RETRIEVALS of C_2H_2 from HIGH-RESOLUTION FTIR SOLAR SPECTRA RECORDED at the JUNGFRAUJOCH STATION (46.5ºN) and COMPARISON with ACE-FTS OBSERVATIONS

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1. INSTRUMENTATION AND OBSERVATIONAL DATABASE

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

More than 3300 high-resolution (0.004 to 0.009 cm^{-1}) IR absorption solar spectra relevant to the present study have been recorded regularly at the Jungfraujoch, essentially since the early 1990s.

Limits (cm^{-1})	Fitted species
3255.180 – 3255.725	C_2H_2, H_2O, solar spectrum
3268.290 – 3268.645	C_2H_2, H_2O, O_3, solar spectrum
3278.000 – 3278.360	C_2H_2, $H_2{}^{18}O$, O_3, solar spectrum
3305.825 – 3305.350	C_2H_2, H_2O, $H_2{}^{18}O$

TABLE 1. Characteristics of the four microwindows simultaneously used to retrieve C_2H_2. Some windows have been enlarged to include additional absorption lines of the major interfering species (i.e. H_2O and $H_2{}^{18}O$), in order to optimize their adjustments. Per-spectrum initial guess profiles for water vapor are determined from pre-fits performed in the same spectral region.

2. RETRIEVAL STRATEGY

All retrievals have been performed with the SFIT-2 algorithm (v3.91) which is based on a semi-empirical implementation of the Optimal Estimation Method formalism of Rodgers [1]. This code allows to determine information on the vertical distribution of most of the species accessible to the ground-based FTIR technique.

The HITRAN-2004 spectroscopic compilation used here includes the latest August 2006 updates for water vapor. The acetylene (C_2H_2) a priori information (vertical distribution and covariance matrix assuming an inter-layer correlation of 4 km for extra diagonal elements) is essentially based on ATMOS occultation measurements which were performed in the 39-49ºN latitude range during the ATLAS-3 mission of November 1994.

There are exploitable C_2H_2 absorption lines in both the so-called MCT and INSB spectral domains, more precisely in the 750-780 and 3250-3305 cm^{-1} ranges. Although all these lines are weak (~1% absorption), they can be used for the retrieval of C_2H_2 from ground-based high-resolution IR spectra. In the present study, a systematic and careful look at ISSJ "dry and wet observations" has allowed to identify 5 candidate lines close to 3251, 3255, 3268, 3278 and 3305 cm^{-1}. The major interferences are water vapor (H_2O) and one of its less abundant isotopologues ($H_2{}^{18}O$). Absorptions by solar spectrum, O_3 and HCN have also to be accounted for.

Each of the identified lines has been characterized in terms of information content using a representative subset of spectra. Mean values for the degree of freedom for signal (DOFS) range from 1.2 to 1.5. Fits to the 3268 cm^{-1} line are less satisfactory, hence this feature was subsequently discarded from the list of candidate lines.

A strategy using simultaneously four C_2H_2 lines has therefore been developed and validated; the final adopted settings are summarized in TABLE 1.

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BACKGROUND INFORMATION ON ACETYLENE (C_2H_2)

Acetylene (C_2H_2) is among the nonmethane hydrocarbons (NMHCs) accessible to infrared remote sensing technique.

As a product of combustion and biomass burning, it is emitted at the Earth's surface and further transported and mixed into the troposphere.

Destruction by OH is the main removal process. The average tropospheric lifetime of C_2H_2 is estimated at about 1 month on the global scale; at mid-latitudes, it varies between 20 days in summer to 160 days in winter.

This compound is appropriate to study tropospheric pollution and transport, and is often used in conjunction with other tracers of fires.

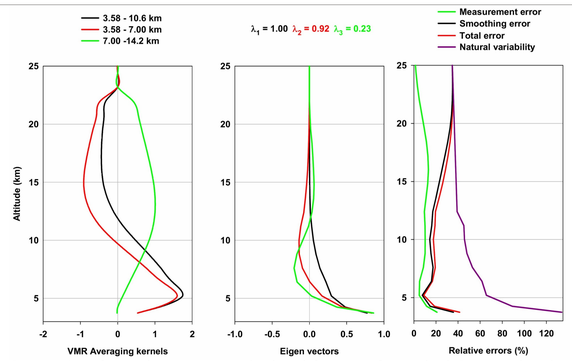

FIGURE 1. Information content calculated for vmr, for typical C_2H_2 retrievals at the Jungfraujoch station. Left frame shows vmr averaging kernels for selected merged layers. The three first eigenvectors are reproduced in the middle frame. Right frame gives the corresponding error budget, with identification of the main error components, together with the assumed variability.

3. CHARACTERIZATION OF THE RETRIEVED PRODUCTS

Information content and error budget have been carefully evaluated. FIGURE 1 displays typical results computed for vmr.

vmr averaging kernels for merged layers are displayed on the left for the 3.58-10.6, 3.58-7.0 and 7.0-14.2 km altitude ranges. The two first eigenvectors and corresponding eigenvalues (see middle frame, in black and red, respectively) show that information on the C_2H_2 tropospheric column is only coming from the retrieval, and that discrimination between partial columns below and above 7 km is essentially coming from the retrieval (92%). Further vertical resolution is available as indicated by the third eigenvector (in green), but with a predominant contribution from the a priori (77%).

The error budget affecting the retrieved vmrs below 25 km is given in the right panel of FIGURE 1. Total and individual error profiles are provided and identified (see color codes). These typical errors correspond to 4% of the tropospheric column and to 12% of the 7-14.2 km partial column.

References

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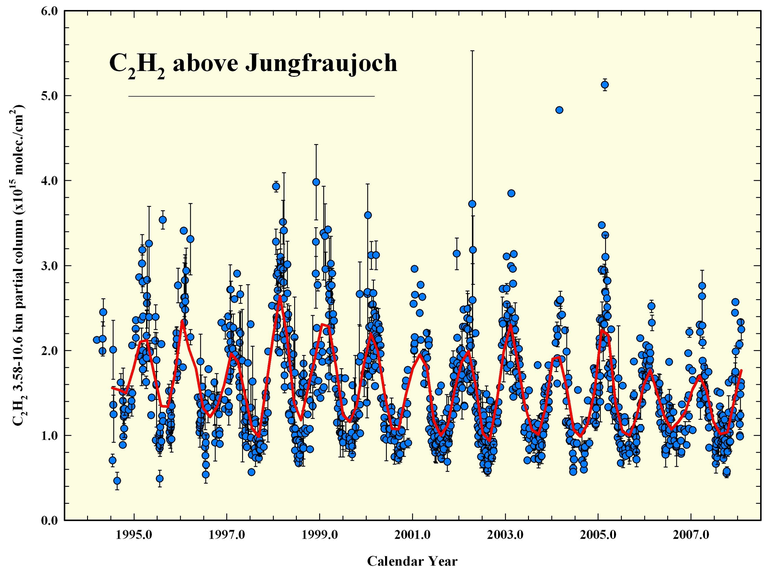

FIGURE 2. Time series of daily mean tropospheric columns of C_2H_2 above the Jungfraujoch station. Error bars correspond to the standard deviations around the daily means, they show that very high intra-day variability can be observed in some instances.

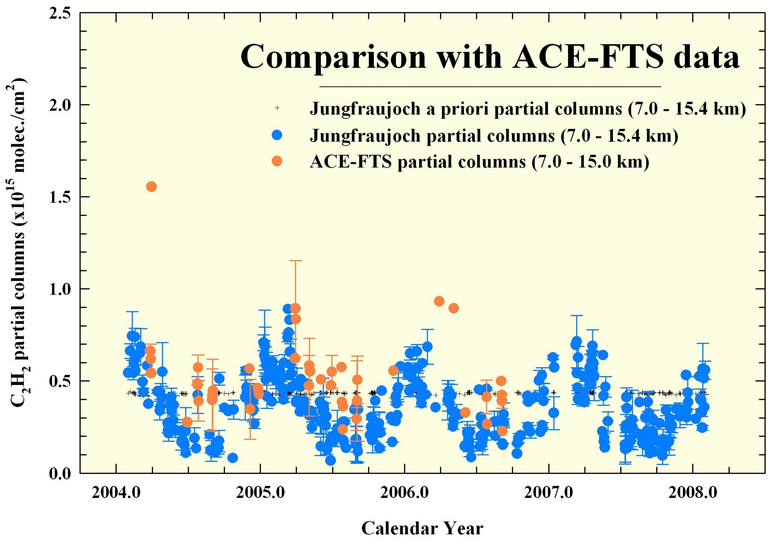

FIGURE 3. Comparison between the Jungfraujoch and ACE-FTS partial column time series, computed over the 7 to 15 km altitude range. Error bars give the standard deviations around the daily means.

4. C_2H_2 TROPOSPHERIC COLUMN TIME SERIES

The retrieval approach described here has been applied to all available Jungfraujoch spectra from 1994 onwards. Resulting time series of daily mean tropospheric columns (from 3.58 to 10.6 km) is displayed in FIGURE 2.

Among striking features, we notice high tropospheric columns observed in 1998 correlated with documented high values of carbon monoxide resulting from important biomass burning [2]. This is also consistent with record-high troposheric columns of HCN and C_2H_6 observed the same year at the Jungfraujoch.

Our measurements allow to characterize the strong seasonal variation of C_2H_2, with maximum columns generally observed around mid-February. On average, the peak-to-peak amplitude amounts to nearly 90% of the mean yearly column.

Trend analysis and correlation with other tropospheric species (e.g. C_2H_6, CO, HCN) are ongoing and will be the subject of future presentations.

5. COMPARISON WITH ACE-FTS DATA

The ACE-FTS is a Canadian instrument which was launched onboard the SCISAT satellite on 12 August 2003. Since the beginning of routine operations on 21 February 2004, this Fourier Transform Spectrometer has recorded up to 15 sunrise and sunset occultations per day (about every 90 minutes), with a maximum spectral resolution of 0.02 cm^{-1} in the broad 750-4400 cm^{-1} spectral region [3].

ACE-FTS C_2H_2 products used here correspond to an update of the standard version 2.2. Vertical profile distributions are retrieved using 14 lines from both the MCT and INSB domains. Over northern mid-latitudes, vmr values are generally available from 17 km down to at best 7 km. Corresponding ACE-FTS partial columns can be compared to Jungfraujoch data, since the ground-based retrievals are sensitive to that range, as shown by the information content analysis.

FIGURE 3 compares the Jungfraujoch and the ACE-FTS data sets. The latter includes all occultation measurements obtained between 41 and 51ºN latitude and extending down to 7 km.

The Jungfraujoch time series (in blue) is characterized by a clear seasonal modulation, with partial columns ranging from about 1 to $9x10^{14}$ molec./cm^2. The Jungfraujoch and ACE-FTS data sets (in orange) agree reasonably well, although the latter seems to be slightly biased high, especially during summertime. An extension in time of the ACE-FTS data set is needed to confirm these first conclusions.