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# CHANGES in ATMOSPHERIC COMPOSITION DISCERNED from LONG-TERM NDACC MEASUREMENTS: TRENDS in DIRECT GREENHOUSE GASES DERIVED from INFRARED SOLAR ABSORPTION SPECTRA RECORDED at the JUNGFRAUJOCH STATION

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## TIME SERIES OF GHGs TARGETED BY THE KYOTO PROTOCOL

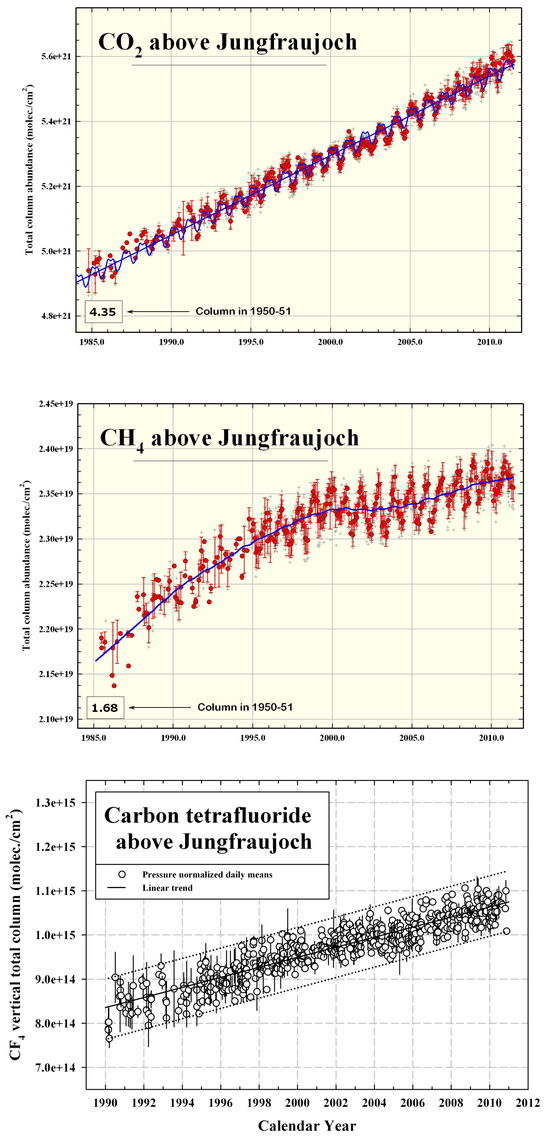


**FIGURE 1.** Total column time series of greenhouse gases targeted by the Kyoto Protocol derived from regular FTIR observations performed at the Jungfraujoch station. When available, mean total columns deduced from pioneering observations performed at the same site in the early 1950s are indicated in the lower left corner of each frames.
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.

## 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 $cm^{-1}$) IR solar absorption spectra recorded year-round since 1984. 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. Among the two dozen gases actually retrieved from Jungfraujoch observations, 10 are direct greenhouse gases (GHGs): water vapor, $CO_2$, $CH_4$, $N_2O$, $CF_4$, $SF_6$, $CCl_3F$, $CCl_2F_2$, $CHClF_2$ and $CCl_4$. Corresponding time series are shown here, we further report about their long-term trends.
- All retrievals have been performed either with the SFIT-1 or the SFIT-2 algorithm (v3.91). The latter code is based on a semi-empirical implementation of the Optimal Estimation Method formalism of Rodgers [1990]. This code allows in most cases to determine information on the vertical distribution of the species accessible to the ground-based FTIR technique. 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).

## TIME SERIES OF GHGs TARGETED BY THE MONTREAL PROTOCOL

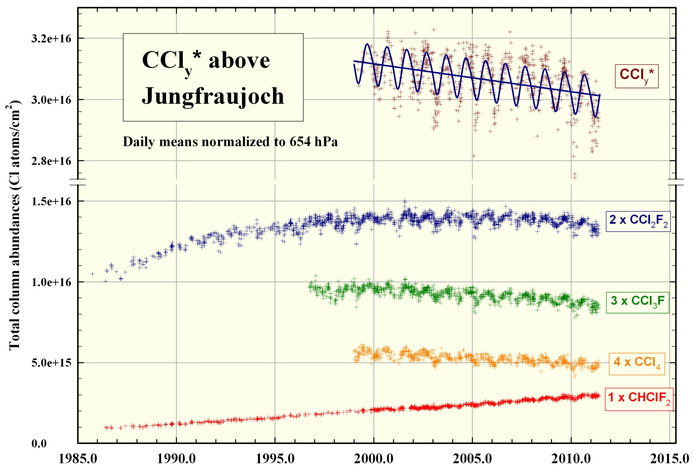


**FIGURE 2.** Temporal evolution of the long-lived chlorinated source gases retrieved from the Jungfraujoch observational data base, weighted by the number of Cl atoms in each species (notice the vertical scale break). Data points correspond to daily means normalized to 654 hPa. The upper data set is 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.

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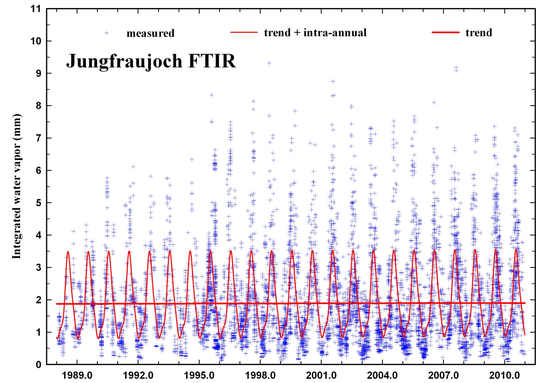


**FIGURE 3.** Long-term time series of integrated water vapor above the Jungfraujoch station, updated from Sussmann et al. [2009]. The red curve shows a fit to the individual measurements reproduced as blue crosses, it indicates a very strong seasonal cycle, with maximum water columns observed during summertime. The linear component of the fitted function is also reproduced as a thick red line, a long-term trend of (0.03 ± 0.04) mm/decade is computed for the whole data set, i.e. not statistically significant at the 95% uncertainty level. In contrast, significant positive and negative trends are deduced when considering only the summer or winter months, respectively (see Table 1).

| Greenhouse gas | Time period | Trend ± 2σ * | |
|---|---|---|---|
| $H_2O$ (water vapor) | 1988-2010, year-round data | +(0.03 ± 0.04) mm/decade | +(1.6 ± 2.1) %/decade |
| | 1988-2010, Jun. to Aug. data | +(0.36 ± 0.10) mm/decade | +(13.2 ± 3.8) %/decade |
| | 1998-2010, Dec. to Feb. data | -(0.13 ± 0.04) mm/decade | -(11.4 ± 3.4) %/decade |
| $CO_2$ (carbon dioxide) | 1984-2011/05 | +(2.45 ± 0.02) x$10^{19}$ molec./cm$^2$ | +(0.50 ± 0.01) %/yr |
| $CH_4$ (methane) | 2000-2004 | -(0.59 ± 0.91) x$10^{16}$ molec./cm$^2$ | -(0.03 ± 0.04) %/yr |
| | 2005-2011/05 | +(5.52 ± 0.56) x $10^{16}$ molec./cm$^2$ | +(0.24 ± 0.02) %/yr |
| $N_2O$ (nitrous oxide) | 1984-2011/05 | +(1.04 ± 0.03) x $10^{16}$ molec./cm$^2$ | +(0.27 ± 0.01) %/yr |
| $CF_4$ (carbon tetrafluoride) | 1990-2010 | +(1.14 ± 0.04) x $10^{13}$ molec./cm$^2$ | +(1.25 ± 0.05) %/yr |
| $SF_6$ (sulfur hexafluoride) | 2001-2011/05 | +(4.59 ± 0.16) x $10^{12}$ molec./cm$^2$ | +(6.75 ± 0.24) %/yr |
| $CCl_3F$ (CFC-11) | 2001-2011/05 | -(2.80 ± 0.15) x$10^{13}$ molec./cm$^2$ | -(0.88 ± 0.05) %/yr |
| $CCl_2F_2$ (CFC-12) | 2001-2011/05 | -(1.75 ± 0.26) x $10^{13}$ molec./cm$^2$ | -(0.25 ± 0.04) %/yr |
| $CHClF_2$ (HCFC-22) | 2001-2011/05 | +(8.79 ± 0.13) x $10^{13}$ molec./cm$^2$ | +(4.26 ± 0.06) %/yr |
| $CCl_4$ (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.**

**References**

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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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