

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

Luc Delbouille (em.), Philippe Demoulin, Pierre Duchatelet, Mélanie Kessels, Emmanuel Mahieu, Ginette Roland (em.), Thomas Rulmont, Christian Servais (project leader), Rodolphe Zander (em.)

Jacqueline Bosseloirs, Guy Buntinx, Vincent Van De Weerd, Diane Zander

Project description:

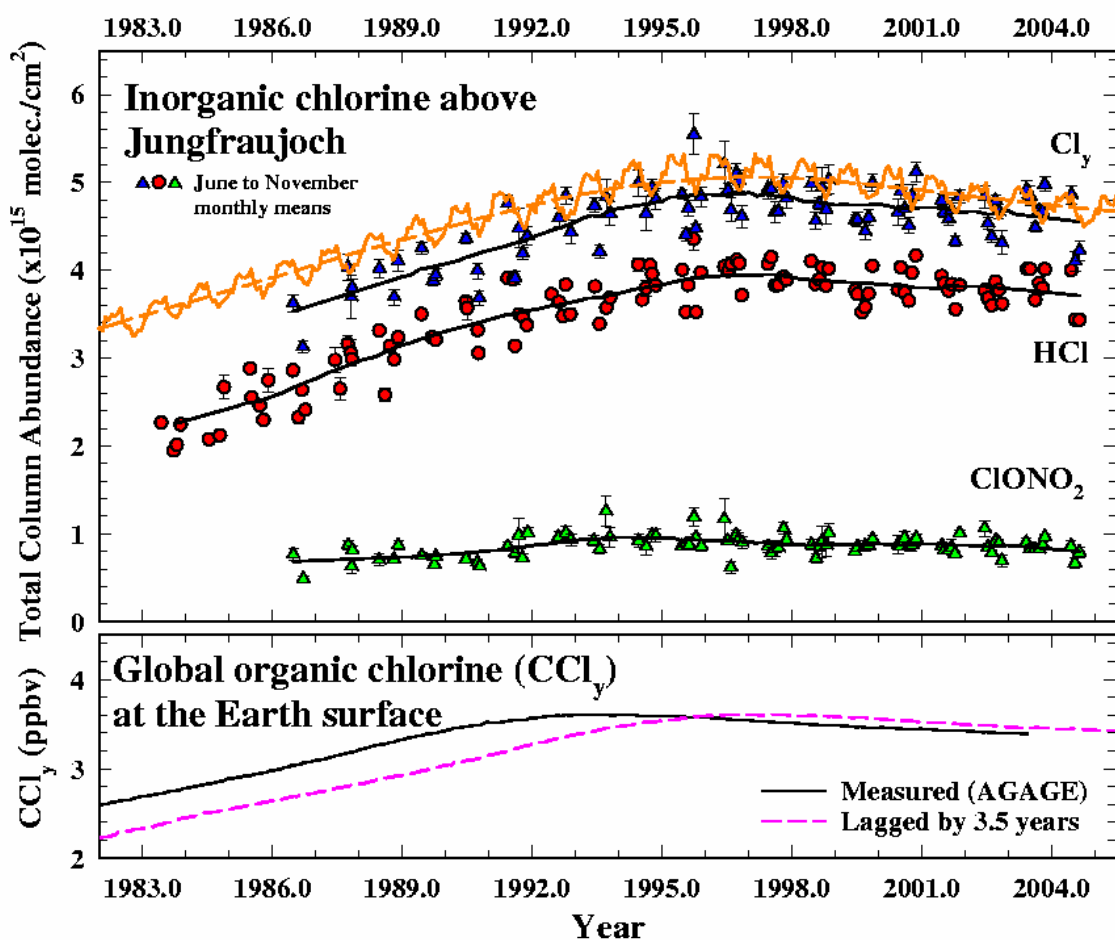
The main activity of the Liège group at the Jungfraujoch was the continuation of the long-term monitoring of the Earth atmosphere. The observations achieved by the two high-performance infrared spectrometers allow to routinely derive abundances of more than 20 constituents related to the erosion of the ozone layer in the stratosphere (HCl, ClONO<sub>2</sub>, HNO<sub>3</sub>, NO, NO<sub>2</sub>, HF, COF<sub>2</sub>, O<sub>3</sub>, ...), 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>, ...) or 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 2004, Liège observers spent 238 days at the Jungfraujoch. Unfortunately, weather conditions enabled observations on 93 days only, which is proportionally less than for the previous years.

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 to determine partial columns (e.g. to distinguish between tropospheric and stratospheric contents) as well as more accurate total columns.

Within the frame of the NDSC (Network for the Detection of Stratospheric Change), the total vertical column abundances of HCl and ClONO<sub>2</sub>, by far the two most important inorganic chlorine (Cl<sub>y</sub>) reservoirs at northern mid-latitudes, have been further monitored above the Jungfraujoch station. Their temporal evolution indicates that, after a steady increase at a mean rate of 3.8 %/yr during the 1986-1993 period, the total stratospheric inorganic chlorine loading has begun a progressive stabilization, with a maximum observed during the second half of 1996. Since then, a slight Cl<sub>y</sub> decrease has been observed, at a mean yearly rate close to -0.7% over the 1997-2003 period (see Figure 1) [Mahieu et al., 2004].

Overall, these findings are in very good agreement with the 2D-model calculations at University of Leeds that, after smoothing of the seasonal variations (see upper dashed curve in the top frame of Figure 1), also indicate a Cl<sub>y</sub> peak during the second half of 1996.



norg\_Cl\_2004

ULg-GIRPAS / 10-Sep-2004

**Figure 1:** Top panel: total column abundances of HCl and ClONO<sub>2</sub> above the Jungfraujoch. Their sum (blue triangles) is a good estimate for the total inorganic chlorine Cl<sub>y</sub>. The upper trace (in orange) corresponds to the sum of the HCl and ClONO<sub>2</sub> columns calculated by a 2D-model at University of Leeds. The lower panel displays the evolution of total organic chlorine based on in situ surface measurements performed by the AGAGE network.

The Cl<sub>y</sub> long-term evolution is also consistent with the temporal development of the organic chlorine (CCl<sub>y</sub>) loading measured at the surface, when accounting for a time delay of about 3.5 years for tropospheric air to propagate and mix into the mid-latitude stratosphere (Figure 1, lower frame). After a peak occurrence in 1993, CCl<sub>y</sub> also exhibits a slow decrease of about 0.7%/yr, in excellent agreement with the Cl<sub>y</sub> observations and model calculations.

These results demonstrate the appropriateness and proper implementation of the phase out scenarios adopted by the Montreal Protocols and its subsequent enforcements.

To evaluate the worldwide evolution of the inorganic chlorine and fluorine in the Earth's middle atmosphere, our group coordinates the Cl<sub>y</sub>-F<sub>y</sub> project, involving 23 NDSC FTIR stations. Data from the *in situ* surface networks as well as measurements from the satellite experiments HALOE, ENVISAT, ACE and AURA will also be included in the analysis.

In the frame of the SOGE (System for Observation of halogenated Greenhouse gases in Europe) project, sulphur hexafluoride (SF<sub>6</sub>) concentrations have been deduced from the infrared spectra recorded at the Jungfraujoch. Mainly produced by man-made activities, SF<sub>6</sub>, with its long lifetime of about 3200 years, is one of the most efficient greenhouse gases present in the atmosphere, with a global warming potential of 22 200 relative to CO<sub>2</sub> over a 100-year time horizon.

Our analysis shows a linear SF<sub>6</sub> increase of  $0.24 \pm 0.01$  pptv per year between 1986 and 2002. Extrapolation of the Jungfraujoch data predicts tropospheric SF<sub>6</sub> concentrations of about 15 pptv in 2050 (compared this to the 2.0 pptv concentration measured in 1988) [Krieg et al., 2005]

Measurements of carbon monoxide total column amounts above the Jungfraujoch reveal increased CO abundance during summer and autumn periods of 2002 and 2003 in comparison with the previous two years. Similar increases in the northern hemisphere were observed from other ground-based stations in the Arctic and Europe, as well as from the space-based MOPITT experiment onboard the Terra satellite. Moreover, the CMDL and GAW networks also observed increased CO concentrations near the surface. It is most likely that strong boreal forest fires are responsible for this hemispheric CO build up [Yurganov et al., 2004b].

During 2004, we also continued to provide 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 8376 total column 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>, deduced from Jungfraujoch observations between July 2002 and October 2003. Vertical distributions of HNO<sub>3</sub> and N<sub>2</sub>O for 199 days between July 2002 and March 2004 were also produced for specific validation of MIPAS profiles.

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 2004:

---

### Refereed journal articles

Krieg, J., J. Notholt, E. Mahieu, C.P. Rinsland, and R. Zander, Sulphur hexafluoride (SF<sub>6</sub>): comparison of FTIR-measurements at three sites and determination of its trend in the northern hemisphere, *J. Quant. Spectrosc. Radiat. Transfer*, **92**, 383-392, 2005.

Rinsland, C. P., E. Mahieu, R. Zander, A. Goldman, S. W. Wood, and L. S. Chiou, Free Tropospheric Measurements of Formic Acid (HCOOH) from Infrared Ground-Based Solar Absorption Spectra: Retrieval Approach, Evidence for a Seasonal Cycle, and Comparison with Model Calculations, *J. Geophys. Res.*, **109**, D18308, doi:10.1029/2004JD004917, 2004.

Yurganov, L.N., T. Blumenstock, E.I. Grechko, F. Hase, E.J. Hyer, E.S. Kasischke, M. Koike, Y. Kondo, I. Kramer, F.-Y. Leung, E. Mahieu, J. Mellqvist, J. Notholt, P.C. Novelli, C.P. Rinsland, H.E. Scheel, A. Shultz, A. Strandberg, R. Sussmann, H. Tanimoto, V. Velazco, R. Zander, and Y. Zhao, A Quantitative Assessment of the 1998 Carbon Monoxide Emission Anomaly in the Northern Hemisphere Based on Total Column and Surface Concentration Measurements, *J. Geophys. Res.*, **109** (D15), D15305, doi:10.1029/2004JD004559, 2004a.

Yurganov, L.N., P. Duchatelet, A.V. Dzhola, D.P. Edwards, F. Hase, I. Kramer, E. Mahieu, J. Mellqvist, J. Notholt, P.C. Novelli, A. Rockmann, H.E. Scheel, M. Schneider, A. Schulz, A. Strandberg, R. Sussmann, H. Tanimoto, V. Velazco, J.R. Drummond, and J.C. Gille, Increased Northern Hemispheric carbon monoxide burden in the troposphere in 2002 and 2003 detected from the ground and from space, *Atmos. Chem. Phys. Discuss.*, **4**, 4999-5017, 2004b.

### Book sections

Zander, R., and M. De Mazière, Atmospheric composition changes: Causes and processes at work, in "Assessment and Integration Report on Global Change-Related Projects Funded by the OSTC in the Area of Natural Sciences During 1990-2002", *in press*, 2004

### Conference papers

Camy-Peyret, C., G. Dufour, S. Payan, H. Oelhaf, G. Wetzell, G. Stiller, Th. Blumenstock, C. E. Blom, T. Gulde, N. Glatthor, A. Engel, M. Pirre, V. Catoire, G. Moreau, M. De Mazière, C. Vigouroux, E. Mahieu, U. Cortesi, and F. Mencaraglia, Validation of MIPAS N<sub>2</sub>O Profiles by Stratospheric Balloon and Ground-based Measurements, *in Proceedings of the Second Workshop on the Atmospheric Chemistry Validation of ENVISAT (ACVE-2), ESA-ESRIN, Frascati, Italy, 3-7 May 2004 (ESA SP-562, August 2004) EMI02CC1.*

De Mazière, M., B. Barret, T. Blumenstock, M. Buchwitz, R. De Beek, P. Demoulin, H. Fast, A. Gloudemans, A. Griesfeller, D. Griffith, D. Ionov, K. Janssens, N. Jones, E. Mahieu, J. Mellqvist, R.L. Mittermeier, J. Notholt, C. Rinsland, H. Schrijver, A. Shultz, D. Smale, A. Strandberg, K. Strong, R. Sussmann, T. Warneke, and S. Wood, Comparisons between SCIAMACHY Scientific Products and Ground-based FTIR Data for Total Columns of CO, CH<sub>4</sub> and N<sub>2</sub>O, *in Proceedings of the Second Workshop on the Atmospheric Chemistry Validation of ENVISAT (ACVE-2), ESA-ESRIN, Frascati, Italy, 3-7 May 2004 (ESA SP-562, August 2004) ESC02MDM.*

De Mazière, M., B. Barret, C. Vigouroux, T. Blumenstock, F. Hase, I. Kramer, C. Camy-Peyret, P. Chelin, T. Gardiner, M. Coleman, P. Woods, K. Ellingsen, M. Gauss, I. Isaksen, E. Mahieu, P. Demoulin, P. Duchatelet, J. Mellqvist, A. Strandberg,

V. Velazco, A. Shulz, J. Notholt, R. Sussmann, W. Stremme, and A. Rockmann, Ground-based FTIR measurements of O<sub>3</sub>- and climate-related gases in the free troposphere and lower stratosphere, in *Ozone, Vol. I, Proceedings of the XX Quadrennial Ozone Symposium, Kos, Greece, 1-8 June 2004, ISBN 960-630-103-6, pp. 529-530, 2004.*

Lambert, J-C., M. Allaart, S. B. Andersen, T. Blumenstock, G. Bodeker, E. Brinksma, C. Cambridge, M. De Mazière, P. Demoulin, P. Gerard, M. Gil, F. Goutail, J. Granville, D. V. Ionov, E. Kyrö, M. Navarro-Comas, A. Piters, J-P. Pommereau, A. Richter, H. K. Roscoe, H. Schets, J.D. Shanklin, T. Suortti, R. Sussmann, M. Van Roozendael, C. Varotsos, T. Wagner, S. Wood, and M. Yela, First Ground-Based Validation of SCIAMACHY V5.01 Ozone Column, in *Proceedings of the Second Workshop on the Atmospheric Chemistry Validation of ENVISAT (ACVE-2), ESA-ESRIN, Frascati, Italy, 3-7 May 2004 (ESA SP-562, August 2004) ESC01JL1.*

Lambert, J-C., T. Blumenstock, F. Boersma, A. Bracher, M. De Mazière, P. Demoulin, I. De Smedt, H. Eskes, M. Gil, F. Goutail, J. Granville, F. Hendrick, D. V. Ionov, P. V. Johnston, I. Kostadinov, K. Kreher, E. Kyrö, R. Martin, A. Meier, M. Navarro-Comas, A. Petritoli, J-P. Pommereau, A. Richter, H. K. Roscoe, C. Sioris, R. Sussmann, M. Van Roozendael, T. Wagner, S. Wood, and M. Yela, Geophysical Validation of SCIAMACHY NO<sub>2</sub> Vertical Columns: Overview of Early 2004 Results, in *Proceedings of the Second Workshop on the Atmospheric Chemistry Validation of ENVISAT (ACVE-2), ESA-ESRIN, Frascati, Italy, 3-7 May 2004 (ESA SP-562, August 2004) ESC01JL2.*

Mahieu, E., P. Duchatelet, R. Zander, P. Demoulin, C. Servais, C.P. Rinsland, M.P. Chipperfield, and M. De Mazière, The evolution of inorganic chlorine above the Jungfraujoch station: An update, in *Ozone, Vol. II, Proceedings of the XX Quadrennial Ozone Symposium, Kos, Greece, 1-8 June 2004, pp. 997-998, 2004.*

#### **Edited books**

Zander, R., E. Mahieu, P. Duchatelet, P. Demoulin, F. Mélen, and C. Servais, Monitoring of the Variability and Long-term Evolution of Tropospheric Constituents by Infrared Solar Absorption Spectrometry at the Jungfraujoch, Switzerland, in *"Sounding the Troposphere from Space: A New Era for Atmospheric Chemistry", P. Borrell, P.M. Borrell, J.P. Burrows, and U. Platt, Eds., ISBN 3-540-40873-8, Springer-Verlag Berlin Heidelberg, pp. 407-416, 2003.*

Address:

---

Institut d'Astrophysique et de Géophysique - Université de Liège  
allée du VI août, 17 - Bâtiment B5a  
B-4000 Sart Tilman (Liège, Belgique)

Contacts:

---

Luc Delbouille	Tel. +32 4 342 2594	e-mail: delbouille@astro.ulg.ac.be
Philippe Demoulin	Tel. +32 4 366 9785	e-mail: demoulin@astro.ulg.ac.be
Pierre Duchatelet	Tel. +32 4 366 9786	e-mail: duchatelet@astro.ulg.ac.be
Emmanuel Mahieu	Tel. +32 4 366 9786	e-mail: mahieu@astro.ulg.ac.be
Ginette Roland	Tel. +32 4 342 2594	e-mail: roland@astro.ulg.ac.be
Christian Servais	Tel. +32 4 366 9784	e-mail: servais@astro.ulg.ac.be
Rodolphe Zander	Tel. +32 4 366 9756	e-mail: zander@astro.ulg.ac.be

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

