

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:

Contribution to the long-term monitoring of the Earth's atmosphere has remained the central activity of the Liège group. Regular observations carried out at the Jungfraujoch with high-performance Fourier-transform infrared (FTIR) spectrometers allow to derive abundances of more than 25 constituents affecting our climate and monitored in the frame of the Kyoto protocol ( $N_2O$ ,  $CH_4$ ,  $CO_2$ ,  $SF_6$ ,  $CF_4$ ...); related to the erosion of the ozone layer in the stratosphere and therefore linked to the Montreal Protocol (HCl,  $ClONO_2$ ,  $HNO_3$ , NO,  $NO_2$ , HF,  $COF_2$ ,  $O_3$ ,  $CCl_2F_2$ ,  $CHClF_2$ ,  $CCl_3F$ ,  $CCl_4$ ...); or altering the oxidization processes in the troposphere ( $CO$ ,  $C_2H_2$ ,  $C_2H_6$ , OCS, HCN,  $H_2CO$ ,  $H_2CO_2$ ...). The resulting databases allow the precise determination of the short-term variability, seasonal modulations, inter-annual as well as long-term changes affecting most of these species.

During 2012, Liège observers were present during 245 days at the Jungfraujoch and recorded 2115 high-resolution FTIR solar spectra on 116 different days, including 20 days with remote operation from Liège. Here is a selection of key 2012 results derived by our team from Jungfraujoch infrared spectra:

**Ethane ( $C_2H_6$ )**

Ethane is the most abundant non-methane hydrocarbon of the Earth's atmosphere, with a lifetime of approximately 2 months.  $C_2H_6$  has both anthropogenic and natural emission sources such as biomass burning, natural gas loss and bio-fuel consumption.

Thanks to new spectroscopic parameters (Harrison *et al.*, 2010), ethane retrievals have been significantly improved and two partial columns (lower troposphere and upper-troposphere/lower-stratosphere UTLS) can now be derived from Jungfraujoch FTIR spectra. Figure 1 displays the retrieved  $C_2H_6$  total and partial columns above the Jungfraujoch from Septembre 1994 onwards. Overall decrease of ethane since 1994 amounts to -14 %, -9 % and -39 %, respectively for total, lower troposphere and UTLS columns. A strong seasonal variation is observed, with a maximum in February and peak-to-peak amplitude of 50 % for the total columns. See Bader *et al.* [1] for more details.

**Carbonyl sulfide (OCS)**

Carbonyl sulfide is the most abundant sulfur-containing trace gas in the atmosphere and is believed to account for a substantial portion of the sulfur in the stratospheric aerosol layer which influences the Earth's radiation budget and stratospheric ozone chemistry. The major identified OCS sources are oceans and anthropogenic emissions, while atmospheric loss and uptake by vegetation and soils constitute the main OCS sinks. The uptake by vegetation strongly influences the distribution and seasonality of OCS throughout most of the Northern Hemisphere.

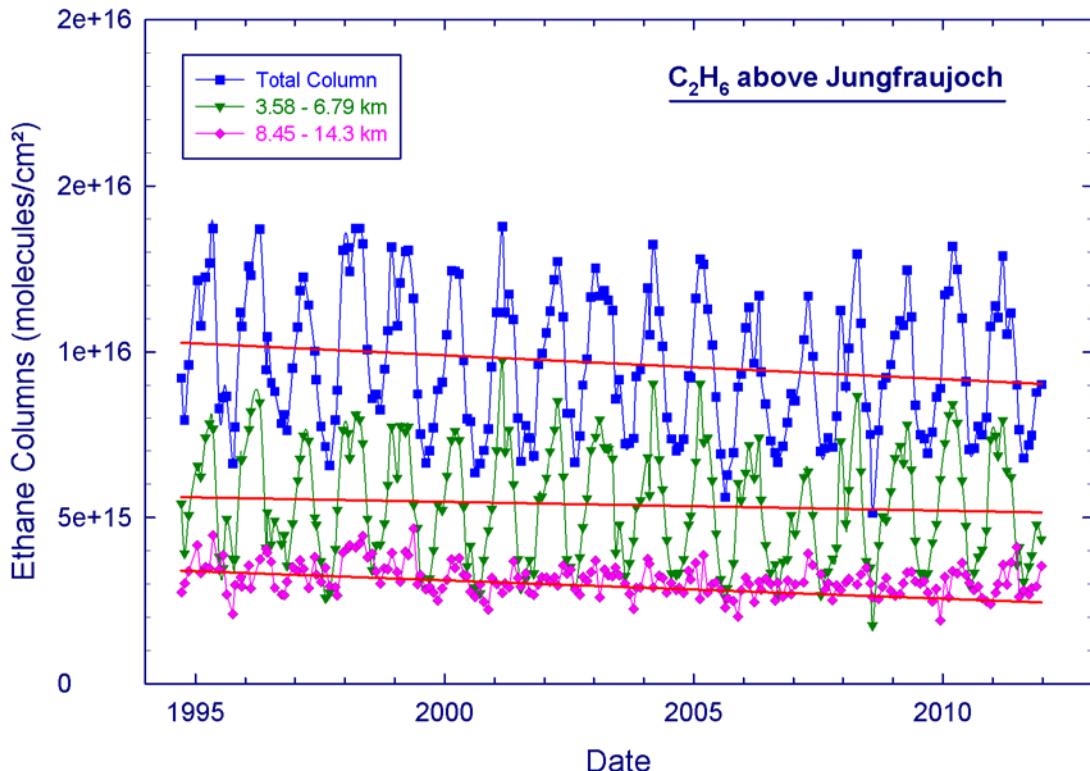


Figure 1: Time series of  $C_2H_6$  monthly mean total (in blue), low-tropospheric (3.58-6.79 km, in green) and UTLS (8.45-14.3 km, in pink) columns above the Jungfraujoch. Red lines are linear trends.

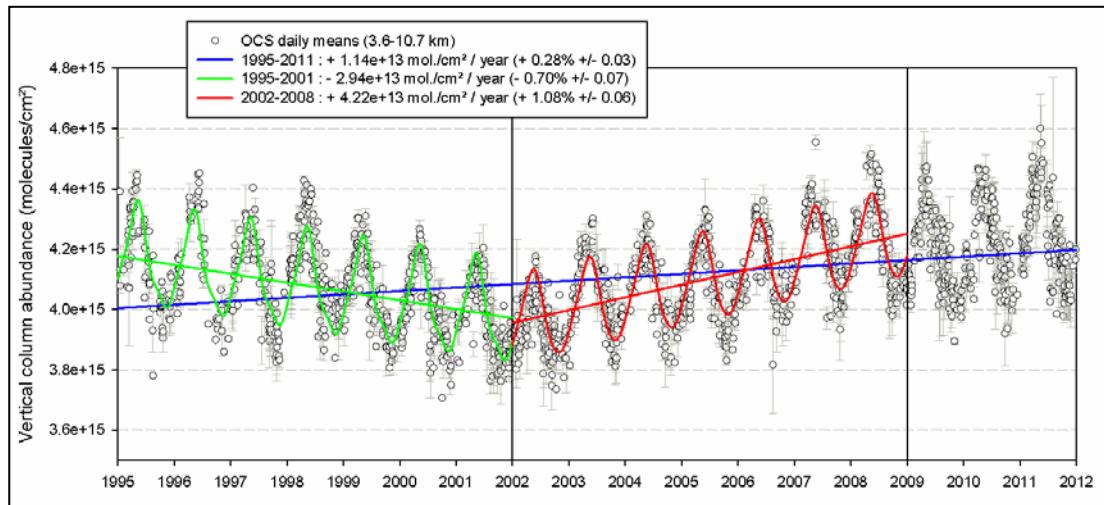


Figure 2: Time series of OCS tropospheric columns above the Jungfraujoch (daily means values). From 1995 to 2001, carbonyl sulfide decreased at a rate of  $0.70 \pm 0.07\%$  per year; from 2002 to 2009, it increased at  $1.08 \pm 0.06\%$  per year.

There is still a lack of understanding of the strength of some components in the atmospheric OCS budget. It seems that uptake by plants has been strongly underestimated, suggesting that additional significant OCS sources still have to be identified. The study of this gas is thus important and it will help improving our current estimates of OCS budget. Furthermore, recent works suggest that OCS could provide a valuable constraint on photosynthesis and so improve studies of carbon cycle processes.

A new retrieval strategy has been developed to retrieve OCS from Jungfraujoch FTIR spectra: it uses 4 micro-windows in the 2047-2055 cm<sup>-1</sup> spectral domain ( $v_3$  band), and the

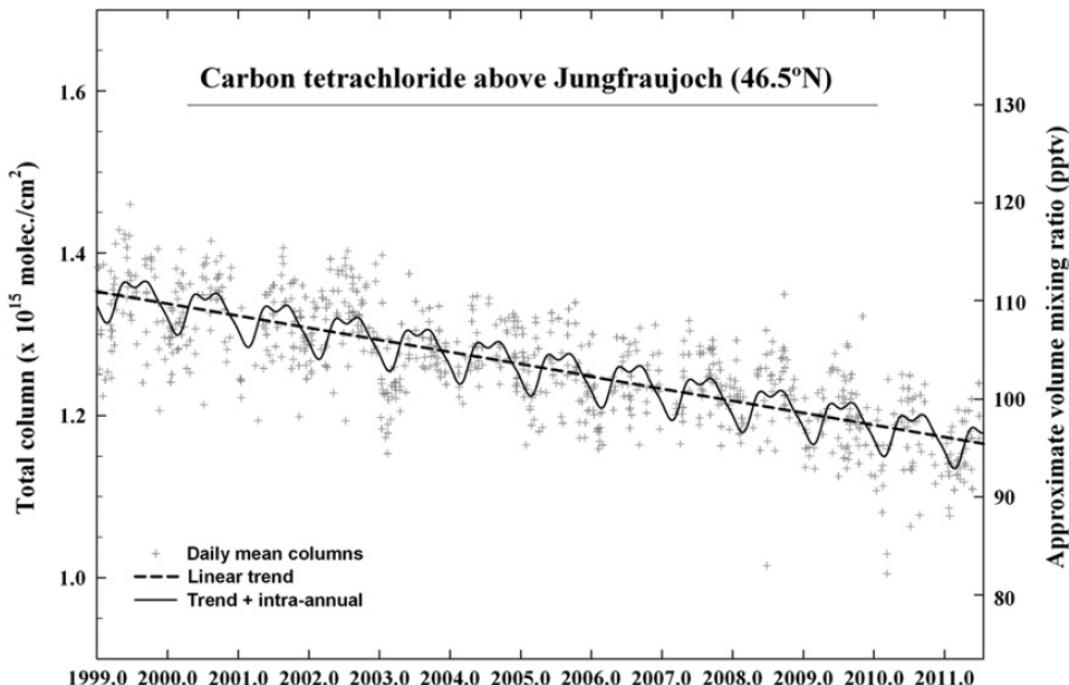
information analysis for this retrieval indicates a degree of freedom of about 2.6, meaning that we can derive at least 2 significant partial columns from the spectra.

Figure 2 shows the tropospheric OCS columns above the Jungfraujoch: after a few years of decrease, these columns have been increasing since 2002. Reasons of this increase have been investigated, with possible sources being the intensification of emissions from coal combustion and from aluminium production (respectively 30 % and 65 % of the estimated anthropogenic direct OCS emissions). See Lejeune *et al.* [4] for more information.

### Carbon tetrachloride $\text{CCl}_4$

The carbon tetrachloride gas, emitted at the ground, has been and remains a key component of the stratospheric chlorine budget, still contributing over 10% to the total chlorine loading, and to the stratospheric ozone depletion by a similar percentage. It is also a potent greenhouse gas with a global warming potential relative to  $\text{CO}_2$  of 1400, on a 100-year horizon. Monitoring its atmospheric evolution remains, therefore, of relevance to both the Kyoto and the Montreal Protocol.

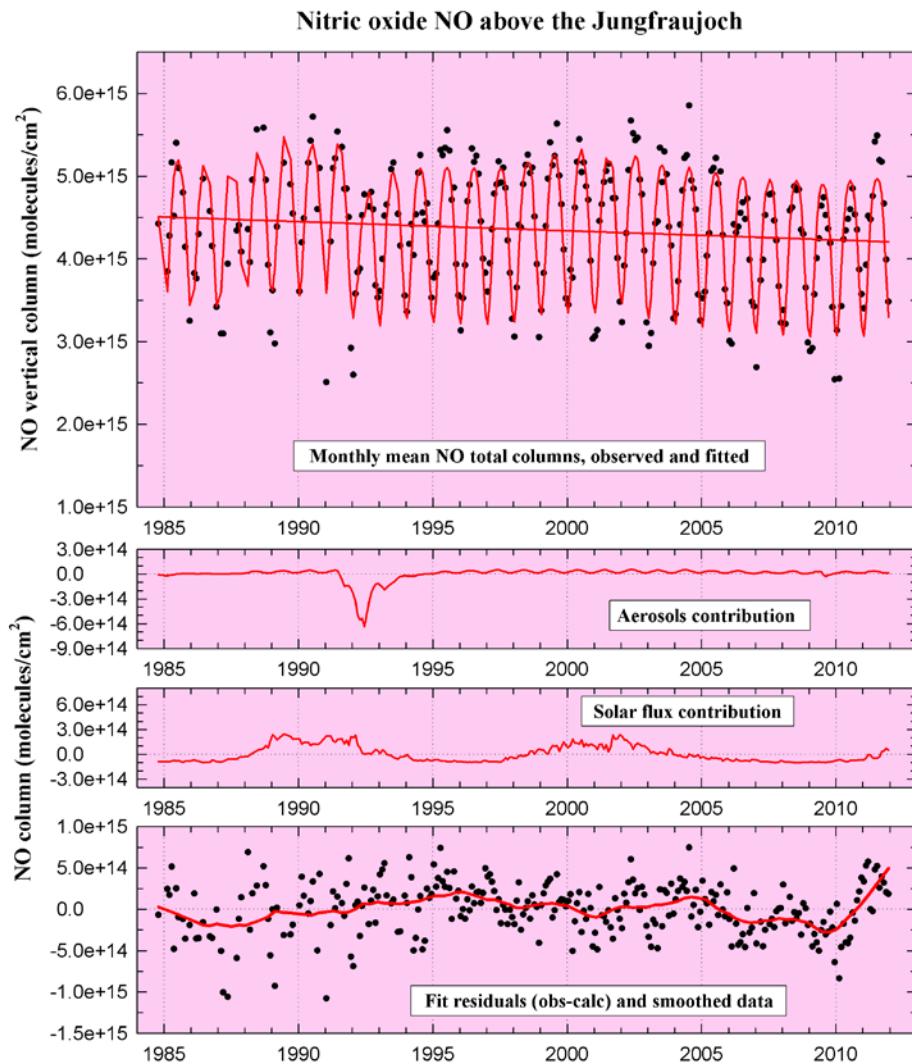
The long-term trend of the atmospheric  $\text{CCl}_4$  burden has been retrieved from FTIR spectra recorded at the Jungfraujoch between January 1999 and June 2011 (Figure 3). Total columns were derived from spectrometric analysis of the strong  $\text{CCl}_4$   $\nu_3$  band at  $794 \text{ cm}^{-1}$ . A significant improvement in the fitting residuals and in the retrieved  $\text{CCl}_4$  columns was obtained by taking into account line-mixing in a strong interfering  $\text{CO}_2$  Q-branch. A fit to the  $\text{CCl}_4$  daily mean total column time series returns a statistically-significant ( $2\sigma$ ) long-term trend of  $-1.49 \pm 0.08 \times 10^{13} \text{ mol/cm}^2$  per year. This corresponds to an annual decrease of  $-1.31 \pm 0.07$  pptv for the mean free tropospheric volume mixing ratio. Furthermore, the total column data set reveals a weak seasonal cycle with a peak-to-peak amplitude of 4.5 %, with minimum and maximum values occurring in mid-February and mid-September, respectively. This small seasonal modulation is attributed primarily to the residual influence of tropopause height changes throughout the year. The negative trend of the  $\text{CCl}_4$  loading reflects the continued impact of the regulations implemented by the Montreal Protocol and its strengthening amendments and adjustments. See Rinsland *et al.* [2] for details.



*Figure 3: Daily mean  $\text{CCl}_4$  total columns above the Jungfraujoch station are shown as plus symbols. The dashed line shows the linear component of the function fitted to all available daily means. The solid curve reproduces the derived seasonal modulation.*

### Total reactive nitrogen $\text{NO}_y$

The  $\text{NO}_y$  family of gases, defined as  $\text{NO} + \text{NO}_2 + \text{NO}_3 + 2\times\text{N}_2\text{O}_5 + \text{HNO}_3 + \text{HNO}_4 + \text{ClONO}_2 + \text{BrONO}_2$ , plays an important role in the ozone depletion (NO catalytic cycle, Crutzen 1970). At the Jungfraujoch observatory, FTIR spectrometers measure since 1984 the four most abundant members of  $\text{NO}_y$  i.e.  $\text{NO}$ ,  $\text{NO}_2$ ,  $\text{HNO}_3$  and  $\text{ClONO}_2$ . Their sum is a good proxy for  $\text{NO}_y$  (the most important missing gas being  $\text{N}_2\text{O}_5$ ). The trends of these four gases have been derived with a multiple regression model that includes linear trend, seasonal variation, stratospheric aerosol optical depth, solar flux... The apparent discrepancy between the  $\text{NO}_y$  trend and the trend of its source gas  $\text{N}_2\text{O}$  probably arises, according to the models, from the strengthening of the Brewer-Dobson circulation, the stratospheric cooling and the changes in stratospheric chlorine loading. Figures 4 to 7 show some results derived from this study. See Demoulin *et al.* [3] and Hendrick *et al.* [5] for more details.



*Figure 4: Upper frame: monthly mean NO total columns derived from Jungfraujoch FTIR spectra (black dots), together with best fit of the regression model and linear trend (red lines). A mean NO decrease of  $-0.25 \pm 0.12 \text{ \%}/\text{yr}$  has been derived for the 1985-2012 time period. Middle frames give the contribution of the aerosol optical depth and of the solar flux to the NO total columns. The large aerosol perturbation from the Mt. Pinatubo eruption on June 15, 1991, is clearly visible. The eleven-year solar cycle influences NO total columns by a few percents. Lower frames show the residuals of the fit (measured - model) (black dots) together with a smoothed curve (red lines) of these residuals.*

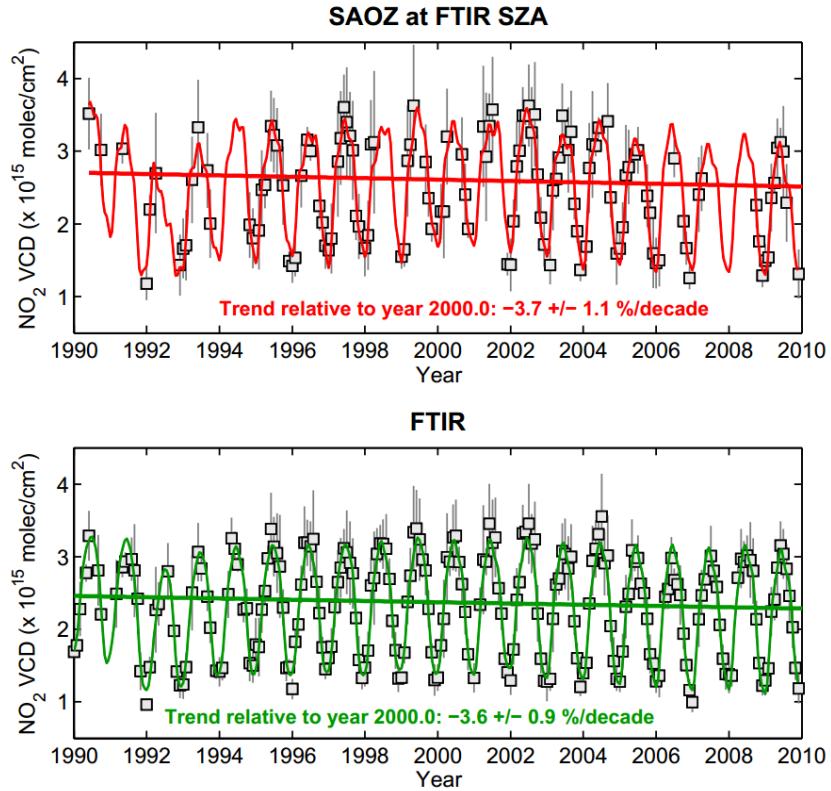


Figure 5: This figure shows the NO<sub>2</sub> vertical column time series of 2 co-located NDACC instruments: the ULg FTIR solar spectrometer and the BIRA-IASB SAOZ UV-vis instrument, both operating at the Jungfraujoch observatory. As NO<sub>2</sub> is varying during the day, twilight SAOZ measurements have been reported, for this comparison exercise, to the solar zenith angles of FTIR observations. Colored lines correspond to the linear trend (thick line) and to the NO<sub>2</sub> columns recalculated using the multiple linear regression model (thin line). Trends derived from both datasets,  $-3.7 \pm 1.1$  and  $-3.6 \pm 0.9$  %/decade, respectively for FTIR and SAOZ instruments, are in very good agreement.

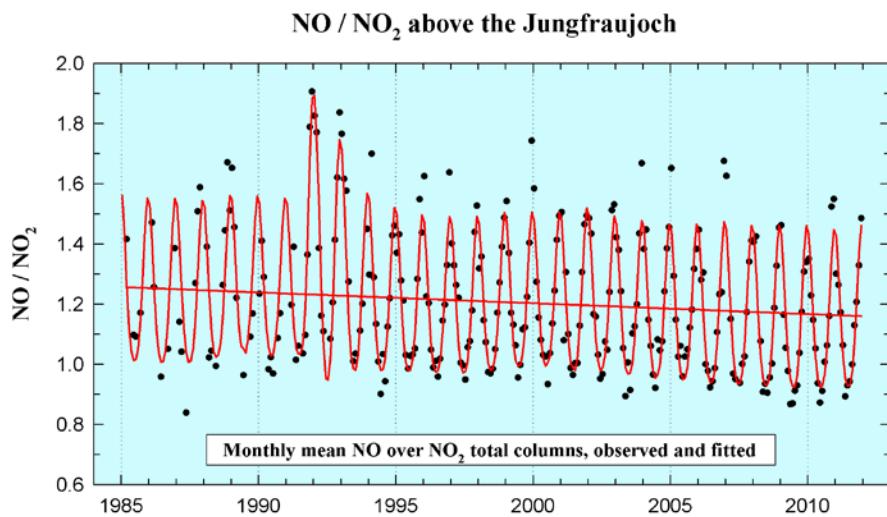


Figure 6: The NO/NO<sub>2</sub> ratio is slowly decreasing above the Jungfraujoch, at a rate of  $-0.29 \pm 0.12$  %/year. This decrease is due to increased chlorine loading in the atmosphere, which increases the rate of the reaction  $[NO + ClO \rightarrow NO_2 + Cl]$ . The NO/NO<sub>2</sub> ratio is expected to increase in the 21<sup>st</sup> century, as a result of the decreasing chlorine loading and of CO<sub>2</sub>-induced stratospheric cooling, which slows the temperature-dependent reaction  $[NO + O_3 \rightarrow NO_2 + O_2]$  (Revell 2012).

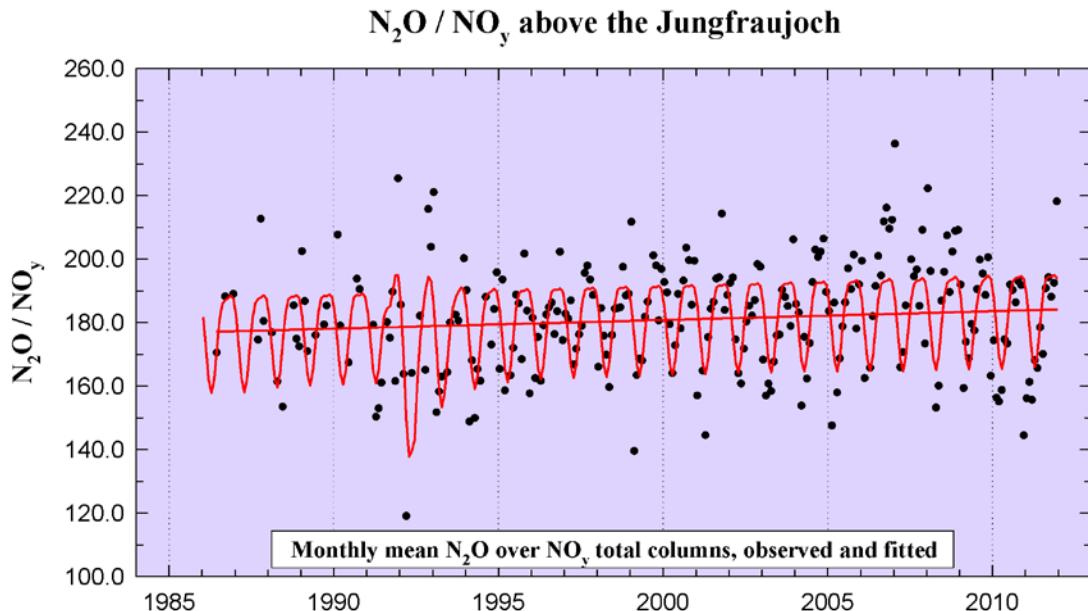


Figure 7: Although  $N_2O$  is the source of  $NO_y$ ,  $NO_y$  is not increasing at the same rate ( $0.31 \pm 0.02\%/\text{year}$  for  $N_2O$ ,  $0.11 \pm 0.13\%/\text{year}$  for  $NO_y$ ). This difference is probably due to increasing  $CO_2$  concentrations cooling the stratosphere (Rosenfield and Douglass 1998) and to ozone and halogens changes in the stratosphere (McLinden 2001).

### Varia

Results derived from the Jungfraujoch FTIR spectra are regularly archived at the NDACC data center (<ftp://ftp.cpc.ncep.noaa.gov/ndacc/station/jungfrau/>), usually within less than one year. However, our group is now involved in the NORS project, where we will store a first evaluation of our data at <ftp://ftp.cpc.ncep.noaa.gov/ndacc/RD/jungfrau/> (« Rapid Data Delivery », i.e. typically within less than 1 month).

In June 2012, our group organized at Wengen the annual infrared working group of the NDACC network. This meeting was attended by about 50 scientists from around the world, specialists in Fourier transform infrared spectroscopy applied to the long term study of the Earth atmosphere. It was followed by a visit to the Jungfraujoch laboratories.

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### Key words:

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

Internet data bases:

<ftp://ftp.cpc.ncep.noaa.gov/ndacc/station/jungfrau/>, <http://www.nilu.no/nadir/>  
<ftp://ftp.cpc.ncep.noaa.gov/ndacc/RD/jungfrau/>

Collaborating partners/networks:

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

Scientific publications and public outreach 2012:

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**

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## Conference papers

Bader, W., E. Mahieu, B. Bovy, B. Lejeune, P. Demoulin, and C. Servais, First retrievals of methanol ( $\text{CH}_3\text{OH}$ ) above Jungfraujoch (46.5°N): Optimization of the retrieval strategy and information content, poster presentation at the “Journée des doctorants UNITER”, Université Libre de Bruxelles, Belgique, December 10, 2012.  
<http://hdl.handle.net/2268/135878>

[1] Bader, W., A. Perrin, D. Jacquemart, J.J. Harrison, G.C. Toon, K. Sudo, O.A. Søvde, P. Demoulin, C. Servais, and E. Mahieu, Retrievals of ethane from ground-based high-resolution FTIR solar observations with updated line parameters: determination of the optimum strategy for the Jungfraujoch station, poster presentation at the “11th Atmospheric Spectroscopy Applications” meeting (ASA 2012), united with the “12th HITRAN Conference”, Reims, France, August 29-31, 2012.  
<http://hdl.handle.net/2268/129289>

Bader, W., A. Perrin, D. Jacquemart, K. Sudo, H. Yashiro, M. Gauss, P. Demoulin, C. Servais, and E. Mahieu, Retrievals of ethane from ground-based high-resolution FTIR solar observations with updated line parameters: determination of the optimum strategy for the Jungfraujoch station, poster presentation at the “EGU 2012 General Assembly”, Vienna, Austria, April 22-27, 2012 and at the 4th Symposium on METEOrology and CLIMatology for PhD students - MeteoClim2012, 2012.  
<http://hdl.handle.net/2268/117067>

Bader, W., A. Perrin, D. Jacquemart, K. Sudo, H. Yashiro, O.E. Søvde, P. Demoulin, C. Servais, and E. Mahieu, Retrievals of ethane from ground-based high-resolution FTIR solar observations with updated line parameters: determination of the optimum strategy for the Jungfraujoch station, poster presentation at the “NDACC-IRWG Annual Meeting”, Wengen, Switzerland, June 11-15, 2012.  
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[4] Lejeune, B., E. Mahieu, P. Demoulin, C. Servais, W. Bader, B. Bovy, O. Flock, R. Zander, and G. Roland, Trend evolution of carbonyl sulfide above Jungfraujoch deduced from ground-based FTIR and ACE-FTS satellite observations, oral presentation at the “ACE Science Team Meeting”, University of Waterloo, Waterloo, ON, Canada, May 23-24, 2012.  
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Mahieu, E., the GIRPAS- and the ACE-teams, Post-peak trend in inorganic chlorine ( $\text{Cl}_y$ ) from Jungfraujoch, ACE-FTS and HALOE infrared solar observations, oral presentation at the “ACE / SAGE III-ISS International Workshop”, Old Dominion University, Norfolk, VA, USA, October 15-17, 2012.  
<http://hdl.handle.net/2268/132600>

Mahieu, E., W. Bader, B. Bovy, B. Lejeune, C. Vigouroux, P. Demoulin, C. Servais, G. Roland, and R. Zander, Retrieval of methanol ( $\text{CH}_3\text{OH}$ ) from the high-altitude Jungfraujoch station (46.5°N): preliminary total column time series, long-term trend and seasonal modulation, poster presentation at the “NDACC-IRWG Annual Meeting”, Wengen, Switzerland, June 11-15, 2012.  
<http://hdl.handle.net/2268/124069>

Mahieu, E., W. Bader, B. Lejeune, C. Vigouroux, P. Demoulin, C. Servais, G. Roland, and R. Zander, Seeking for the optimum retrieval strategy of methanol ( $\text{CH}_3\text{OH}$ ) using ground-based high-resolution FTIR solar observations recorded at the high-altitude Jungfraujoch station (46.5°N), poster presentation at the “EGU 2012 General Assembly”, 22-27 April 2012, Vienna, Austria, April 22-27, 2012.  
<http://hdl.handle.net/2268/116957>

*International Foundation HFSJG*  
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Pastel, M., S. Godin-Beckmann, E. Mahieu, P. Demoulin, and K. Hocke, A new methodology for integrating ground-based ozone profile data, poster presentation at the Quadrennial Ozone Symposium QOS 2012, Toronto, Canada, August 27–31, 2012.

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Servais C. and Roland G., Soixante ans de spectroscopie solaire et atmosphérique depuis le Jungfraujoch, Swiss Academy of Sciences, Les chercheurs de l'extrême, Interlaken, October 25-26, 2012.

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### **Edited books**

Schneider, M., P. Demoulin, R. Sussmann, and J. Notholt, Fourier Transform Infrared Spectrometry, Chapter 6 in Monitoring Atmospheric Water Vapor: Ground-Based Remote Sensing and In-Situ Methods, ISSI Scientific Report Series, **vol. 10**, ISBN 978-1-4614-3908-0, Springer New York, 2012.

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