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Column abundance measurements of formaldehyde above the Jungfraujoch

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This manuscript reports on the variability and long-term trend in the vertical column abundance of formaldehyde, H_2CO, above the Jungfraujoch station (Swiss Alps, 46.5°N latitude, 8.0°E longitude, 3.58 km altitude, primary NDSC station). The analysis is based on high resolution and high signal-to-noise ratio solar absorption spectra recorded with two infrared Fourier transform spectrometers (one built at the University of Liège and operated at the Jungfraujoch since 1984, and the other being a commercial Bruker 120HR in use since 1990). For this specific study, InSb detectors and optical wide-band filters isolating the 1900-3200 cm^{-1} domain, in which the most intense H_2CO features occur, were used. Typical spectral resolution achieved was about 0.005 cm^{-1}. To increase the S/N, the spectra have been binned over 7 solar elevation intervals (airmass step of 1.6) and averaged over 2-month periods, leading to average spectra with resulting S/N up to 20000.

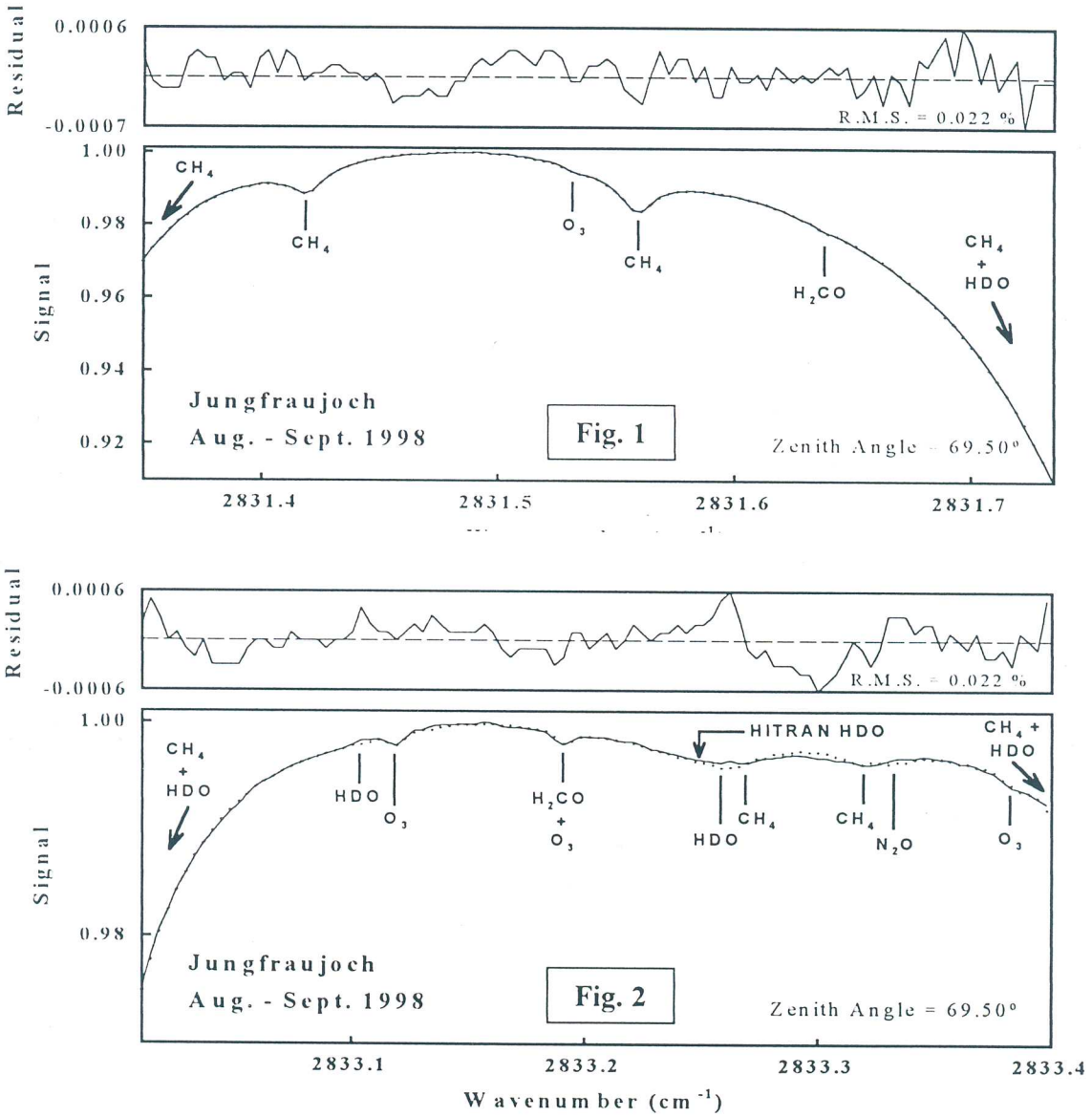
The vertical columns have been derived with the SFIT 1.09e algorithm (Rinsland et al., 1991), which fits spectra in a non-linear least squares iterative procedure, by scaling the mixing ratio of up to 5 target molecules. The line-by-line calculations use a 29-layer atmospheric model extending from the surface to 100 km. A Voigt line shape is assumed and there is an option to include pressure shifts. The spectroscopic parameters are read from the HITRAN-96 database (Rothman et al., 1998). Vertical pressure and temperature profiles, for the atmosphere above the Jungfraujoch, are obtained from the National Centers for Environmental Prediction (NCEP) archive.

Microwindows selection

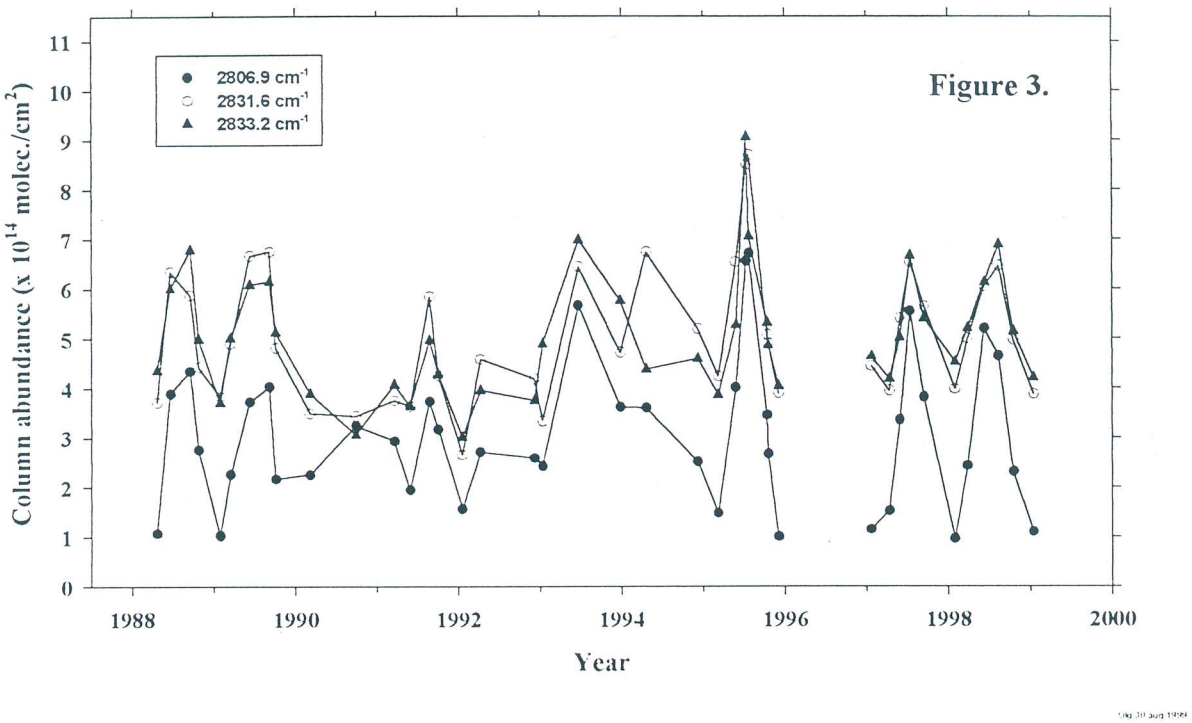
An extensive search for the strongest, free of interferences H_2CO absorptions was carried out over the 2720 – 2930 cm^{-1} spectral domain. The coherence among the H_2CO columns derived from the microwindows investigated was also adopted as a selection criterion.

Finally, the best domains, together with the main H_2CO absorptions and interference species, retained for the current study are the following:

- 2831.35 – 2831.74 cm^{-1} (H_2CO at 2831.6 cm^{-1}; also fitted: CH_4, HDO, O_3) (Fig. 1)
- 2833.01 – 2833.40 cm^{-1} (H_2CO at 2833.2 cm^{-1}; also fitted: CH_4, N_2O, HDO, O_3) (Fig. 2)
- 2806.37 – 2807.04 cm^{-1} (H_2CO at and 2806.5 and 2806.9 cm^{-1}; also fitted: CH_4, N_2O, H_2O, O_3) (Fig. 4)



Figures 1 and 2. Two examples of domains selected to retrieve H_2CO in the Jungfraujoch spectra, averaged from August – September 1998 observations. In each figure, the lower panel shows the computed spectrum (dotted trace) fitted to the observed one (continuous trace); the residuals (observed minus computed) are reproduced in the upper panel. The HDO line at 2833.246 cm^{-1} (position in HITRAN-96) had to be moved to 2833.258 cm^{-1}.



Results

The vertical column abundances of formaldehyde above the Jungfraujoch, as retrieved from the 3 best spectral domains selected in this study, are reproduced in Fig. 3. The time base extends from 1988 to 1999, with retrievals averaged over 2-month periods. The results among the 3 microwindows show a good relative consistency.

The mean bias between the columns from the 2806.9 cm^{-1} domain with respect to the two other ones could be caused by either the presence of unidentified interfering lines, or some inconsistency among the H_2CO spectroscopic line parameters, in particular their intensities.

Because the current atmospheric loading of H_2CO produces extremely weak absorptions (generally < 0.1 %), the main sources of error are the limited S/N of the spectra and the presence of numerous unidentified weak absorption features (see example in Fig. 4).

Estimation of the uncertainty in the H_2CO column retrievals is based on the comparison between the results from the 3 microwindows. We notice that the average of the H_2CO column differences between the 2831.6 and the 2833.2 cm^{-1} regions is negligible (within 1.2 %), while it amounts to 42 % for the 2831.6 and the 2806.9 cm^{-1} regions. The corresponding standard deviations on these differences are respectively 13 % and 19 %.

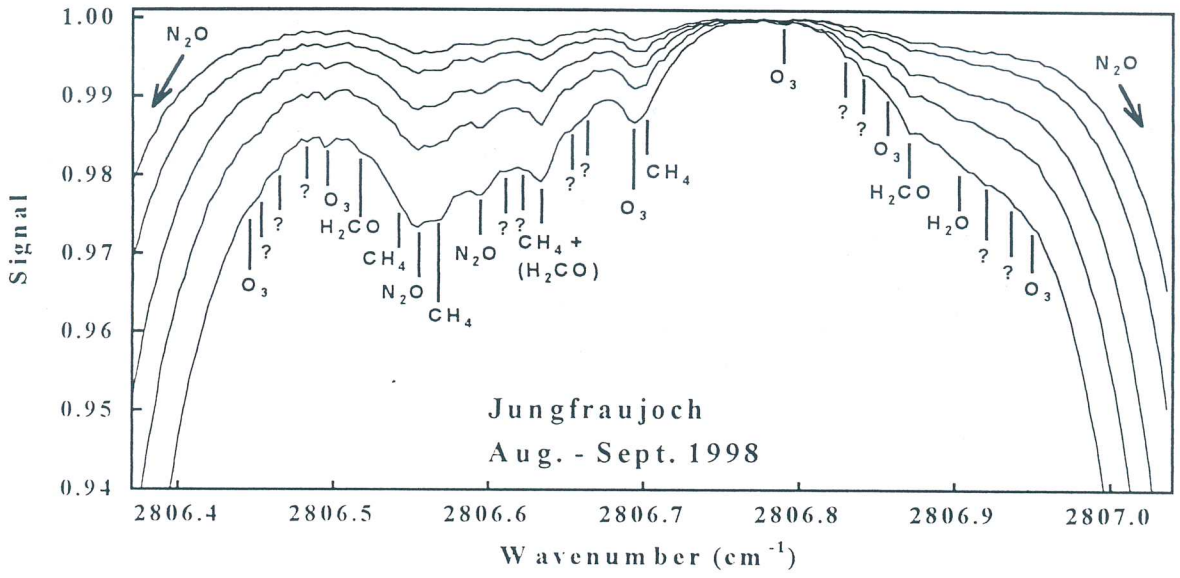


Figure 4. One of the spectral domains selected to retrieve H_2CO, averaged from August – September 1998 observations. The 5 tracings correspond to mean solar zenith angles of 69.50, 76.04, 81.39, 84.15 and 86.41 degrees (top to bottom). The position of identified lines of H_2CO, CH_4, N_2O, H_2O and O_3, as well as numerous unidentified features ("?" symbols) are indicated by tick marks. Such interferences render the quantification of atmospheric formaldehyde difficult.

Consequently, and on the basis of the 3 domains considered here, it is estimated that absolute formaldehyde columns can not be retrieved to better than 40 % but that the precision on derived columns is not worse than 20 %. Additional laboratory work on both H_2CO and interfering features remain mandatory to improve H_2CO retrievals.

Because of the important variability of H_2CO and the high uncertainties in the retrievals, no statistically significant trend emerges from our current database. However, a seasonal variation of H_2CO, with lower columns in winter, appears to be present.

Acknowledgments:

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Reference:

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