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LONG-TERM TRENDS of a DOZEN DIRECT GREENHOUSE GASES DERIVED from INFRARED SOLAR ABSORPTION SPECTRA RECORDED at the JUNGFRAUJOCH STATION

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INSTRUMENTATION, OBSERVATIONAL DATABASE AND TOOLS

- Two high-resolution Fourier Transform Infrared (FTIR) spectrometers are operated under clear-sky conditions at the high-altitude International Scientific Station of the Jungfraujoch (ISSJ, 46.5ºN, 8.0ºE, 3580m a.s.l.), within the framework of the Network for the Detection of Atmospheric Composition Change (NDACC, http://www.ndacc.org).

- We use high-resolution (0.003 to 0.007 cm^{-1}) broadband IR solar absorption spectra recorded year-round between 2 and 16 microns. Since 1984, more than 45000 FTIR spectra have been collected, using either HgCdTe or InSb detectors. The measurement density has increased since the early 1990s after the installation of a second interferometer, a Bruker IFS-120HR. Signal-to-noise ratios are often larger than 1000, reaching 3500-4000 in the most favorable cases.

- Since the FTIR technique allows recording broadband spectra, features of numerous atmospheric species are available. At present, about two dozen gases are retrieved from Jungfraujoch observations. Among them, 11 are direct greenhouse gases (GHGs): water vapor, CO_2, CH_4, N_2O, CF_4, SF_6, CCl_3F, CCl_2F_2, $CHClF_2$, CCl_4 and tropospheric ozone. Corresponding time series are shown here, we further report about their long-term trends.

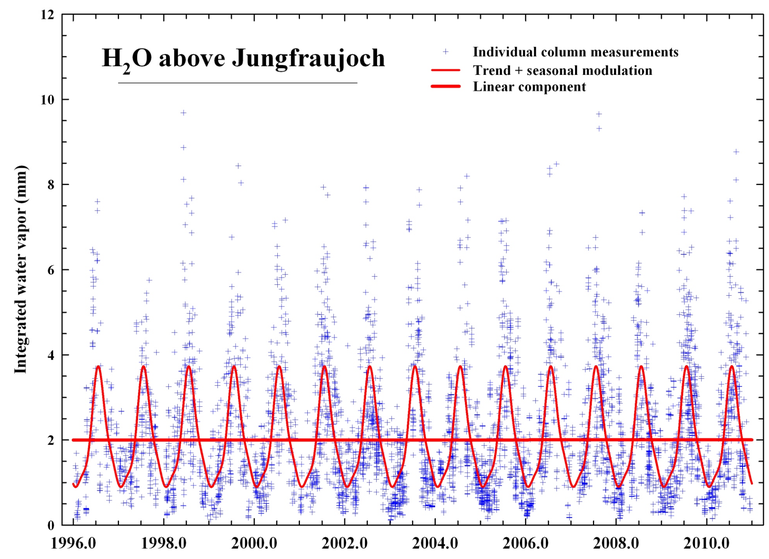
- All retrievals have been performed either with the SFIT-1, SFIT-2 or PROFFIT-9 algorithm, allowing producing long-term total column time series of the target gases. In addition, information on their vertical distributions with altitude can generally be deduced when using the two latter codes which both implement the Optimal Estimation Method formalism of Rodgers [1990]. Various versions of the HITRAN spectroscopic line parameter compilations have been used here [e.g. Rothman et al., 2009]. Also, cross-sections available for some of the target gases (e.g. the CFCs) have been converted to pseudolines, compatible with the SFIT codes, by G.C. Toon (NASA-JPL).

	Greenhouse gas	Time period	Trend ± 2σ *	
	H_2O (water vapor)	1996-2010, year-round data	+(0.002 ± 0.074) mm/decade	+(0.01 ± 0.37) %/decade
		1996-2010, Jun. to Aug. data	-(0.006 ± 0.18) mm/decade	-(0.02 ± 0.53) %/decade
		1996-2010, Dec. to Feb. data	-(0.09 ± 0.10) mm/decade	-(0.80 ± 0.86) %/decade
	Tropospheric ozone (O_3)	1995-2010	+(1.04 ± 0.52) x10^{16} molec./cm^2	+(0.12 ± 0.06) %/yr
KYOTO	CO_2 (carbon dioxide)	1984-2011/05	+(2.45 ± 0.02) x10^{19} molec./cm^2	+(0.50 ± 0.01) %/yr
KYOTO	CH_4 (methane)	2000-2004	-(0.59 ± 0.91) x10^{16} molec./cm^2	-(0.03 ± 0.04) %/yr
KYOTO		2005-2011/05	+(5.52 ± 0.56) x 10^{16} molec./cm^2	+(0.24 ± 0.02) %/yr
KYOTO	N_2O (nitrous oxide)	1984-2011/05	+(1.04 ± 0.03) x 10^{16} molec./cm^2	+(0.27 ± 0.01) %/yr
KYOTO	CF_4 (carbon tetrafluoride)	1990-2010	+(1.14 ± 0.04) x 10^{13} molec./cm^2	+(1.25 ± 0.05) %/yr
KYOTO	SF_6 (sulfur hexafluoride)	2001-2011/05	+(4.59 ± 0.16) x 10^{12} molec./cm^2	+(6.75 ± 0.24) %/yr
MONTREAL	CCl_3F (CFC-11)	2001-2011/05	-(2.80 ± 0.15) x10^{13} molec./cm^2	-(0.88 ± 0.05) %/yr
MONTREAL	CCl_2F_2 (CFC-12)	2001-2011/05	-(1.75 ± 0.26) x 10^{13} molec./cm^2	-(0.25 ± 0.04) %/yr
MONTREAL	$CHClF_2$ (HCFC-22)	2001-2011/05	+(8.79 ± 0.13) x 10^{13} molec./cm^2	+(4.26 ± 0.06) %/yr
MONTREAL	CCl_4 (carbon tetrachloride)	1999-2011/05	-(1.49 ± 0.08) x 10^{13} molec./cm^2	-(1.10 ± 0.06) %/yr

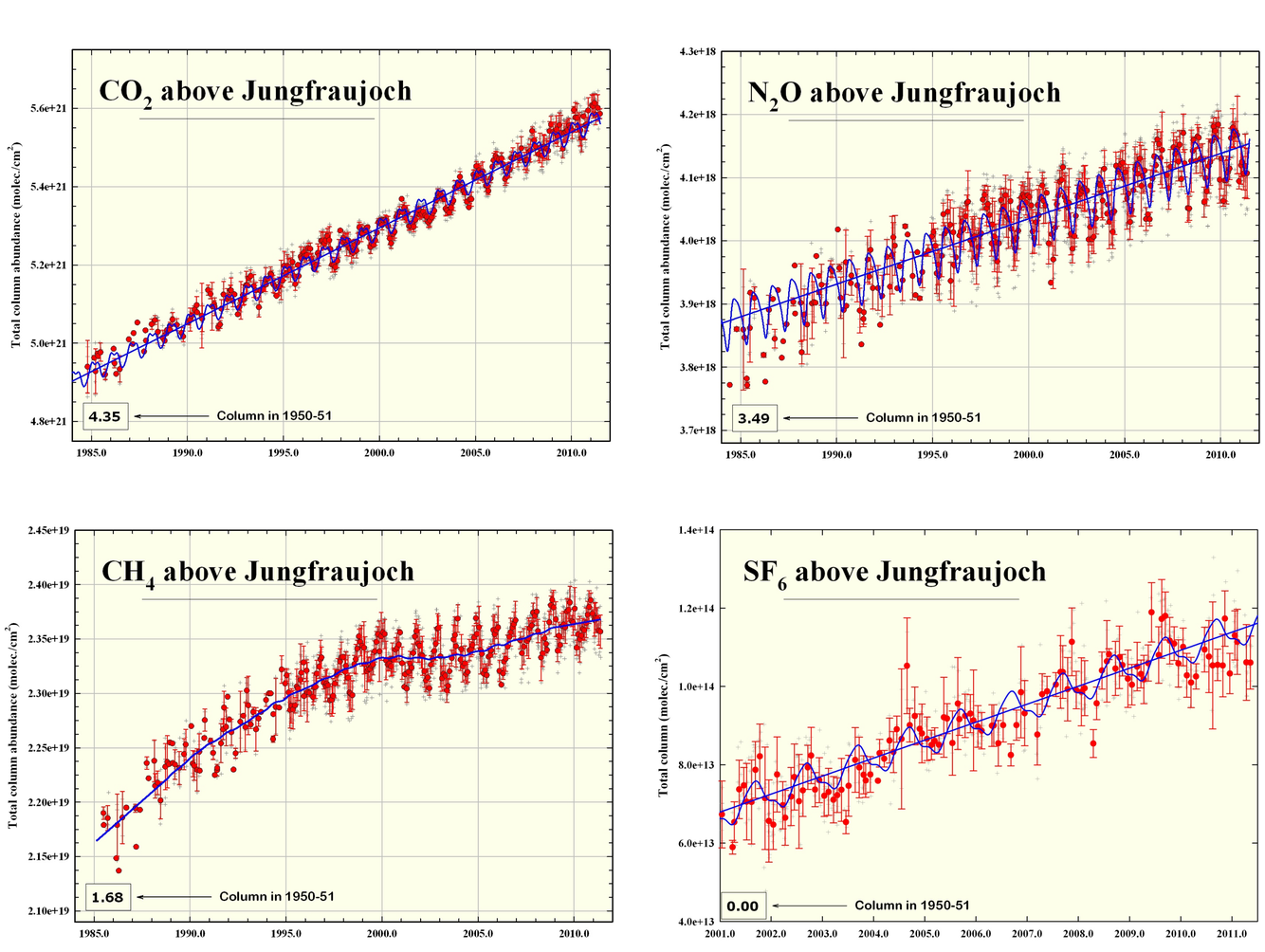
* For relative trends, the abundance of the gas for the first year of the time period under consideration is taken as reference.

TABLE 1.

FIGURE 2. Right-hand side: time series of integrated water vapor above the Jungfraujoch station, derived within the framework of the MUSICA project. [e.g. Schneider et al., 2010]. The red curve shows a fit to the individual measurements reproduced as blue crosses, it indicates a very strong seasonal cycle, with maximum water columns observed during summertime. The linear component of the fitted function is also reproduced as a thick red line, a long-term trend of (0.002 ± 0.005) mm/decade is computed for the whole data set, i.e. not statistically significant at the 95% uncertainty level. The same conclusion is verified when considering only the summer or winter months, respectively (see Table 1).



TIME SERIES OF GHGs TARGETED BY THE KYOTO PROTOCOL



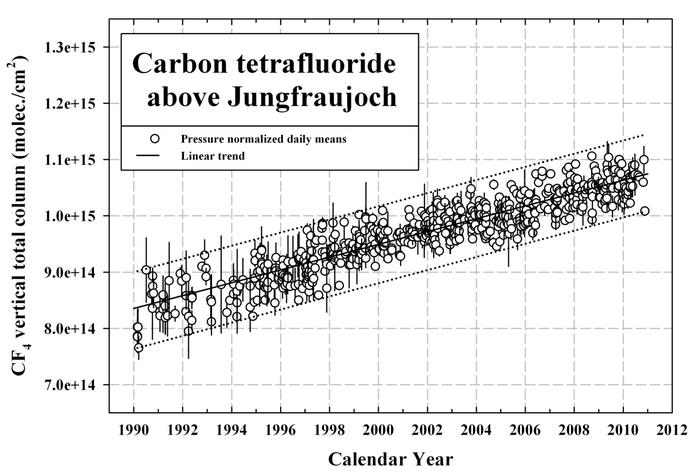


FIGURE 1. Total column time series of greenhouse gases targeted by the Kyoto Protocol and derived from regular FTIR observations performed at the Jungfraujoch station. When available, mean total columns deduced from pioneering observations performed at the same site in the early 1950s are indicated in the lower left corner of each frame.
With the exception of methane which exhibits a non-monotonic accumulation in the Earth's atmosphere, the four other gases show a somewhat regular build up of their abundance. In each case, we note significant increase of the total columns over the time period under consideration. Table 1 reports the numerical values deduced from a fitting to all available daily means, computed with a bootstrap resampling statistical tool described in Gardiner et al. [2008], combining a linear function with a 3rd order Fourier series.

TIME SERIES OF GHGs TARGETED BY THE MONTREAL PROTOCOL

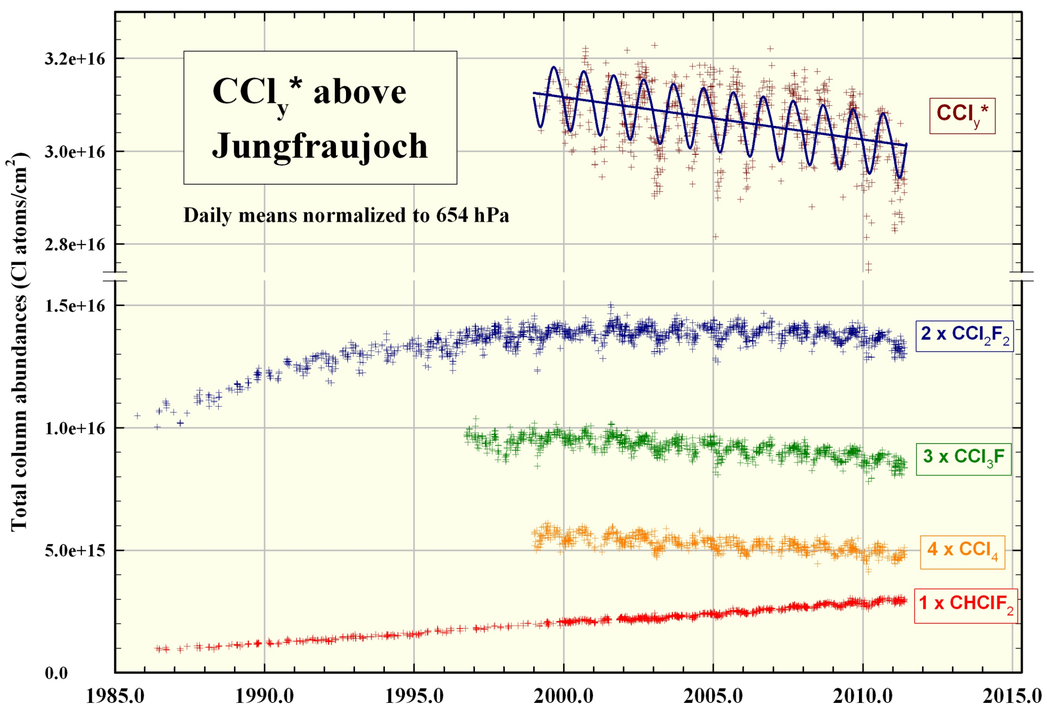


FIGURE 3. Temporal evolution of the long-lived chlorinated source gases retrieved from the Jungfraujoch observational data base, weighted by the number of Cl atoms in each species (notice the vertical scale break). Data points correspond to daily means normalized to 654 hPa. The upper data set is the 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). The seasonal signals seen in most time series essentially result from the tropopause height changes throughout the year.
Although the overall CCl_y* trend is negative, it is important to realize that the largest negative contribution of CFC-11 (-8.4 x10^{13} Cl atoms/cm^2 per year) is currently balanced by the steady accumulation of HCFC-22 (+8.8x10^{13} Cl atoms/cm^2 per year).

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The water vapor data used here are produced within the framework of the MUSICA project.

This communication is dedicated to our colleague and friend Curtis Rinsland who passed away in April 2011. We have enjoyed and benefited from a close and steady collaboration with him, over nearly 30 years for some of us, within the framework of the ATMOS, ACE and NDACC programs and missions. We will deeply miss him.

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