

Overview of existing high-level data products derived from high-resolution Fourier Transform Infra-Red (FTIR) spectra recorded at the Jungfrauoch station, typical results and their valorization

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A bit of history

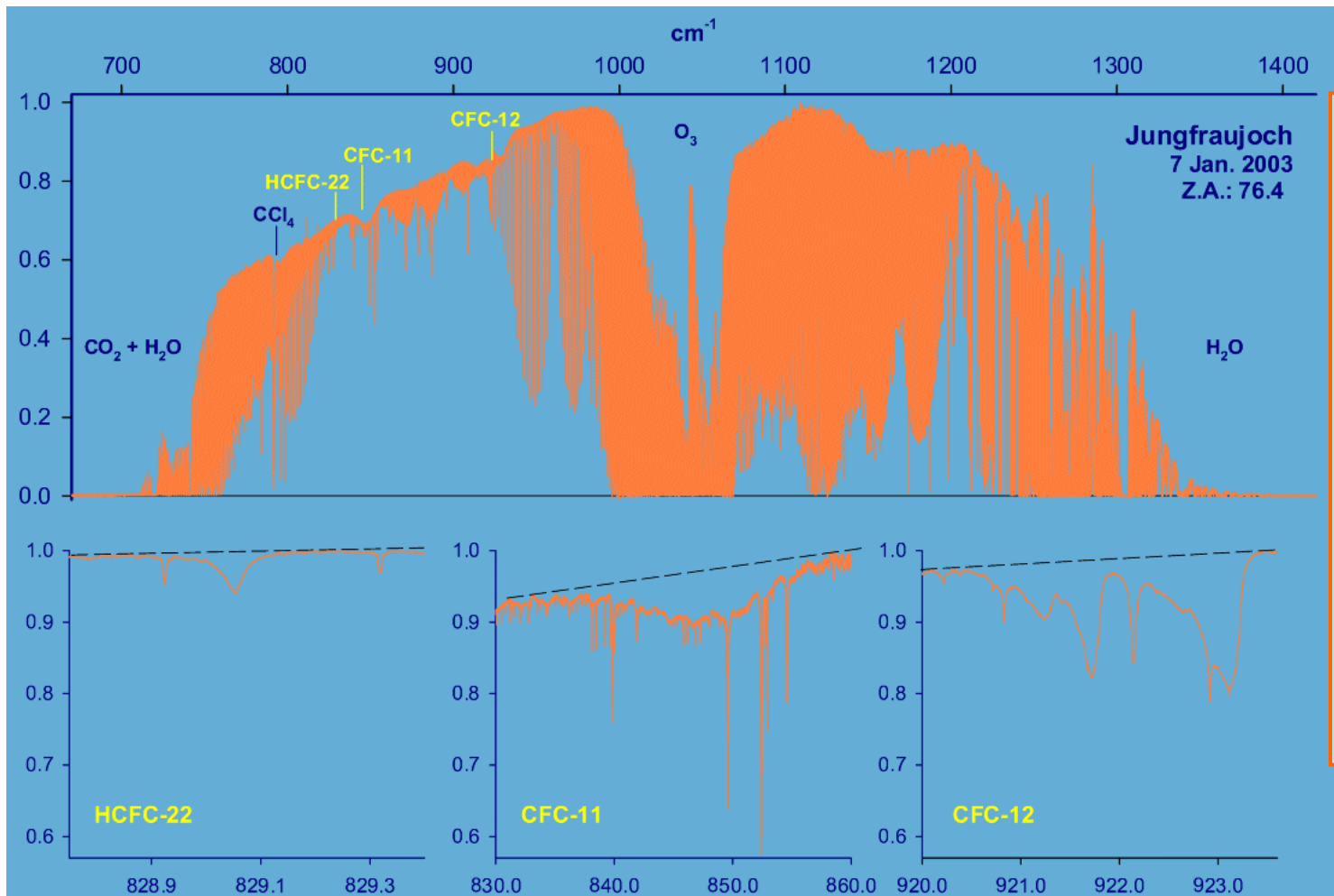
- The Liege group is present at the site since the early 1950s. Pioneering InfraRed atmospheric observations have been performed by Pr Migeotte with a prism-grating spectrometer, encompassing absorptions of e.g. CH₄, N₂O, CO₂. These recordings have been digitized and re-analyzed in the 1990s, allowing to derive mean total column of these minor constituents in 1950-51, very useful for comparison with their contemporary abundances.
- Atmospheric oriented observations [for solar spectrum measurements and related investigations, see G. Roland's talk] have started again in the mid seventies, aiming at monitoring the evolution of stratospheric species [NO₂, HCl, HF] as well as of some important source gases [N₂O, CH₄, CO], using a double-pass grating spectrometer.
- Two FTIR instruments have been installed at the Jungfrauoch, a homemade and a Bruker IFS-120HR, they perform routine measurements since 1984 and 1990, respectively [for current status of the instrumentation, its development and remote control operation, see C. Servais' talk].
- The two FTIR instruments are affiliated to the Network for the Detection of Atmospheric Composition Change, NDACC (previously Network for the Detection of Stratospheric Change, NDSC), within this framework and to meet our commitments toward the network (e.g. frequency of observations), routine operation of the instruments on site is performed as often as possible, with observers present for about 2 days over three.

Observational database

- The two FTIR instruments are equipped with InSb and HgCdTe detectors allowing to record solar absorption spectra in between 1 and 14 μm , in complementary domains, using a set of carefully selected optical filters
- This technique allows to simultaneously produce atmospheric observations combining high-resolution (down to 0.002 cm^{-1}), wide spectral range (several hundreds of cm^{-1}) and high S/N (>1000)
- Since 1984, nearly 42000 atmospheric FTIR spectra have been recorded at the Jungfraujoch, without any significant interruption, which makes of this observational database a unique set worldwide (length and density)
- Density of observations has significantly increased after 1990; over the last ten years, we have spectra available for typically 120 days/yr (clear sky conditions mandatory), i.e. about 1 day of observations over three

Sample spectrum in the MCT domain ($\sim 14 - 7 \mu\text{m}$), recorded in ~ 6 minutes

Encompasses numerous absorption features associated to important tropospheric and stratospheric species, involved in processes affecting our environment (ozone depletion, climate change, pollution)



A dozen species are studied in this range:

H_2O , CO_2 , CH_4 ,
 O_3 , N_2O ,

several CFCs,
HCFCs and
HFCs, SF_6 , CCl_4

C_2H_2 , HCOOH ,
 HNO_3 , ClONO_2 ,
 COF_2

Molecules currently studied in Jungfraujoch FTIR solar spectra

Table 1 – Gaseous constituents monitored at the Jungfraujoch based on the spectrometric analysis of solar spectra recorded with FTIR instruments

Reference gas	N ₂ (1%)
Minor gases	CO ₂ (2%), N ₂ O (2%), CH ₄ (2%), CO (3%), O ₃ (4%)
Trace gases	
Halogenated species	CCl ₂ F ₂ (4%), CHClF ₂ (5%), CCl ₃ F (12%), CCl ₄ (15%), SF ₆ (25%), HCl (3%), ClONO ₂ (15%), HF (3%), COF ₂ (10%)
Nitrogenated species	NO (5%), NO ₂ (6%), HNO ₃ (4%)
Others	C ₂ H ₆ (5%), C ₂ H ₂ (20%), HCN (7%), OCS (6%), H ₂ CO ₂ (12%) [CH ₃ Cl, H ₂ CO, HNO ₄ , H ₂ O; various isotopes]

Percentages between parentheses indicate typical precisions with which individual total column abundances have been retrieved routinely above the Jungfraujoch during the past two decades. Species typed in *italic* are primarily present in the stratosphere, while the others are tropospheric source gases. Species between brackets have not been evaluated routinely. Notice that brominated compounds have too weak absorption features to be observable in the infrared under their present concentrations in the atmosphere.

Zander et al., *Sci. Total Environ.*, 391, 184-195, 2008

About two dozen are routinely retrieved, using essentially the SFIT-2 software, implementing the OEM formalism of Rodgers

In most cases, information on the vertical distribution of the species can be derived and characterized

Budget for e.g. the chlorine and nitrogen families can be established, allowing to determine partitioning between corresponding reservoirs

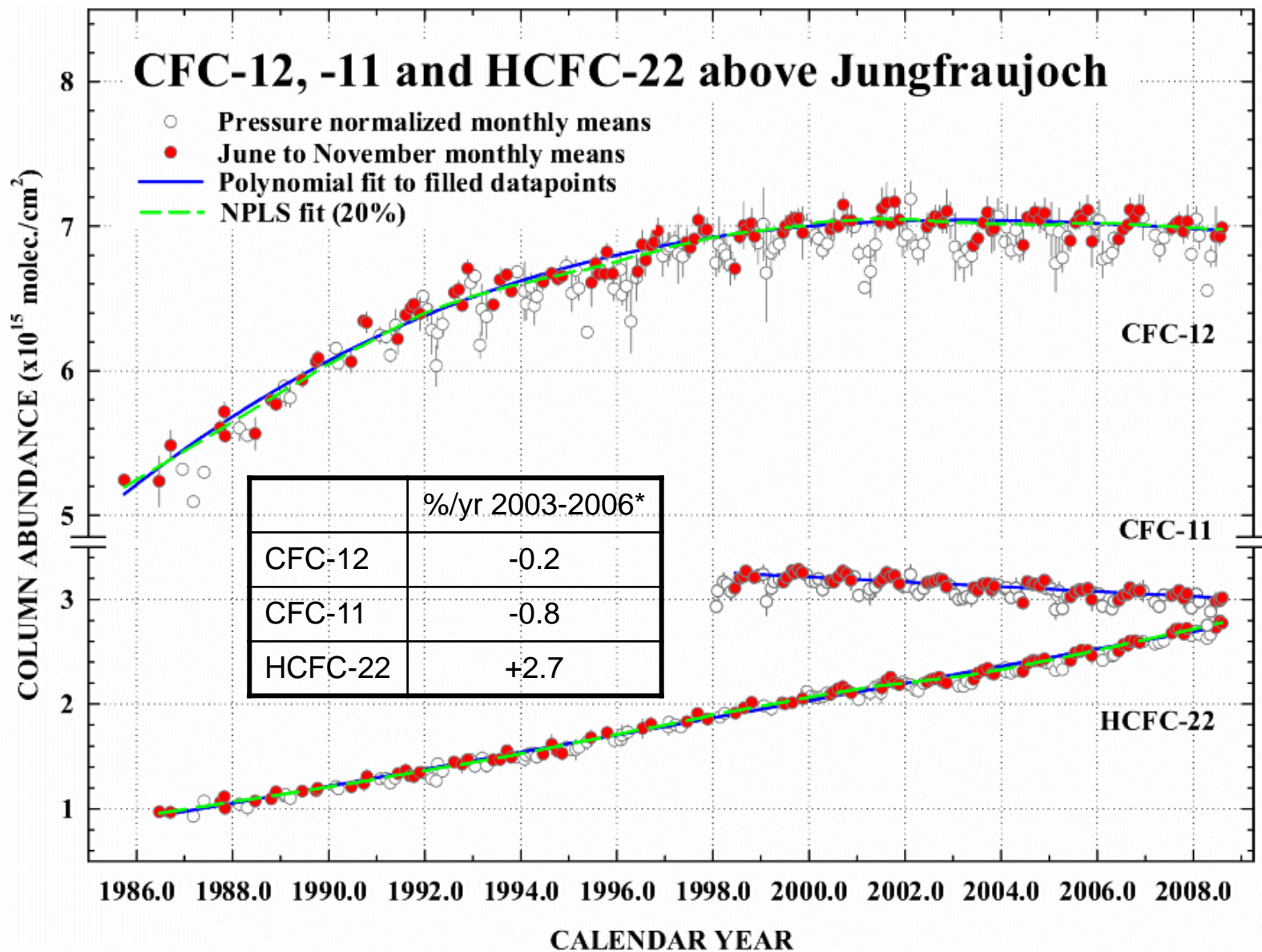
New species are regularly added to this list

H₂O investigations in progress
[see talk by Ph. Demoulin]

Key results

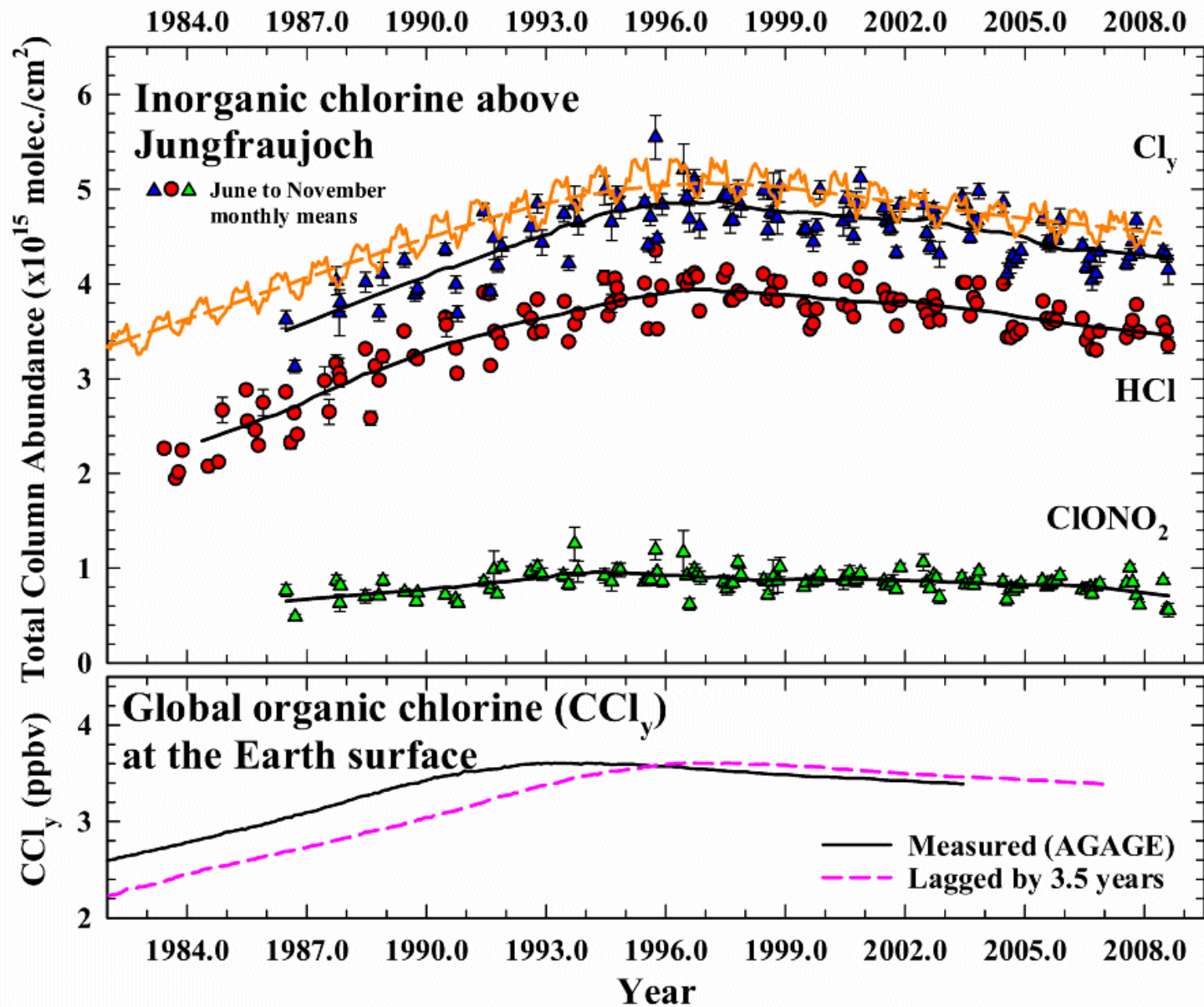
Long-term monitoring of the atmospheric composition and change, trend determinations:

- In support of the Montreal Protocol, source gases (CFCs, HCFC-22, CCl_4) and major reservoir species (inorganic chlorine, Cl_y : HCl , ClONO_2 ; inorganic fluorine, F_y : HF and COF_2) [for COF_2 : see talk by P. Duchatelet]
- Several GHGs regulated under the Kyoto Protocol are under surveillance, with two-decade time series available for CO_2 , CH_4 , N_2O , SF_6 , and even reference abundances derived from early Jungfraujoch observations performed in the 1950s



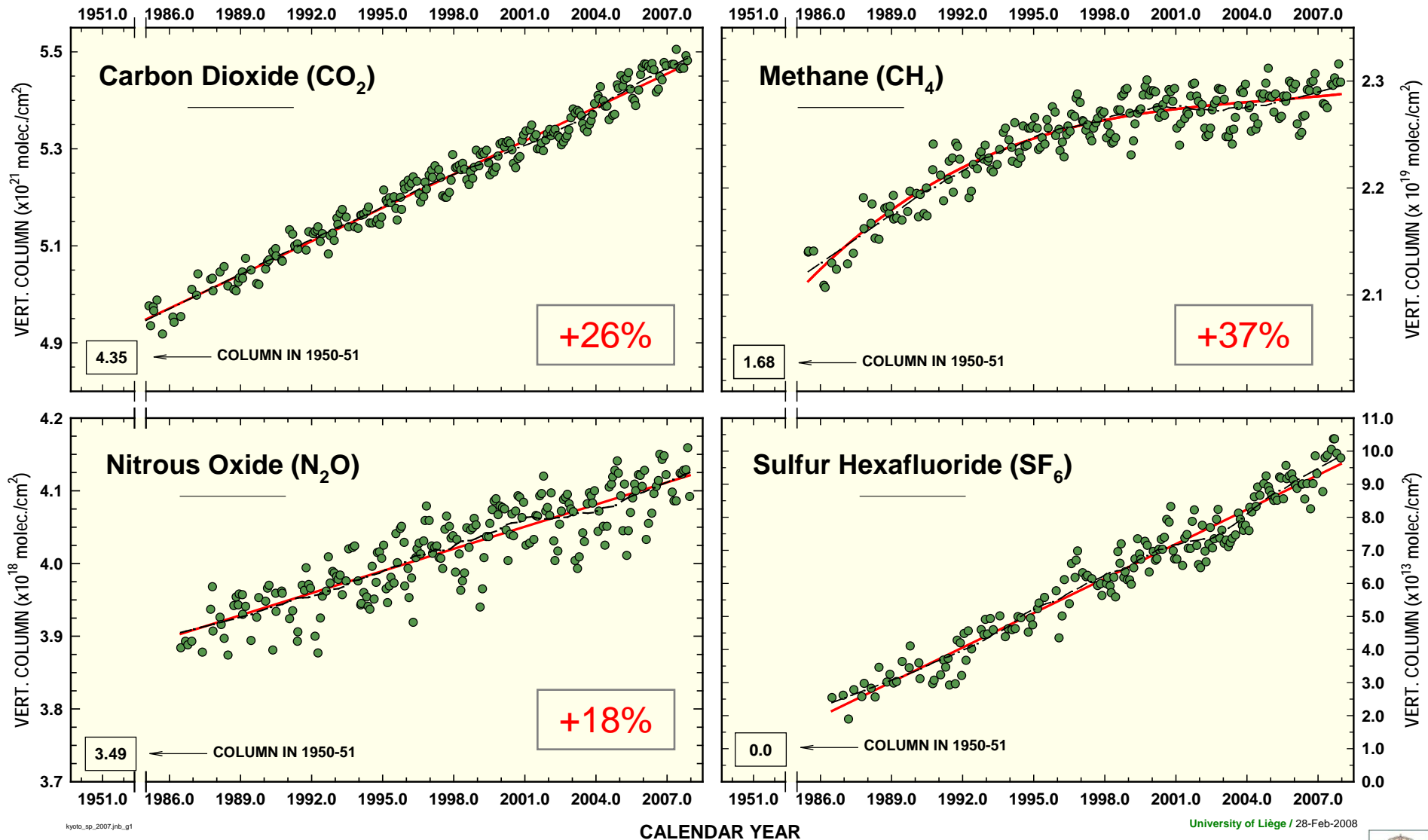
* Shown to be in excellent agreement with trend values derived from EMPA *in situ* measurements at the Jungfrauoch, see Reimann et al., *Sci. Total Environ.*, 391, 2008

ULg-GIRPAS / 25-Sep-2008



- $[HCl] + [ClONO_2]$ a good proxy for Cl_y
- In the 1980s and early 1990s, increase of Cl_y at $\sim 4\%/yr$
- Turn over in 1996, i.e. ~ 3.5 years later than in the troposphere
- Decrease since 1999 at $-1.1 \pm 0.2\%/yr$, in good agreement with model calculations [SLIMCAT, in orange in upper panel; KASIMA, not shown here]

KYOTO-PROTOCOL RELATED MEASUREMENTS AT THE JUNGFRAUJOCH



kyoto_sp_2007_jfb_g1

University of Liège / 28-Feb-2008

Calibration and Validation of space-based sensors

- Over the last 6 years, ground-based FTIR measurements at the Jungfraujoch have contributed to the validation of 7 space-based sensors, for as many as 10 different stratospheric or tropospheric species
- Ready to support the validation of the IASI/Metop instrument
- Two recent examples shown here:
 - MIPAS (e.g. ClONO₂, but also N₂O and HNO₃ [Vigouroux et al., 7, 2007])
 - ACE-FTS (e.g. HCl, but also HF, CH₄, N₂O, O₃, HNO₃, ClONO₂, CO)

Validation of MIPAS ClONO₂ columns

Fig. 13. Comparison between MIPAS (red) and FTIR (green) daily mean column amounts versus time for the collocation criterion $\Delta d_{\max}=800$ km, $\Delta t_{\max}=8$ h, and $\Delta p v_{\max}=3 \times 10^{-6}$ Km² kg⁻¹ s⁻¹ at 475 K.

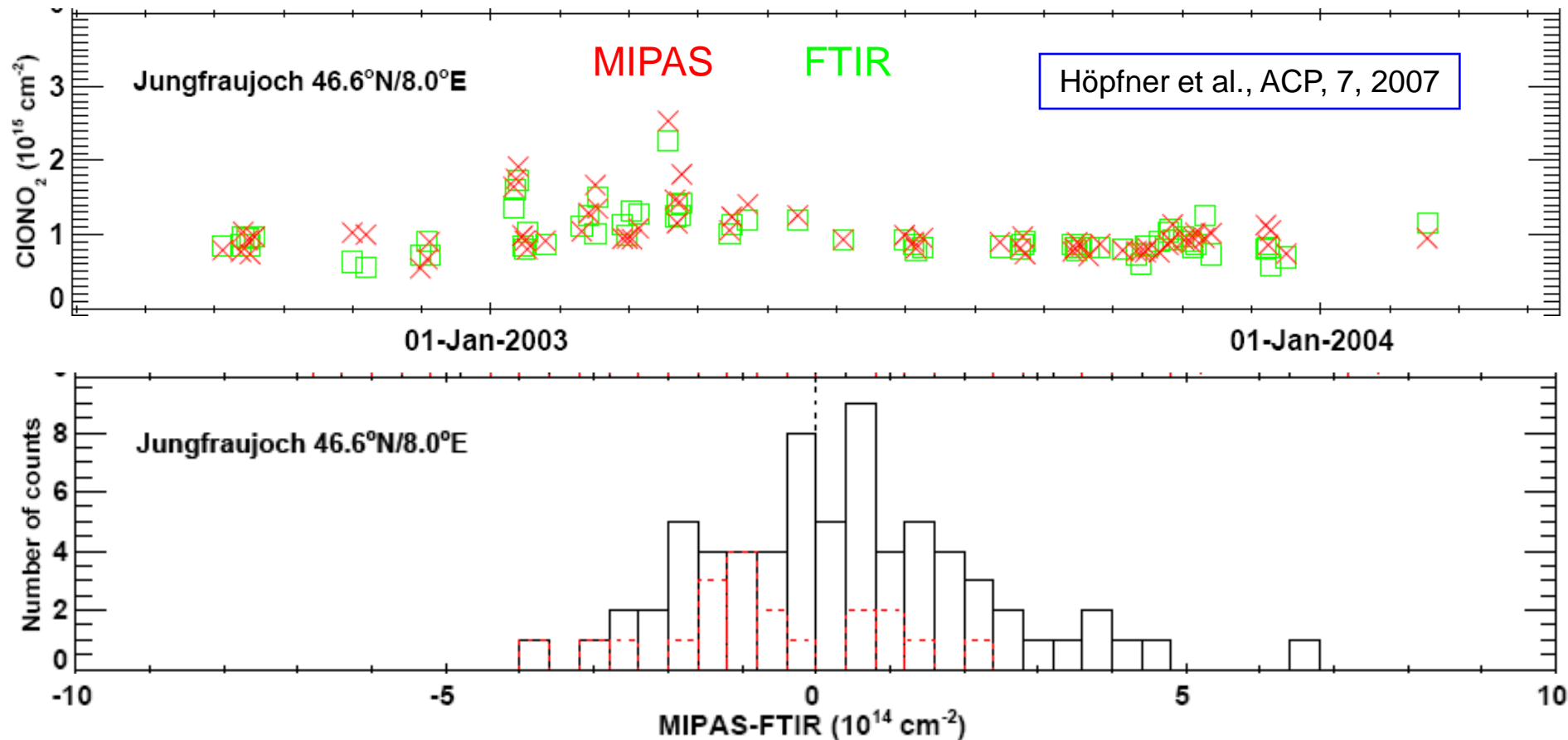
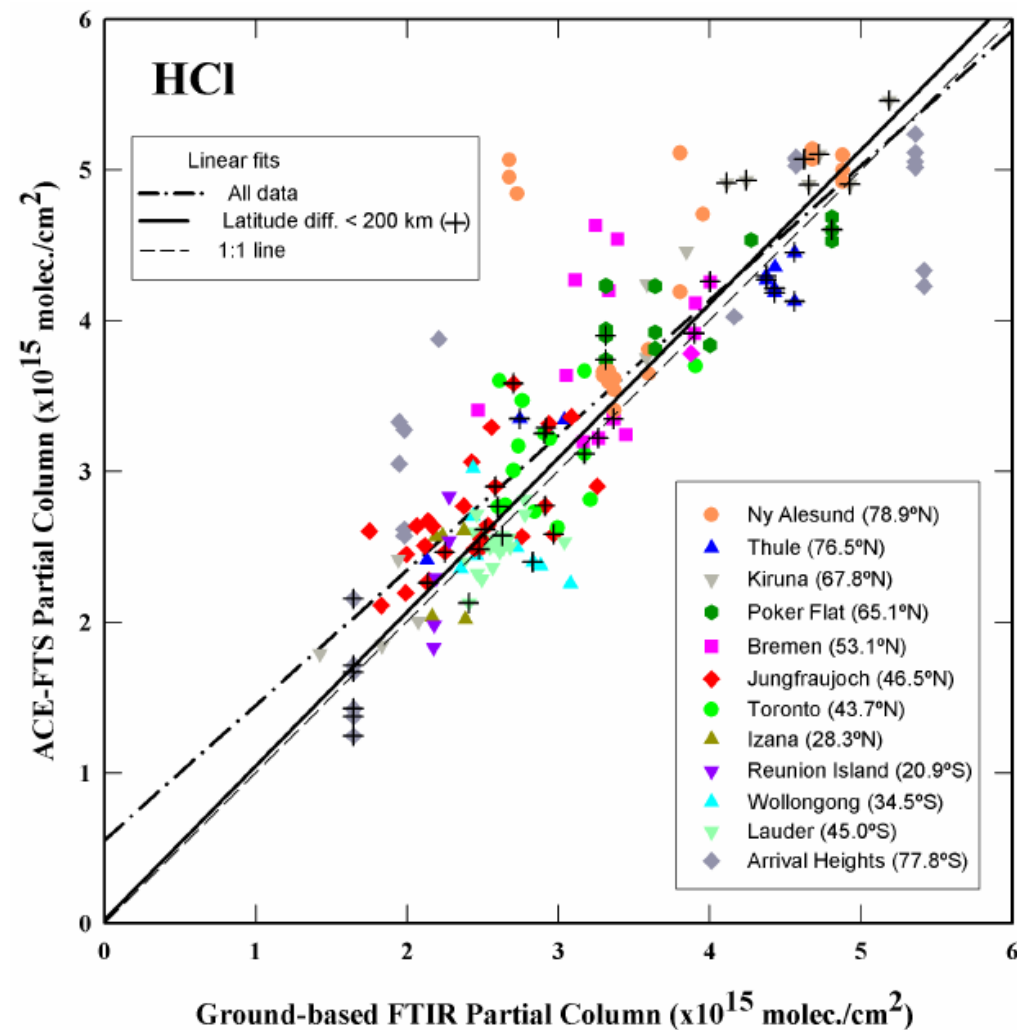


Fig. 15. Histograms of the column amounts daily differences for the collocation criterion $\Delta d_{\max}=800$ km, $\Delta t_{\max}=8$ h, and $\Delta p v_{\max}=3 \times 10^{-6}$ Km² kg⁻¹ s⁻¹ at 475 K (black solid) and $\Delta d_{\max}=400$ km, $\Delta t_{\max}=4$ h, and $\Delta p v_{\max}=3 \times 10^{-6}$ Km² kg⁻¹ s⁻¹ at 475 K (red dotted).

Validation of ACE-FTS HCl columns

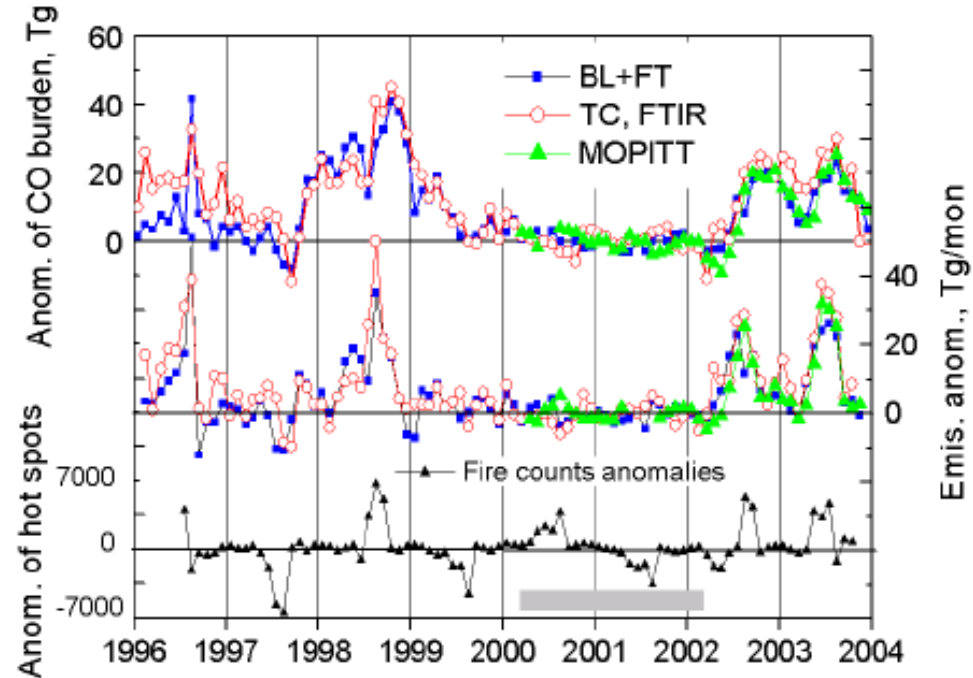
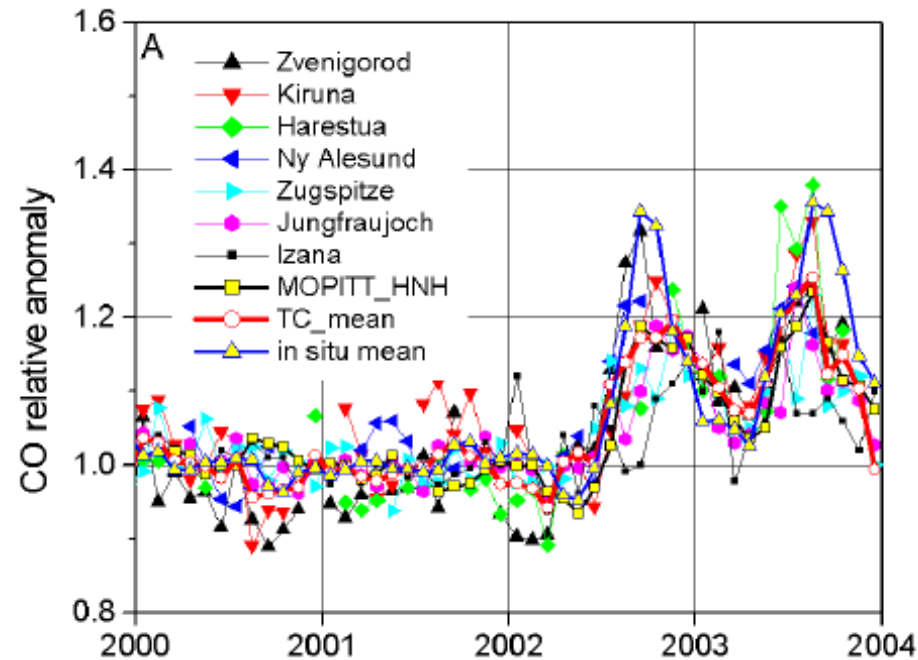


	Mean rel. dif. (%), 1 σ	Std. Er. (%)	# coinc.
All data (12 sites) < 1000 km	6.95 15.95	1.21	174
Lat. diff. < 200 km	2.03 11.74	1.81	42

Fig. 8. Scatter plot of the ACE-FTS partial columns versus the ground-based coincident measurements, taken within ± 24 h and 1000 km (restricted to 500 km for Kiruna and Thule, relaxed to 1200 km for Reunion Island, see text). See inserted legend for identification of the sites. Linear fit to all data points is reproduced as a dash-dotted black line. When restricting latitudinal difference to less than 200 km (see symbols with pluses), the correlation is improved, with a linear fit (continuous black line) close to the 1:1 line (dashed line).

Mahieu et al., ACP, 8, 2008.
 Includes corresponding results for HF

Impact of biomass burning on atmospheric composition



Yurganov et al., ACP, 5, 2005.

- Looking at – and quantifying – CO emission anomalies between 30-90°N
- Study including data from *in situ*, ground-based FTIRs and MOPITT/Terra
- Additional emissions of 95 and 130 Tg in 2002 and 2003, respectively
- Associated to strong boreal forest fires that occurred mainly in Russia

Biomass burning

- Yurganov et al. [2005] about CO anomalies
- In addition to CO, many other species typically present in biomass burning plumes are accessible to gb-FTIR, e.g. HCN, C₂H₂, C₂H₆, C₂H₄, HCOOH, OCS, CH₄, [CH₃OH], allowing to calculate emission ratios; for some of these species, we have good vertical information available, which still increase the value of these measurements
- Related ongoing activity: investigation of CO long-term trend and comparison with EMPA *in situ* measurements, in collaboration with B. Dils et al. [BIRA-IASB] and M. Steinbacher et al. [EMPA] [see talk by M. De Mazière]

Valorisation

- Publications, a complete list of all publications published since 1986 is available from <http://girpas.astro.ulg.ac.be>
- For the 2003-2008 time period, 28 peer-reviewed publications have included results from FTIR measurements at the Jungfraujoch
- In addition, results have been included in several Assessment Reports (6 over 2003-2008), at the international [WMO, IPCC, SPARC-ASAP] and Belgian national levels [A&I, EEW2006, MIRA-T], which are essential tools for decision makers

Archiving: current status and perspectives

- Essential for promoting the use of the data in the scientific community
- Actually, Jungfraujoch FTIR data are essentially archived in NASA-Ames at the NDACC data center (1989-2008/08) daily mean total columns of HCl, HF, ClONO₂, COF₂, CFC-12, HCFC-22, NO, NO₂, HNO₃, O₃), and in hdf at the ENVISAT CAL/VAL database (2002-2004 individual total columns measurements of CO, CH₄, CO₂, O₃, N₂O, NO, NO₂, HNO₃)
- NDACC will adopt soon the hdf format: significant improvement allowing to archive in self-described files the retrieves profiles, the a priori state, error covariance and averaging kernel matrices and many ancillary useful informations
- Our team is now ready to provide data in hdf, using the template defined within the NDACC-InfraRed Working Group



groundbased_ftir.ch4_ulg002_jungfrauoch_o2_20040130t095652z_001.hdf

Profile	Normal	Detail	
+ DATETIME			2.2 Kb ▲
+ LATITUDE.INSTRUMENT			8 bytes
+ LONGITUDE.INSTRUMENT			8 bytes
+ ALTITUDE.INSTRUMENT			8 bytes
+ PRESSURE.SURFACE_INDEPENDENT			2.2 Kb
+ TEMPERATURE.SURFACE_INDEPENDENT			2.2 Kb
+ ALTITUDE			328 bytes
+ ALTITUDE.BOUNDARIES			328 bytes
+ PRESSURE_INDEPENDENT			89.0 Kb
+ TEMPERATURE.AIR_INDEPENDENT			89.0 Kb
+ CH4.MIXING.RATIO_ABSORPTION.SOLAR			89.0 Kb
+ CH4.MIXING.RATIO_ABSORPTION.SOLAR_APRIORI			89.0 Kb
+ CH4.MIXING.RATIO_ABSORPTION.SOLAR_AVK			3.6 Mb
+ CH4.MIXING.RATIO_ABSORPTION.SOLAR_INTEGRATION.TIME			2.2 Kb
+ CH4.MIXING.RATIO_ABSORPTION.SOLAR_UNCERTAINTY.RANDOM			3.6 Mb
+ CH4.MIXING.RATIO_ABSORPTION.SOLAR_UNCERTAINTY.SYSTEMATIC			3.6 Mb
+ CH4.COLUMN.VERTICAL.PARTIAL_ABSORPTION.SOLAR			89.0 Kb
+ CH4.COLUMN.VERTICAL.PARTIAL_ABSORPTION.SOLAR_APRIORI			89.0 Kb
+ CH4.COLUMN.VERTICAL_ABSORPTION.SOLAR			2.2 Kb
+ CH4.COLUMN.VERTICAL_ABSORPTION.SOLAR_APRIORI			2.2 Kb
+ CH4.COLUMN.VERTICAL_ABSORPTION.SOLAR_AVK			89.0 Kb
+ CH4.COLUMN.VERTICAL_ABSORPTION.SOLAR_UNCERTAINTY.RANDOM			2.2 Kb
+ CH4.COLUMN.VERTICAL_ABSORPTION.SOLAR_UNCERTAINTY.SYSTEMATIC			2.2 Kb
+ ANGLE.SOLAR_AZIMUTH			2.2 Kb
+ ANGLE.SOLAR_ZENITH.ASTRONOMICAL			2.2 Kb
+ H2O.MIXING.RATIO_ABSORPTION.SOLAR			89.0 Kb
+ H2O.COLUMN.VERTICAL_ABSORPTION.SOLAR			2.2 Kb ▼