

1. Introduction • The Ethane ( $C_2H_6$ ) is the most abundant Non-Methane HydroCarbon (NMHC) in the Earth's atmosphere, with a lifetime of approximately 2 months.  $C_2H_6$  main sources are biomass burning, natural gas loss and biofuel consumption. Oxidation by the hydroxyl radical is by far the major  $C_2H_6$  sink as the seasonally changing OH concentration controls the strong modulation of the ethane abundance throughout the year. Ethane reduces Cl atom concentrations in the lower stratosphere and is a major source of peroxyacetyl nitrate (PAN) and carbon monoxide (by reaction with OH). Involved in the formation of tropospheric ozone and in the destruction of atmospheric methane through changes in OH, C<sub>2</sub>H<sub>6</sub> is an indirect greenhouse gas with a net-global warming potential of 5.5 (100-yr horizon).

• All the spectra analyzed here have been recorded at the International Scientific Station of the Jungfraujoch (46.5°N, 8°E, 3580 m 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

• Two Fourier Transform Infrared (FTIR) instruments (a commercial Bruker IFS-120HR and a homemade spectrometer) have been put into regular operation in 1984 and 1991, respectively. They allow to record wide-band high-resolution IR solar spectra either with a MCT or InSb detector. Since 1991, the two FTIR instruments are affiliated to the framework of the Network for the Detection of Atmospheric Composition Change (NDACC, visit http://www.ndacc.org).

# **2. Retrieval Strategy**

Parameters have been settled down on the basis of tests on a full year minimizing residuals and maximizing DOFS. Microwindows Selection



**Figure 1** - Synthetic spectra of our three fitted  $\mu$ -windows (see limits on Table I) for all gases (in black) as well as for the individual contributors (C<sub>2</sub>H<sub>6</sub> in blue, H<sub>2</sub>O in cyan, CH<sub>4</sub> in green, O<sub>3</sub> in dark pink, CH<sub>3</sub>Cl in pink and solar lines in orange) to the absorption in this spectral interval. For clarity, the contributions of each species have been vertically displaced. Simulated solar zenith angle: 60°.

A priori Volume Mixing Ratio (VMR) profiles



Figure 2 - The a priori profiles tested (left panel) and their associated relative standard deviation (right panel) are illustrated. The first adopted a priori C<sub>2</sub>H<sub>6</sub> profile (grey crosses) is a zonal mean (for the 41-51°N latitude band) of 771 occultations recorded by the ACE-FTS instrument between the 2nd of November in 2004 and the 8th of February in 2011 extending from 8.5 to 20 km. The profile extension down to 3.58 km is based on EMEP in situ measurements (bottom panel) while the upper extension to 100 km is based on the WACCM model climatology (Whole Atmosphere Community Climate, http://waccm.acd.ucar.edu). Pink crosses is the a priori profile issued from the CHASER 3-D Chemical Transport Model developed at the Center for Climate System Research (CCSR), University of Tokyo/National Institute for Environmental Studies (NIES). Chemical Transport Model v.2 and v.3 from the University of Oslo (Berntsen et al., 1997) are plotted in dark and light green crosses, respectively.

# References

\* Berntsen T., and I. S. A. Isaksen (1997), A global 3-D chemical transport model for the troposphere, 1, Model description and CO and Ozone results., J. Geophys. Res., 102, 21239-21280. \* Bray C., A. Perrin, D. Jacquemart et al. (2011), The v1, v4 and 3v6 bands of methyl chloride in the 3.4 µm region: Line positions and intensities, J. Quant. Spectrosc. Radiat. Transfer, 112, 2446–2462. \* 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, 6719-6727. <sup>+</sup> Harrison J. J., N. D. C. Allen and P. F. Bernath (2010), Infrared cross sections for ethane (C<sub>2</sub>H<sub>6</sub>) in the 3 μm region., J. Quant. Spectrosc. Radiat. Transfer, 111, 357-363. \* Rinsland C. P., N. B. Jones, and B. J. Connor (1998), Northern and southern hemisphere ground-based infrared spectroscopic measurements of tropospheric carbon monoxide and ethane, J. Geophys. Res., 103 (D21), 28197 \* Rothman L.S., D. Jacquemart, A. Barbe et al. (2010), The HITRAN 2008 molecular spectroscopic database, J. Quant. Spectrosc. Radiat. Transfer, 110, 533-572. \* Sudo K., M. Takahashi, J. Kurokawa et. al. (2002), CHASER : A global chemical model of the troposphere. 1. Model Description, J. Geophys. Res., 107, 4339.

	µ-windows	1- <sup>P</sup> Q <sub>3</sub>	u2	u3	Globa
	1- Hitran-08	0.431	0.206	0.471	0.424
	2- Harrison + 1	0.171	0.158	0.173	0.179
	3- Chelin + 2	0.169	0.158	0.172	0.179
	4- Bray + 3	0.153	0.151	0.152	0.163



interfering gases are based on the WACCM model climatology.

µ-windows	Limits (cm <sup>-1</sup> )	In
1 - <sup>P</sup> Q <sub>3</sub>	2976.66 - 2977.059	$C_2H_6$ ,
2	2983.2 - 2983.5	$C_2H_6$ ,
3	2986.43 - 2986.85	$C_2H_6$ ,



We selected the a priori VMR among four ethane profiles (see Figure 2) giving retrieved profiles with the least oscillations and least negative VMR values. We also tested those profiles adjusted on EMEP measurements made at the Rigi station (47°N, 8°E, 1031 m a.s.l.). Based on these criteria we sélected the a priori profile their and uncertainties issued from the Chemical AGCM for Study of atmospheric Environment and Radiative Forcing (CHASER) Model (Sudo et al., 2002). Residuals were reduced by 7.6 % while DOFS is improved by 11.1 and comparison to the Oslo's CTM v2 and v3 a priori profiles,

> Bruxelles are further acknowledged for observational activities support. Jungfraujoch and Gornergrat (HFSJG, Bern) for supporting the facilities contribution from all the Belgian colleagues in performing the Jungfrauj Pasadena) for the conversion of the ethane cross sections into pseudolines and O. A. Søvde for providing synthetic data.



Poster presented at the EGU General Assembly 2012 Vienna, Austria, 22-27 April 2012. Permalink : http://hdl.handle.net/2268/117067

lungfrauj	och	• The Figure 5 displays our retrieved $C_2H_6$ total column and both partial columns (low-tropospheric and UTLS) above Jungfraujoch. We computed an overall decrease in ethane concentrations since 1994 of -14, -9 and -39 %						
201		concentrations since 1994 of -14, -9 and -39 % resp. for our three columns. Trends have been determined using the boostrap resampling tool developed by Gardiner (2008) (see Table III). • The decrease of ethane remains smaller than the seasonnal amplitude. Our measurements allow to characterize strong seasonal variations of $C_2H_6$ total and low-tropospheric columns with a maximum generally observed around mid- February. On average, the peak-to-peak amplitudes respectively amount to 50 and 76 % of the 1994 reference column. • Whereas the seasonnal change of ethane UTLS column is less obvious with a peak-to-peak amplitude of 25 % (Ref : 1994 column). Therefore, the observed overall decrease of 39 % prevails in						
I UTLS	Time	Series	Total Column	3.58 - 6.79 km	8.45 - 14.3 km			
above	Junfi (1994	raujoch 4-2011)	-0.47 ± 0.35 % (1994)	-0.92 ± 0.30 % (1994)	-0.69 ± 0.24 % (1994)			
	Jungf	fraujoch	-1.06 ± 0.31 %					
ТМ	(1994 CH	4-2008) ASER	(1994) 0.55 ± 0.18 %					
12	(1994 Junaf	4-2008) frauioch	(1994) -1.58 ± 0.68 %	-0.79 ± 0.98 %	-2.59 ± 0.89 %			
ns	(1998) Oslo	8-2005) 's CTM	(1998) -0 89 + 0 56 %	(1998) -0 72 + 0 57 %	(1998) -1 26 + 1 23 %			
	(1998	8-2005)	(1998)	(1998)	(1998)			
	Tak and	ble III - Andread IIIII - Andread III - Andread III - Andread III - Andread III - Andr	nnual Change ( erence year fo	in %), its 2-σ u or Jungfraujoc	ncertainties h, CHASER			
+ -	col	umns.		s time series it	or our three			
	0	Tabla III						
	• On within	$2-\sigma$ bety	we notice a ween Oslo's C	TM computed t	trends and the			
	• On Figure 6, we compare monthly means of an averaged							
year of our Jung issued from the		e CHASER 3D-Model and Oslo's CTM v.3.						
	ethane	e in the	atmosphere. In	the case of O	slo's model, it			
m	may be explained by the use of undervalued atmosp pressures.				d atmospheric			
	<ul> <li>C<sub>2</sub>H<sub>6</sub> seasonnal change has been evaluated for CHASER and Oslo's CTM data with a peak-to</li> </ul>							
	amplit respec	ude of 3 ctively f	85 % (Ref. : 199 or ethane tota	and of 44 % (Ref. : 1998), columns. While our low-				
Ť	tropos the CH	spheric p IASER N	beak-to-peak ar lodel and for 5	nplitude amou 1 % for Oslo's (	nts to 67 % for CTM.			
12								
lungfra colum colum	aujoch o ns are r ns are o	observati especitve computed	ons (in blue), re ely averaged on d over the 2007-	esults from CHA the 1994-2011, 2009 time perio	SER Model (in 1970-2008 and d.			
d to Hitran 2008 compilation improve the retrieval of ethane in ontent ; as well as Chelin's $O_3$ and Bray's $CH_3Cl$ updates. The combined of the least negative profiles with best								
e determined a significant decrease in its concentration over the a seasonnal change in total and tropospheric columns of ethane ease. In the UTLS the long-term decrease is as the same order of								
he study of the interannual variations as well as of the seasonnal nd in the UTLS region. troscopy, instrumental line shape, geometry in order to refine our								
PRODEX program funded by the Belgian Federal Science Policy Office iate with the F.R.S. – FNRS. The FRS-FNRS and the Fédération Wallonie Ve thank the International Foundation High Altitude Research Stations needed to perform the observations. We further acknowledge the vital och observations used here. We further thank G.C. Toon (NASA-JPL which can be used by our retrieval algorithm. We finally thank K. Sudo								