The current budget of NO_y above the Jungfraujoch as derived from IR solar observations

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INTRODUCTION

This paper reports on an investigation of a series of compounds of the NO_y family, based on high resolution infrared solar observations made at the ISSJ (International Scientific Station of the Jungfraujoch), Switzerland (46.55°N, 7.99°E, 3580 m a.s.l.). These observations are part of a long-term monitoring effort undertaken by the Liège group since the mid-1970s, and integrated more recently as a contribution to the Network for the Detection of Stratospheric Change (NDSC).
Currently, vertical column abundances of over 20 molecules are retrieved from solar spectra recorded under clear sky conditions as regularly as possible, using two high resolution Fourier transform infrared (2 to 15 μm) spectrometers [1].
The columns are retrieved from the spectra by non-linear least squares spectral fitting, using the SFIT 1.09c algorithm; a discussion of the retrieval procedure can be found in [2].

NO_y BUDGET

NO_y, the total reactive nitrogen, is defined as the sum of the following species: $NO + NO_2 + NO_3 + 2(N_2O_5) + HNO_3 + HO_2NO_2 + ClONO_2 + BrONO_2$. For the purpose of deriving the NO_y column trend above the Jungfraujoch, monthly mean columns of those species that can easily be measured from the ground, i.e. NO, NO_2, HNO_3 and $ClONO_2$, have been calculated from the consistent database spanning from 1985 to present. This procedure of computing monthly means avoids giving excessive weight to months with high-density observations. These mean columns are shown as dots in frames A to D of Figure 1; they have been modeled with linear+sinusoidal functions (continuous curves). Trend values, referred to 1990.0, are also indicated with their 1 σ deviations.
The sum of these monthly mean columns, displayed in Figure 1, frame E, represents the most important part of the total NO_y as derived from the ISSJ data. The missing NO_3, N_2O_5, HO_2NO_2 and $BrONO_2$, not easily observable from the ground, represent less than 5 % of the total NO_y [3]
As for the individual species, the NO_y trend has been simulated with a linear+sinusoidal function (continuous curve); the resulting trend of (0.3 ± 0.3) % per year, although barely significant, is consistent with the trend of (0.35 ± 0.04) % per year, found for the NO_y gas source N_2O [4] (Figure 2, frame F). The important uncertainty in the NO_y trend is mainly due to the high variability of the HNO_3 columns, which is predominant during the November to April months (circulation and heterogeneous processes).
Notice that the constituents considered in the present investigation show seasonal variations summarized in the following table:

	NO	NO_2	HNO_3	$ClONO_2$
Peak-to-peak variation (%)	34	74	28	37
Occurrence of maximum	June-July	June-July	Feb.-March	March
Occurrence of minimum	January	January	August	September

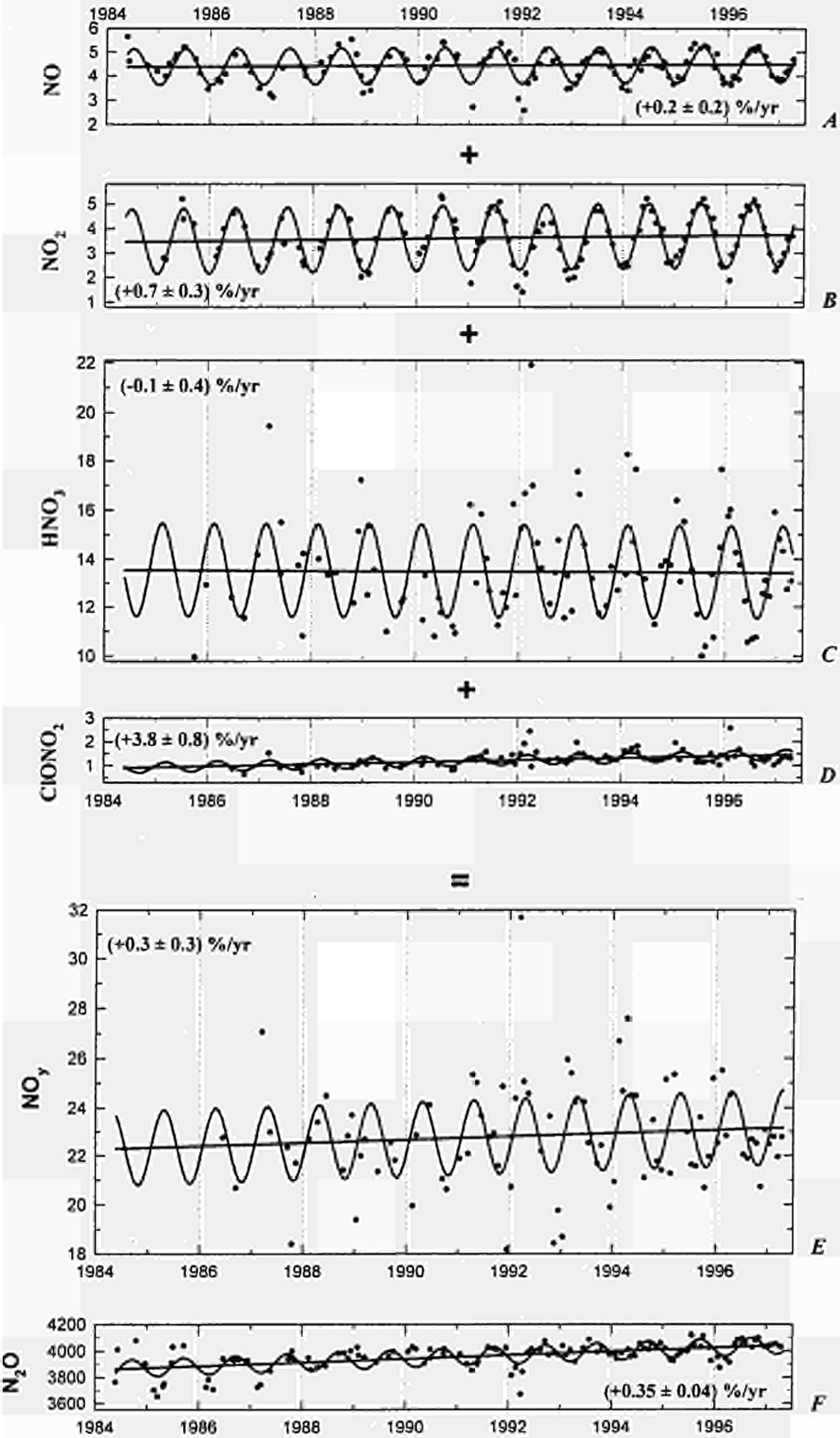

Figure 1: column abundances above ISSJ, expressed in 10^{15} molec/cm^2

HNO_3 AND HF COLUMNS CORRELATION.

In Figure 2, we have correlated the HNO_3 columns with those of HF (with the HF trend removed and reported to 1990.0) obtained on same days. Most of the points cluster about an oblique line: the points on the left correspond to typical summer conditions, whereas those to the right, generally correspond to air masses originating from the higher latitudes, enriched in both HF and HNO_3, as a consequence of the latters' latitudinal distribution.

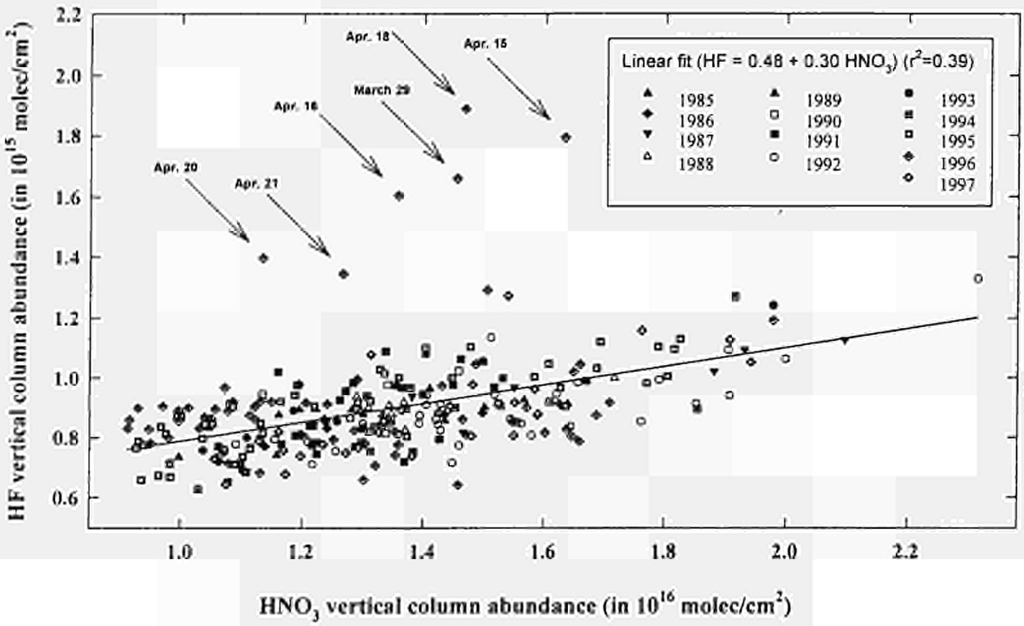

Figure 2: HNO_3 and HF columns correlation

An interesting episode in March - April 1996 is identified in Fig.2 by arrows and dates; during that period, extremely high values of HF (the highest ever recorded above ISSJ) have been observed.

At the end of March, the polar vortex was centered over Scandinavia and was still well defined. Switzerland was near the edge of the vortex (on the 31st March, potential vorticity was 40×10^{-6} $Km^2kg^{-1}s^{-1}$ at the 475 K level). Air masses above the Jungfraujoch originated from the polar regions and were thus enriched in HF; Figure 3 shows an example of back trajectories for the 29th March 1996, ending at the closest rawinsonde station (Payerne, 85 km North-West of the Jungfraujoch). The vortex then slowly dissolved: by mid-April, only two fragments remained, one centered over East Siberia, the other over Central Europe. Potential vorticity reached again high values (38×10^{-6} $Km^2kg^{-1}s^{-1}$ at the 475 K level on the 18th April) above the Jungfraujoch.

During that time period, no specially high values of HNO_3 were observed, as we could have expected from the usual relationship between HF and HNO_3 shown in Figure 2: based on the latter, we should have measure HNO_3 values around 5×10^{16} molec./cm^2 instead of the values around 1.5×10^{16} molec./cm^2 actually observed. This suggests that denitrification occurred in these air parcels.

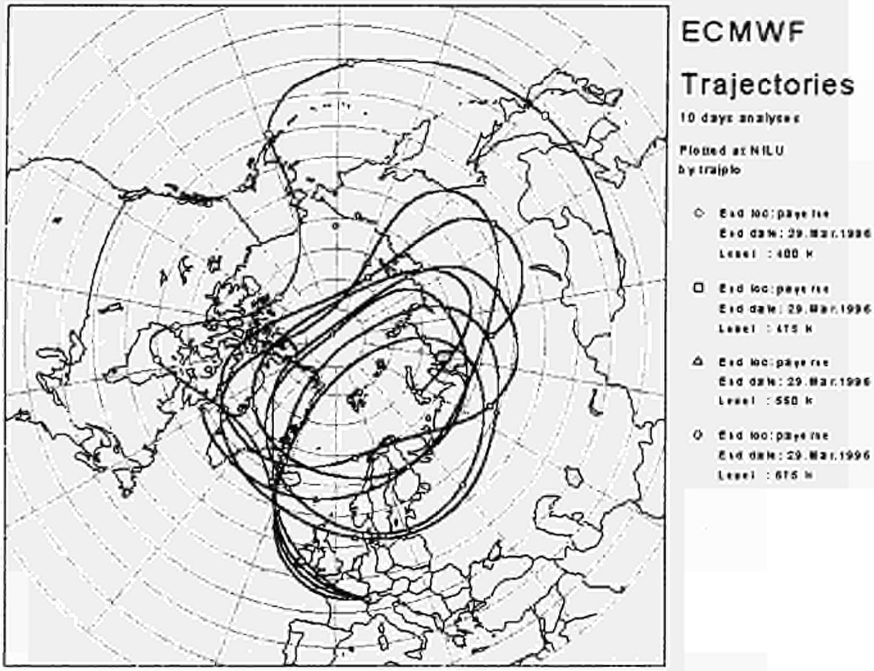

Figure 3: back trajectories ending at Payerne, on the 29th March, 1996

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