TREND ANALYSIS OF STRATOSPHERIC NO_2 ABOVE JUNGFRAUJOCH (46.5°E, 8°E) AND HARESTUA (60°N, 11°E) USING LONG-TERM GROUND-BASED UV-VISIBLE, FTIR, AND SATELLITE OBSERVATIONS

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I.INTRODUCTION

Nitrogen dioxide (NO_2) plays an important role in controlling ozone abundances in the stratosphere, either directly through the NO_x ($NO+NO_2$) catalytic cycle, or indirectly by converting active chlorine, bromine, and hydrogen into their reservoir forms, reducing their availability for ozone-destroying catalytic cycles.

Ground-based zenith-sky UV-visible measurements of the stratospheric NO_2 column have been conducted at the NDACC (Network for the Detection of Atmospheric Composition Change) stations of Jungfraujoch (46.5°E, 8°E) and Harestua (60°N, 11°E) since 1990 and 1994, respectively. Also available at both stations are coincident satellite observations from the ERS-2/GOME, ENVISAT/SCIAMACHY (nadir and limb), and METOP/GOME-2 instruments, as well as in case of Jungfraujoch and since the mid-1980s, ground-based Fourier transform infrared (FTIR) solar absorption measurements.

Here we present the results of trend analyses of stratospheric NO_2 by applying a multiple linear regression model to the ground-based and satellite monthly mean NO_2 columns time series. The regression model includes forcing mechanisms for solar flux, quasi-biennial oscillation (QBO), and aerosol loading, in addition to a linear trend. The aerosols term is essential since both UV-visible and FTIR observations at Jungfraujoch started before the Mount Pinatubo eruption. The consistency between inferred NO_2 trend values at both stations using the different platforms are investigated. The observed NO_2 trends are compared to the increase of nitrous oxide (N_2O), usually considered as the main source of NO_x in the stratosphere.

II. GROUND-BASED DATA

UV-vis

- SAOZ instrument operated at Jungfraujoch by BIRA since 1990
- UV and visible spectrometers operated by BIRA at Harestua since 1994 (continuously since 1998)
- Zenith radiance spectra analyzed using the DOAS method:
  - Fitting window: 430-470 nm
  - Fitted species: NO_2, O_3, O_4, H_2O, Ring effect
  - NO_2 XS: Vandaele et al. (1998) at 220 K
  - Daily reference spectra
- NO_2 vertical columns retrieved by applying an OEM-based profiling technique to sunrise and sunset zenith-sky NO_2 slant columns (Hendrick et al., 2004)

FTIR

- 2 high resolution FTS instruments operated by the University of Liège (1 homemade and 1 Bruker IFS-120HR)
- Column retrieval (Hendrick et al., 2012):
  - SFIT-2 algorithm (v3.91): OEM + HITRAN database
  - 2 microwindows: 2914.6 to 2914.7 cm^{-1} and 2915 to 2915.11 cm^{-1}
  - P, T profiles: NCEP
  - Interfering species: H_2O, CH_4, H_2CO and O_3
  - A priori NO_2 profiles: SLIMCAT model, as for UV-vis

III. SATELLITE DATA

NADIR INSTRUMENTS

- GOME, SCIAMACHY, and GOME-2 stratospheric NO_2 columns (200km overpasses) from the KNMI/BIRA TEMIS NO_2 algorithm
- Based on a three-step approach:
  - NO_2 slant column retrieval using the DOAS method
  - Estimation of the stratospheric component of the NO_2 slant columns through data assimilation in the TM4 CTM
  - NO_2 VCDs obtained by dividing the assimilated stratospheric slant columns by a simple geometrical airmass factor
  - More details in Boersma et al. (2004) and Dirksen et al. (2011)

SCIAMACHY LIMB

- IUP-Bremen scientific product (v3.1):
  - Level 1 data: ESA version 6.03
  - Differential two-step inversion approach
  - Spectral range: 420-470 nm
  - Reference tangent height: ~ 43km
  - Additional information on pressure and temperature from ECMWF
  - More details at http://www.iup.physik.uni-bremen.de/~sciaproc/CDI/DOCU/PSD_NO2_v3_1.pdf

IV. TREND MODEL

- Linear least squares regression model (adapted from Bodeker et al., 1998):

$$\Omega(t) = A\,(N_A{=}2) + B\,(N_B{=}2) \times t + C\,(N_C{=}2) \times \mathrm{QBO}_{30hPa}(t) + D\,(N_D{=}2) \times \mathrm{QBO}_{50hPa}(t) + E\,(N_E{=}0) \times \mathrm{Solar}(t) + F\,(N_F{=}1) \times \mathrm{Aerosols}(t)$$

with A-F coefficients expanded as:

$$A = A_0 + \sum_{k=1}^{N_A}\left[A_{2k-1}\sin(2\pi kt) + A_{2k}\cos(2\pi kt)\right]$$

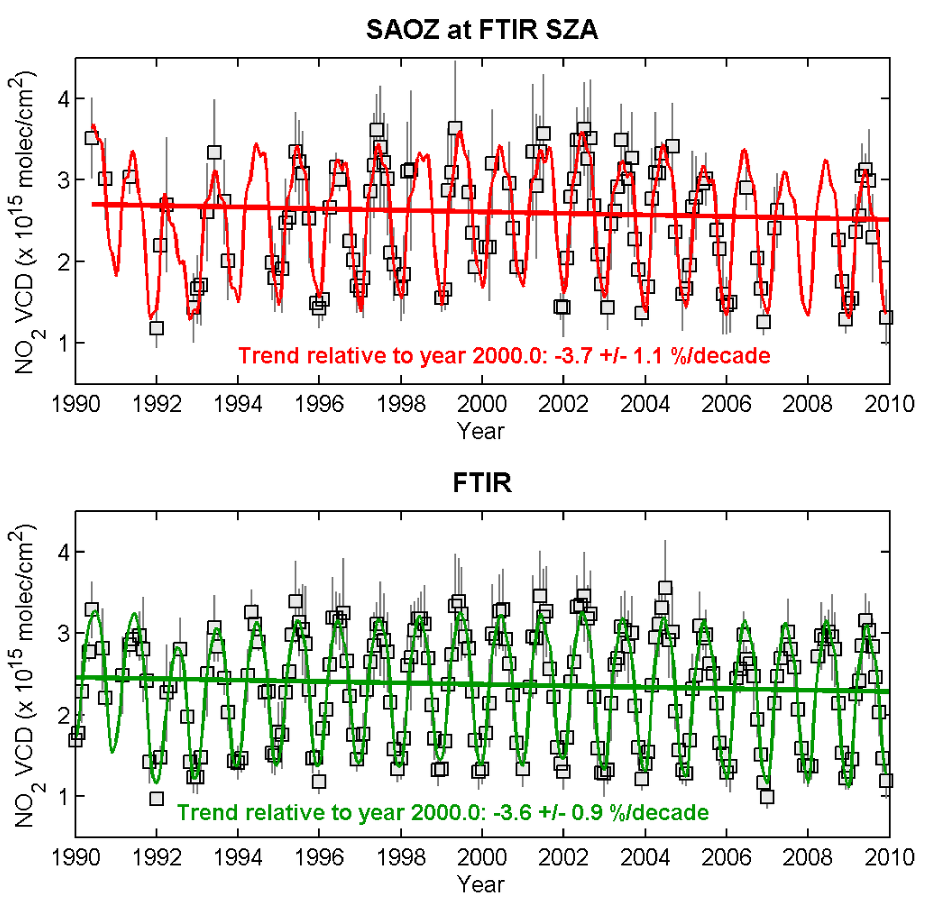
to fit seasonality

- QBO basis functions based on the Singapore monthly mean zonal winds (http://www.geo.fu-berlin.de/met/ag/strat/produkte/qbo/index.html)
- Solar cycle basis function: based on the radio-frequency F10.7 cm solar flux (ftp://ftp.ngdc.noaa.gov/STP/SOLAR_DATA/SOLAR_RADIO/FLUX)
- Aerosols basis function: based on the stratospheric aerosol optical depth (AOD) climatology of Vernier et al. (2011) created from SAGE II, CALIPSO, and ENVISAT/GOMOS observational data sets

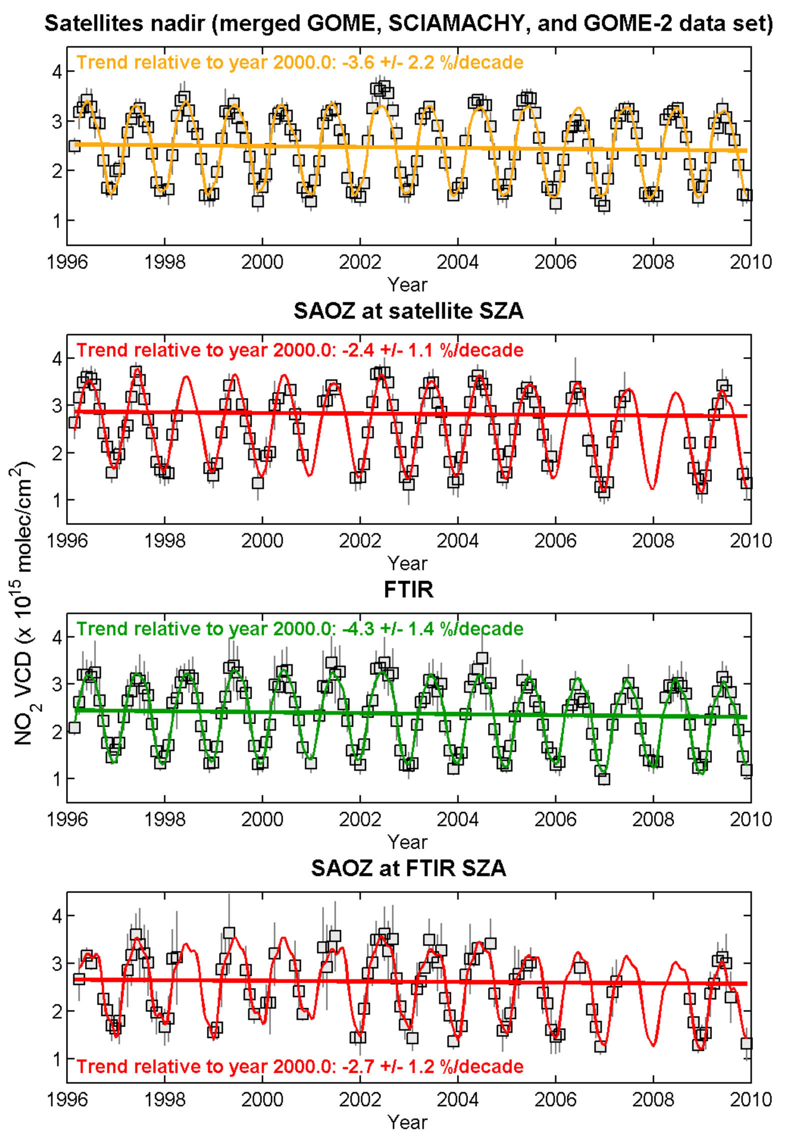
V. TREND ANALYSES

Jungfraujoch (46.5°N, 8°E)

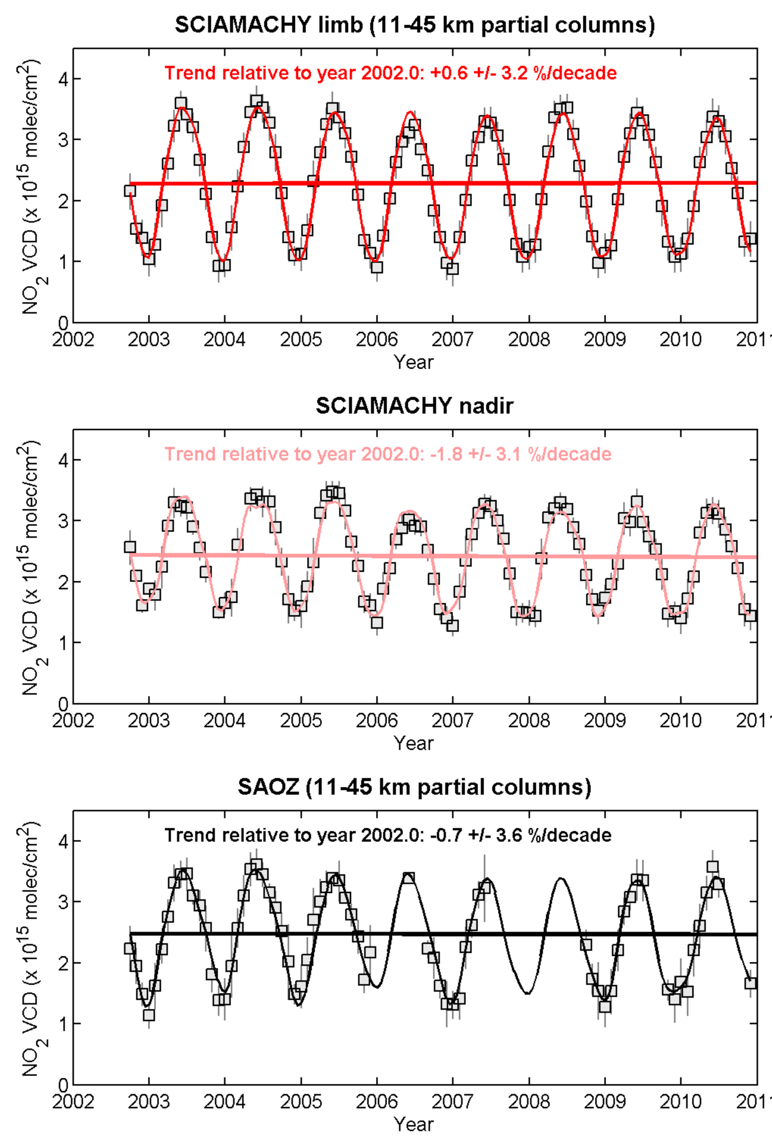
1990-2009



1996-2009

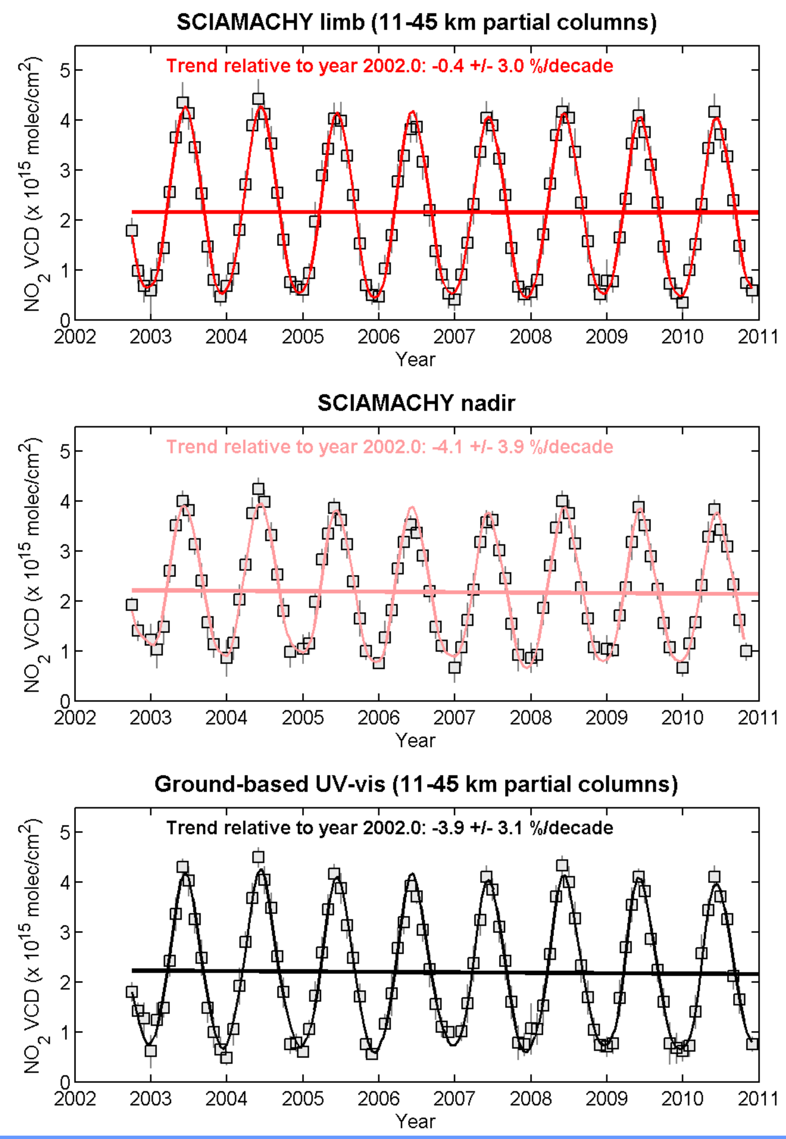


2002-2010



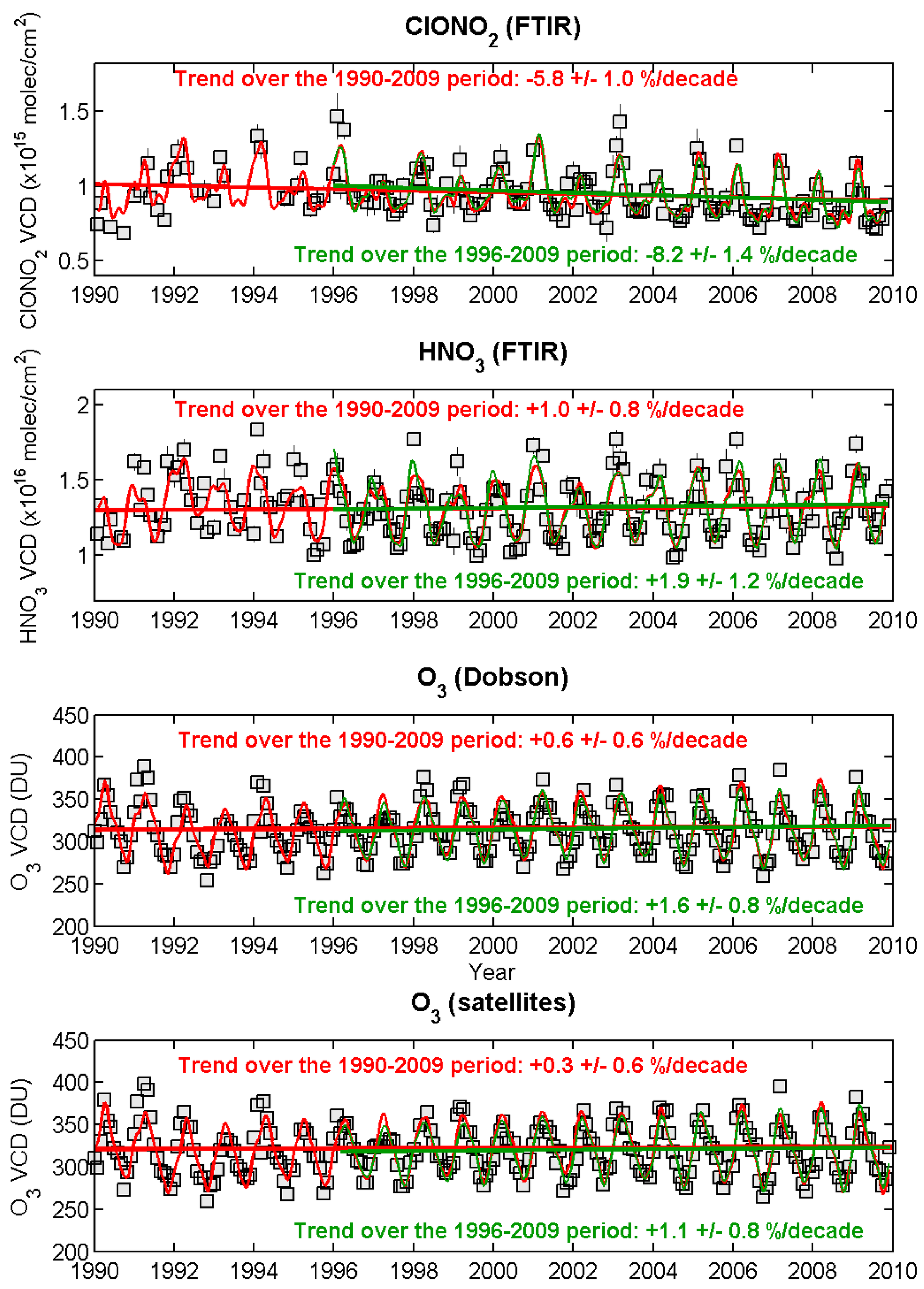
Harestua (60°N, 11°E)

2002-2010



VI. DISCUSSIONS

Jungfraujoch (46.5°N, 8°E)



	Jungfraujoch (46.5°N, 8°E)			Harestua (60°N, 11°E)
	1990-2009 (%/decade)	1996-2009 (%/decade)	2002-2010 (%/decade)	2002-2010 (%/decade)
UV-vis at FTIR SZA	-3.7 ± 1.1	-2.7 ± 1.2	-	-
FTIR	-3.6 ± 0.9	-4.3 ± 1.4	-	-
UV-vis at satellite SZA	-	-2.4 ± 1.1	-0.7 ± 3.6	-3.9 ± 3.1
Satellites nadir	-	-3.6 ± 2.2	-1.8 ± 3.1 (SCIAMACHY)	-4.1 ± 3.9 (SCIAMACHY)
SCIAMACHY limb	-	-	+0.6 ± 3.2	-0.4± 3.0

- Jungfraujoch: good consistency between ground-based UV-vis, FTIR, and satellite observations with negative NO_2 column trend values of about –3%/decade over the 1990-2009 and 1996-2009 periods and trend statistically not significantly different from zero over 2002-2010 period
- Harestua: Negative trends are also obtained over the 2002-2010 periods; however, the trend values are statistically not significant within the 95% confidence level.
- These results further confirm that the trend of stratospheric NO_2 does not necessarily follow the trend of N_2O (~ +2.5%/decade; WMO, 2007) considered as the main source of NO_x in the stratosphere. According to investigations made at the Jungfraujoch station, it could be due to:
  - A change in the NO_x partitioning in favor of NO, due to possible stratospheric cooling (not investigated here) and the decline of chlorine content in the stratosphere through the $ClO + NO \rightarrow Cl + NO_2$. The Cl decline is further confirmed by the observed decrease in $ClONO_2$ (see Figure on the left)
  - A positive trend of stratospheric O_3 (see Figure on the left) consistent with a decrease of NO_2 through the $NO_2 + O_3 \rightarrow NO_3 + O_2$ reaction
  - A strengthening of the Dobson-Brewer circulation allowing less time in the stratosphere for the conversion of N_2O into reactive nitrogen

VII. CONCLUSIONS

- The trend of the stratospheric NO_2 column has been assessed at the NDACC stations of Jungfraujoch and Harestua using satellite, ground-based UV-visible and FTIR datasets
- Depending on the station and time period, trend values are found to be negative or statistically not significantly different from zero.
- These results further confirm that the trend of stratospheric NO_2 does not necessarily follow the trend of N_2O considered as the main source of NO_x in the stratosphere. Investigations made at the Jungfraujoch station suggests that it could be due to a change in the NO_x partitioning and a simultaneous increase of stratospheric O_3
- More details on this study can be found in Hendrick et al. (2012)

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