Investigation of NO_2 vertical distribution from satellite data by using two NO_2 DOAS retrievals

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1 Motivation & Introduction

- NO_2 is one of the most important air pollutants
- catalyses ozone production, causes summer smog, acid rain, and adds local radiative forcing
- NO_x emission sources and their horizontal distribution are well known from satellite measurements
- knowledge of the vertical NO_2 distribution is only limited

- DOAS: Differential Optical Absorption Spectroscopy
  - based on Lambert Beer's law: $\mathbf{I}(\lambda,\mathbf{s}) = \mathbf{I_0}\exp(-\sigma(\lambda)\rho\mathbf{s})$
    - λ: wavelength; σ: absorption cross-section; ρ: amount of absorbers
    - method to calculate the absoprtion of light travelling through the atmosphere
  - can be used for ultra violet (UV) and visible (vis) light
  - amount of trace gases can be derived from the absorption => slant columns (SCs) can be calculated

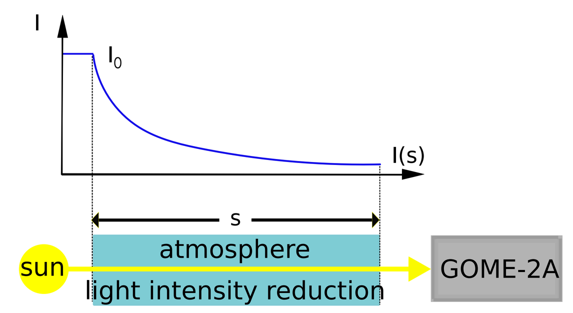

Figure 1: Illustration of the DOAS method
I: intensity at the detector
I0: intensity of the light directly from the sun
s: light path

- Rayleigh scattering in the atmosphere depends on the wavelength
  - larger wavelength: larger penetration depth than smaller wavelength => vertical sensitivity of NO_2 measurements
  - NO_2 SCs derived from the visible are mostly higher than from the UV spectral range
- vertical sensitivities, expressed as box air mass factors (BAMFs) can be calculated with the radiative transfer model SCIATRAN to investigate the differences between the UV and visible spectral ranges
  - above 9km, the sensitivity is slightly higher in the UV, while below 9km, the sensitivity is considerably higher in the vis

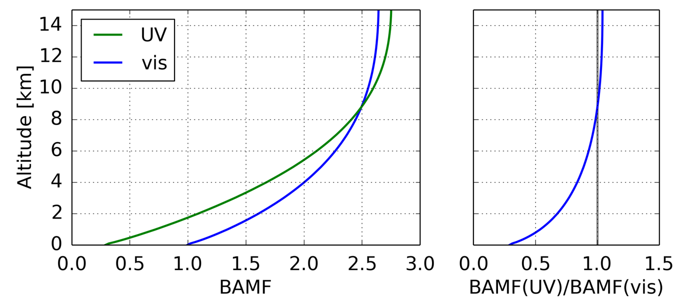

Figure 2: BAMF for both spectral ranges
BAMF for 352 nm (UV) and 438 nm (vis) with a solar zenith angle of 50°, a surface spectral reflectance of 0.04 (UV) and 0.06 (vis) and a US standard atmosphere profile.

=> to get additional knowledge about NO_2 vertical distribution: develop new NO_2 retrieval for UV spectral range based on satellite observations from GOME-2 on board of EUMETSAT's MetOp-A

2 The NO_2 DOAS retrievals and datasets

- fit settings of NO_2 retrievals:

	UV NO_2 fit	vis NO_2 fit
fitting window	342 – 361.5nm	425 – 450nm
polynomial degree	5	3
cross sections	O_3, NO_2, O_4, BrO, HCHO, Ring	O_3, NO_2, O_4, H_2O, Ring
instrumental function	Zeta	–

- the dataset is from 2007 to 2015
- UV NO_2 VCs have a larger spread (7.4×10^{14} molec cm^{-2}) compared with vis NO_2 VCs (2.1×10^{14} molec cm^{-2})

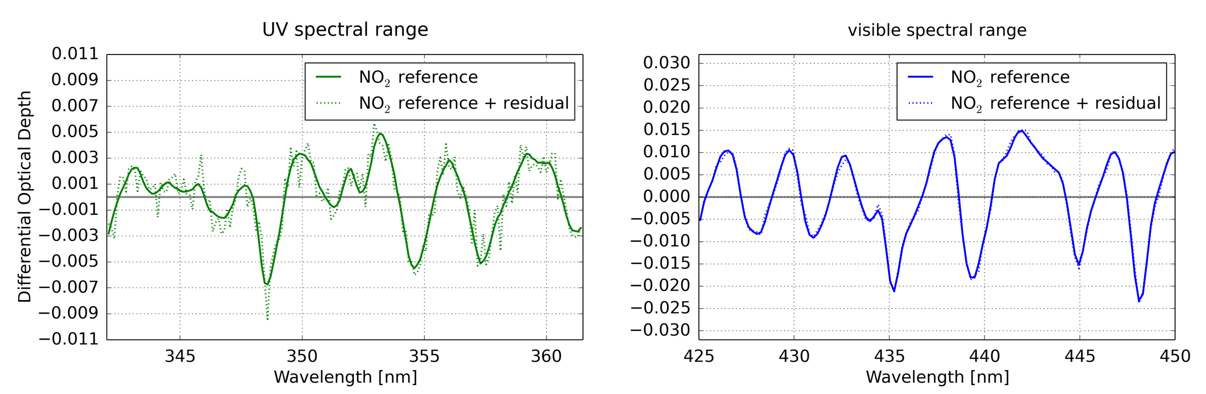

Figure 3: Comparison of NO_2 fits for the UV and visible spectral ranges
The NO_2 reference and reference plus residual for the UV (left) and visible (right) spectral ranges for one pixel of an orbit above Teheran (35.38°N, 51.47°E) on January 22, 2008. The retrieved SC for this pixel for the UV spectral range is 6.31×10^{16} molec cm^{-2} with a fit error of 4.3%. The SC for this pixel for the visible spectral range is 9.33×10^{16} molec cm^{-2} with a fit error of 0.8%. Note the different y-axes.

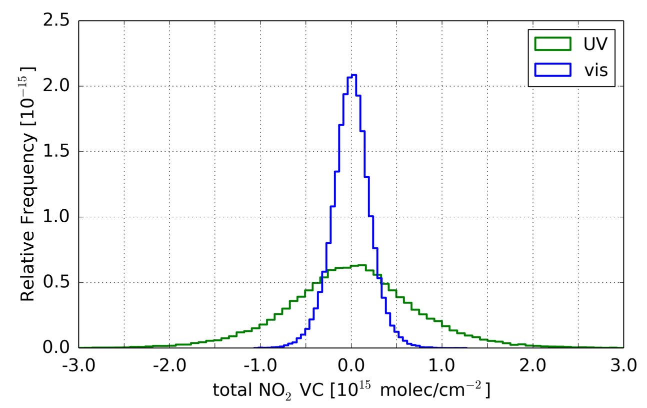

Figure 4: Scatter of NO_2 retrievals
Distribution of NO_2 VCs (January 2008) over a clean area (located over the Pacific: 5°S – 5° N, 150° – 210°E). It is assumed that there is no NO_2 in the troposhere. Therefore, a stratospheric AMF was applied to calculate NO_2 VCs. The curves are normalised and centred on zero.

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3 Comparison of global NO_2 SCs

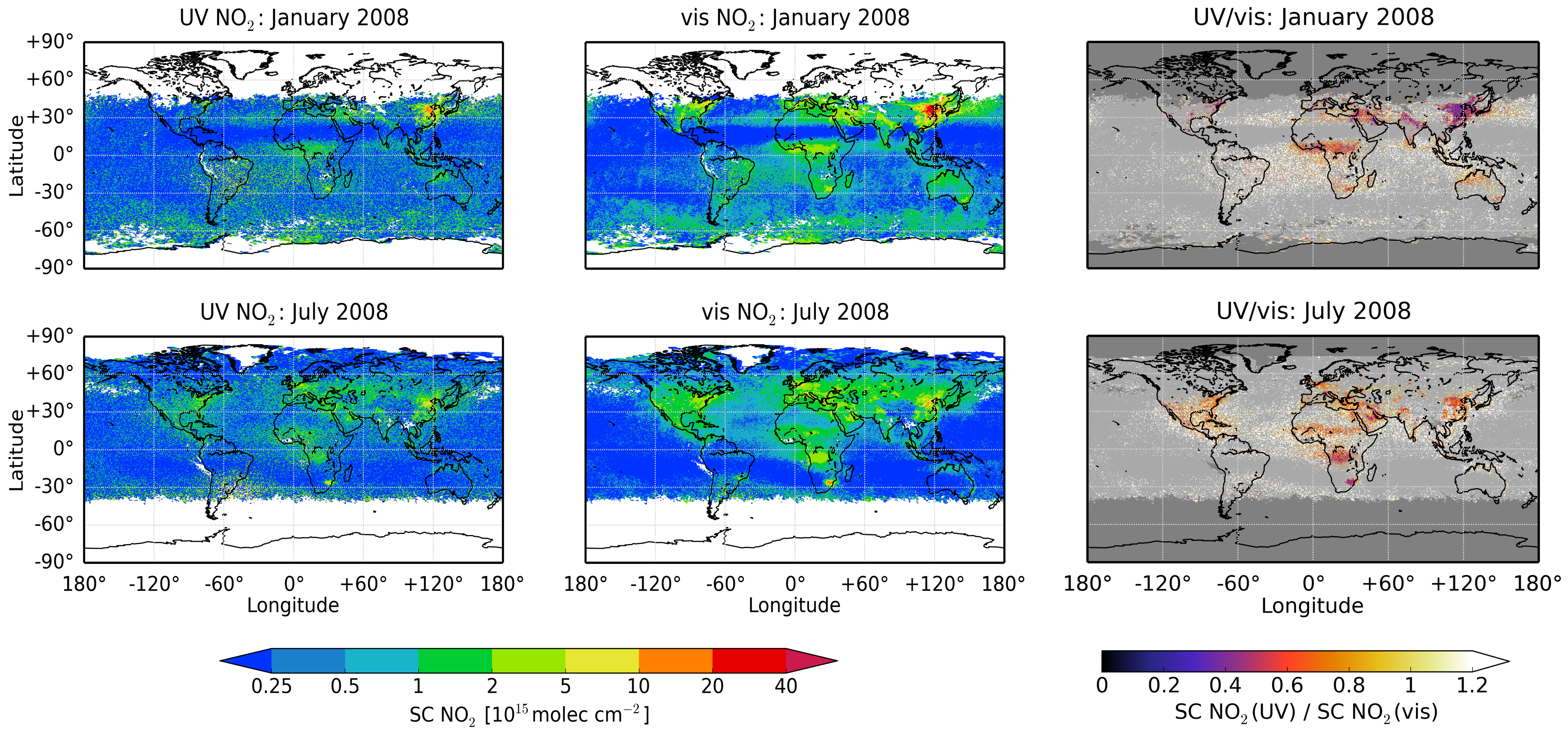

Figure 5: Global distribution of monthly mean tropospheric NO_2 SCs for both spectral ranges and their ratio
SCs (left and center columns): Similiar patterns in UV and visible spectral ranges can be found. Also in the UV spectral range, well known NO_2 signals above highly polluted areas can be observed. Ratio (right column): Dark gray shaded areas indicate that there are no NO_2 values available. Lighter gray shaded areas indicate that the NO_2 values are below one standard deviation of a smoothed latitude-dependent threshold over the reference sector.

- only pixels with cloud fraction < 0.2 are used (FRESCO+ version 6)
- for stratospheric correction: reference sector (180° – 210°E) method

NO_2 SCs:
- with the UV NO_2 retrieval both anthropogenic and natural air pollution can be detected
  - anthropogenic: e.g., China and Highveld Plateau region in South Africa
  - natural: biomass burning regions, e.g., Africa south of the Equator
- NO_2 signals in the UV spectral range are lower than the NO_2 signal in the visible spectral range
- larger differences between NO_2 retrievals above highly polluted areas, especially in the winter hemisphere

Ratio of SCs:
- to assess vertical sensitivity of NO_2, the ratio of two NO_2 retrievals is calculated
- in the winter hemisphere the values of the NO_2 ratio are smaller
- highly polluted areas are clearly visible as local minima of the SC ratio

4 Comparison of time series of NO_2 SCs

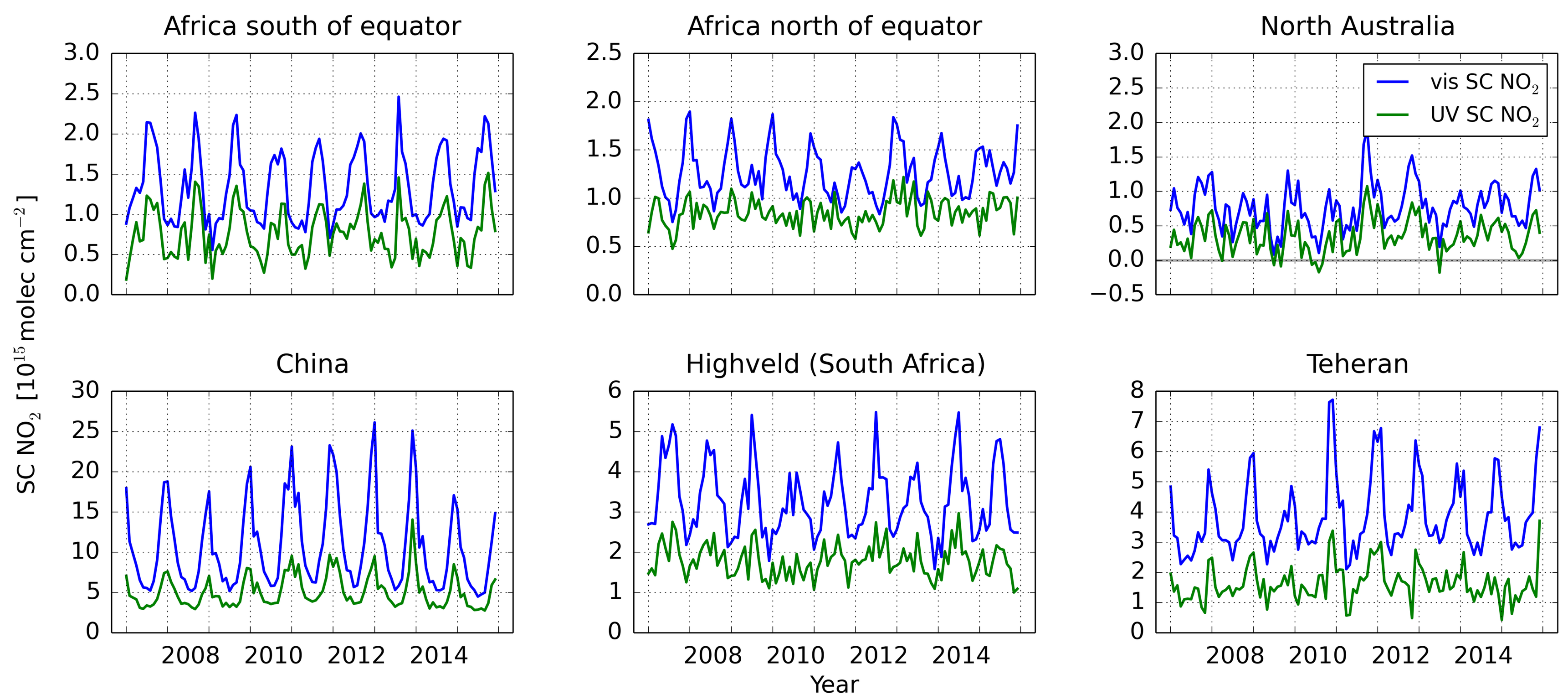

Figure 7: Time series of tropospheric NO_2 SCs for the visible (blue) and UV (green) spectral ranges
Time series from 2007 to 2015 for different regions. The upper three plots are for biomass burning regions and the lower plots are regions with high anthropogenic air pollution. Note the different y-axes of the individual subplots.

Natural air pollution:
- seasonal cycles of the three biomass burning areas differ, because of the shift in the intertropical convergence zone
- Africa south of Equator and north Austraila: similar seasonal cycle for UV and visible spectral range
- Africa north of Equator: clear seasonal cycle in vis, but no intra-annual variability in UV
  - possible reasons:
    - the NO_2 values are to small to be detected
    - the NO_2 is close to the ground, i.e., difficult to detect in UV
    - introduced due to stratospheric correction or cloud filter artefacts
- slightly negative values in north Australia are due to stratospheric correction, which introduces an offset

Anthropogenic air pollution:
- higher values in the respective winter season
- stronger differences between UV and vis NO_2 values in anthropogenically polluted areas compared to naturally polluted areas
- stronger seasonal cycle in the visible spectral range than in UV

5 Summary & Outlook

- we provide a NO_2 DOAS fit in the UV spectral range for GOME-2/MetOp-A satellite data
- the patterns of SCs derived from vis and UV spectral ranges agree well, however UV NO_2 values are smaller than vis NO_2 values
- NO_2 retrieval in the visible spectral range is more sensitive to the lower troposphere => possibility to assess vertical distribution of NO_2
- large differences are mostly located in areas with high anthropogenic air pollution
- next step: comparison of NO_2 VCs, we can get additional information about height dependency => if the a priori model simulates the air pollution profile with correct height dependency, there should be no differences in NO_2 vertical columns for the two wavelength ranges

References & Acknowledgement

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