

H₂O retrievals from Jungfrauoch infrared spectra: some spectroscopic problems.

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Since 1949, solar absorption spectra have been acquired at the Jungfrauoch observatory (Swiss Alps, 46.5°N, 8.0°E, 3580 m a.s.l.), first with grating spectrometers, then with Fourier transform infrared (FTIR) spectrometers.

Most observations collected with these instruments encompass water vapor absorptions, even the narrow spectral domains regularly recorded with the grating instruments to monitor the evolution of important atmospheric constituents. The aim of this work is to analyze all available spectra containing usable H₂O absorption lines, to derive a long-term record of its total column in the atmosphere above the Jungfrauoch.

In addition, more recent FTIR broadband spectra, with better signal-to-noise ratio and resolution, will be analyzed with state of the art algorithm, in order to derive information on the vertical distribution of H₂O.

Instrumentation

Since the fifties, various instruments were installed and operated at the Jungfrauoch observatory by the Institute of Astrophysics of the University of Liège:

- 1950-1951: 1-m focal length infrared Pfund-type grating spectrometer
- 1958-1989: 7.2-m focal length grating Ebert-Fastie spectrometer; installed in 1958, it has been modified in a double-pass instrument in 1963
- 1974-1976: near-infrared stepping Fourier transform spectrometer
- from 1984 onwards: home-made fast scanning FTIR spectrometer
- from 1990 onwards: commercial Bruker IFS-120HR FTIR spectrometer

The FTIR spectrometers are now an essential component of the primary Alpine Station of the NDACC (Network for the Detection of Atmospheric Composition Change, <http://www.ndacc.org>).

Observational database

Most of the old observations have been preserved and are archived in Liège. A few hundreds of grating spectra, recorded on paper from 1950 to 1967, are still available. But the bulk of grating observations consists of around 8000 spectra recorded digitally from 1968 to 1989. These observations were achieved to study the solar spectrum and to record, on the driest days, two solar atlases, one in the near infrared, the other one covering the visible spectrum from 300 nm to the near infrared [Delbouille 1995]. The H₂O line at 694.38 nm was systematically recorded to evaluate the dryness of the atmosphere. From 1976 to 1989, regular atmospheric infrared observations were also carried out with the grating instrument, in narrow spectral windows (about 10 cm⁻¹ wide, resolution ≈ 15 mK) encompassing lines of atmospheric interest. Appropriate H₂O lines contained in these intervals will be used to derive corresponding total columns of water vapor.

A few dozen of near-infrared spectra were recorded from 1974 to 1976 with the stepping Fourier transform spectrometer [Malbrouck 1977]. In respectively 1984 and 1990, two high-resolution FTIR spectrometers were installed at the Jungfrauoch and are still in operation today. They regularly record solar absorption spectra of the atmosphere in the infrared, between 2 and 14 micrometers (5000 to 700 cm⁻¹). More than 40 000 high resolution spectra have been obtained with these two instruments, at typical resolution from 2.85 mK to 6.10 mK (i.e. optical path difference of 175.4 cm to 81.9 cm).

Search for H₂O lines

Numerous water vapor lines are present in the spectral range accessible by the Jungfraujoch FTIR spectrometers. The first part of this work consisted in finding appropriate H₂O lines, temperature-insensitive, free of interferences... in different spectral regions. A first selection, based on the results of a line finding algorithm [Notholt 2006], provided 10278 lines between 700 and 4300 cm⁻¹. In a second selection, only 63 micro-windows were kept (see Table 1). Combination of several micro-windows with strong and weak lines should allow to increase the quantity of information retrieved from the FTIR spectra, in particular the information characterizing the vertical distribution of water vapor. However, when several micro-windows are simultaneously used, the quality of the fittings of the spectra deteriorates, which probably results from inadequacies in the spectroscopic parameters describing the different lines (Figure 1).

In the case of the grating spectra, the narrow spectral domains recorded for regular atmospheric trend studies contain very few H₂O lines and we do not have many choices in the lines selection.

One major difficulty associated to the present work will be to carefully intercalibrate the different H₂O lines selected for retrieval in different spectral regions, because the spectroscopic parameters of these lines are not always known with high precision and biases regularly exist between intensities of lines from different bands. Fortunately the different instruments were always operated with some overlap (except in 1950 !) and it should therefore be possible to intercalibrate the lines in different spectral domains by using observations simultaneously performed by two instruments.

Spectral domain (cm ⁻¹)	Number of selected microwindows
700 – 1300	16
1900 – 2200	8
2500 – 3100	10
3100 – 3500	17
4000 – 4300	12

Table 1

Analysis

The grating spectra are analyzed with a former version of the retrieval algorithm SFIT-1. This particular version uses a gaussian function as instrumental line shape, instead of the sinc function used for the FTIR spectra. The program computes a synthetic spectrum and adjusts it to the observed spectrum in a non linear least squares iterative procedure, by applying multiplicative factors to the *a priori* vertical mixing ratios of the different gases absorbing in the analyzed micro-windows (example in Fig. 2). With this version of SFIT, only total H₂O columns may be retrieved. The FTIR spectra are analyzed with the retrieval software SFIT-2 v3.91, which uses the optimal estimation method [Rogers 2000] to fit the observed and the computed spectra. With this version, the shape of the vertical profile of the different gases may be adjusted, thus providing some information about their distribution versus altitude.

Information content analyses show that the sensitivity of FTIR spectrometers for H₂O retrievals at Jungfraujoch is good throughout most of the troposphere (Figure 3 and 4). At least 3 independent atmospheric layers (DOFS, Degree Of Freedom for Signal) can be deduced from the spectra: typically from 3.58 to 4.3 km, from 4.3 to 6.5 km and from 6.5 to 11 km. In the most favorable cases, some information on water vapor in the upper troposphere/lower stratosphere region can also be retrieved.

Both SFIT-1 and SFIT-2 programs use a Voigt function as line profile. The HITRAN 2004 spectroscopic database, including the latest updates for H₂O (August 2006, www.hitran.com), has been used for the analysis. Bad fittings are often achieved when fitting the different chosen microwindows, because of the poor quality of the H₂O spectroscopic parameters in these domains. Line parameters (position, strength and half-width) have been adapted in some cases to improve the quality of the fittings. Some examples of the spectroscopic problems encountered in this study are presented below.

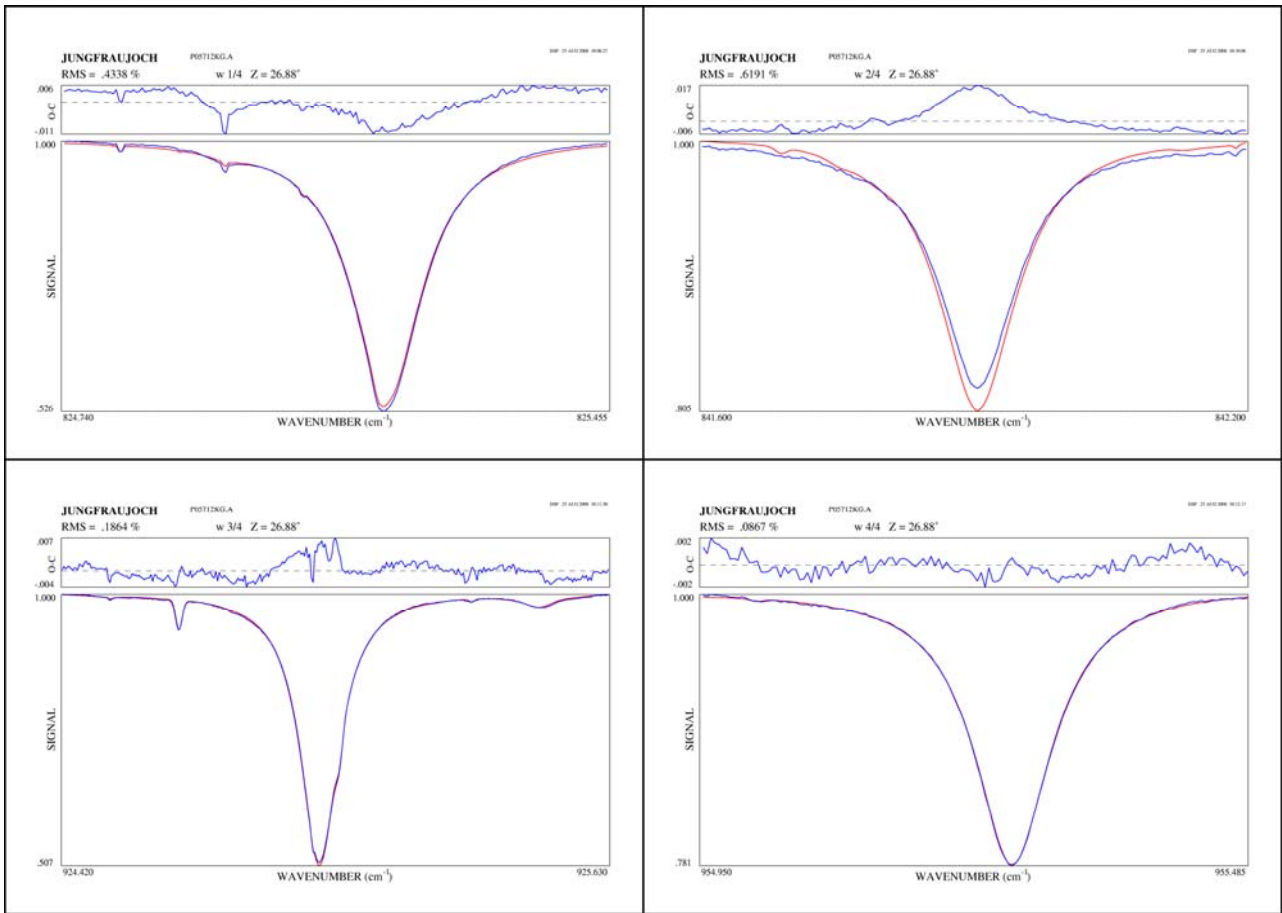


Figure 1. Simultaneous fit of 4 H₂O micro-windows (DOFS = 2.89) from a FTIR absorption spectrum recorded at the Jungfraujoch on July 12, 2005, at a solar zenith angle of 26.88°. Blue curves correspond to observed spectrum, red curves to computed spectrum. The upper frames of each micro-window show the residuals (observed-computed spectrum). Notice the vertical scales of the residuals and of the spectrum, different for each micro-window. Obviously, the spectroscopic parameters of the 841.9028 cm⁻¹ line (top right panel) need to be revised!

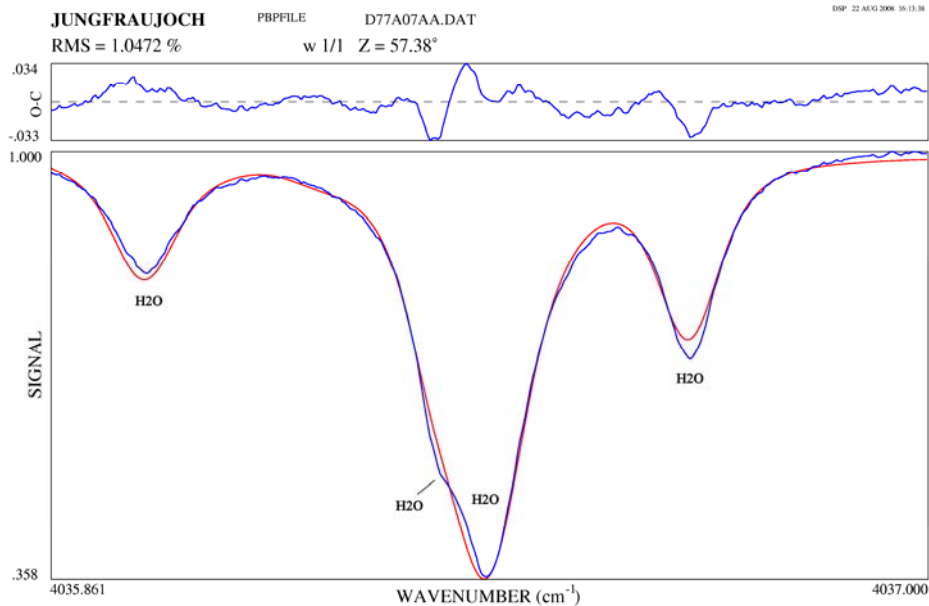
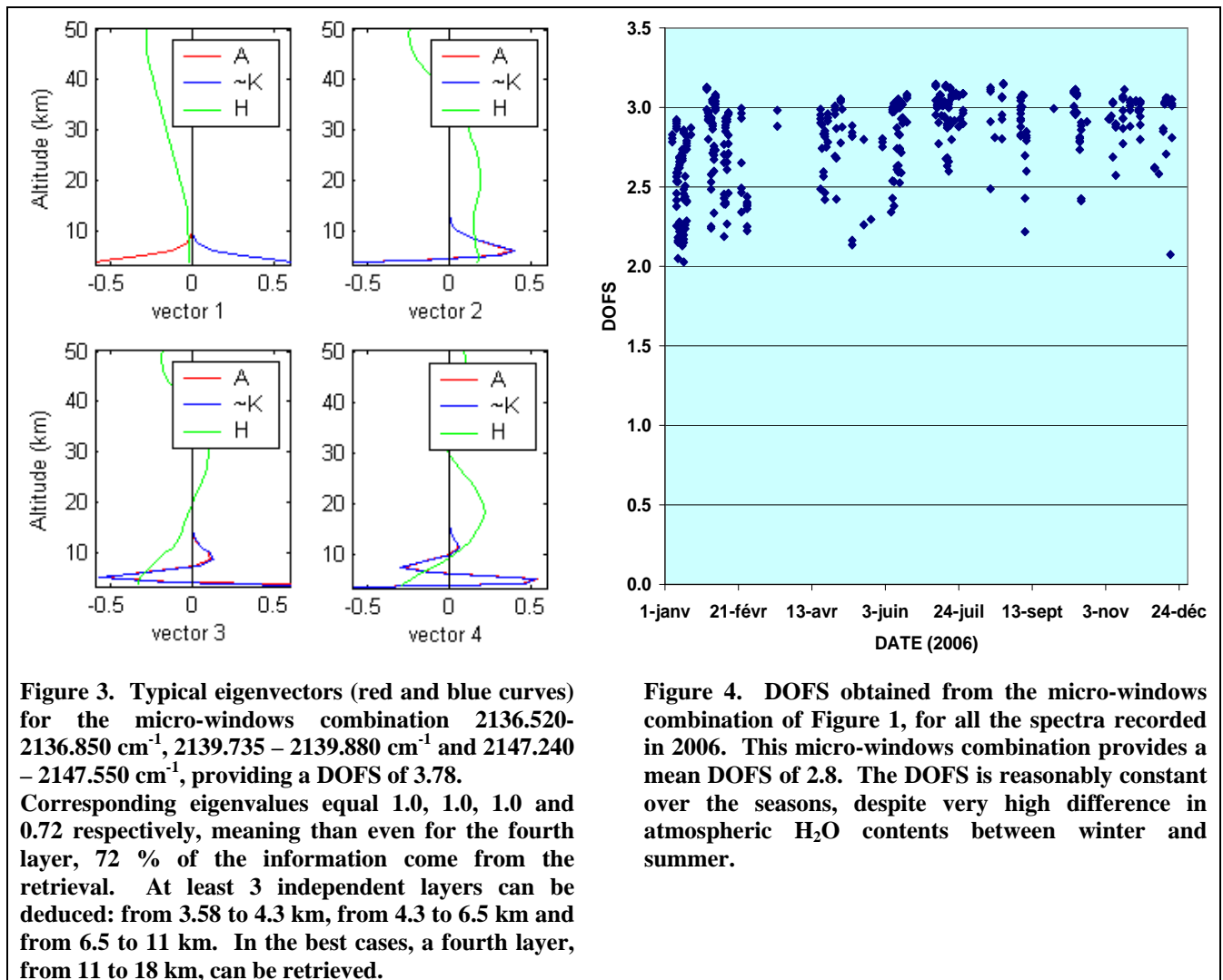


Figure 2. Fit of H₂O lines in the HF window, recorded with the Jungfraujoch grating spectrometer on October 7, 1977. The fit quality improved considerably when position of the first line was changed from 4035.97653 cm⁻¹ to 4035.98276 cm⁻¹ and position and strength of the last line were changed from 4036.69703 cm⁻¹ and 9.521 10⁻²⁴ cm⁻¹/(molec.×cm⁻²) to respectively 4036.69078 cm⁻¹ and 6.990 10⁻²⁴ cm⁻¹/(molec.×cm⁻²). [preliminary results !!]



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