

## **The “Laboratory of Atmospheric and Solar Physics” of the University of Liège at the Jungfraujoch**

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A general description of the laboratory, its history, equipments and research activities has been published in [16] and will only be briefly summarized and updated here.

Research carried out at the Jungfraujoch by the University of Liège (ULg) group during the last decades mainly dealt with the monitoring of the variability and long-term evolution of the chemical composition of the atmosphere above the HFS, and contributed with a recognized success to the characterization of the Earth atmosphere at the mid-latitudes of the northern hemisphere. The activities are supported by the Belgian FNRS and Belgian national programs as well as included in European research efforts closely connected with the programs of the World Climate Research Program and of the International Geosphere-Biosphere Program. They have been, since 1989, integrated in a global context after the recognition of the Jungfraujoch site as one of the essential components of the primary Alpine Station of the NDSC (Network for the Detection of Stratospheric Change). This network plays an important role in the international research effort to follow permanently the changes of the atmospheric composition due to human activities; it now includes five primary stations and more than forty complementary stations, distributed all over the world.

The observations are obtained by Fourier Transform Spectrometry, recognized as the most efficient way to achieve very high resolution and obtain very high quality spectra in the infrared, particularly for atmospheric research [15].

Two Fourier transform spectrometers are installed at the Jungfraujoch.

The first one, developed in Liège, operates routinely since 1984 at the Coudé focus of the 76-cm diameter telescope of the HFS. Two sets of optics can be used: CaF<sub>2</sub> beamsplitter with InSb detector, or ZnSe beamsplitter with HgCdTe detector, allowing observations respectively in the 2 – 5.5 μm or in the 5.5 – 14 μm spectral domain. Both of these detectors work at liquid nitrogen temperature (the liquid N<sub>2</sub> is produced at the HFS).

The second one, a commercial instrument Bruker 120 HR, is installed on the first floor of the Sphinx building. The solar beam is collected on the upper terrace by a computer-controlled coelostat following the Sun automatically by computing at any time its position, taking into account the refraction of the atmosphere when the Sun is close to the horizon. The beamsplitter is made of KBr, allowing observations with both InSb and HgCdTe detectors, also cooled by liquid nitrogen.

Using the Sun as source of background radiation, they provide absorption spectra of the atmosphere in five domains (between 2 and 14 micrometers) containing the "signatures" of more than 20 different gases playing a role in the atmospheric physics and chemistry, as well as many isotopic species of some of these gases.

During the past decades, measurements at the Jungfraujoch have been performed as regularly as possible throughout the year, with observers being present at the site during about 250 days per year.

The analysis of the spectra is performed with the SFIT 1.09 algorithm [14]. This program allows determining the abundances above the Jungfraujoch of the different gases, by computing a synthetic spectrum and adjusting it to the observed spectrum in a non-linear least-squares iterative procedure. The line-by-line calculations use a 29-layer atmospheric model extending from the surface to 100 km. A Voigt line shape is assumed and the spectroscopic parameters are read from the HITRAN database [17]. Pressure and temperature vertical profiles for the atmosphere above the Jungfraujoch are obtained from the National Centers for Environmental Prediction (NCEP).

The University of Liège team is particularly interested in three groups of molecules:

a.- Those permitting to quantify the impact of human activities on the erosion of the ozone layer in the stratosphere, in particular HCl, ClONO<sub>2</sub>, HNO<sub>3</sub>, NO, NO<sub>2</sub>, HF and COF<sub>2</sub>; ozone O<sub>3</sub> is evidently also part of these studies. The subsequent evaluation of the budget of chlorine-containing species will give the possibility to check how adequate are the restrictions imposed by the Montreal Protocol (and its successive amendments) in the production and use of various chlorine-bearing source gases (in particular the CFCs and HCFCs). Proposed substitution species (notably HFCs and FCs) will affect the fluorine budget, whereas the evolution of the budget of nitrogen containing species will reflect the small increase of the tropospheric N<sub>2</sub>O concentration observed since the beginning of the industrial era.

b.- A series of other gases, participating in the greenhouse effect, thus playing a major role in the determination of the Earth future climate, a particularly current concern. Their tropospheric abundances, steadily increasing, must be carefully monitored. Amongst these, a particular interest is given to 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> and CHClF<sub>2</sub>.

c.- Various atmospheric constituents released at the ground, i.e., CO, C<sub>2</sub>H<sub>2</sub>, C<sub>2</sub>H<sub>6</sub>, OCS, HCN, H<sub>2</sub>CO and H<sub>2</sub>O, affecting the oxidization processes in the troposphere and the stratosphere, or appearing as important precursors in tropospheric ozone production.

Measuring the abundances of these species is difficult, due to spectral interferences not always sufficiently well quantified; new laboratory measurements are obviously needed to progress.

One follows also, evidently, the production and release of the new products developed to replace those already banned, or likely to be banned in a near future, in order to detect as soon as possible their “signatures” in the observed spectra and to start monitoring them as well.

After proper data analysis, all the results are archived in two internationally recognized data centers: NILU (the Norwegian Institute for Air Research) and the NDSC center of NOAA (National Oceanic and Atmospheric Administration) in the United States. In this context, we like to mention the participation of our group in campaigns aiming to check the necessary cohesion between all the observations within the NDSC. The recent Jungfraujoch results are regularly reported in the WMO scientific assessments as [18].

Until now, the retrieved data have consisted in vertical column abundances. However, ongoing developments of sophisticated retrieval algorithms will progressively allow to further provide information on the distribution versus altitude for a number of the species listed above; a complete re-analysis of all archived spectra will be undertaken as soon as these retrieval tools have been tested and adopted by the NDSC.

The ULg group has also participated to specific campaigns, such as EASOE, SESAME, THESEO and THESEO-2000 organized by the European Community, generally aiming at the study of chemical and/or dynamic atmospheric processes.

Further activities have been devoted to the validation of various atmospheric space observations: ATMOS and CRISTA on the Shuttle, HALOE and CLAES on board of the UARS satellite, MAPS and TOMS on NASA satellites, GOME on board of the European ERS-2 satellite and the MOPITT experiment on board of the NASA satellite TERRA. We were part of the COSE project, actually in its final phase, devoted to a compilation of atmospheric data obtained from the ground to support observations from space above Europe.

In the near future, specific support is planned in favor of the atmospheric chemistry experiments aboard ENVISAT (GOMOS, MIPAS and SCIAMACHY, in mid-2001), METOP-1 (IASI) and Sci-SAT (ACE).

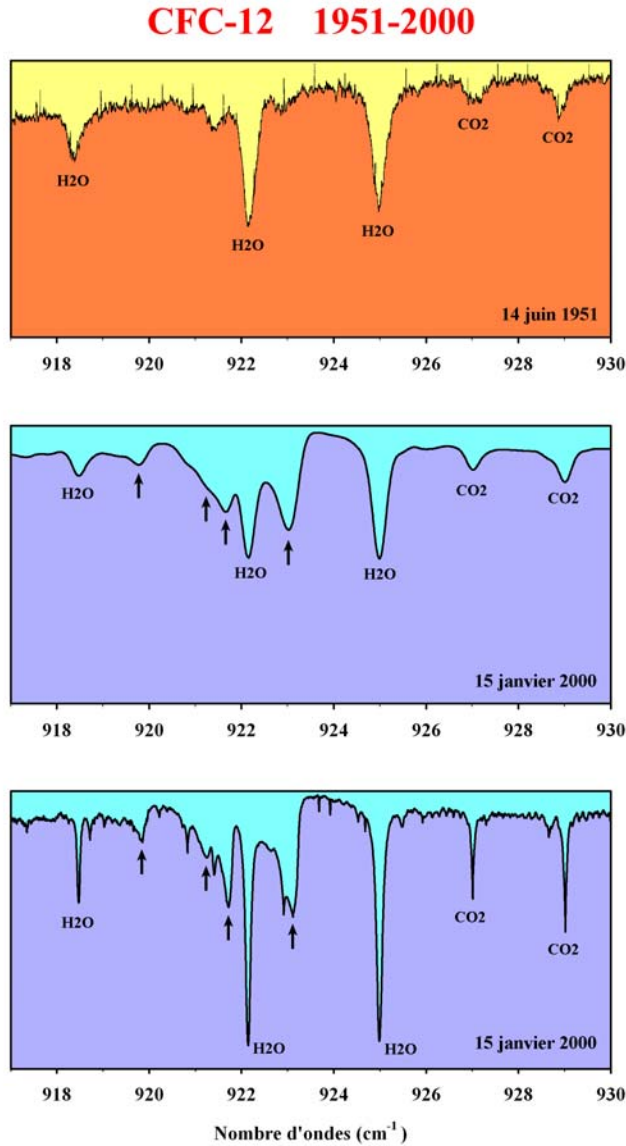
### Activities during 2000

During 2000, the Liège group, helped by colleagues from the Observatoire Royal de Belgique and from the Institut d'Aéronomie Spatiale de Belgique, was present at the Jungfraujoch during 258 days, among which good weather conditions enabled observations on 115 days.

The main activity on the site was to pursue the monitoring of the abundances of the already mentioned key stratospheric constituents, mostly within the frame of the NDSC program. The group also supported the THESEO-2000 campaign, during the entire year.

A symposium "*Observations atmosphériques dans les Alpes suisses en appui au Développement Durable*" has been organized on May 11, in Liège, to celebrate the 50<sup>th</sup> anniversary of the first observations of Marcel Migeotte at the Jungfraujoch, as well as the 50 years of presence of the Liège group in the station. It was also the occasion to stress how atmospheric research is a long tradition amongst the group who discovered the presence in the atmosphere of CO and CH<sub>4</sub> in 1948, of HF in 1974.

Figure 1 shows an example of a 1951 observation compared to a recent one. The upper frame of the figure shows an observation by M. Migeotte in 1951, in a spectral domain where CFC-12 absorbs [CFC-12 (CCl<sub>2</sub>F<sub>2</sub>) is one of the main gases contributing to chlorine release in the stratosphere, with the latter destroying the ozone catalytically]. The lowermost frame is a spectrum of the same domain, recorded in 2000 (note the improvement in the quality). For an easier comparison, the resolution of the 2000 spectrum has been degraded to that of the 1951 observation (middle frame). The obvious new absorptions, indicated by arrows, are produced by the CFC-12, which was essentially absent from the atmosphere in 1951.



**FIGURE 1**

Figure 2 reproduces an excerpt of the databases derived from the analysis of observations carried out at the Jungfraujoch during the past decade; it shows daily averaged vertical column abundances of seven stratospheric species (HF, HCl, ClONO<sub>2</sub>, HNO<sub>3</sub>, NO, NO<sub>2</sub> and O<sub>3</sub>), as well as those of the long-lived source gas N<sub>2</sub>O which is used as a tracer of atmospheric circulation and dynamics. These databases allow to determine seasonal variations (easily seen on the graph for ozone, NO and NO<sub>2</sub>), as well as long-term changes affecting some of these species, e.g. HCl and HF. Short-term variability which occurs mainly during the winter-spring-time period is also noticeable in Figure 2; for example, during the first part of 1999, we have observed intrusions of Arctic polar air over the station, characterized by record-high values for O<sub>3</sub>, HNO<sub>3</sub>, ClONO<sub>2</sub>, HCl and HF, anti-correlated with low N<sub>2</sub>O columns. Details on the timely rates of changes derived from these databases can be found in [5].

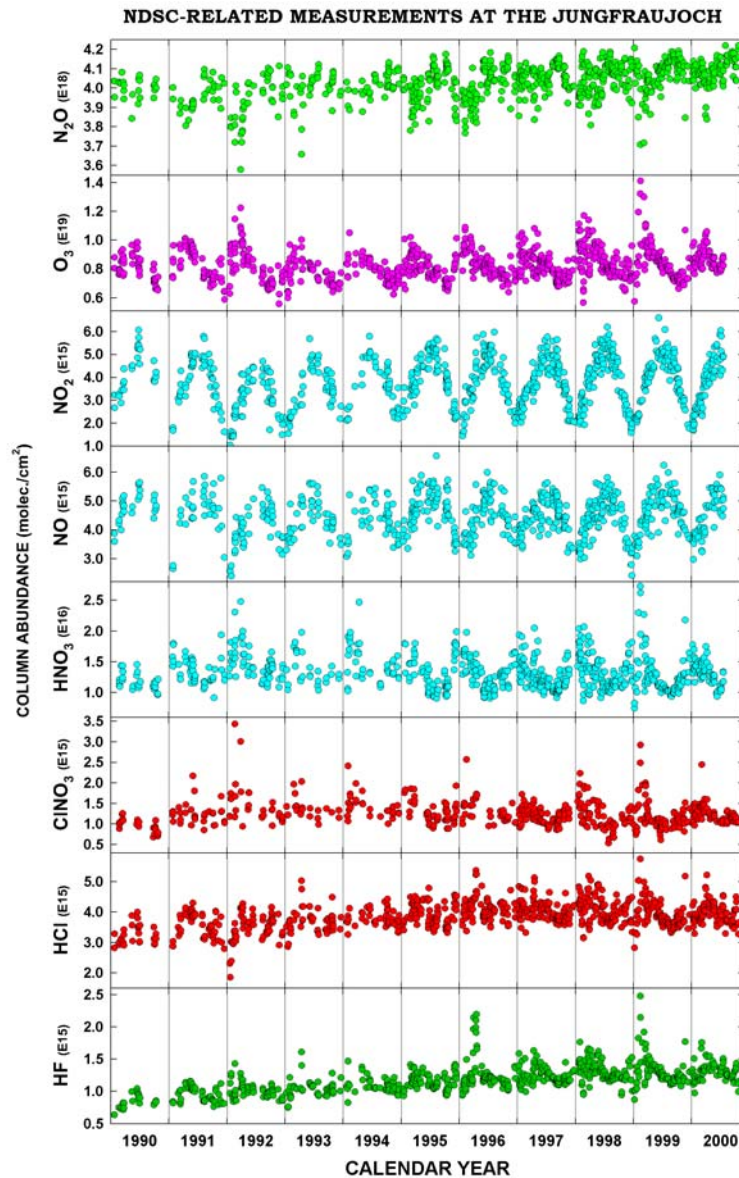


FIGURE 2

To improve long-term trend determinations, we need to remove the effect of short-term variability; this is achieved by considering measurements collected during the "quietest months" of June to November. An example of such an evaluation is given in the Figure 3, extracted from [11]. It shows the evolutions above the Jungfraujoch station of the June to November monthly mean vertical column abundances of the two main chlorine reservoirs, HCl and ClONO<sub>2</sub>. The curves fitted to these data points peak in 1997.1 and 1994.8 respectively for HCl and ClONO<sub>2</sub>. The inorganic chlorine (Cl<sub>y</sub>) budget has been evaluated by summing the HCl, ClONO<sub>2</sub> and ClO individual contributions, the latter being estimated from MLS/UARS measurements at northern mid-latitudes. Altogether, these three species account for nearly 95% of the inorganic chlorine. The Cl<sub>y</sub> curve shows its maximum occurrence around mid-1996, i.e. about 3.5 years after the northern hemisphere organic chlorine (CCl<sub>y</sub>) maximum determined from ground-based *in situ* measurements [WMO 1998, report 44]. See [11] for further details. The decrease of the stratospheric chlorine loading results from the effective application of the regulations adopted by the Montreal Protocol and its

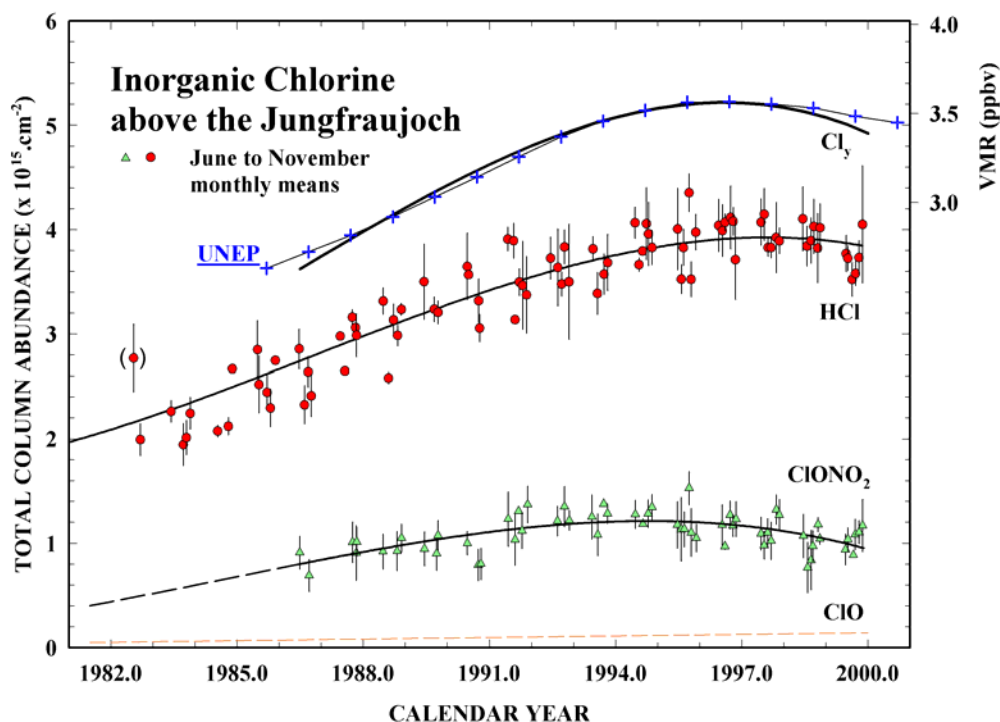


FIGURE 3

successive amendments. Further extension of the time series reproduced in the figure 3 will help to precise the  $Cl_y$  maximum determination.

As mentioned, the increase of the abundances of the gases controlling the greenhouse effect is another major environmental concern. The analysis of our observations allows determining the vertical column abundances of four important species concerned by the Kyoto Protocol, i.e.  $CO_2$ ,  $CH_4$ ,  $N_2O$  and the very long-lived  $SF_6$ . Figure 4 shows the evolution of their abundances during the last fifteen years: all of them are unambiguously on the rise. Description of trend analysis of these time series

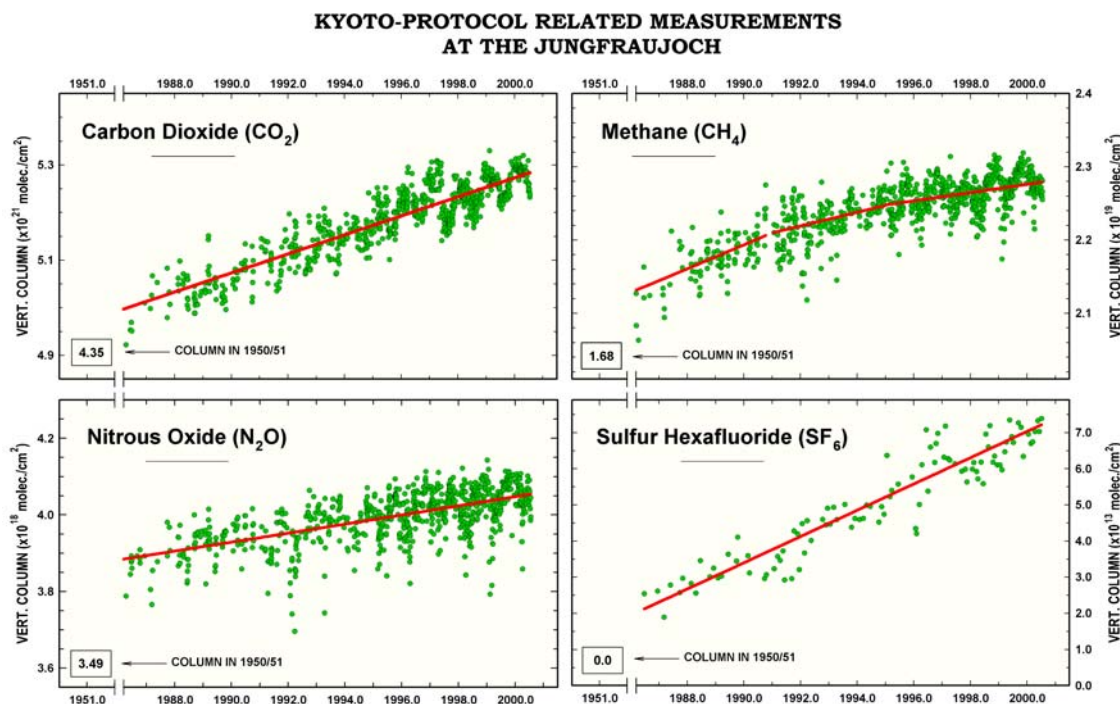


FIGURE 4

can be found in Zander *et al.* [2000a], except for CO<sub>2</sub> whose trend is 0.4%/yr. It is also very interesting to compare the actual abundances of these gases with the ones derived from the pioneering IR observations carried out by M. Migeotte, at the same site, in the early fifties (given in boxes at the lower left part of each frame).

We like to already mention that collaboration with EMPA (the Swiss Federal Institute for Material Testing and Research) has been decided, in the frame of the European Community project SOGE (System for Observation of Greenhouse Gases in Europe), starting in 2001. This project will develop a long-term European observation system for halocarbons and assess impacts of the halocarbons on the climate and on the ozone layer.

We have also developed new drive electronics for the 76-cm telescope located at the Sphinx. Due to the venerable age (35 years) of the previous electronics, this modification was primarily driven by reliability concerns. However, it was a good opportunity to implement at the same time different interesting improvements.

The new drive system uses a PC with a flat monitor and custom synthesizer, interface and power cards. This highly integrated control system uses only a small fraction of the volume needed by the old one and about 2 cubic meters could be reclaimed this way. This small size also made it possible to place the control system close to the observers in the coudé room. The telescope control program takes care not only of the movement of stars but is now capable of following comets, planets, the Sun or the Moon, taking into consideration the air refraction effects close to the horizon. This is a very important improvement, since the telescope is also used down to the horizon as suntracker for one of our IR interferometers.

The driving software has also been designed in such a way that it is remotely possible to verify the status of the telescope in case of problems with the possibility, if further implemented, to control its movement over the Internet from the experiments themselves.

In September, the Jungfraujoch station received the visit of the NDSC Steering Committee. Each group had the pleasure to show its instrumentation and comment its research program, and the visiting colleagues had the opportunity to see that the Sphinx building is now, practically, the location of a complete NDSC station.

#### **Recent publications** (the names of members of the Liège group are underlined)

[1] Barret, B., E. Mahieu, M. Carleer, M. De Mazière, R. Colin, and R. Zander, Tropospheric boundary layer investigations by differential ground-based solar FTIR spectrometry, in *Environmental Sensing and Applications, SPIE Proceedings 3821*, pp. 116-123, 1999.

[2] Delbouille, L. and Zander, R., Contribution to “*Envisat MIPAS –Michelson Interferometer for Passive Atmospheric Sounding- an instrument for atmospheric chemistry and climate research*”, ESA SP-1229, March 2000, Noordwijk, The Netherlands.

[3] De Mazière, M., O. Hennen, M. Van Roozendael, P. Demoulin, and H. De Backer, Daily ozone vertical profile model built on geophysical grounds, for column retrieval from atmospheric high-resolution infrared spectra, *J. Geophys. Res.*, *104*, 23855-23869, 1999.

[4] Ingold, T., B. Schmid, C. Mätzler, Ph. Demoulin, and N. Kämpfer, Modeled and empirical approaches for retrieving columnar water vapor from solar transmittance measurements in the 0.72, 0.82, and 0.94 micron absorption bands, *J. Geophys. Res.*, *105*, 24327-24343, 2000.

[5] Mahieu, E., R. Zander, P. Demoulin, M. De Mazière, F. Mélen, C. Servais, G. Roland, L. Delbouille, J. Poels, and R. Blomme, Fifteen years-trend characteristics of key stratospheric constituents monitored by FTIR above the Jungfraujoch, in *Proceedings of the "Fifth European Symposium on Stratospheric Ozone"*, St. Jean de Luz, France, September 27 - October 1, 1999, N. R. P. Harris, M. Guirlet and G. T. Amanatidis Eds, pp. 99-102, 2000.

[6] Rinsland, C. P., E. Mahieu, R. Zander, P. Demoulin, J. Forrer, and B. Buchmann, Free tropospheric CO, C<sub>2</sub>H<sub>6</sub> and HCN above central Europe: Recent measurements from the Jungfraujoch station including the detection of elevated columns during 1998, *J. Geophys. Res.*, *105*, 24235-24249, 2000.

[7] Zander, R., P. M. Midgley, and M. J. Kurylo, The NDSC in support of satellite data validation and calibration, in *Proceedings of the ESA "European Symposium on Atmospheric Measurements from Space"*, ESTEC-Nordwijk, The Netherlands, January 18-22, 1999, WPP-161, Vol. 2, pp. 649-654, 1999a.

[8] Zander, R., E. Mahieu, P. Demoulin, C. Servais, F. Mélen, G. Roland, and L. Delbouille, Spectrometric solar observations at the Jungfraujoch for long-term atmospheric monitoring, in *Actes du Colloque "Ozone dans la Troposphère: la Recherche et la Politique"*, Bruxelles, 26 juin 1998, SSTC-D/1999/1191/3, pp. 69-76, 1999b.

[9] Zander, R., E. Mahieu, P. Demoulin, C. Servais, and F. Mélen, Long-term evolution of the loading of CH<sub>4</sub>, N<sub>2</sub>O, CO, CCl<sub>2</sub>F<sub>2</sub>, CHClF<sub>2</sub> and SF<sub>6</sub> above Central Europe during the last 15 years, in *Proceedings of the "Second International Symposium on Non-CO<sub>2</sub> Greenhouse Gases - Scientific Understanding, Control and Implementation"*, Noordwijkerhout, The Netherlands, September 8-10, 1999, Sp. Vol. *Environmental Monitoring and Assessment*, 2000 Kluwer Academic Publishers, pp. 211-216, 2000a.

[10] Zander, R., E. Mahieu, F. Mélen, and J. Elkins, An evaluation of the northern mid-latitude tropopause heightening, based on N<sub>2</sub>O column abundance measurements above the Jungfraujoch, in *Proceedings of the "Fifth European Symposium on Stratospheric Ozone"*, St. Jean de Luz, France, September 27 - October 1, 1999, N. R. P. Harris, M. Guirlet and G. T. Amanatidis Eds, pp. 135-138, 2000b.

### **To Appear**

[11] Mahieu, E., R. Zander, F. Mélen, P. Demoulin, C. P. Rinsland, and J. M. Russell III, Monitoring the stratospheric chlorine budget during the past decades: the Montreal Protocol at work, in *Proceedings of the "International Quadrennial Ozone Symposium"*, Sapporo, Japan, 3-8 July 2000, to appear.

[12] Rinsland, C. P., A. Goldman, R. Zander, and E. Mahieu, Enhanced tropospheric HCN columns above Kitt Peak during the 1982-1983 and 1997-1998 El Niño Warm Phases, *J. Quant. Spectrosc. Radiat. Transfer*, submitted for publication.

[13] Zander, R., The Current NDSC Status – Supporting capabilities for European satellite missions, to appear in *Proceedings of the “1999-ATMOS-C1 Workshop”*, Sapporo, Japan, October 11, 1999.

## References

[14] C.P. Rinsland, R. Zander, P. Demoulin. Ground-based infrared measurements of HNO<sub>3</sub> total column abundances: long-term trend and variability. *J. Geophys. Res.* 96, 9379-9389, 1991

[15] Delbouille L. The role of very high resolution FTS in recent determinations of the quantitative composition of the stratosphere. *SPIE Proceedings* 2089, 32-37, 1994

[16] Delbouille L. and G.Roland. High resolution solar and atmospheric spectroscopy from the Jungfrauoch high-altitude station. *Optical Engineering* 34 (9), 2736-2739, 1995

[17] L.S. Rothman et al. The HITRAN molecular spectroscopic database and HAWKS: 1996 edition. *J. Quant. Spectrosc. Radiat. Transfer* 60, 5, 665-710, 1998

[18] Prinn, R. G., R. Zander, J. W. Elkins, P. J. Fraser, M. K. W. Ko, D. Cunnold, A. Engel, M. R. Gunson, E. Mahieu, P. M. Midgley, J. M. Russell III, and R. F. Weiss, Long-lived ozone-related compounds, *Scientific Assessment of Ozone Depletion: 1998*, WMO Report 44, pp. I-1 to I-54, World Meteorological Organization, P.O. Box 2300, Geneva 2, CH 1211, Switzerland, ISBN: 92-807-1722-7, 1999.

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