

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 our two 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 ...), 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 2010, Liège observers spent 228 days at the Jungfraujoch and recorded 2377 high-resolution FTIR solar spectra on 127 different days, including 22 days with spectra remotely recorded from Liège.

Various improvements have been applied to the remote observation system including the possibility to remotely insert a reference absorption cell in the instrument light beam. Regular measurements with this sealed cell containing HBr gas have also been realized, in order to characterize the instrumental line shape. This objectively warrants that the observations are performed consistently at the highest level of quality/performance.

In addition to the routinely retrieved constituents, here are a few examples where emphasis was placed in 2010:

"Kyoto species"

The 1997 "Kyoto Protocol on climate change" specifically targets – among other greenhouse gases – CO_2 , CH_4 , N_2O and SF_6 . They present characteristic infrared absorption features allowing to quantify their atmospheric abundances from the FTIR high-resolution broadband spectra. Analyses of the more recent Jungfraujoch observations – and complete re-investigations using improved retrieval strategies for CH_4 and N_2O – have resulted in updates of the datasets and of their temporal evolutions [Duchatelet 2010c]. Related time series, which cover more than two decades, are reproduced in Figure 1. The production of such long-term time series for carbon tetrafluoride (CF_4), another strong greenhouse gas whose anthropogenic emissions are also targeted by the Kyoto Protocol, is currently under progress.

The first obvious feature is the regular growth of these gases over the 1985-2008 time period. Even methane, which remained stable from 1999 to 2005, seems to be on the rise again.

Comparisons with abundances derived from pioneering observations performed in 1950-1951 at the same site by M. Migeotte indicate that the total columns of CO₂, CH₄ and N₂O have been respectively multiplied by 1.27, 1.41 and 1.19. More specifically, trend determinations reveal a yearly increase of 0.487 ± 0.004 % for CO₂ (reference year: 1984), in excellent agreement with *in situ* surface measurements (e.g. www.cmdl.noaa.gov).

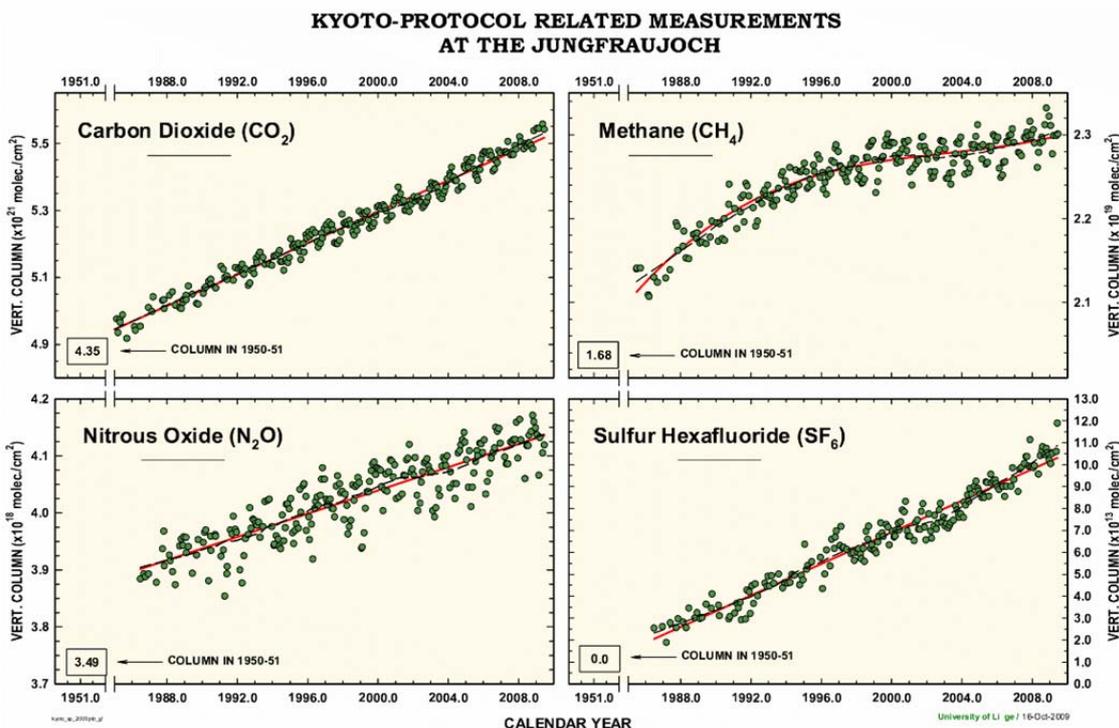


Figure 1. Long-term evolution of four species targeted by the Kyoto Protocol as derived from ground-based observations conducted at the Jungfraujoch station. Notice the different vertical axis units for each frame.

The very long-lived nitrous oxide (120 years) shows a similar behaviour, with an annual linear build up of $(1.07 \pm 0.03) \times 10^{16}$ molec./cm². This corresponds to an increase of 0.28 ± 0.01 %/year (reference year: 1984), also commensurate with *in situ* trend data.

Rapid growth of the total column abundance of SF₆ has been confirmed, with an annual load increase of 6.0 %/year (reference year: 2000) in the last years. 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. Its global warming potential is indeed extremely high, estimated at 22800 (100-year horizon) by IPCC, 2007.

Extrapolation of the Jungfraujoch data predicts tropospheric SF₆ concentrations of about 18 pptv in 2050 and about 32 pptv in 2100 (compare this to the 2.0 pptv concentration measured in 1988). These values are significantly lower than those reported in WMO-report 2002 scenarios, justifying further monitoring of this species to determine its effective future evolution and related climatic impact.

Another striking feature of this figure is the stabilization of the CH₄ loading during recent years. Comparisons with models are ongoing, aiming at the identification of the relative contributions in sources and sinks changes leading to this stabilization. It will be as important to assess the future evolution of methane in the coming years, to see if the current re-increase will continue. The extension of this methane time-series, using grating spectra recorded at Jungfraujoch between 1976 and 1989, is also currently under progress.

Inorganic chlorine and chlorinated source gases

Within the context of the Montreal Protocol, the monitoring of chlorinated source and reservoir gases has been part of our activities. Time series and trends of CFC-11 (CCl₃F), CFC-12 (CCl₂F₂), HCFC-22 (CHClF₂), CFC-10 (CCl₄), HCl and ClONO₂ have been updated (Figure 2 and 3 and Table 1) [Mahieu 2010].

CCl_y^{*}, the sum of the four chlorinated source species retrieved from the Jungfraujoch observations, is decreasing slowly, at a rate of -0.19 %/yr (see top part of Table 1). Over the time period under investigation, the largest negative contribution of CFC-11 is balanced by the accumulation of HCFC-22 (+8.7x10¹³ Cl atoms/cm² per year) whose partial ban by the Montreal Protocol has only started in 2005.

For the chlorinated reservoir gases HCl and ClONO₂, after a build up which lasted until about 1996-1997, at mean rates larger than 3 %/yr, we have then observed a significant decrease in inorganic chlorine. Post-peak trend values are listed in the bottom part of Table 1 and are compared to data derived from time series produced by the 3-D Karlsruhe model KASIMA. This model most often predicts lower rates of decline: possible causes for these discrepancies are currently under investigation.

Annual changes for chlorinated source gases, over 2001 -2010			
		10 ¹³ atoms/cm ²	%
CFC-10		-5.97 ± 0.50	-1.09 ± 0.09
CFC-11		-7.89 ± 0.54	-0.83 ± 0.06
CFC-12		-2.04 ± 0.61	-0.15 ± 0.04
HCFC-22		+8.70 ± 0.15	+4.22 ± 0.07
CCl _y		-6.02 ± 1.67	-0.19 ± 0.05
Annual changes for inorganic chlorine, over 1996 -2009			
HCl	FTIR	-3.59 ± 0.42	-0.87 ± 0.10
	KASIMA	-0.96 ± 0.35	-0.29 ± 0.11
ClONO ₂	FTIR	-0.91 ± 0.27	-0.90 ± 0.27
	KASIMA	-0.55 ± 0.16	-0.72 ± 0.21
Cl _y	FTIR	-4.80 ± 0.72	-0.93 ± 0.14
	KASIMA	-1.67 ± 0.60	-0.41 ± 0.15

Table 1. Trend values derived from the Jungfraujoch daily mean time series, in terms of annual total column (Cl atoms/cm²) or percent change. The time periods considered are 2001-2010 for the source gases and 1996-2009 for the reservoirs. Trend values derived from KASIMA runs (1996-2008) for the reservoir species are also provided. To avoid the impact of sampling, only model data coincident with measurements are included in the trend evaluations. Annual percent changes are computed using 2001 and 1996 total columns as reference, respectively for the chlorinated source gases and for the Cl reservoir species.

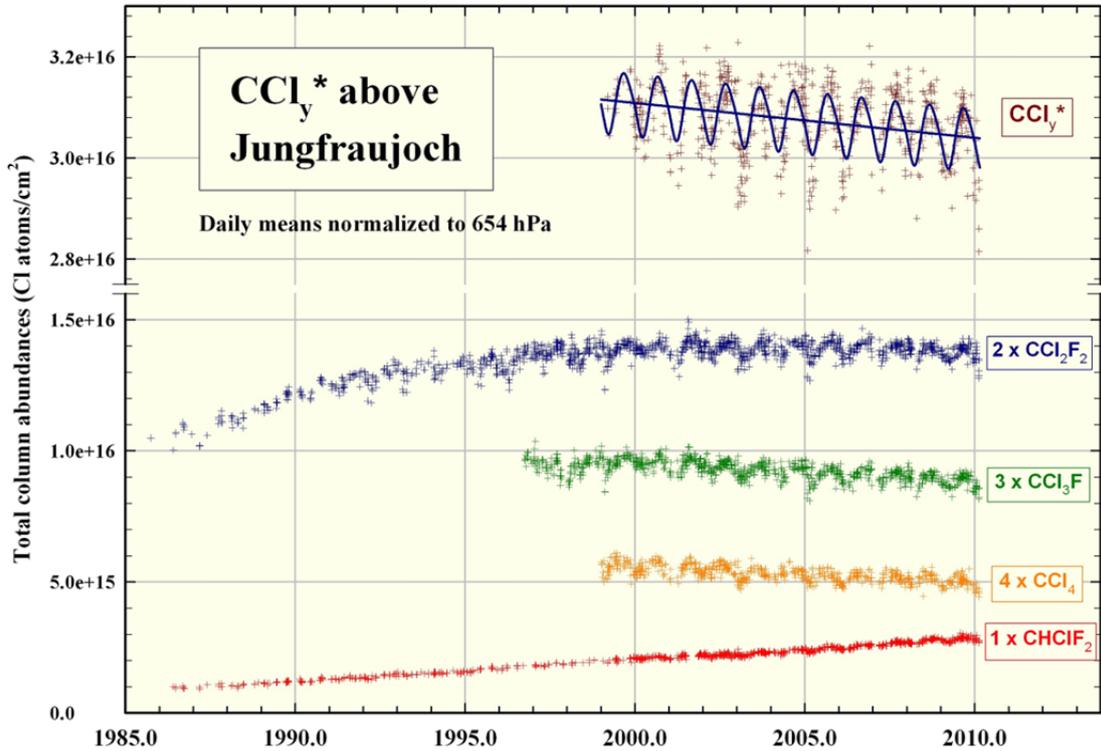


Figure 2. Temporal evolution of the long-lived chlorinated source gases retrieved from the Jungfraujoch FTIR spectra, weighted by the number of Cl atoms in each species (notice the vertical scale break). Data points correspond to daily means normalized to 654 hPa. The upper data set is the sum of the individual contributions, for days with simultaneous measurements available. It is noted CCl_y^* since it does not represent the total organic chlorine in the atmosphere, with two significant contributors missing (CH_3Cl and CFC-113). Altogether, the four species retrieved from Jungfraujoch observations correspond to $\sim 72\%$ of the total CCl_y budget for the year 2004. The seasonal signals seen in most time series essentially result from the tropopause height changes throughout the year.

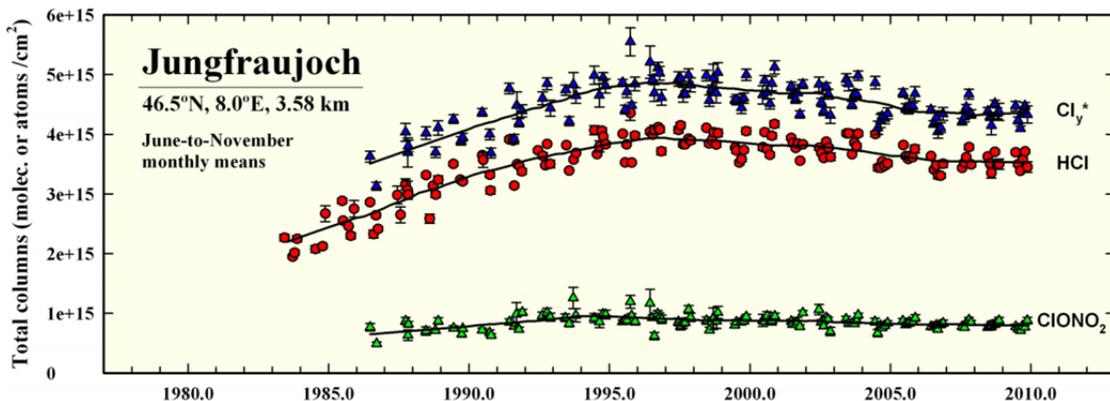


Figure 3. Jungfraujoch time series of monthly mean total columns of HCl (red circles), ClONO_2 (green triangles) and their summation Cl_y^* (blue triangles) which is a good proxy of the total inorganic chlorine Cl_y , since these two reservoir gases represent $\sim 92\%$ of Cl_y at mid-latitude, in absence of chlorine activation. Error bars correspond to the standard deviations around the monthly means. Only June to November months are reproduced here since they are less affected by variability resulting from meridional transport and subsidence events occurring essentially during winter-springtime. Continuous thick curves

correspond to non-parametric least square fits to the monthly means and help appraising the temporal evolution of the loadings of inorganic chlorine above Jungfraujoch.

Hydrogen fluoride HF

FTIR solar spectra recorded between March 1984 and December 2009 at the Jungfraujoch have been reanalyzed to derive time series of hydrogen fluoride (HF) total columns. The spectra have been inverted with the PROFFIT9.5 algorithm, using the optimal estimation method. Intercomparison of HF total columns retrieved with PROFFIT and SFIT2 – the other reference algorithm in the FTIR community – has been performed. The effect of a Galatry lineshape model on HF retrieved total columns and vertical profiles, on the residuals of the fits and on the error budget has also been quantified [Duchatelet 2010a, 2010b]. Information content analysis indicates that in addition to HF total vertical abundance, three independent stratospheric HF partial columns can be derived from the Bruker spectra (approximately from 10 to 17 km, from 17 to 25 km and from 25 to 40 km). A complete error budget has been established and indicates that the main source of systematic error is linked to HF spectroscopy and that the random error affecting the HF total columns does not exceed 2.5 %. Ground based middle and upper stratospheric HF amounts have been compared to satellite data collected by the HALOE or ACE-FTS instruments (Fig. 4).

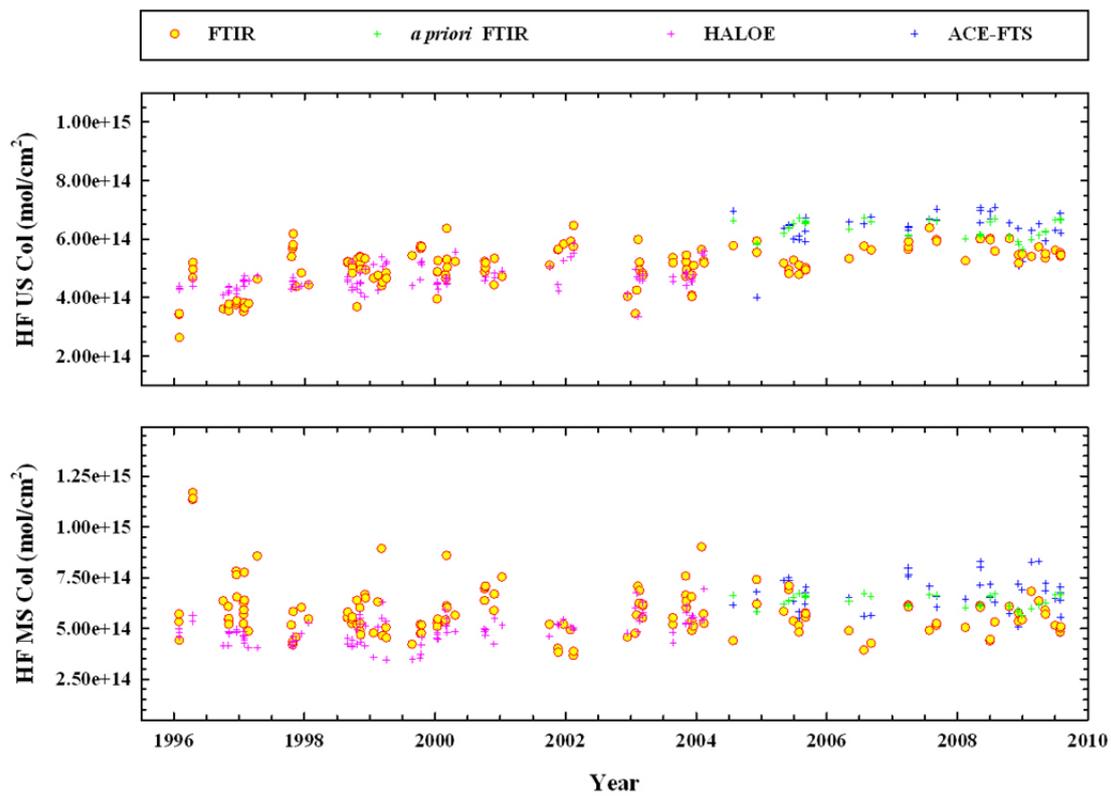


Figure 4. Low-middle stratospheric (MS, approximately from 17 to 25 km) (bottom) and middle-upper stratospheric (US, approximately from 25 to 40 km) (top) HF partial columns as observed at the Jungfraujoch by FTIR technique (yellow dots) and by space occultations (HALOE: pink crosses; ACE-FTS: blue crosses). All data points correspond to daily mean values for coincident measurement days between ground- and space-based observations. Green crosses reproduce initial guess values adopted during FTIR retrievals with the PROFFIT code. FTIR-HALOE relative difference values amount to $21.7 \pm 28.9 \%$ and $4.0 \pm 17.4 \%$ for MS and US regions, respectively, whereas corresponding FTIR-ACE-FTS values are $-17.5 \pm 11.2 \%$ and $-12.0 \pm 11.8 \%$. For a complete discussion about these results, see [Duchatelet 2010a].

Comparisons of measured FTIR HF total and partial columns with data produced by two three-dimensional numerical models (SLIMCAT and KASIMA) have also been performed. Finally, FTIR and model HF total and partial columns time series have been analyzed to derive the main characteristics of their seasonal cycles. These results have been published in [Duchatelet 2010a].

Formic acid H₂CO₂

Total vertical column abundances of formic acid above the Jungfraujoch station have been derived for the September 1985 - September 2007 time period. The investigation is based on the spectrometric fitting of five spectral intervals, one encompassing the H₂CO₂ ν_6 band Q-branch at 1105 cm⁻¹, and four additional ones allowing to account for major temperature-sensitive or timely changing interferences by other atmospheric gases, in particular HDO, O₃, CCl₂F₂ and CHClF₂. The *a priori* H₂CO₂ vertical profile used in the retrievals presents a slope which is consistent with the very short lifetime (from a few hours to a few days) of this reactive species and commensurate with numerical values deduced from earlier studies for northern mid-latitudes.

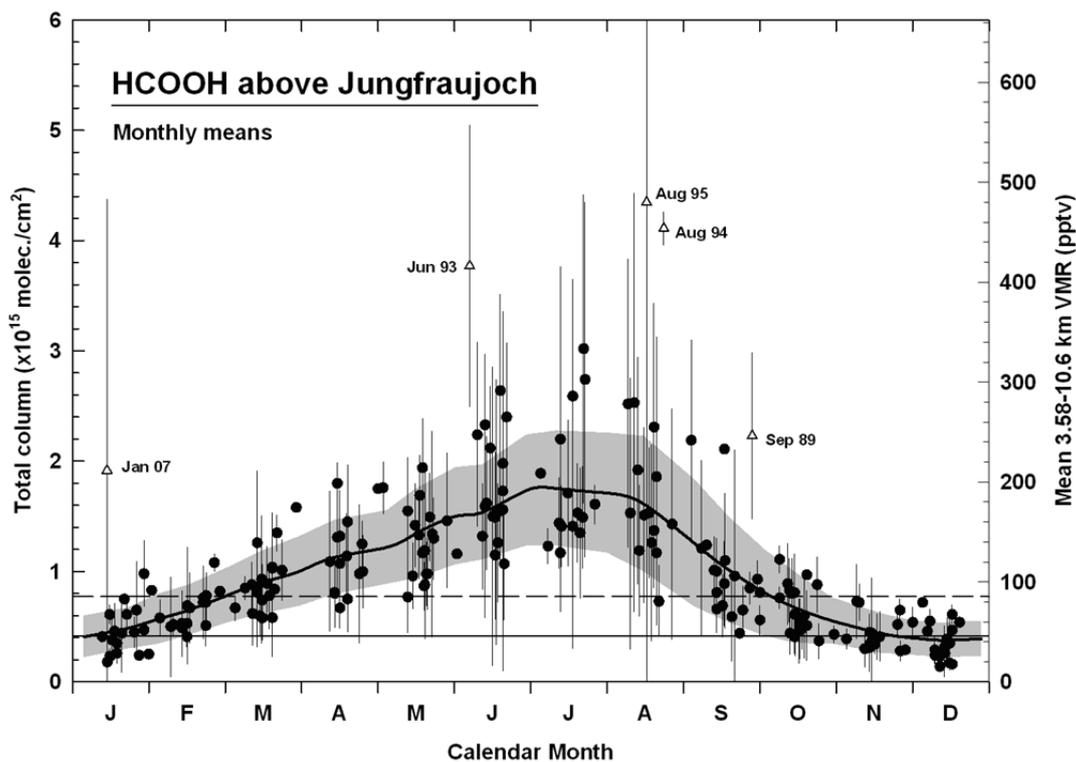


Figure 5. Monthly mean column abundances and associated standard deviation bars, displayed on a one-year time base, with 5 obvious monthly mean outliers identified by triangular symbols. The thick curve corresponds to a running mean fit to the black filled symbols only, with a 15-day step and a 2-month wide integration time. The shaded area visualizes the 1- σ standard deviation associated to the running mean curve and reflects primarily the observed inter-annual variability of H₂CO₂. The right side scale allows converting an observed total H₂CO₂ column abundance read off of the left side scale into a mean VMR concentration (expressed in parts per trillion by volume, i.e., pptv) over the 3.58 to 10.6 km altitude range.

A major improvement in the absolute determination of the atmospheric H₂CO₂ columns has resulted from the adoption of new spectral line intensities for the ν₆ band of trans-formic acid, resulting in retrieved free tropospheric loadings being about a factor of two smaller than those derived with previous spectroscopic parameters.

The main other results derived from this formic acid database indicate that the free tropospheric burden of H₂CO₂ above the Jungfrauoch undergoes important short-term daytime variability, seasonal and inter-annual modulations. For example, Figure 5 reveals an overall seasonal variation, with a broad summer maximum peaking in July and a November-December-January minimum. This result constitutes an important finding, establishing, for the first time, the existence of a seasonal variation of the free tropospheric loading of H₂CO₂ above a continental site at northern mid-latitudes. These findings have been published in [Zander 2010].

Key words:

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/>

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 HYMN (<http://www.knmi.nl/samenw/hymn/>) and GEOMon (<http://geomon.ipsl.jussieu.fr/>) / 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 2010:

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>.

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