http://girpas.astro.ulg.ac.be

This poster participates in OSP

Outstanding Student Poster Contest

Extension of the long-term total column time series of atmospheric methane above Jungfraujoch station : analysis of grating infrared spectra between 1976 and 1989.

Whitney Bader (1), Bernard Lejeune (1), Philippe Demoulin (1), Pierre Duchatelet (1), Ginette Roland (1), Kengo Sudo (2), Hisashi Yashiro (3), and Emmanuel Mahieu (1).

(1) University of Liège, Institute of Astrophysics and Geophysics, LIEGE, Belgium (emmanuel.mahieu@ulg.ac.be)
(2) Graduate School of Environmental Studies, Nagoya University, Nagoya, Aichi, Japan
(3) Japan Agency for Marine-Earth Science and Technology (JAMSTEC), Yokohama, Japan

1. Introduction and context

- Methane (CH_4) is one of the most abundant greenhouse gases in the Earth's atmosphere, with current mean volume mixing ratio close to 1800 ppb. CH_4 has a global warming potential of 25 (100-yr horizon) and an atmospheric lifetime of 12 years, it further affects climate through its participation to tropospheric ozone production and as a source of stratospheric water vapor. Given all these characteristics, the Kyoto Protocol has included it among the species whose emissions have to be regulated to limit global warming. Methane data sets are therefore needed to characterize its build up, to understand its cycle and to help improving its atmospheric budget (sources and sinks).

The Jungfraujoch site

- All the spectra analyzed here have been recorded at the International Scientific Station of the Jungfraujoch, a site located in the Swiss Alps (46,55° N, 8,98° E , 3580m asl.). This high altitude station offers excellent conditions to perform solar observations, in particular in the infrared (IR), because of weak local pollution (no major industries within 20km) and very high dryness. Indeed, the amount of water vapor, a strong interference in the IR, is at least twenty times lower than at sea level.

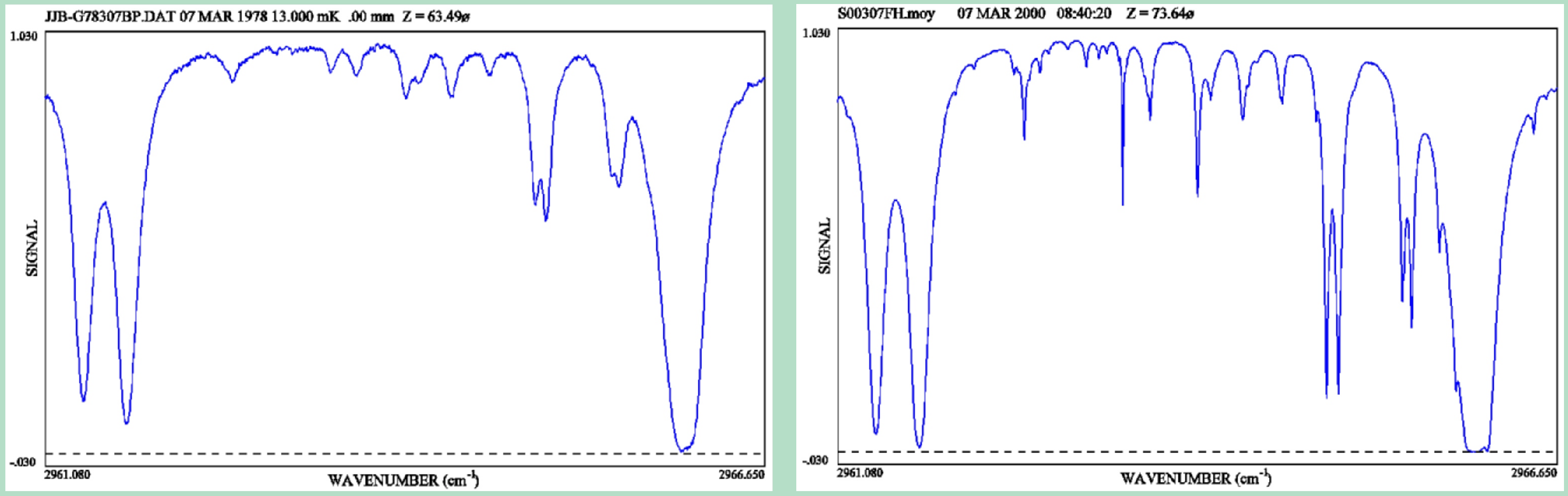


Figure 1 : These two spectra (DPGS-left hand side and FTS-right hand side) show that there has been a real improvement in resolution as they respectively achieve a resolution of around 0,02 cm^{-1} and 0,003 cm^{-1}. Spectra are zoomed on the microwindow n°3 (See Table I).

2. Instruments and observational database

- Early pioneering atmosphere-oriented observations have been recorded at the Jungfraujoch station in 1950, by the University of Liège (ULg) using high-performance grating spectrometers. From 1958 to 1975, the main objective has been the study of the solar photosphere in the visible and near-IR and the production of high-resolution solar atlases (e.g. Delbouille et Roland, 1963). After the detection of HF in the Earth's atmosphere by R. Zander in 1975 (Zander, 1975), the ULg group resumed its atmospheric observations at the Jungfraujoch to monitor the abundance of important constituents of our environment, among which HCl, HF and CH_4.
- We use solar observations recorded by a double-pass grating spectrometer (DPGS) installed at the Jungfraujoch by the ULg in 1976. The DPGS achieved a spectral resolution of about 0.02 cm^{-1} and was equipped with a InSb detector. Narrow-band infrared spectra encompassing isolated lines of e.g. CH_4, HF, HCl, NO_2 have been regularly recorded until 1989.
- From 1984 to present days, regular observations have been carried out at the Jungfraujoch station with two high-performance Fourier-transform infrared spectrometers (FTS) a homemade and a Brucker IFS-120HR. They allow us to derive abundances of more than 25 constituents affecting our climate and monitored in the frame of the Kyoto protocol, related to stratospheric ozone depletion, or altering the oxidation capacity of the troposphere. The resulting time series allow the determination of the short-term variability, seasonal modulations, as well as long-term changes affecting most of these species. As these two FTIR instruments are affiliated to the NDACC Network [http://www.ndacc.org], contribution to the long-term monitoring of the Earth's atmosphere has remained the central activity of the GIRPAS group over the last decades.

3. Retrieval Strategy

- All retrievals have been performed with the SFIT-2 algorithm (v 3.91) (Rinsland et al., 1998) in order to retrieve methane vertical total column. The microwindows have been selected to encompass lines of methane with minimum absorption interference by any other telluric gases or solar lines (see Table 1). The inversion has been performed on a series of about 2500 spectra recorded between 1977 and 1989, with different diffraction orders and assumed resolutions (see in Table 2).
- The CH_4 a priori volume mixing ratio (VMR) profile adopted in all our retrievals is based on MkIV FTIR balloon measurements performed at northern mid-latitudes, it has been adapted to the mean tropopause altitude of the Jungfraujoch by Sussmann et al. (2008), in the framework of the European HYMN project [http://knmi.nl/samen/hymn]. A priori profiles for the interfering gases are based on the WACCM model climatology [The Whole Atmosphere Community Climate Model, http://waccm.acd.ucar.edu].
- In the present runs, HITRAN 2008 line parameters (Rothman et al., 2008) and line parameters produced in the framework of the HYMN project (see Duchatelet et al., 2008) as well as the solar line compilation provided by F. Hase (KIT) have been assumed for the target and interfering absorptions. Adopted temperature and geopotential height data sets are provided by the National Centers for Environmental Prediction (NCEP, Washington DC, USA).

Microwindows	Limits (cm^{-1})	Fitted species
1	2913.4 – 2914.25	CH_4 ; HDO
2	2924.17 – 2924.99	CH_4 ; H_2O ; HDO + solar lines
3	2962.89 – 2963.7	CH_4 ; H_2O ; HCl

Table I : List of microwindows used for our CH_4 inversions, for each of them, the third column provides interfering gases adjusted during the retrieval

Time period covered	Number of spectra	Order	Assumed Resolution (mK)	Microwindows used
From Jul-1977 to Aug-1983	308	1	28	2;3
From Jul-1977 to Dec-1978	9	2	27	3
From Aug-1983 to Sep-1985	610	7	10 ; 8 ; 13	1 ; 2 ; 3
24-Sep-1985	4	5	13	1
From Oct-1984 to Oct -1989	1602	6	13	1 ; 2

Table II : Description of the data set : the number of spectra for each time period as well as the diffracted order, the assumed resolution and the microwindows used (see Table I) are listed in this table.

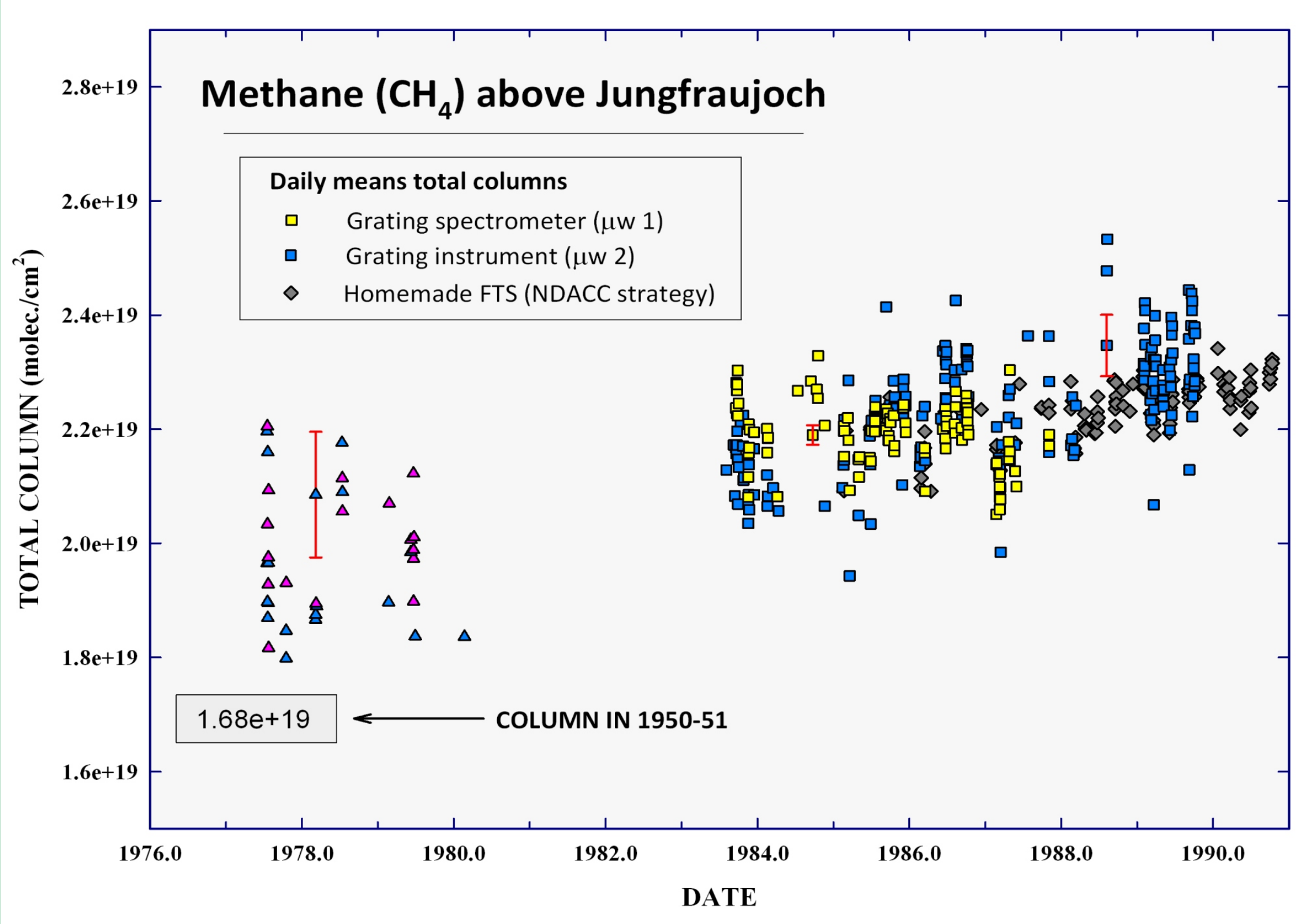


Figure 2 -
This figure displays the harmonized data set of methane daily mean total columns from 1983 to 1989 (blue and yellow squares).

Harmonization between microwindow 1, microwindow 2 and FTS (grey diamonds) time series is based on the difference between the mean methane total column for each series. Raw data covering the 1977-1979 period still have to be further analyzed.

The microwindow 1 is plotted in blue triangles and the microwindow 3 in pink triangles. For each data set, error bars represent typical error values in order to appraise their accuracies.

Error Budget

We are still working on establishing a complete error budget. Nevertheless, we have already quantified the impact on our retrieved CH_4 total columns of the NCEP temperature profiles and of the choice of the assumed resolution. They are within 1% and 4%, respectively.

4. Trend Calculations

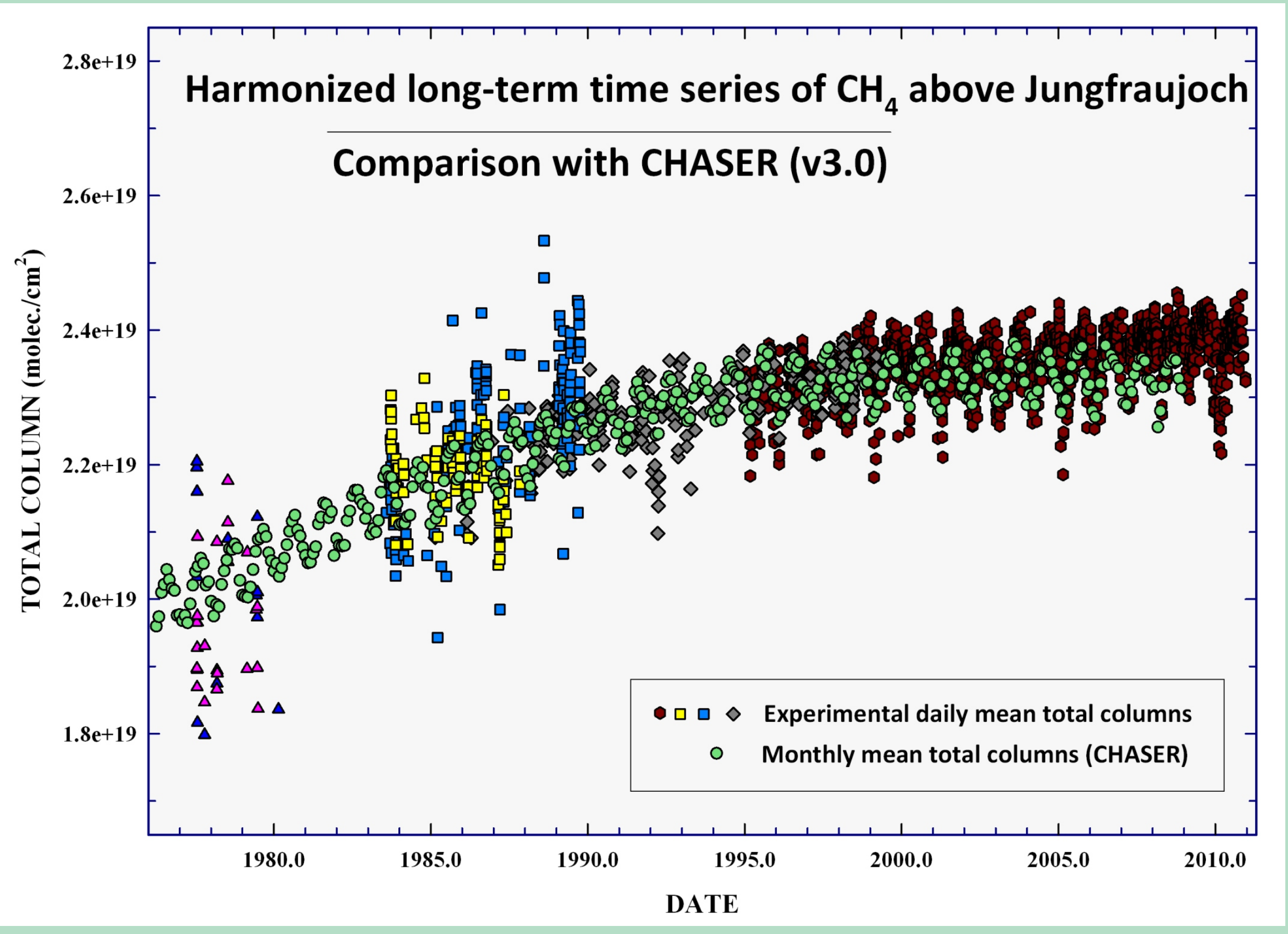
For the trend calculations, we use the statistical tool developped by Gardiner et al. (2008) that employs a bootstrap re-sampling method to determine the long-term change as well as the confidence levels associated to the retrieved quantities. All the uncertainties provided correspond to the 95% confidence interval. The function fitted to the time series is a combination of a linear component and a 3rd order Fourier series, i.e. :

$$F(t,b)=c_0+c(t-t_0)+b_1\cos 2\pi(t-t_0)+b_2\sin 2\pi(t-t_0)+b_3\cos 4\pi(t-t_0)$$
$$+b_4\sin 4\pi(t-t_0)+b_5\cos 6\pi(t-t_0)+b_6\sin 6\pi(t-t_0)$$

where c_0 is the vertical abundance at the reference time t_0 for the linear component (seasonalized data), and c is the annual trend.

CHASER Model (v 3.0)

- The CHemical AGCM for Study of atmospheric Environment and Radiative forcing (CHASER) has been developed at the Center for Climate System Research (CCSR), University of Tokyo/National Institute for Environmental Studies (NIES). The main goal of the Atmospheric General Circulation Model CHASER described by Sudo et al. (2002) is to study tropospheric ozone and related chemistry and their impact on climate. Sudo et al. provided us with a data set of monthly mean methane total columns computed above Jungfraujoch and covering the time period from 1970 to 2008.
- In contrast to the excellent agreement noted between the CHASER and the Jungfraujoch trends for the 1983-1992 time period, the differences observed afterwards are possibly due to the comparison between daily and monthly means. We notice a decrease in the CH_4 abundance over the fourth time period derived from CHASER. K. Sudo indicated that this might be due to high emission from wet land area in 2007 not included in the model.



Time Period	Reference Column (Year)	Annual Change (molec/cm²/an)	Annual Change (%/year)
1983 – 1992	$2{,}17 \times 10^{19}$ $2{,}14 \times 10^{19}$ (1983)	$1{,}56 \pm 0{,}23 \times 10^{17}$ $1.70 \pm 0.07 \times 10^{17}$	0,72 ± 0,11 0.79 ± 0.04
1993 – 1999	$2{,}29 \times 10^{19}$ 2.31×10^{19} (1993)	$8{,}40 \pm 1{,}58 \times 10^{16}$ $3.34 \pm 1.15 \times 10^{16}$	0,36 ± 0,07 0.15 ± 0.05
2000 – 2004	$2{,}35 \times 10^{19}$ 2.33×10^{19} (2000)	$4{,}83 \pm 0{,}17 \times 10^{15}$ $1.00 \pm 1.60 \times 10^{16}$	0,02 ± 0,07 0.04 ± 0.07
2005 - 2010 2005 - 2008	$2{,}36 \times 10^{19}$ 2.33×10^{19} (2005)	$3{,}54 \pm 1{,}45 \times 10^{16}$ $-2.98 \pm 2.64 \times 10^{16}$	0,15 ± 0,06 -0.13 ± 0.11

Figure 3 -
This figure compares the harmonized long-term time series of methane with modeled data derived from CHASER Model (green circles).

The Jungfraujoch time series includes different observational data sets : the DPGS-μw1 (yellow squares), DPGS-μw2 (blue squares), DPGS-μw3 (pink triangles), FTS-homemade (grey diamonds) and the FTS-Brucker (dark circles).

Table III -
Absolute (third column) and relative (fourth column) trend values computed over four successive time periods.
Blue : Jungfraujoch observations.
Green : CHASER Model (v 3.0).

Conclusions and Perspectives

- Inversion of DPGS measurements appears to be a reliable method in order to extend the long-term total column time series of atmospheric methane.
- We expect to improve the inversion strategy for the 1977-1979 time period in order to harmonize our time series from 1977.
- We will soon evaluate the impact of spectroscopy, a priori profiles of methane and interfering gases to complete our error budget.
- Since infrared observations above Jungfraujoch are available from the mid-seventies, the method could be extended to other important atmospheric trace gases (e.g. HCl, HF,...), providing 35-year long experimental time series for comparison with numerical models.

References

Delbouille L. and Roland G. (1963), Photometric atlas of the solar spectrum from λ 7498 to λ 12016. Mém Soc Roy Sci Liège, Special Vol 4.
Duchatelet, P., E. Mahieu, P. Demoulin, et al. (2008), Impact of different spectroscopic datasets on CH4 retrievals from Jungfraujoch FTIR spectra, in the Proceedings of the "8th Atmospheric Spectroscopy Applications" meeting (ASA2008), 27 30 August, Reims, France, pp. 80-83.
Gardiner T., A. Forbes, M. de Mazière et al. (2008), Trend analysis of greenhouse gases over Europe measured by a network of ground-based remote FTIR instruments, Atmos. Chem. Phys., 8, 67196727.
Rinsland, C.P., N.B. Jones, B.J. Connor, et al. (1998), Northern and southern hemisphere ground-based infrared spectroscopic measurements of tropospheric carbon monoxide and ethane, Journal of Geophysical Research, 103, 28197-28218.
Rothman, L. S., I. E. Gordon, A. Barbe, et al. (2009), The HITRAN 2008 molecular spectroscopic database, Journal of Quantitative Spectroscopy and Radiative Transfer, 110, 533-572.
Sudo K., M. Takahashi, J. Kurokawa, et al. (2002), CHASER : A global chemical model of the troposphere. 1. Model description, Journal of geophysical research, 107, 4339.
Sussmann, R., F. Foster, T. Borsdorff, M. De Mazière, et al. (2008), Satellite validation of column-averaged methane on global scale: ground-based data from 15 FTIR stations versus last generation ENVISAT/SCIAMACHY retrievals, poster presented at the "IGAC 10th International Conference", 7 12 September 2008, Annecy, France.
Zander, R. (1975), Presence de HF dans la stratosphere supérieure, C. R. Acad. Sci. Paris, Ser. B 281, 213-214.

Acknowledgments

Work at the University of Liège was supported by the Belgian Federal Science Policy Office (SSD and PRODEX Programs, respectively), the FRS-FNRS, both in Brussels, as well as by the GAW-CH program of MeteoSwiss. We thank the International Foundation High Altitude Research Stations Jungfraujoch and Gornergrat (HFSJG, Bern) and the University of Liège for supporting the facilities needed to respectively perform the observations and their analyses. We further acknowledge the vital contribution from all the colleagues in performing the observations used here.

Contact information:
whitney.bader@student.ulg.ac.be
http://girpas.astro.ulg.ac.be

Permanent Link :
http://hdl.handle.net/2268/88180