THE CURRENT BUDGET OF NO_y ABOVE THE JUNGFRAUJOCH AS DERIVED FROM IR SOLAR OBSERVATIONS.

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RESEARCH CONTEXT

Site:

International Scientific Station of the Jungfraujoch (ISSJ)
(primary NDSC station)
Location: Swiss Alps (46.55°N, 7.99°E, 3580 m a.s.l.)

Intruments:

Two high resolution (0.0025 and 0.0010 cm^{-1}) Fourier transform infrared spectrometers, using the Sun as source of radiation, operated by the University of Liège.
Spectral domain: 2 to 15 μm.

Database:

Primarily from 1985 to 1997, vertical columns of more than 20 molecules (O_3, HCl, HF, CFCs, NO, NO_2, HNO_3, $ClONO_2$, N_2O, COF_2, …).
Column retrieval: non-linear least squares spectral fitting, using the SFIT 1.09c algorithm.
An excerpt of the ISSJ database is given in Figure 1.

Sample vertical column abundances above ISSJ

1990 1991 1992 1993 1994 1995 1996 1997

J M M J S N J M M J S N J M M J S N J M M J S N J M M J S N J M M J S N J M M J S N J M

N_2O (E18) 4.2 4.0 3.8 3.6

O_3 (E18) 12 10 8 6

NO_2 (E15) 6.0 5.0 4.0 3.0 2.0

NO (E15) 6.0 5.0 4.0 3.0

HNO_3 (E16) 2.5 2.0 1.5 1.0

$ClONO_2$ (E15) 3.0 2.0 1.0

HCl (E15) 5.0 4.0 3.0 2.0

HF (E15) 2.0 1.6 1.2 0.8

J M M J S N J M M J S N J M M J S N J M M J S N J M M J S N J M M J S N J M M J S N J M

1990 1991 1992 1993 1994 1995 1996 1997

19 sept 1997

Figure 1: excerpt of the ISSJ database. The vertical columns retrieved from the infrared spectra recorded between 1990 and present are displayed for a number of NDSC-related key species.

Notice the increases of N_2O, $ClONO_2$, HCl and HF, as well as the important seasonal variations of O_3, NO_2, NO, HNO_3 and $ClONO_2$.

For the constituents considered in the present NO_y investigation, their seasonal variation can be summarized as follows:

	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	Sept.

NOy budget above ISSJ

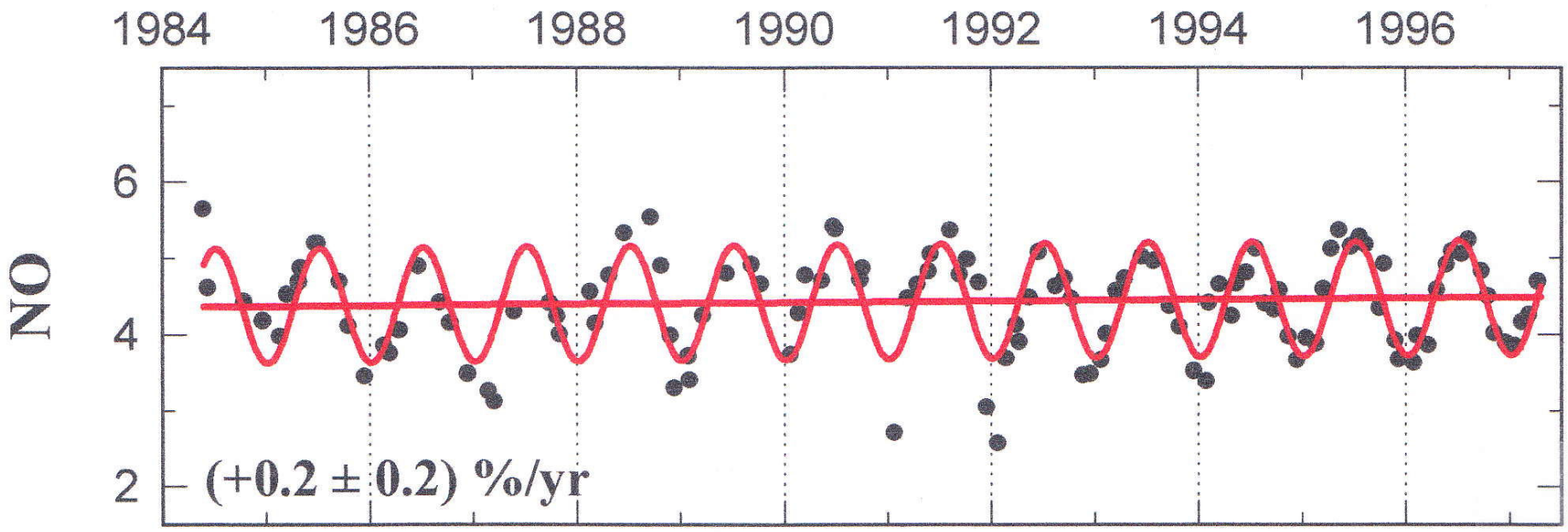

Figure 2a.

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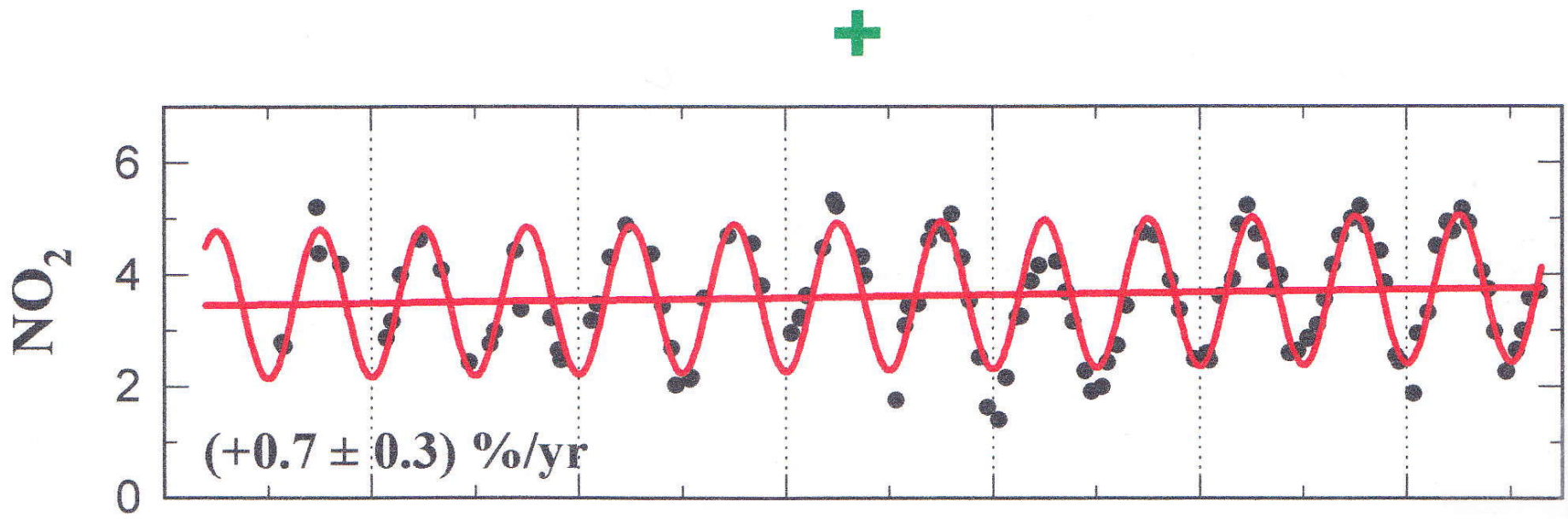

Figure 2b.

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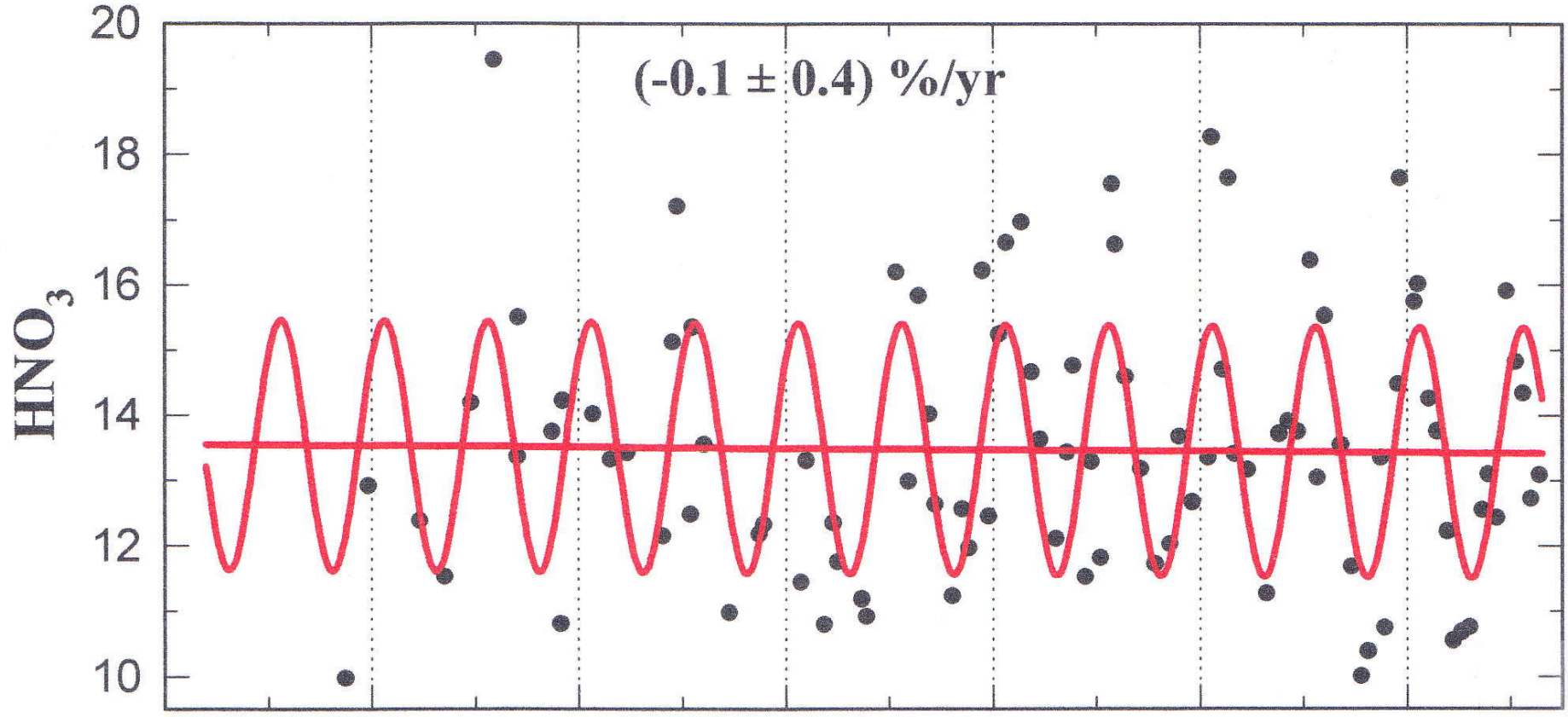

Figure 2c.

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Figure 2d.

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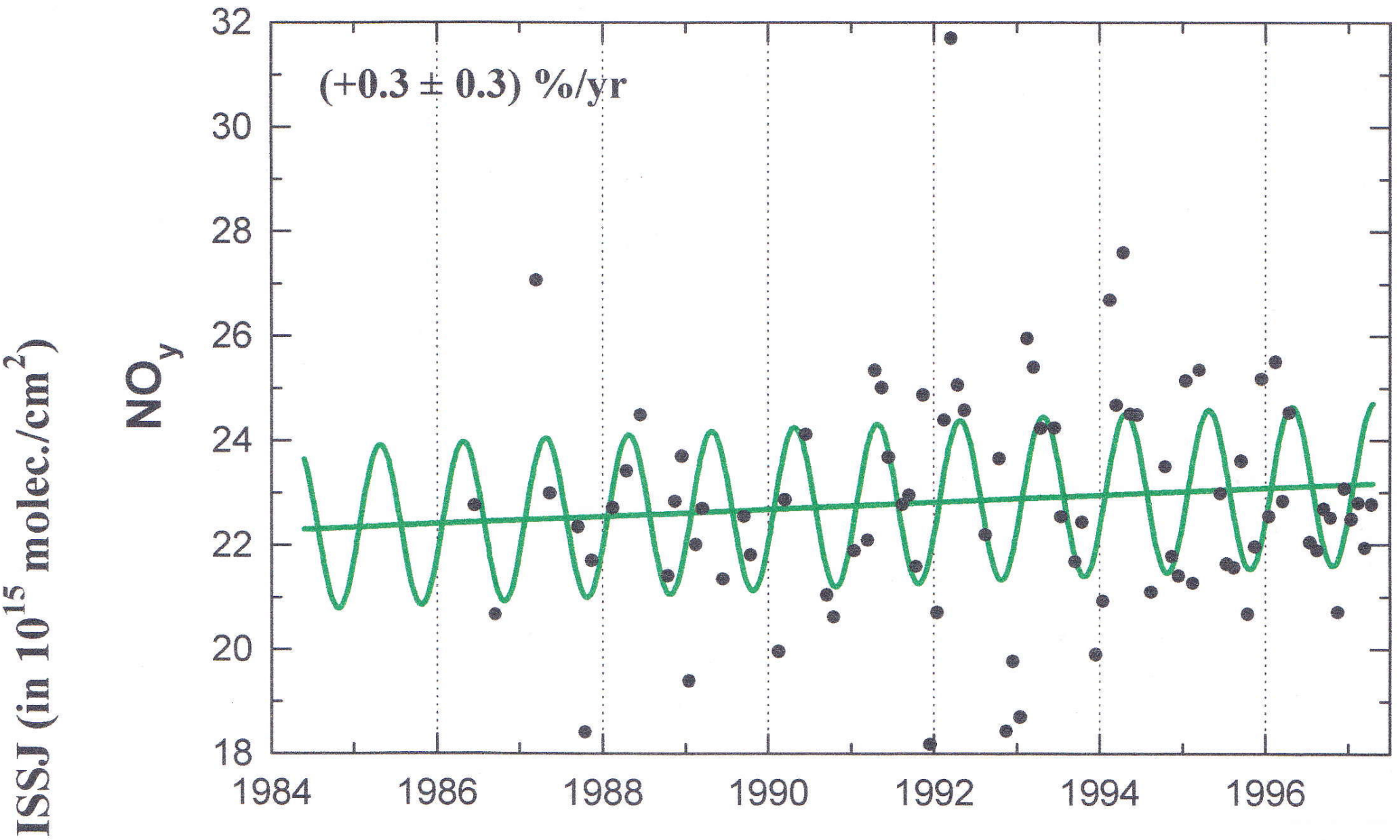

Figure 2e

N_2O trend:

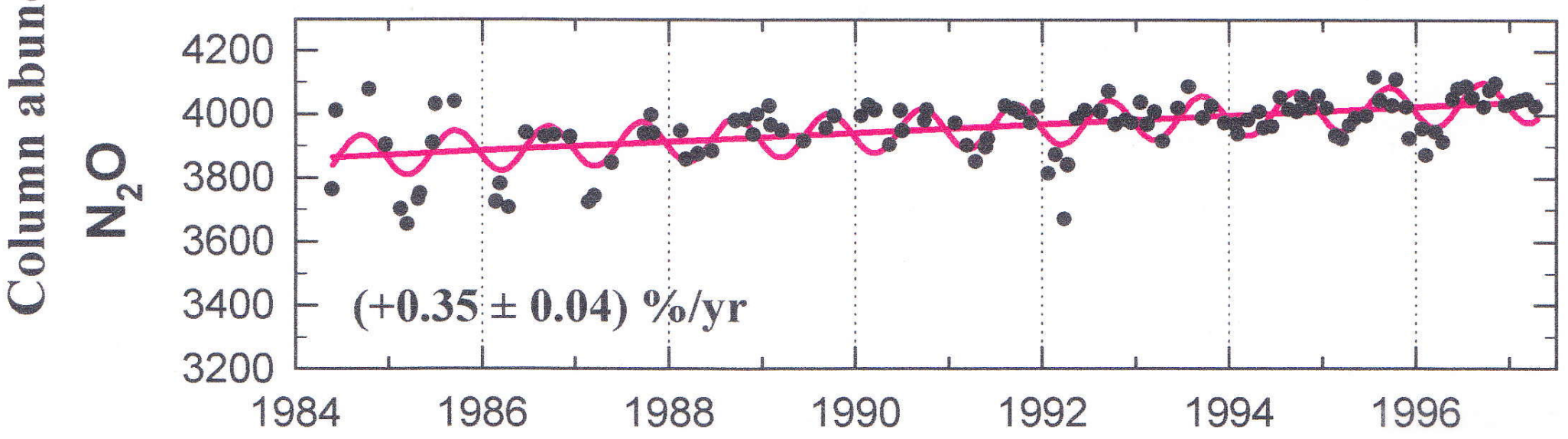

Figure 2f

Conclusion : consistency between NO_y and N_2O trends

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Figure 2: NO_y budget. Monthly mean vertical columns of NO (Fig. 2a), NO_2 (Fig. 2b), HNO_3 (Fig. 2c) and $ClONO_2$ (Fig. 2d) are shown as dots. The trends of these molecules have been modeled with (linear + sinusoidal) functions (red curves); trend values, reported to 1990.0, are also indicated.

The sum of these monthly mean columns, shown in Figure 2e represents the "total" NO_y as derived from the ISSJ data. The missing N_2O_5, not easily observable from the ground, represents about 5 to 10 % of the total NO_y.

As for the individual species, the NO_y trend has been simulated with a (linear + sinusoidal) function (green curve); the resulting trend of (0.3 ± 0.3) % per year, is consistent with the trend of (0.35 ± 0.04) % per year, obtained from the NO_y gas source, N_2O (Fig. 2f).

The important uncertainty in the NO_y trend is mainly due to the high variability of the HNO_3 columns (circulation).

HNO_3 & HF above ISSJ correlation

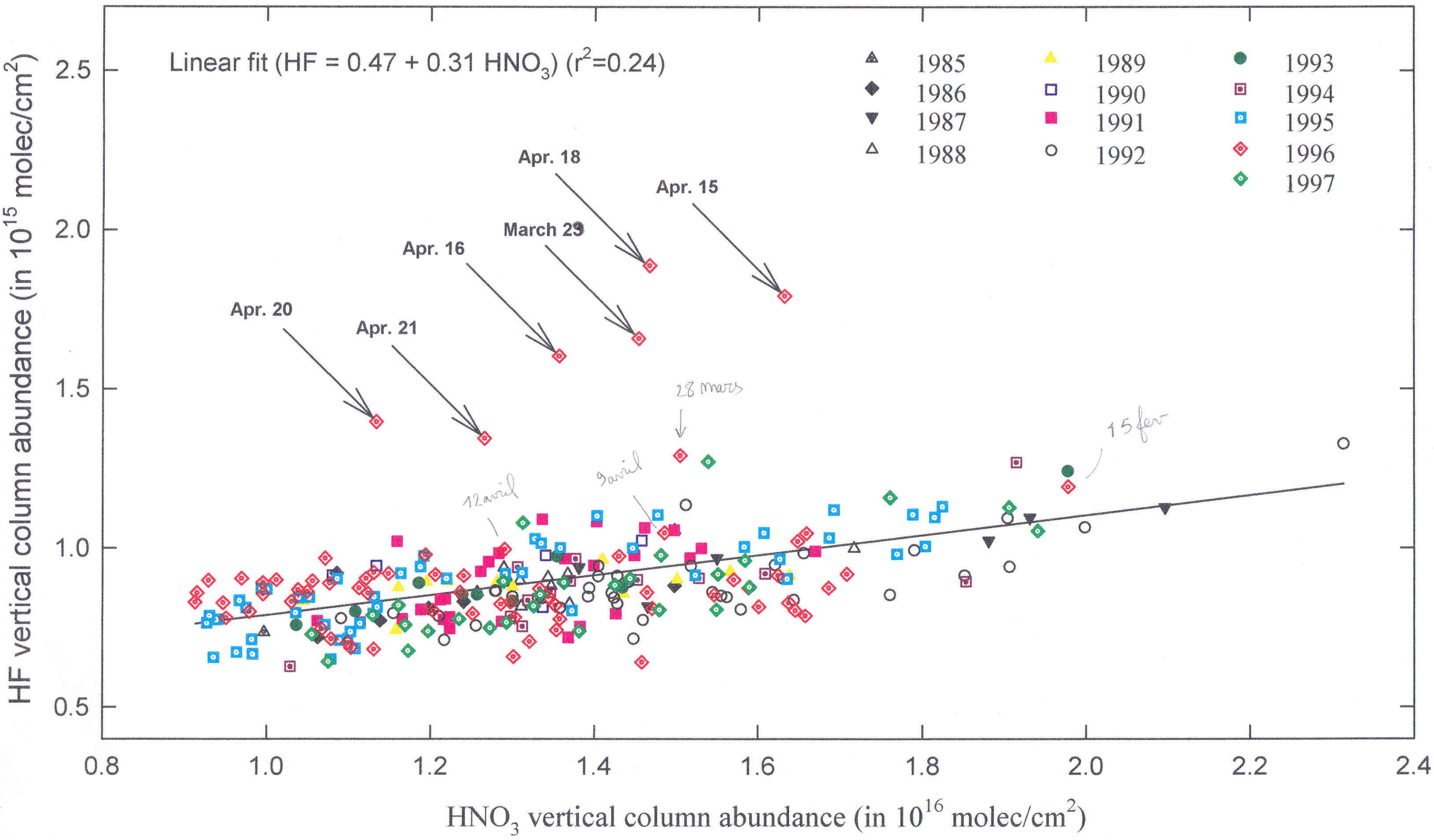

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Figure 3: HNO_3 and HF columns correlation.
HF columns (with the HF trend removed and reported to 1990.0) have been plotted here as a function of the HNO_3 columns obtained on the same day. Most of the points cluster about an oblique line: the points on the left correspond to typical summer conditions, whereas those to the right generally correspond to air masses originating from the higher latitudes.
Figures 4 and 5 give two examples of back trajectories ending at Payerne, Switzerland and illustrating well this latitudinal effect: on January 11th, 1997, high latitude air contained high quantities of HF ($1.40\ 10^{15}$ molecules/cm^2) and of HNO_3 ($1.94\ 10^{16}$ molecules/cm^2), whereas on January 24th, 1997, sub-tropical air brought lower quantities of HF ($1.03\ 10^{15}$ molecules/cm^2) and of HNO_3 ($1.17\ 10^{16}$ molecules/cm^2) above the Jungfraujoch.

An interesting episode in March - April 1996 is indicated by arrows and dates: air masses above the Jungfraujoch originated from the polar regions during that period (see example of trajectory in Figure 6) and extremely high values of HF (the highest ever recorded above ISSJ) have been observed, together with relatively low values of HNO_3 (denitrification).
Figure 7 shows the evolution of some key species during that episode, as well as the potential vorticity at 475 K and the temperature at 25 km.

ECMWF Trajectories

10 days analyses

Plotted at NILU
by trajplo

○ End loc: payerne
End date: 11.Jan.1997
Level : 475 K

Figure 4.

ECMWF

Trajectories

10 days analyses

Plotted at NILU
by trajplo

- End loc: payerne
  End date: 24.Jan.1997
  Level : 475 K

Figure 5.

ECMWF

Trajectories

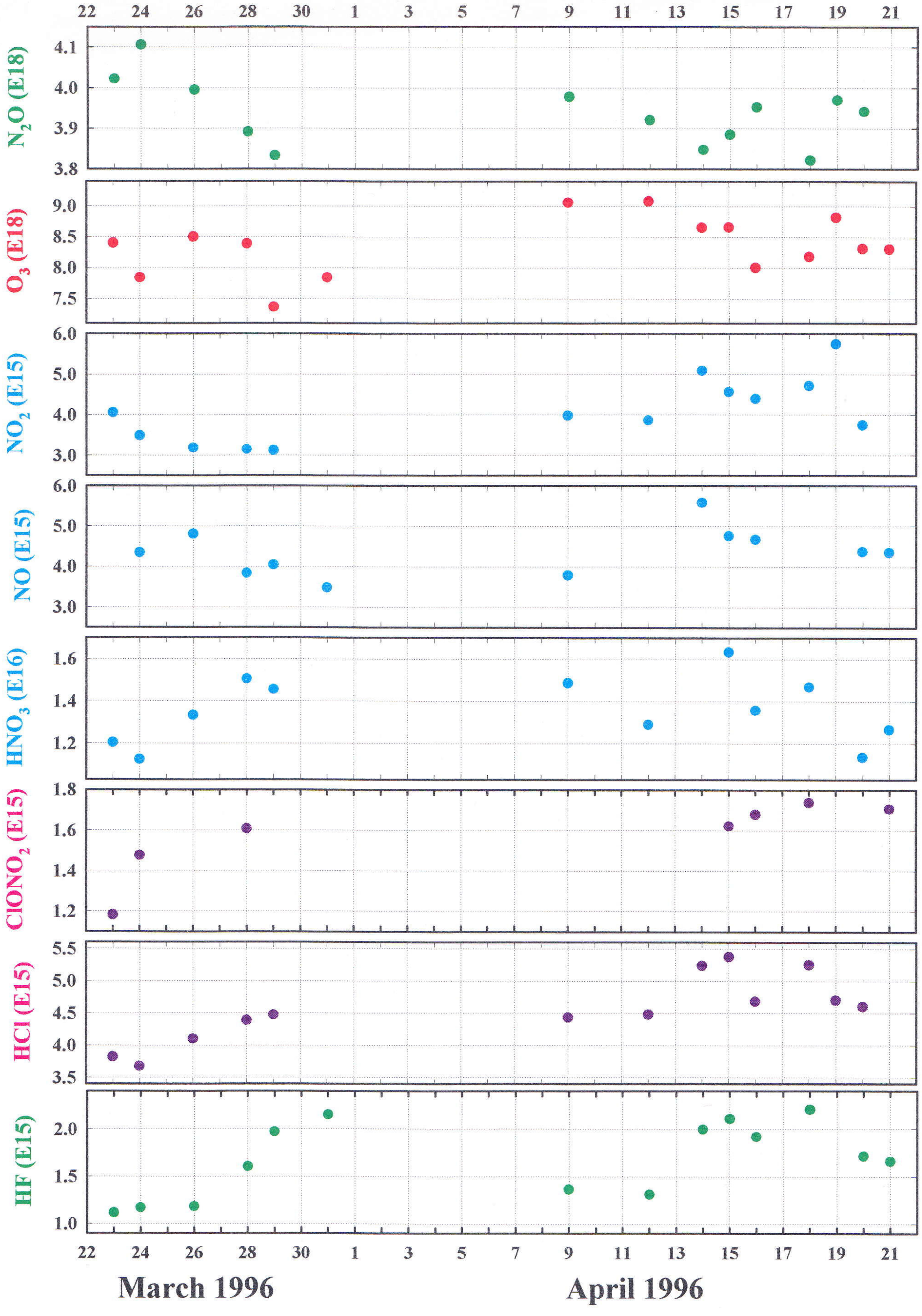
10 days analyses

Plotted at NILU
by trajplo

○ End loc: payerne
End date: 29.Mar.1996
Level : 475 K

Figure 6.

Vertical column abundances above ISSJ : March - April 1996



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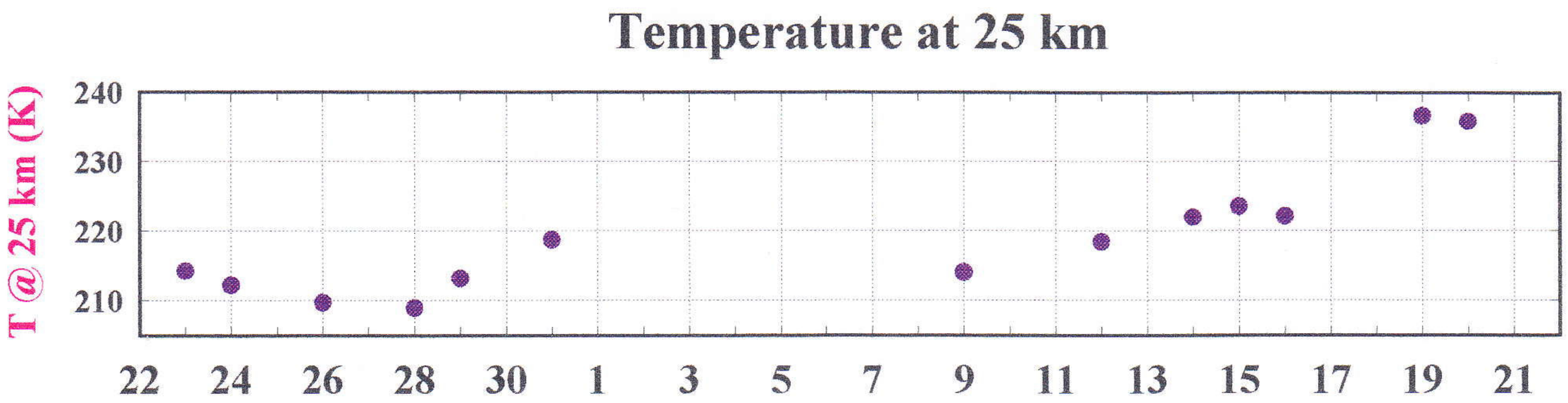

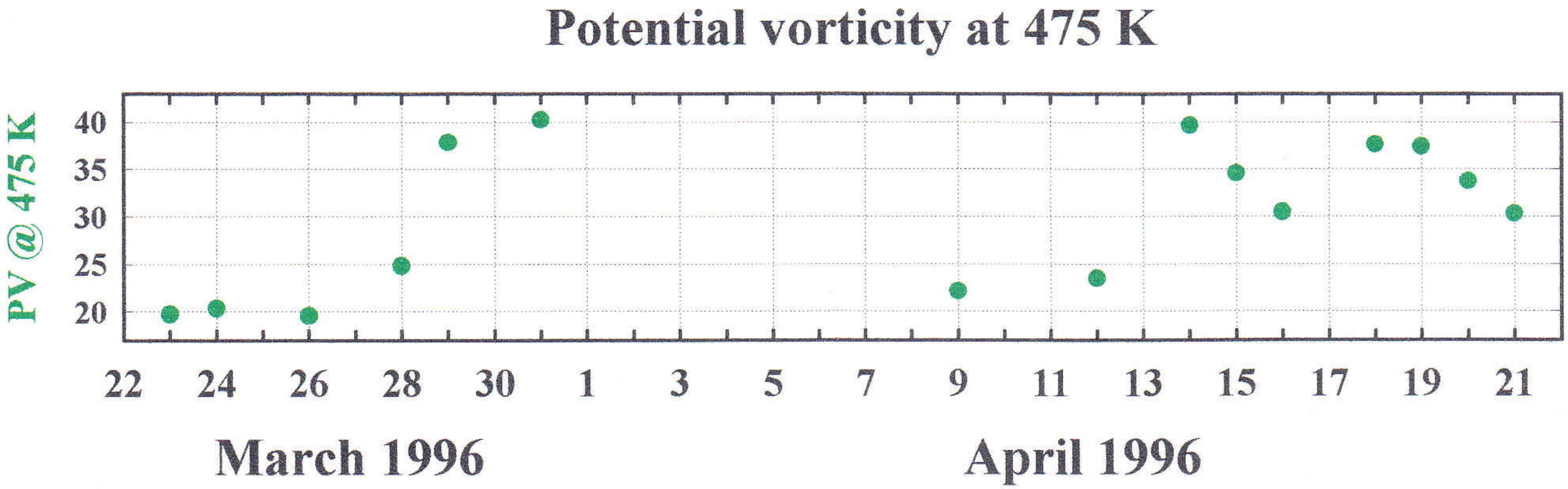

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Figure 7.