NEW NDACC RECOMMENDATIONS FOR THE RETRIEVAL OF STRATOSPHERIC NO_2 COLUMNS FROM GROUND-BASED ZENITH-SKY UV-VISIBLE OBSERVATIONS

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I.INTRODUCTION

Stratospheric nitrogen dioxide (NO_2) column measurements are daily performed at sunrise and sunset using ground-based zenith-sky UV-visible spectrometers deployed all over the world in about 35 stations, most of them belonging to the Network for the Detection of Atmospheric Composition Change (NDACC). Despite several cross evaluation exercises, it has been recognized that the NO_2 data records still suffer from residual inconsistencies mainly due to (1) differences in the DOAS settings, in particular the temperature dependence of the NO_2 absorption cross sections and (2) a lack of homogeneity in the air mass factors (AMFs) applied to the measured NO_2 slant columns for their conversion into vertical columns.

Recently, the NDACC UV-visible Working Group has formulated new recommendations aiming at improving the homogeneity of the stratospheric NO_2 column measurements. Regarding the spectral analysis, a list of recommended settings has been established including fitting intervals, absorption cross sections data sets and temperature dependence, as well as methods for wavelength calibration and residual amount in the reference spectrum determination. In case of AMFs, look-up tables (LUTs) have been built from NO_2 profile climatologies based on the harmononic decomposition of the HALOE, SAGE-II and POAM-III data records for the stratosphere and on SAOZ balloon observations for the UTLS, allowing accounting for the dependence of the AMF on the latitudinal and seasonal variations of the NO_2 vertical profile at sunrise and sunset. The calculated LUTs, only suitable for background aerosols conditions, depend on latitude, day of year, wavelength, SZA, surface albedo, and station altitude. The impact of those recommendations is investigated through their application to measurements from a selection of stations of the NDACC/UV-visible network. The consistency of the new ground-based UV-visible data sets with correlative observations from the ERS-2/GOME, ENVISAT/SCIAMACHY, EOS-AURA/OMI, and METOP-A/GOME-2 satellite nadir instruments is investigated.

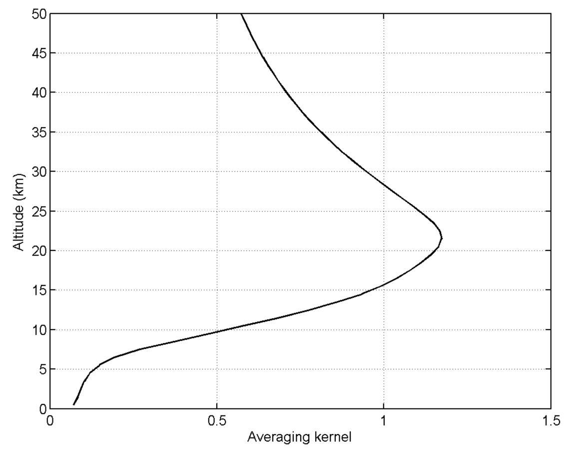
II. NO_2 COLUMN RETRIEVAL

$$VCD(\theta) = \frac{DSCD(\theta) + RCD}{AMF(\theta)}$$

- ***VCD(θ)***: vertical column density at SZA θ
- ***DSCD(θ)***: differential slant column density at SZA θ; direct product of the spectral analysis
- ***RCD***: Residual NO_2 amount in the reference measurement
- ***AMF(θ)***: air mass factor at SZA θ

MEASUREMENT SENSITIVITY:

Typical example of column averaging kernel representative of the NO_2 vertical column at 90°SZA PM at 45°N in May (475 nm)



III. DOAS SETTINGS

Parameter	Recommendation
Fitting interval	425-490 nm
Wavelength calibration	Calibration based on reference solar atlas (Chance and Kurucz, 2010)
Cross-sections	
NO_2	Vandaele et al. (1997), 220°K
O_3	Bogumil et al. (2003), 223°K
H_2O	Hitran 2004 (Rothman et al., 2005)
O_4	Hermans (http://spectrolab.aeronomie.be/o2.htm)
Ring effect	Chance and Spurr (1997)
Molecular and aerosol scattering	Polynomial of order 3 to 5
AMF calculation	BIRA-IASB NO_2 AMF look-up tables
Determination of residual amount in reference spectrum	Chemically modified Langley plot
SZA range for twilight averaging of vertical columns	86-91°SZA

IV. AMF LOOK-UP TABLES

- 2 AMF look-up tables (1 for sunrise and 1 for sunset conditions) based on the harmonic climatology of stratospheric NO_2 profile developed by Lambert et al. (1999, 2000). This climatology consists of a Fourier harmonic decomposition of UARS HALOE v19 and SPOT-4 POAM-III v2 NO_2 profile data records.
- The Lambert et al.'s climatology of NO_2 profiles covers the 20-60 km altitude range. Between 12 and 17 km, the NO_2 profiles are complemented by a climatology derived from SAOZ balloon observations (F. Goutail, personal communication). The NO_2 concentration is set to zero below 12 km altitude.
- NO_2 AMFs calculated using the UVSPEC/DISORT RTM (multiple scattering in a pseudo-spherical geometry). This model is initialized with O_3 and temperature profiles from AFGL and TOMS V8 (TV8) climatology, respectively, and with an aerosol extinction profile corresponding to background conditions.

Parameter	Value
Lambert et al.'s NO_2 profile climatology	– Latitude: 85°S to 85°N step 10° – Month: 1 (Jan) to 12 (Dec) step 1 – Sunrise and sunset conditions – Altitude range: 20-60 km
SAOZ balloon NO_2 profile climatology	– Latitude: tropics, mid– and high latitudes – Resolved in season (spring, summer, fall, winter) – Altitude range: 12-20 km
Wavelength	350 to 550 nm step 40 nm
Surface albedo	0 and 1
Altitude output	0 and 4 km
SZA	10, 30, 50, 70, 80, 82.5, 85, 86, 87, 88, 89, 90, 91, and 92°

V. SATELLITE NO_2 PRODUCTS

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 NO2 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
  - Data available for GOME, SCIAMACHY, GOME-2, and OMI satellite instruments (200 km overpasses)
  - More details in Boersma et al. (2004) and Dirksen et al. (2011)

IUP-Bremen NO_2 algorithm

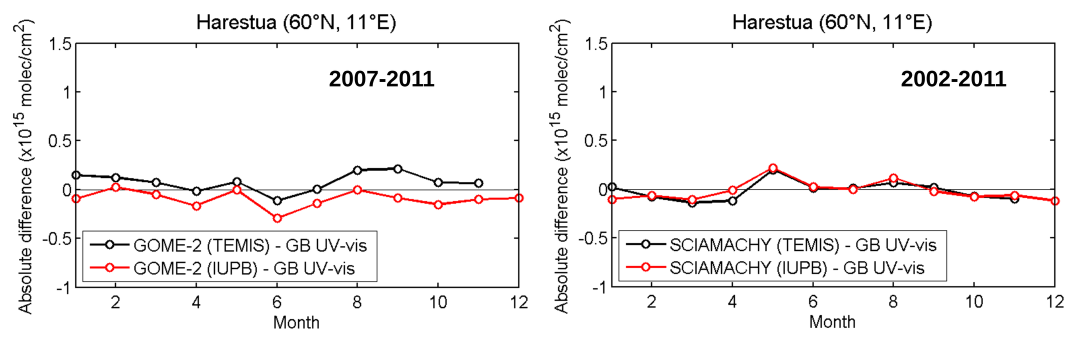
- NO_2 VCDs obtained by dividing NO_2 total slant columns by stratospheric AMFs.
- Strong pollution events partially removed by selecting smallest NO_2 column values within a radius of 200 km around the stations
- Data available for GOME, SCIAMACHY, and GOME-2 satellite instruments
- More details in Richter et al. (2011)

VI. COMPARISON WITH SATELLITE NADIR OBSERVATIONS

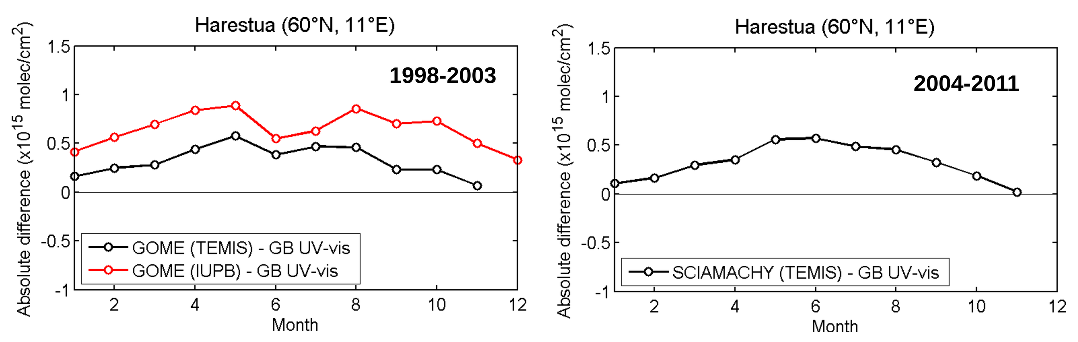
POLAR REGION

Harestua (60°N, 11°E)

GOME-2 SCIAMACHY



GOME OMI

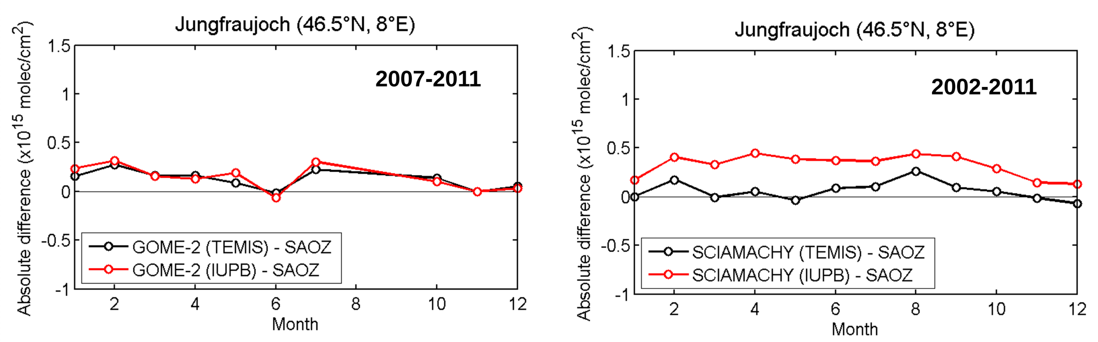


- Ground-based data photochemically converted to satellite overpass SZA
- GOME-2 and SCIAMACHY: good agreement with ground-based UV-visible observations; no marked seasonality in the satellite - ground-based difference; good consistency between TEMIS and IUPB products
- GOME and OMI: larger positive biases are obtained with both products but this effect is significantly larger with IUPB (tropospheric contamination issue ?); more marked seasonality in the satellite - ground-based difference

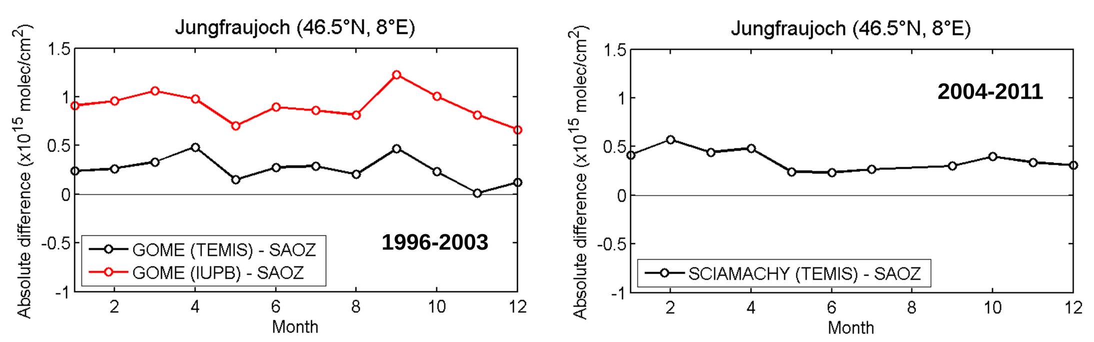
MID-LATITUDE

Jungfraujoch (46.5°N, 8°E)

GOME-2 SCIAMACHY



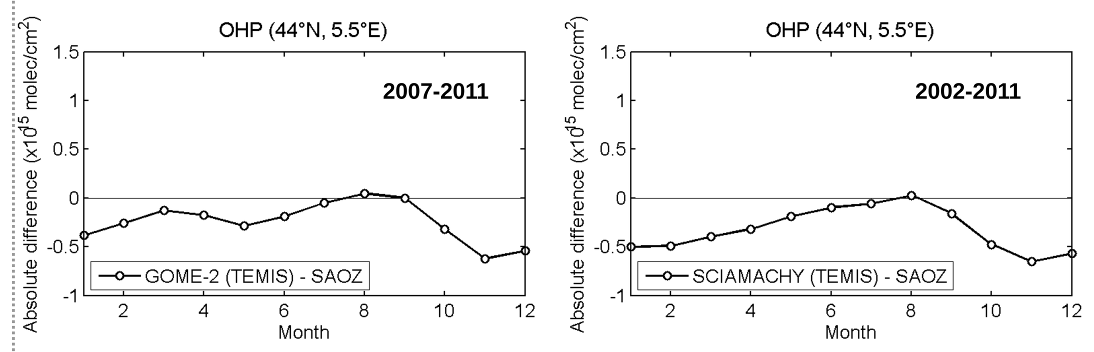
GOME OMI



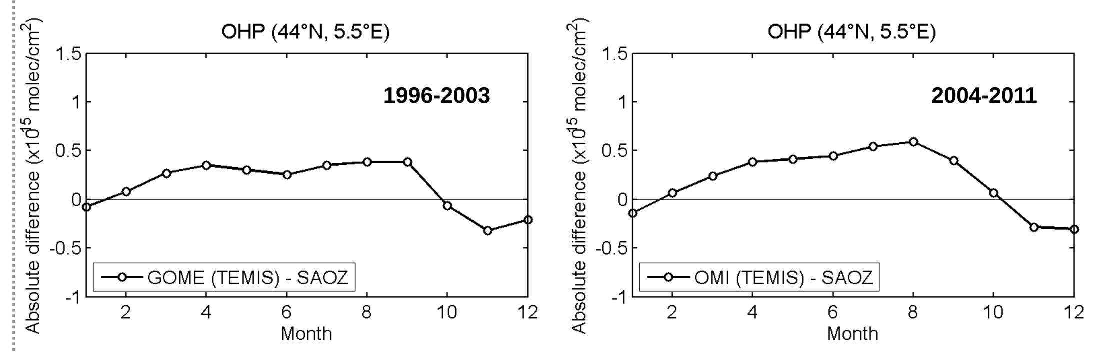
- Ground-based data photochemically converted to satellite overpass SZA
- GOME-2: good agreement with ground-based UV-visible observations; no marked seasonality in the satellite - ground-based difference; good consistency between TEMIS and IUPB products
- SCIAMACHY: larger positive bias obtained with the IUPB product
- GOME and OMI: larger positive biases are obtained with both products but this effect is significantly larger with IUPB (GOME); no marked seasonality in the satellite - ground-based difference

OHP (44°N, 5.5°E)

GOME-2 SCIAMACHY



GOME OMI

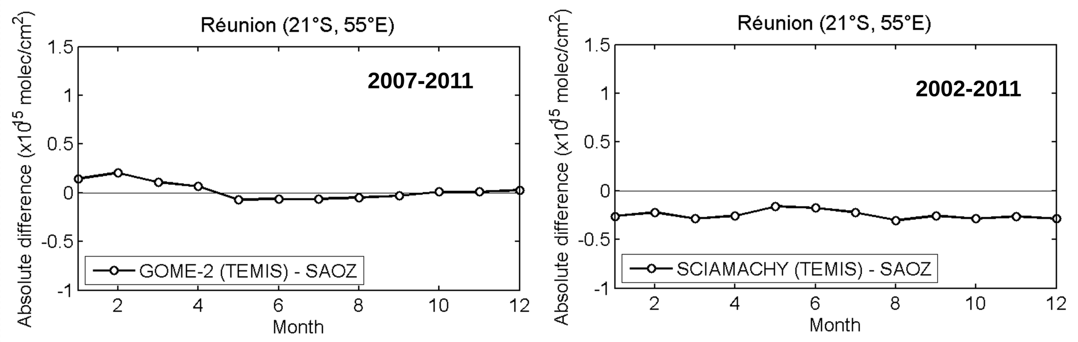


- Ground-based and satellite data photochemically converted to 12h local time
- GOME-2 and SCIAMACHY: larger differences and more marked seasonality than for similar comparison at Jungfraujoch
- GOME and OMI: more marked seasonality than for similar comparison at Jungfraujoch
- Since both stations are very close, these results suggest a possible issue with ground-based UV-visible observations; this is currently under investigation.

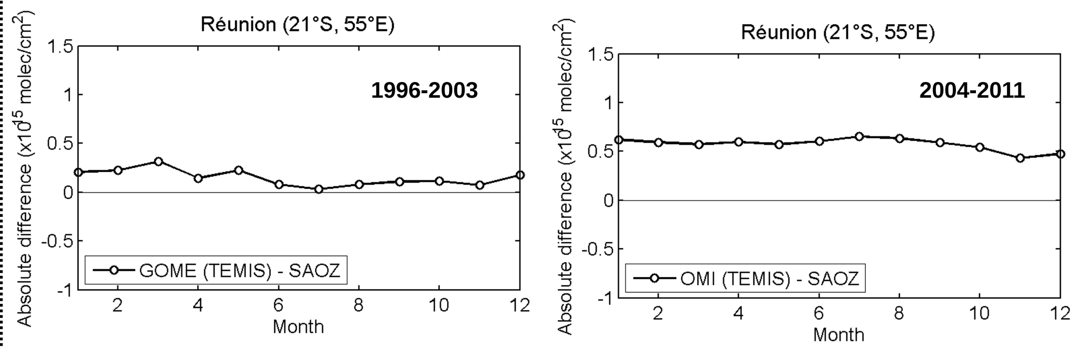
TROPICS

Réunion (21°S, 55°E)

GOME-2 SCIAMACHY



GOME OMI



- Ground-based and satellite data photochemically converted to 12h local time
- GOME-2, GOME, and SCIAMACHY: good agreement with ground-based observations; no seasonality in the satellite - ground-based difference
- OMI: larger differences but no marked seasonality as for the three other satellite instruments

VII. CONCLUSIONS

- The NDACC UV-visible Working Group has made recommendations for improving and homogenizing the retrieval of stratospheric NO_2 columns from twilight zenith-sky visible spectrometers.
- So far the new spectral analysis settings and NO_2 AMFs have been applied to the SAOZ network as well as to the UV-visible spectrometers operated by BIRA-IASB (Jungfraujoch (46.5°N, 8°E) and Harestua (60°N, 11°E)). The evaluation of the error budget on the AMFs as well as on the spectral analysis is currently under progress.
- We started a comparison between satellite nadir and the new ground-based UV-visible NO_2 data sets. The level of consistency depends on the station, satellite instrument, and satellite product. However, it is found that the agreement is generally significantly better with GOME-2 and SCIAMACHY instruments. The interpretation of these results is currently under progress.
- The new recommendations are available at http://uv-vis.aeronomie.be/ground-based.

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