

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 two high-performance infrared spectrometers allowed to routinely derive total abundances of more than 20 constituents: gases 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>, ...), greenhouse gases 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>, CCl<sub>2</sub>F<sub>2</sub>, CHClF<sub>2</sub>, ...) and gases affecting 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.

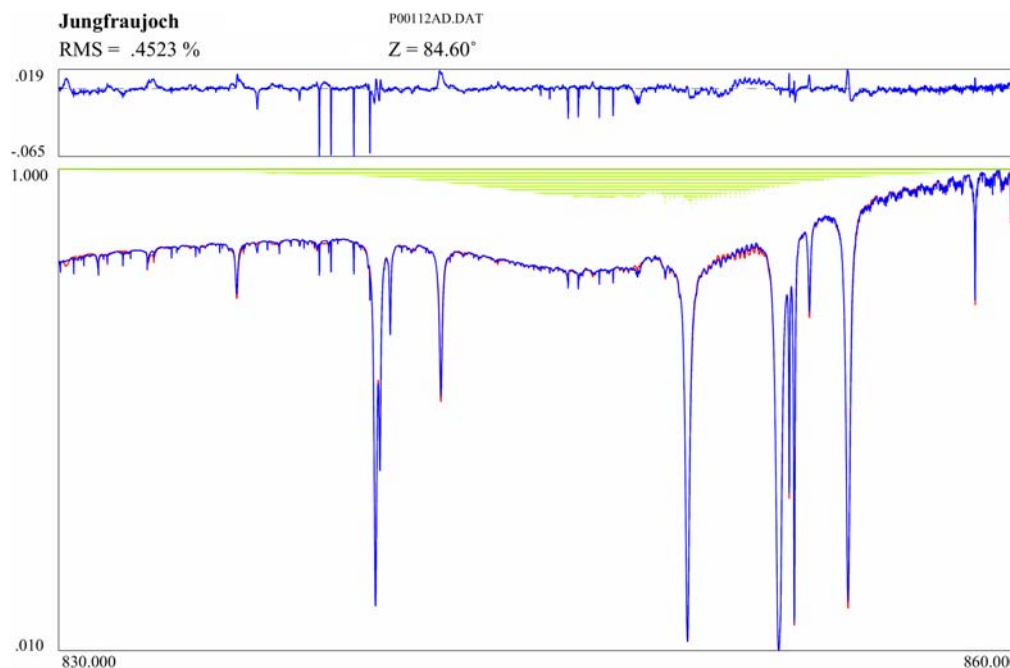
During 2003, 221 days were spent at the Jungfraujoch by observers from Liège and from the Institute of Space Aeronomy of Brussels. Good weather conditions enabled observations on 123 days.

We carried on evaluating the budget of chlorine- fluorine- and nitrogen-containing species. The analysis confirms, for example, that the stratospheric chlorine contents (Cl<sub>y</sub>) has leveled off during the late 1990s, as a result of the restrictions imposed by the Montreal Protocol and its successive amendments on the production of chlorine-bearing source gases. Further measurements will help to assess the expected subsequent slow Cl<sub>y</sub> decrease.

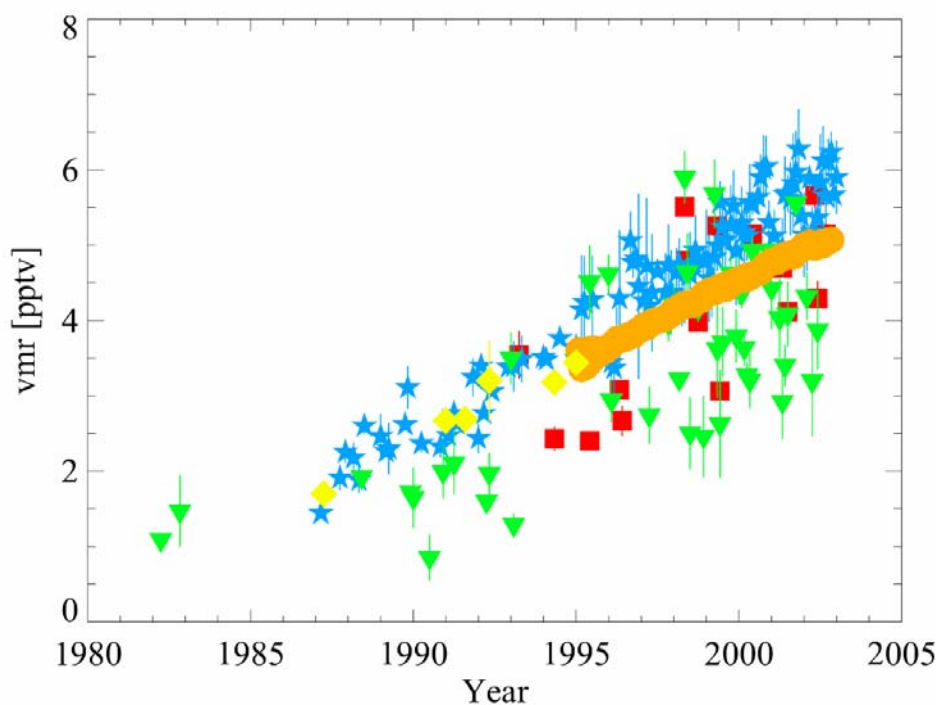
For a number of the species listed above, a complete re-analysis of the archived spectra is currently under way with SFIT-2, a new retrieval algorithm that provides some information on the distribution of the molecules versus altitude. This tool leads to more accurate total columns evaluation. For some gases, it allows to perform partial columns retrievals (e.g. to distinguish between tropospheric and stratospheric contents).

In order to perform retrievals of additional heavy gases like CFCs, HCFCs and other new substitutes showing broad absorption features in the infrared, several modifications to the SFIT-2 analysis algorithm have been implemented successfully. In particular, changes to the source code have been made to significantly increase the number of spectral points that can be handled: wide intervals of up to 50 cm<sup>-1</sup> can

now be fitted. Intensive tests have been performed to identify the best retrieval settings leading to reliable CFC-11 ( $\text{CCl}_3\text{F}$ ) and  $\text{CCl}_4$  total columns determinations. Figure 1 shows the selected spectral domain for the retrieval of CFC-11.



**Figure 1:** sample fit of CFC-11 over the selected spectral domain. Lower frame: observed (blue), computed (red) and CFC-11-only computed (green) spectra. Upper frame: residuals (observed minus computed spectra). Interferences by solar OH quadruplet lines not modeled by the analysis software and located around  $838$  and  $846\text{ cm}^{-1}$  are clearly visible in the upper frame of the figure.



**Figure 2:** mixing ratios of  $\text{SF}_6$ , derived from measurements above Jungfraujoch (stars), Ny Ålesund (Norway, squares) and Kitt Peak (USA, triangles). Additionally, in situ measurements from NOAA/CMDL (circles, from <http://www.cmdl.noaa.gov>) averaged over the northern hemisphere and other values from the literature (diamonds) are shown.

In the frame of the EU-project SOGE (System for Observation of Halogenated Greenhouse Gases in Europe), in which EMPA Dübendorf is also involved, emphasis has been put on the analysis of halogenated greenhouse gases. As an example, Figure 2 show the SF<sub>6</sub> time series retrieved from IR spectra recorded at the Jungfraujoch. SF<sub>6</sub> retrievals are particularly difficult due to strong and close interferences by water vapour and CO<sub>2</sub> absorptions. Intercomparison of the SF<sub>6</sub> time series obtained at three NDSC stations (Jungfraujoch, Ny Ålesund, 78.9°N, 20 m a.s.l. and Kitt Peak 31.9°N, 2090 m a.s.l.) has been performed to verify the consistency among the related data sets.

During 2003, we also continued to provide data for the calibration/validation of 3 instruments (MIPAS, SCIAMACHY and GOMOS) aboard the European satellite Envisat. As a result, we supplied to the calibration team 3037 total abundances of O<sub>3</sub>, N<sub>2</sub>O, CO, CH<sub>4</sub>, NO, NO<sub>2</sub>, HNO<sub>3</sub> and CO<sub>2</sub>, measured at the Jungfraujoch over 63 days between January and August.

Key words:

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

Internet data bases:

<http://www.nilu.no/nadir/>, <ftp://ndsc.wwb.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/niluweb/services/soge>] / NASA Langley Research Center / NASA JPL / University of Oslo / IMK (Forschungszentrum Karlsruhe) / satellite experiments: MOPPIT, ENVISAT and ACE validation / ...

Scientific publications and public outreach 2003:

**Refereed journal articles**

Barret, B., M. De Mazière, and E. Mahieu, Ground-based FTIR measurements of CO from the Jungfraujoch: characterisation and comparison with in situ surface and MOPITT data, *Atmos. Chem. Phys.*, **3**, 2217-2223, 2003.

Rinsland, C. P., A. Goldman, T. M. Stephen, L. S. Chiou, E. Mahieu, and R. Zander, SF<sub>6</sub> ground-based infrared solar absorption measurements : long-term trend, pollution events, and a search for SF<sub>5</sub>CF<sub>3</sub> absorption, *J. Quant. Spectrosc. Radiat. Transfer*, **78**, 41-53, 2003.

Rinsland, C. P., E. Mahieu, R. Zander, N. B. Jones, M. P. Chipperfield, A. Goldman, J. Anderson, J. M. Russell III, P. Demoulin, J. Notholt, G. C. Toon, J.-F. Blavier, B. Sen, R. Sussmann, S. W. Wood, A. Meier, D. W. T. Griffith, L. S. Chiou, F. J. Murcray, T. M. Stephen, F. Hase, S. Mikuteit, A. Schulz, and T. Blumenstock, Long-term trends of inorganic chlorine from ground-based infrared solar spectra: Past increases and evidence for stabilization, *J. Geophys. Res.*, **108(D8)**, 4252, ACH10, doi:10.1029/2002JD003001, 2003.

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with model calculation, *J. Geophys. Res.*, 108(D15), 4437, ACL1, doi:10.1029/2002JD002965, 2003.

### Conference papers

De Mazière, M., T. Coosemans, B. Barret, T. Blumenstock, A. Griesfeller, P. Demoulin, H. Fast, D. Griffith, N. Jones, E. Mahieu, J. Mellqvist, R. L. Mittermeier, J. Notholt, C. Rinsland, A. Schulz, D. Smale, A. Strandberg, R. Sussmann, S. Wood, M. Buchwitz, Validation of ENVISAT-1 Level-2 Products Related to Lower Atmosphere O<sub>3</sub> and NO<sub>y</sub> Chemistry by a FTIR Quasi-global Network, in the *Proceedings of Envisat Validation Workshop*, Frascati, Italy, 9-13 December 2002, ESA SP-531, 2003

Duchatelet, P., E. Mahieu, R. Zander, P. Demoulin, B. Barret, and C. P. Rinsland; Updating the Jungfraujoch database: current status, in *Proceedings of the "Sixth European Symposium on Stratospheric Ozone"*, Göteborg, Sweden, September 2-6, 2002, EUR 20650, ISBN 92-894-5484-9, pp. 136-139, 2003.

Mahieu, E., C. P. Rinsland, R. Zander, P. Duchatelet, C. Servais, and M. De Mazière, Tropospheric and stratospheric carbonyl sulfide (OCS): long-term trends and seasonal cycles above the Jungfraujoch station, in *Proceedings of the "Sixth European Symposium on Stratospheric Ozone"*, Göteborg, Sweden, September 2-6, 2002, EUR 20650, ISBN 92-894-5484-9, pp. 309-312, 2003.

### Data books and reports

Montzka, S. A., P. J. Fraser, J. H. Butler, D. Cunnold, J. Daniel, D. Derwent, P. S. Connell, S. Lal, A. McCulloch, D. E. Oram, C. E. Reeves, E. Sanhueza, P. Steele, G. J. M. Velders, and R. Zander, Controlled substances and other source gases, Chapter 1 of *WMO Scientific Assessment of Ozone Depletion: 2002*, WMO Report No. 47, pp. I-1 to I-83, World Meteorological Organization, P.O. Box 2300, Geneva 2, CH 1211, Switzerland, 2003.

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URL: <http://sunset.astro.ulg.ac.be/girpas/girpasf.html>