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 infrared spectrometers allow to routinely derive abundances of more than 20 constituents related to the erosion of the ozone layer in the stratosphere (HCl, $ClONO_2$, HNO_3, NO, NO_2, HF, COF_2, O_3…), monitored in the frame of the Kyoto protocol (N_2O, CH_4, CO_2, SF_6, CCl_2F_2, $CHClF_2$, CCl_3F…) or affecting 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.

During 2005, Liège observers spent 247 days at the Jungfraujoch. Good weather conditions enabled observations on 124 days.

For a number of the species listed above, a complete re-analysis of the archived spectra is currently under way with SFIT-2, a recent 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.
In the frame of the EC project UFTIR (http://www.nilu.no/uftir), a homogenised optimal retrieval strategy has been developed for the inversion of O_3, N_2O, C_2H_6, HCFC-22, CO and CH_4. The corresponding Jungfraujoch spectra between 1995 and 2004 have been reprocessed, after implementation of this new strategy, and the resulting time series are being archived on the UFTIR database at NILU. The UFTIR outcomes are shared with the NDSC infrared community.
As an example, Figure 1 shows the results of the FTIR retrievals for carbon monoxide CO, compared to the Jungfraujoch *in situ* CO measurements by EMPA.
The agreement between both datasets is rather good, showing similar seasonal and short term variations. CO anomalies observed in the northern hemisphere in 1998-1999, 2002 and 2003 are clearly visible. These anomalies have recently been investigated [Yurganov *et al.*, 2005] and the corresponding extra CO emissions have been evaluated to 95 and 130 Tg, respectively in 2002 and 2003. Strong boreal fires that occurred in Russia during these two years are the most likely causes for the observed CO burden increases.

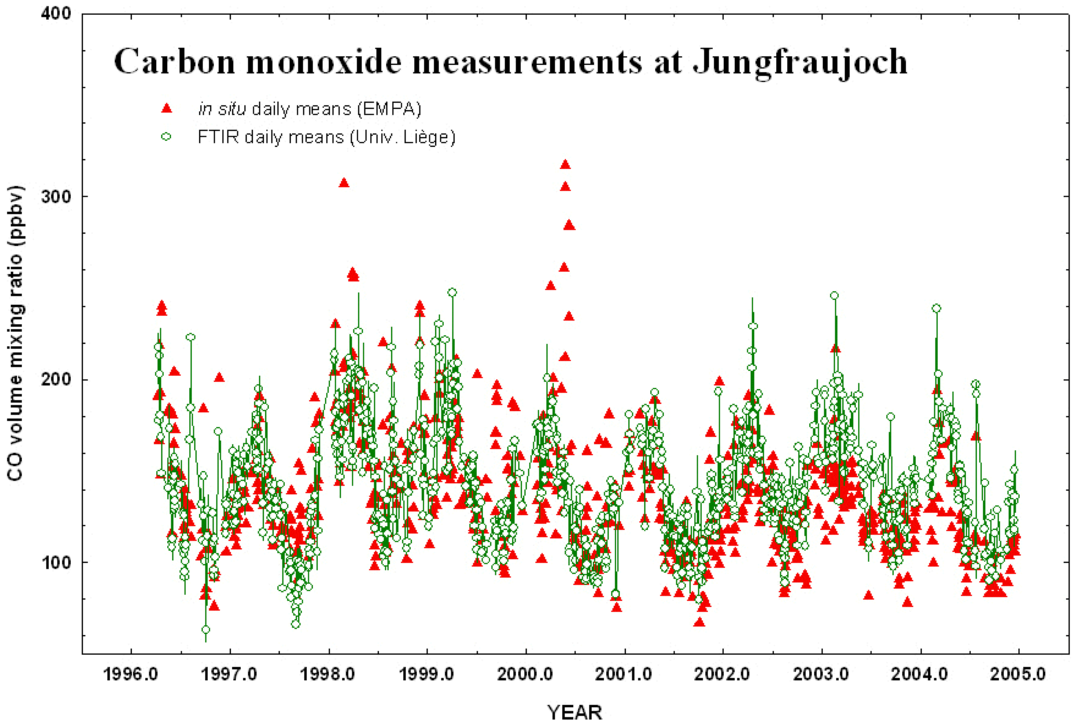

Figure 1: *Daily mean carbon monoxide volume mixing ratio (VMR) at the Jungfraujoch, derived from FTIR spectra (circles) and from EMPA in situ measurements (triangles). FTIR data correspond to the mean VMR retrieved in the lowest layers (i.e. between 3.58 and 5.5 km, corresponding to one independent piece of information). EMPA data have been obtained from the WDCGG (World Data Centre for Greenhouse Gases, http://gaw.kishou.go.jp/wdcgg.html).*

Within the context of the Montreal Protocol, monitoring of chlorinated source gases has been part of our activities. Time series of CFC-12 (CCl_2F_2) and HCFC-22 ($CHClF_2$) have been updated while modifications implemented to the SFIT-2 retrieval algorithm have allowed to perform retrievals of two additional species, i.e. CFC-11 (CCl_3F) and CCl_4. Current monthly mean time series of three of these source gases are shown in Figure 2. Although year round data points are reproduced here, only averaged total columns of the quietest June to November months (i.e. exhibiting less variability, see filled symbols) have been used to characterise the long-term evolutions of these compounds. Corresponding trends and annual column changes are available in Zander *et al.* [2005]. It is interesting to point out here the contrasted evolutions of these source gases: (i) the Montreal-controlled CFC-11 and CFC-12 are decreasing/stabilising as expected from their respective lifetimes (45 and 100 years); (ii) progressive phase out of HCFC-22 (an important CFC substitute) has begun in 2004 and its production is supposed to reach zero in developed countries in 2030; its concentration is expected to continue rising until about 2010 before stabilisation and rapid decrease thereafter. Our measurements indicate a steady increase of the HCFC-22 total columns with recent rate of change on the order of 3 %/year.

Comparisons of the above results with findings deduced from *in situ* measurements performed by the AGAGE and NOAA/CMDL networks indicate very good agreement in terms of trends and atmospheric concentrations for these various species.

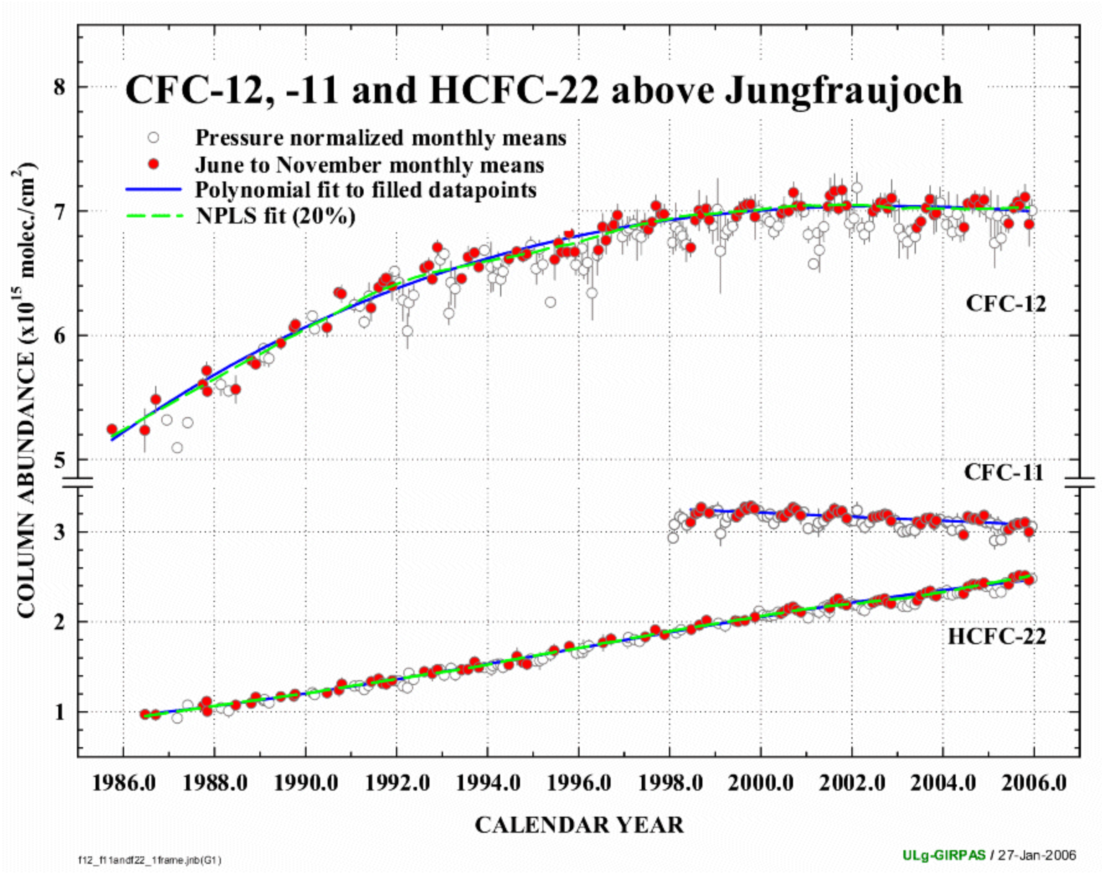

Figure 2: *Contrasted evolutions of the monthly mean total vertical column abundances of CFC-12, CFC-11 and HCFC-22 above the Jungfraujoch.*

In addition to the important greenhouse gases regulated by the Montreal Protocol, the 1997 "Kyoto Protocol on climate change" specifically targets CO_2, CH_4, N_2O and SF_6 which present characteristic infrared absorption features allowing to quantify their atmospheric abundances. Regular analyses of all Jungfraujoch observations have resulted in updates of their temporal evolutions [Zander *et al.*, 2005]. Related time series, which cover now two decades, are reproduced in Figure 3.

The first obvious feature is the regular growth for three of these gases over the 1985-2004 time period, only methane exhibiting a stabilisation over recent years.

Comparisons with abundances derived from pioneering observations performed in 1950-1951 at the same site by M. Migeotte indicate that the total columns of CO_2, CH_4 and N_2O have been respectively multiplied by 1.25, 1.35 and 1.17. More specifically, trend determinations have indicated a yearly increase of 0.42 % for CO_2, in excellent agreement with *in situ* surface measurements (e.g. www.cmdl.noaa.gov). The very long-lived nitrous oxide (120 years) shows a similar behaviour, with an annual linear build up of 1.06 x 10^{16} molec./cm^2; this corresponds to an increase of 0.26 %/year, commensurate with *in situ* trend data.

Rapid increase of the total column abundance of SF_6 has been confirmed over recent years, with an annual load increase still exceeding 4 %/year in 2004. It is important to limit emissions of this compound to the atmosphere because it combines a very strong

absorption of infrared radiation on a per-molecule basis with a very long lifetime of several thousand years.
Extrapolation of the Jungfraujoch data predicts tropospheric SF_6 concentrations of about 15 pptv in 2050 and about 25 pptv in 2100 (compared this to the 2.0 pptv concentration measured in 1988) [Krieg *et al.*, 2005]. These values are significantly lower than those reported in recent scenarios [WMO 2003], justifying future monitoring of this species to determine its effective future evolution and related climatic impact.
A striking feature of this figure is the stabilisation of the CH_4 loading during recent years, that will deserve future comparisons with models, to identify the relative contributions of changes in sources and sinks leading to this stabilisation.

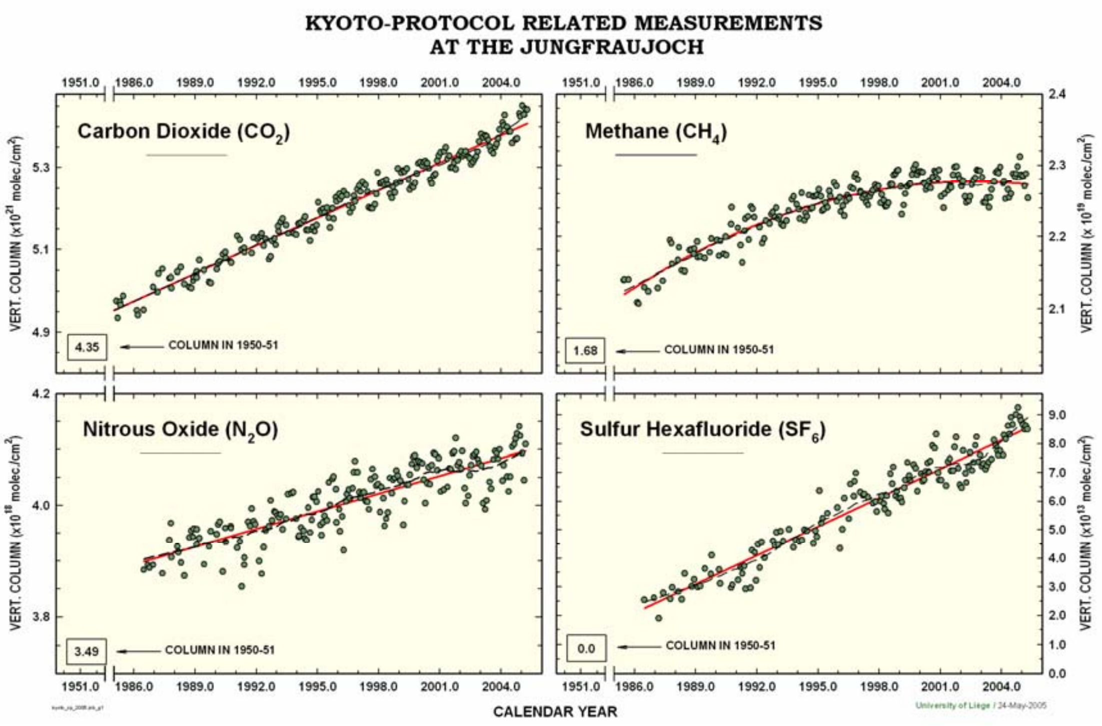

Figure 3: *Long-term evolution of four species targeted by the Kyoto Protocol as derived from ground-based remote observations conducted at the Jungfraujoch station. Notice the different vertical axis units for each frame.*

During 2005, we provided additional data for the calibration/validation of 3 instruments (MIPAS, SCIAMACHY and GOMOS) aboard the European satellite Envisat. On the whole, we supplied to the calibration team 12784 total column abundances of O_3, N_2O, CO, CH_4, NO, NO_2, HNO_3 and CO_2, deduced from Jungfraujoch observations between July 2002 and December 2004. More elaborated products consisting in 4329 vertical distributions and related partial columns of HNO_3, CH_4 and N_2O for 201 days between July 2002 and March 2004 were also produced for specific validation of MIPAS profiles and SCIAMACHY columns.

The Canadian ACE-FTS instrument was launched in August 2003 and has been in "post commissioning" operation since February 2004 [Bernath *et al.*, 2005]. Specific observational campaigns were organized at the Jungfraujoch in support to the

validation of the ACE-FTS spectrometer to record as many coincident measurements as possible.
Scientific and validation papers using the first "Version 1" of level 2 data have been published recently in a GRL special issue. In particular, a study dealing with comparisons between stratospheric columns of HCl and $ClONO_2$ measured up to October 2004 by ACE and by ground-based FTIR instruments operated at five northern latitude NDSC sites (including the Jungfraujoch) has been led by ULg [Mahieu et al., 2005]. Main conclusions were that: (i) ACE is able to identify for both targeted gases distribution features characteristic of geographical, dynamical, seasonal and chemical changes occurring in the atmosphere; (ii) excellent agreement was found when considering the only sets of coincident measurements obtained around the Thule site (76.5ºN), with mean partial column ratio (ACE/Thule) equal to 1.04 and 0.99 respectively for HCl and ClONO2; (iii) good agreement is generally found for other sites (in particular for the Jungfraujoch), even if systematic differences between some data sets deserve further investigations based on additional coincident ACE "Version 2.2" and ground-level remote FTIR measurements.

On the hardware side, the opening of the heliostat has been electrified, a first step towards its complete automation. The previously tedious manual opening of the heliostat is now performed with a simple remote control.

Key words:

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

Internet data bases:

http://www.nilu.no/nadir/, ftp://ndsc.ncep.noaa.gov/pub/ndsc/jungfrau/ftir/

Collaborating partners/networks:

Main collaborations: IASB (Institut d'Aéronomie Spatiale de Belgique) / NDSC (Network for the Detection of Stratospheric Change) / SOGE partners (e.g. EMPA) [http://www.nilu.no/soge/] / NASA Langley Research Center / NASA JPL / University of Oslo / University of Leeds / IMK (Forschungszentrum Karlsruhe) / satellite experiments: MOPPIT, ENVISAT and ACE validation / …

Scientific publications and public outreach 2005:

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Radio and television

"La sentinelle du soleil", interview of Pierre Duchatelet, Belgian Local Television No Télé (Hainaut), 19 October 2004.

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