based on two wide spectral intervals (992-1008 and 1029-1037 cm^{-1}) encompassing features of the fundamental C-O stretching mode ν_8 of methanol centered at 1033 cm^{-1}. Major interferences are due to strong absorptions by ozone, secondary absorbers include less abundant ozone isotopologues, water vapor and CO_2. The retrieval is sensitive to the whole troposphere and very low stratosphere.

Methanol has been systematically retrieved from all available Bruker spectra with solar zenith angles in the 65-80º range, from 1995 onwards. We evaluated the trend of CH_3OH over these 17 years to (-1.3±2.7) x 10^{13} molec./cm^2, or (-0.2±0.4) %/yr, i.e. a non-significant value at the 2-σ level of confidence. Figure 1 shows the CH_3OH daily means over one year. A strong seasonal modulation is obvious, with minimum total columns and variability in winter (December-February), maximum values in summer (June-August), as a result of important emissions associated with plant growth. The peak-to-peak amplitude amounts to ~130 % of the CH_3OH yearly mean column. More information can be found in Bader et al. (2013).

Further work will include comparisons with *in situ* campaign-type measurements performed at Jungfraujoch by Empa in 2005 as well as with UTLS partial columns derived from ACE-FTS solar occultation observations since 2004.

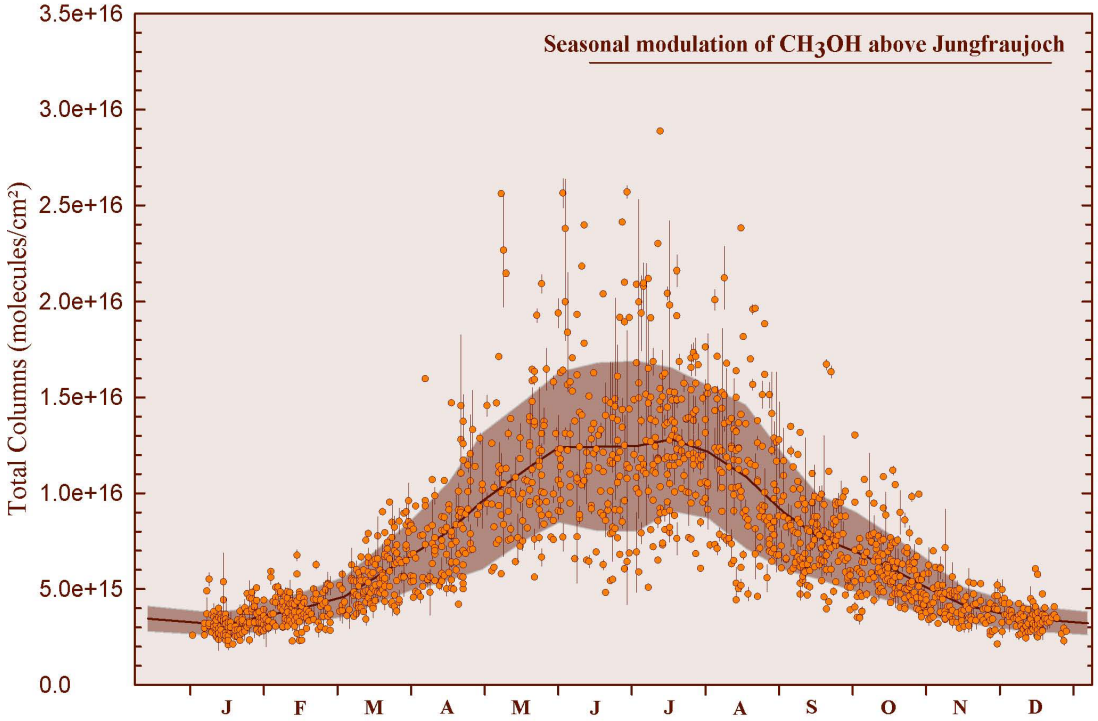

Figure 1. Methanol daily mean total columns reported over a 1 year time base. Vertical error bars represent the standard deviation around the daily means. The thick curve corresponds to a running mean fit to all data points, with a 15-day step and a 2-month wide integration time. The area corresponds to the 1-sigma standard deviation associated to the running mean curve (Bader et al., 2013).

Carbon tetrafluoride (CF_4)

Carbon tetrafluoride (CF_4) is a very strong greenhouse gas targeted by the Kyoto Protocol. It is by far the longest-lived perfluorocarbon, with a lifetime of more than 50000 years; its global warming potential amounts to 7390. It is primarily emitted during anode effect episodes occurring in aluminum production plants. It is also released by manufacturing of semiconductors and other electronic devices. The only natural source is of lithospheric origin.

We have set up an approach allowing retrieving CF_4 from ground-based FTIR spectra, using the strongest infrared features of its ν_3 band centered at 1283 cm^{-1}. A systematic analysis of the home-made and Bruker FTIR spectra from 1989 onwards has been performed. After a careful selection, we retained 3014 spectra recorded on 1272 days. The data set reveals a continuous increase of CF_4 characterized by two regimes, with yearly total column changes of (1.38 ± 0.11) and (0.98 ± 0.02) x 10^{13} molec./cm^2, for the periods 1989-1997 and 1998-2012, respectively. These changes require CF_4 emission rates equal to (15.8 ± 1.25) Gg/yr and (11.1 ± 0.2) Gg/yr for the above mentioned periods. The observed reduction is attributed to efforts of the aluminum industry to limit its emissions despite increasing production.

Our results have been compared with *in situ* and remote-sensing measurements. Figure 2 shows the various data sets, including CF_4 surface measurements performed by Empa at the Jungfraujoch. A very good agreement is observed, when accounting for the uncertainties affecting the retrieved quantities and time needed for a thorough mixing of CF_4 in the atmosphere. Figure 2 also shows that the anthropogenic contribution to the atmospheric CF_4 loading has overtaken in the late 90s the natural background level, estimated at ~35 ppt. See Mahieu et al. (2013a) for more details.

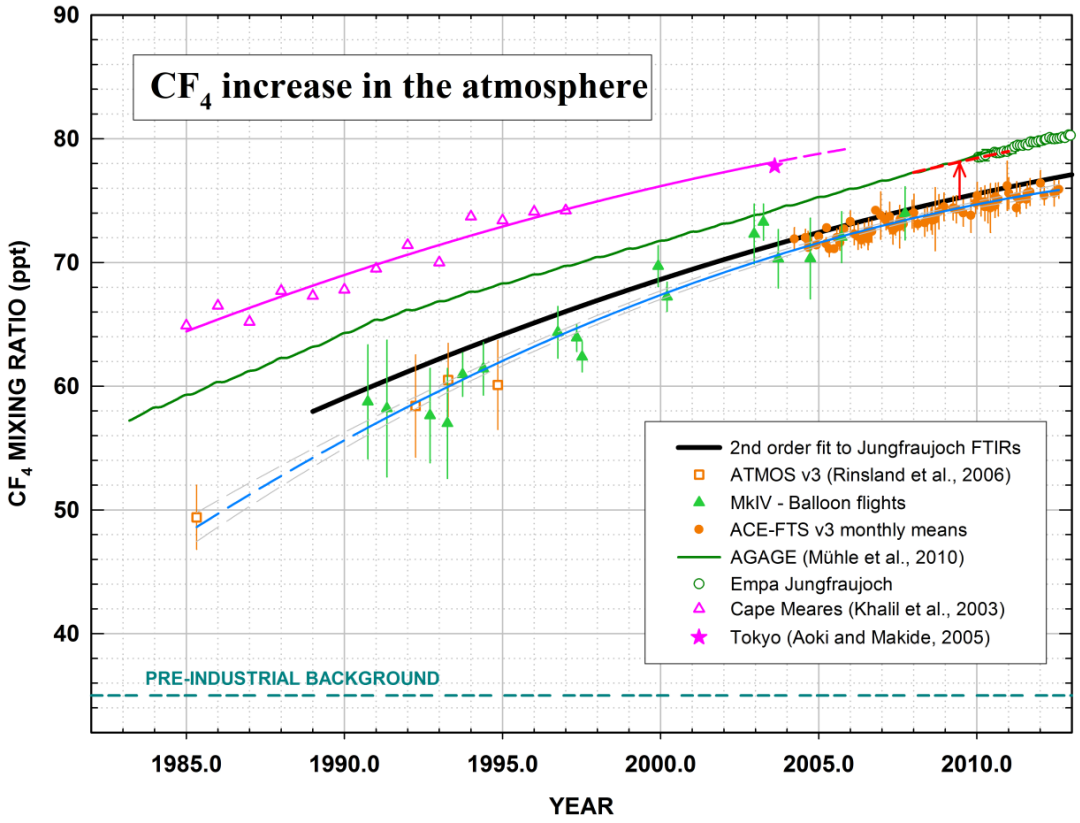

Figure 2. Long-term evolution of the CF_4 atmospheric mixing ratio as inferred from various techniques and locations (see inserted legend for their identification).

Formaldehyde (H_2CO)

Formaldehyde is a key target allowing characterizing air quality. This is a very short-lived species (lifetime of a few hours) resulting from the photochemical oxidation of methane and other carbonic compounds, it is also released by biomass burning and fossil fuel combustion. Identified sinks consist in photolysis, oxidation by the hydroxyl radical and deposition.
Up to now, the retrieval of H_2CO from the high altitude Jungfraujoch station was based on a single narrow window (2833.07-2833.35 cm^{-1}). A 6-window strategy set up for Reunion Island has been successfully implemented for the retrieval of this species from the Jungfraujoch, with the advantage of using a common strategy across the NDACC network (visit http://www.ndacc.org). A Tikhonov regularization is used, providing sensitivity in the lower troposphere. A preliminary time series updated from Franco et al. (2013) has been produced, including all spectra in the 1995-2012 time range. A strong seasonal modulation is confirmed, with maximum abundance in June-August and minimum columns in December-February. This time series has provided the first long-term trend of H_2CO, revealing a significant decrease of this species above our site, at a rate of -1.2±0.2 %/yr (2-σ). Future work will include detailed comparisons with coincident MAX-DOAS measurements performed by our colleagues from BIRA-IASB as well as with 3D-CTM simulations by the IMAGE and GEOS-Chem models.

Key words:

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

Internet data bases:

General website: http://girpas.astro.ulg.ac.be
Consolidated geophysical data are available from NDACC:
ftp://ftp.cpc.ncep.noaa.gov/ndacc/station/jungfrau/
Rapid delivery data are also available from:
ftp://ftp.cpc.ncep.noaa.gov/ndacc/RD/jungfrau/hdf/ftir

Collaborating partners/networks:

Main collaborations: BIRA-IASB (Institut d'Aéronomie Spatiale de Belgique) / NDACC (Network for the Detection of Atmospheric Composition Change; http://www.ndacc.org) / GAW-CH / partners of the EC-project NORS (http://nors.aeronomie.be) / ACE science team / NASA JPL / EMPA / University of Leeds / IMK (Forschungszentrum Karlsruhe) / satellite experiments: IASI, AURA, OMI, ENVISAT / …

Scientific publications and public outreach 2013:

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

Duflot, V., D. Hurtmans, L. Clarisse, Y. R'honi, C. Vigouroux, M. De Mazière, E. Mahieu, C. Servais, C. Clerbaux, and P.-F. Coheur, Measurements of hydrogen cyanide (HCN) and acetylene (C_2H_2) from the Infrared Atmospheric Sounding Interferometer (IASI), Atmos. Meas. Tech., **6**, 917-925, doi: 10.5194/amt-6-917-2013 2013.
http://hdl.handle.net/2268/132381

Mahieu, E., R. Zander, G.C. Toon, M.K. Vollmer, S. Reimann, J. Mühle, W. Bader, B. Bovy, B. Lejeune, C. Servais, P. Demoulin, G. Roland, P.F. Bernath, C.D. Boone, K.A. Walker, and P. Duchatelet, Spectrometric monitoring of atmospheric carbon tetrafluoride (CF_4) above the Jungfraujoch station since 1989: evidence of continued increase but at a slowing rate, Atmos. Meas. Tech., **6**, 7535-7563, doi:10.5194/amtd-6-7535-2013, 2013a.
http://hdl.handle.net/2268/154767

Conference papers

Bader, W., E. Mahieu, B. Bovy, B. Lejeune, P. Demoulin, C. Servais, and J.J. Harrison, Evolution of methanol (CH_3OH) above the Jungfraujoch station (46.5ºN): variability, seasonal modulation and long-term trend, poster presentation at the "EGU 2013 General Assembly", Vienna, Austria, April 07-12, 2013.
http://hdl.handle.net/2268/145478

Franco, B., E. Mahieu, M. Van Roozendael, F. Hendrick, T. Stavrakou, I. De Smedt, G. Pinardi, B. Bovy, P. Demoulin, and C. Servais, Parallel measurements of formaldehyde (H_2CO) at the Jungfraujoch station: preliminary FTIR results and first comparison with MAX-DOAS data, oral presentation at the NORS second progress meeting, Uccle, Belgium, October 17, 2013.
http://hdl.handle.net/2268/158375

Henne, S., M. Steinbacher, E. Mahieu, W. Bader, T. Blumenstock, E. Cuevas-Agulló, D. Brunner, and B. Buchmann, Comparison of ground-based remote sensing and in-situ observations of CO, CH_4 and O_3, accounting for representativeness uncertainty, oral presentation at the "EGU 2013 General Assembly", Vienna, Austria, April 07-12, 2013.

Kolonjari, F., K.A. Walker, E. Mahieu, R.L. Batchelor, P.F. Bernath, C.D. Boone, S. Conway, L. Dan, D. Griffin, A. Harrett, Y. Kasai, A. Kagawa, R. Lindenmaier, K. Strong, and C. Whaley, Validation of ACE-FTS measurements of CFC-11, CFC-12 and HCFC-22 using ground-based FTIRs, poster presentation at the "AGU Fall Meeting 2013", San Fransisco, CA, USA, December 09-13, 2013.

Mahieu, E., B. Bovy, W. Bader, P. Demoulin, B. Franco, B. Lejeune, C. Servais, and C. Vigouroux, Overview of the geophysical data derived from long-term FTIR monitoring at the Jungfraujoch NDACC site (46.5°N), poster presented at the 6th International GEOS-Chem Meeting, Harvard University, Cambridge, MA, USA, May 6-9, 2013b.
http://hdl.handle.net/2268/147824

Mahieu, E., S. O'Doherty, S. Reimann, M. Vollmer, W. Bader, B. Bovy, B. Lejeune, P. Demoulin, G. Roland, C. Servais, and R. Zander, First retrievals of HCFC-142b from ground-based high-resolution FTIR solar observations: application to high-altitude Jungfraujoch spectra, poster presentation at the "EGU 2013 General Assembly", Vienna, Austria, April 07-12, 2013c.
http://hdl.handle.net/2268/144709

Vigouroux, C., M. De Mazière, P. Demoulin, C. Servais, F. Hase, T. Blumenstock, M. Schneider, R. Kohlhepp, S. Barthlott, O. Garcia, J. Mellqvist, G. Persson, M. Palm, J. Notholt, J. Hannigan, and M. Coffey, Ozone tropospheric and stratospheric trends (1995-2012) at six ground-based FTIR stations (28ºN to 79ºN), poster presentation at the "EGU 2013 General Assembly", Vienna, Austria, April 07-12, 2013.

Edited books

Mahieu, E., R. Zander, P.F. Bernath, C.D. Boone, and K.A. Walker, Recent trend anomaly of hydrogen chloride (HCl) at northern mid-latitudes derived from Jungfraujoch, HALOE and ACE-FTS Infrared solar observations, in *The Atmospheric Chemistry Experiment ACE at 10: A Solar Occultation Anthology*, P.F. Bernath (Ed.), ISBN 978-0-937194-54-9, A. Deepak Publishing, 239-249, 2013.

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