Updating the Jungfraujoch FTIR databases : current status

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Introduction

Since the middle of the 20th century, the University of Liège has been active at the International Scientific Station of the Jungfraujoch (ISSJ) in the Swiss Alps (46.5°N, 8.0°E, 3580m asl) to study the chemical composition of both the solar photosphere and the Earth's atmosphere. Since 1989, the ISSJ is an accepted site of the northern midlattitude primary Alpine station of the NDSC (Network for the Detection of Stratospheric Change). This acceptance resulted from earlier monitoring activities by Ulg of a large number of atmospheric constituents [1]. Within the NDSC frame, a special attention and many efforts have been devoted to the monitoring of the most important atmospheric constituents involved both in the erosion of stratospheric ozone and in the greenhouse capacity of the troposphere, after it became clear that human activities have a direct impact on these two processes. Using two high resolution Fourier transform infrared (FTIR) spectrometers, over 25000 wide-band solar spectra encompassing nearly 1700 days since the mid 1980s have been recorded and analyzed to study, quasi simultaneously and repeatedly, local, seasonal and secular variations of some 20 gaseous telluric species. So far, most results have been reported, demonstrating the power of infrared spectrometric solar observations to characterize the chemical composition of the atmosphere. These datas, archived in terms of total vertical column abundances (e.g., at the NDSC-Data Host Facility; http://www.ndsc.ws), are expressed in number molecules per cm^2 above the site using best-know input parameters (i.e. spectroscopic-, instrumental-, environmental-) in the retrieval procedure. Meanwhile, more sophisticated algorithms, based on the "Rodgers" optimal estimation method, have been developed, allowing to derive partial tropospheric- and stratospheric columns for various species, including HCl, $ClONO_2$, O_3, HF, CO, N_2O, CH_4, HCN, OCS. This contribution reports related results for HCl and $ClONO_2$.

Procedure

All data bases displayed here have been derived from high-quality infrared solar observations performed regulary at the ISSJ, using high-resolution, broad-band FTIRs. The retrieved data consist of vertical column abundances (expressed in number molecules per cm^2) derived from the solar spectra using the SFIT2 version 3.7-C algorithm, developed jointly at the NASA-Langley Research Center (LaRC, VA, USA) and at the National Institute of Water and Atmospheric Research (NIWA, New-Zealand). SFIT2 is a radiative transfer- and profile retrieval algorithm for use with solar absorption spectra recorded with FTIRs. The retrieval method (Optimal Estimation Method ; see [2]) consists in fitting a calculated spectrum to the observed one by discrete adjustment of the *a priori* concentrations in the 29 layers encompassing the atmosphere above the Jungfraujoch, out to 100 km altitude. The adjustment sequence ends when the residuals between retrieved and observed spectra are minimum. One or two gas profiles as well as total column amounts of interfering species, instrumental back-ground parameters, wavelenght calibration factors and an instrumental lineshape parameter may be retrieved. One or more spectral windows can be fitted simultaneously. Inputs for the fittings dealt with here include : (i) FTIR spectra recorded at the ISSJ; (ii) most realistic Volume Mixing Ratio profiles for both the target and the interfering gases; (iii) spectroscopic parameters taken from the HITRANtype line parameter compilations and "pseudo-lines" calculated by G.C. Toon (private communication, 2001) from spectra produced by Birk and Wagner [3]; (iv) daily p,T-model atmospheres obtained from the National Centers for Environmental Prediction (NOAA, Washington, D.C., USA); (v) instrumental parameters characterizing the individual observations; (vi) *a priori* Volume Mixing Ratio profiles used for HCl have been derived from HALOE/UARS observations between 1992 and 2000; (vii) the Instrumental Line Shape is derived from an O_3 absorption line at 2775.83 cm^{-1} (near the HCl windows), assuming O_3 profiles resulting from the combination of ozone soundings measurements at Payerne and microwave measurments at Bern (both located in the vicinity of ISSJ).Three spectral intervals belonging to the 1-0 band of HCl were used and fitted simultaneously : the R1 line at 2925.90 cm^{-1}, the P5 line at 2775.76 cm^{-1} and the P7 line at 2727.78 cm^{-1}. The corresponding averaging kernels are shown in Figure 1.

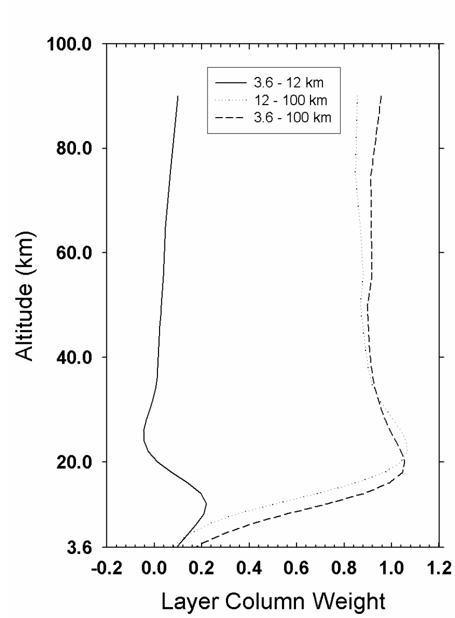

Figure 1 - HCl averaging kernels for atmospheric layers defined in the legend. Results are shown for a retrevial with three spectral lines (R1, P5 and P7). Sensitivity for the stratospheric- and total columns are very good, while the response from the tropospheric column is less satisfactory, considering the adopted a priori profile, in particular its lower tropospheric characteristic.

Analysis

Figure 2 presents an exemple of an HCl spectrum and profile measured at the ISSJ. Notice that all three spectral microwindows are fit reasonably well, even if small systematic features centered on each lines do dominate the residuals. Vertical profile retrievals are also encouraging.

Figure 3 displays HCl and $ClONO_2$ partial columns (tropospheric- and stratospheric contributions) derived from a subset (1990 to mid-2002) of the observationnal time bases available at ISSJ. In the case of $ClONO_2$, only the stratospheric column is reported as the tropospheric contribution is insignificant due to the choice of the initial profile. The red curves reproduce the adjustment (Non parametric Least-Square, NPLS) over monthly mean columns (red dots) deduced from observations made each year between June and November, which corresponds to the quietest period in terms of atmospheric dynamics. The dashed black curves take into account all the months (red and blue dots) of each year. Error bars represent the partial monthly mean column uncertainties, assuming precisions of 5% and 20 %, respectively for HCl and $ClONO_2$ individual retrievals.

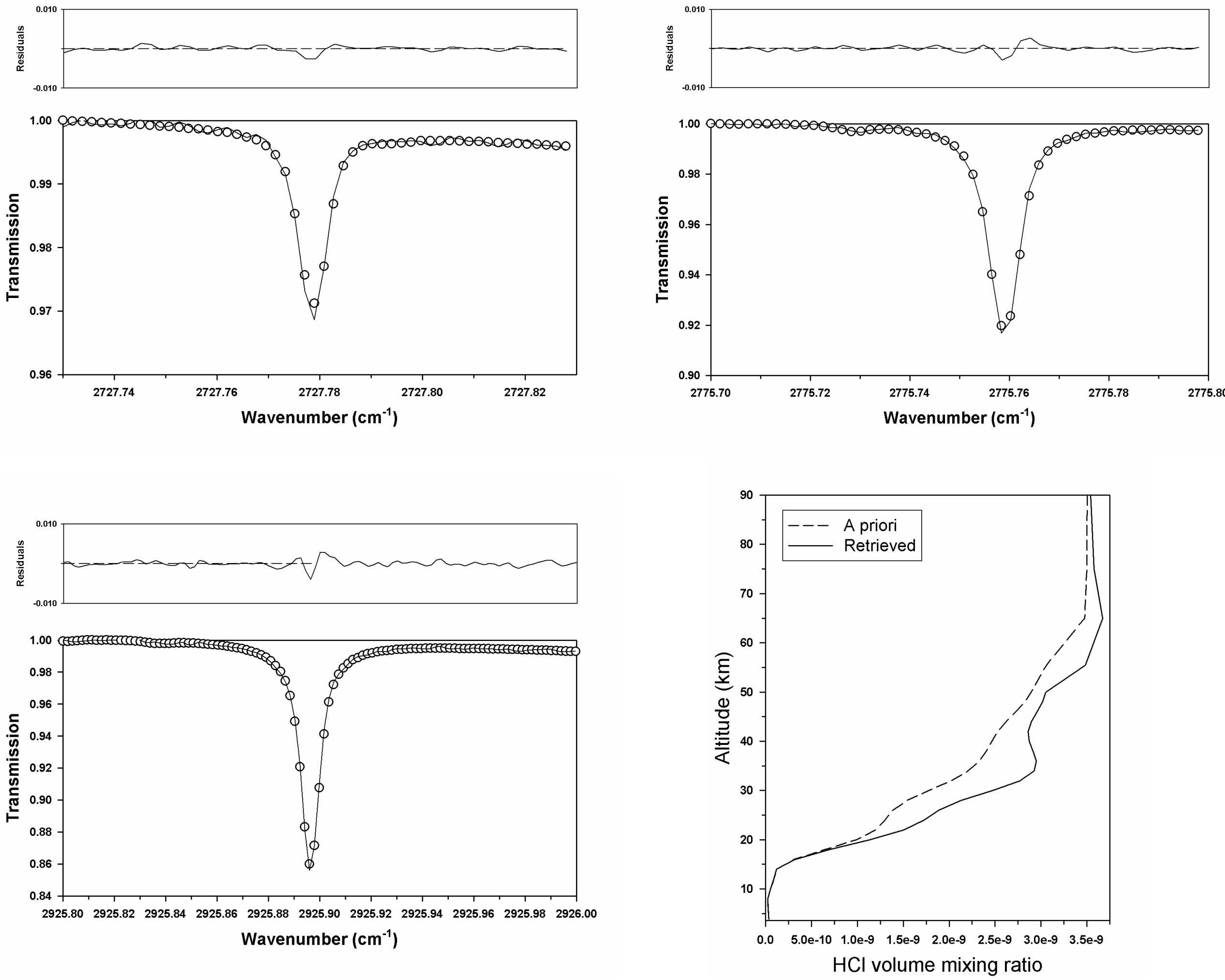

Figure 2 – An HCl retrieval from a 2.85 mK resolved spectra recorded the 16 July 2002 at the ISSJ, Switzerland at a zenital angle of 27.81°. The signal/noise ratio is about 2000. The three microwindows presented here were fitted simultaneously to derive the HCl vertical profile shown in the lower right panel. Small panels above each microwindows plot the difference between calculated and observed spectra.

The time series of Cl_y, defined as the sum of stratospheric HCl and $ClONO_2$ contributions, show a rapid increase till the mid 1990s, with a subsequent stabilization. First signs of a Cl_y decrease appear at the end of the 1990s and confirm the effectiveness of the Montreal Protocol. This "decrease", at least for the June to November months (red Cl_y curve), continues till the end of the time series plotted here.
Notice the increasing divergence between the black and red fittings to the stratospheric HCl columns after about the mid 1990s (and its corresponding impact on Cl_y) : a possible cause of that behaviour, which should be investigated with the help of model calculations, may be changes in atmospheric transport/circulation of winter/spring, versus summer/fall.

Current status

HCl, $ClONO_2$: see Figure 3.

O_3 : for more informations, please refer to B. Barret, M. De Mazière and P. Demoulin, Retrieval and characterization of ozone profiles from solar infrared spectra at the Jungfraujoch, accepted for publication in *J. Geophys. Res.*, 2002.

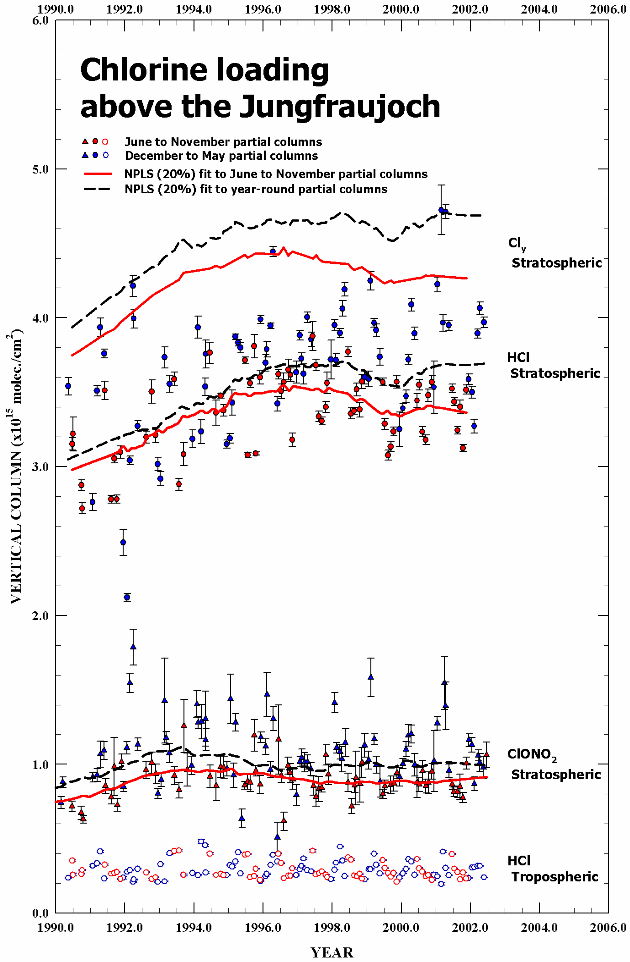

Figure 3 – Evolution versus calendar year of HCl and $ClONO_2$ monthly mean partial columns (see also text). The Cl_y time series correspond to the sum of stratospheric HCl and $ClONO_2$ contributions (the corresponding dots haven't been plotted for clarity).

HF : procedure demonstrated. For more detailed informations, please refer to B. Barret, M. De Mazière, P. Demoulin, E. Mahieu, F. Mélen, B.J. Connor and N. Jones, Investigation of height-resolved information in ground-based, high-resolution Fourier infrared solar spectra above the Jungfraujoch, in *Proceedings of the "Quadriennial Ozone Symposium"*, Sapporo, Japan, 3-8 July 2000, pp. 301-302, 2000.

CO : routine application. For more detailed informations, please refer to C.P. Rinsland, E. Mahieu, R. Zander, P. Demoulin, J. Forrer and B. Buchmann, Free tropospheric CO, C_2H_6 and HCN above central Europe : recent measurements from the Jungfraujoch station including the detection of elevated columns during 1998, *J. Geophys. Res., 105*, 24235-24249, 2000.

N_2O : under investigation.

CH_4 : under investigation.

HCN : routine application. For more detailed informations, please refer to Rinsland et al., Free tropospheric CO, C_2H_6 and HCN above central Europe : recent measurements from the Jungfraujoch station including the detection of elevated columns during 1998, *J. Geophys. Res., 105*, 24235-24249, 2000.

OCS : see abstract Mahieu et al.

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[2] Rodgers, C.D., Retrieval of atmospheric temperature and composition from remote measurements of thermal radiation, *Reviews of Geophysics and Space Physics*, 14(4), 609-624, 1976.

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