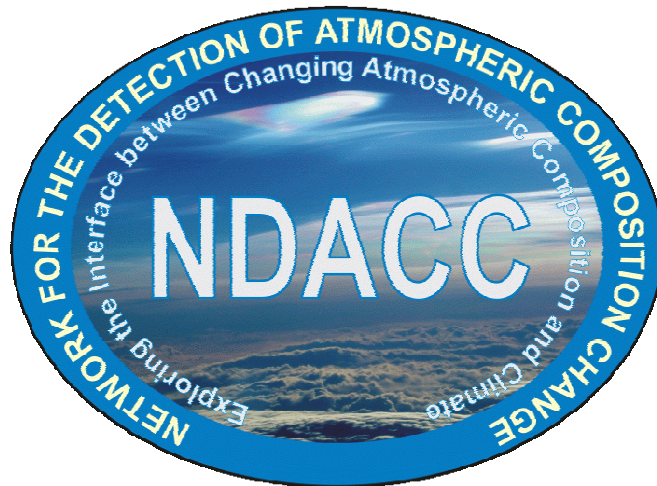


# Network for the Detection of Atmospheric Composition Change (NDACC) UV-VIS Working Group



## Recommendations for NO<sub>2</sub> column retrieval from NDACC zenith-sky UV-VIS spectrometers

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## 1. Introduction

One of the goals of the NDACC is to ensure that long term high quality data sets of a number of important atmospheric parameters are regularly delivered and made available to the scientific community. Protocols for data acquisition, data evaluation and quality assessment are defined as part of the activities of the different instrument working groups. Satellite validation studies have shown that the UV-Vis zenith-sky NO<sub>2</sub> column data sets currently submitted to the NDACC data base still suffer from residual inconsistencies due to (1) differences in the DOAS settings applied by the different data providers, in particular as regards the source and temperature of the NO<sub>2</sub> absorption cross-sections, and (2) a lack of homogeneity in the air mass factors applied to convert NO<sub>2</sub> slant columns into vertical columns. The aim of the present note and of its accompanying material is to provide the recommendations, tools and input data sets that are needed to improve the overall homogeneity of the UV-Vis (stratospheric) NO<sub>2</sub> column measurements. The guidelines described here are preliminary and open for discussion (see in particular section 5). The aim is to converge towards new NDACC standards for NO<sub>2</sub> column retrievals, to be applied for future measurements as well as for reprocessing of historical data sets.

## 2. DOAS retrieval settings

The following recommendations are based on experience from recent intercomparison exercises, in particular from The Cabauw Intercomparison campaign for Nitrogen Dioxide Measuring Instruments (CINDI) in 2009 (Roscoe et al., 2010; Piters et al., 2012).

|                                      | RECOMMENDED SETTINGS                       | COMMENTS   |
|--------------------------------------|--|--|
| <b>Fitting interval</b>              | 425-490 nm                                 | Settings adequate for simultaneous NO <sub>2</sub> and O <sub>4</sub> retrieval. The large water vapor absorption band at 510 nm is avoided.   |
| <b>Wavelength calibration method</b> | Calibration based on reference solar atlas | For optimal DOAS retrievals, spectra should be aligned on an accurate atlas of solar lines. A good reference is Chance and Kurucz (2010).  |
| <b>Cross-sections</b>                |  |  |
| <b>NO<sub>2</sub></b>                | Vandaele et al. (1997), 220° K             | The low temperature reference data set of Vandaele et al. (220° K) is adequate for stratospheric NO <sub>2</sub> retrievals. Fine adjustment of temperature effects can eventually be performed in a post-processing step.   |
| <b>O<sub>3</sub></b>                 | Bogumil et al. (2003), 223° K              | This data set (obtained from SCIAMACHY flight-model measurements) is characterized by an excellent signal/noise ratio in the Chappuis bands and a good consistency with values in the Huggins bands.   |
| <b>H<sub>2</sub>O</b>                | Hitran                                     | We recommend using the most recent updates of the HITRAN data base as a source. Alternatively the measured data set of Harder and Brault (1997) is also a good choice if limited to the the NO <sub>2</sub> spectral region.   |
| <b>O<sub>4</sub></b>                 | Hermans et al. (2003)                      | Current O <sub>4</sub> absorption cross-section reference for the visible and near UV regions.   |
| <b>Ring effect correction method</b> | Chance and Spurr (1997)                    | The Ring effect is due to inelastic Raman scattering by air molecules (N <sub>2</sub> and O <sub>2</sub> ). It is responsible for a smoothing effect that mostly affects the highly structured solar Fraunhofer lines. Ring effect is usually corrected as part of the DOAS fit using a pseudo cross-section. Note that this approach neglects the impact of the Ring effect on the NO <sub>2</sub> absorption itself (molecular Ring effect), which leads to a systematic underestimation of the NO <sub>2</sub> slant columns by about 5%. |
| <b>Polynomial term</b>               | Polynomial of order 3 to 5 maximum         | Filters out the broadband atmospheric attenuation due to scattering by air and particles. High orders should be avoided to   |

|  |   |   |
|--|---|---|
|  |   | minimize the cross-correlation with molecular absorption differential structures.   |
| <b>Intensity offset correction</b>                               | Slope                                     | The intensity offset term corrects for possible additive biases in measured intensities, such as can be produced by spectral stray-light or incorrect dark-current correction. It also implicitly corrects for the wavelength dependence of the Raman scattering probability (not accounted for in the simple Ring correction outlined above).  |
| <b>AMF calculation</b>   | NDACC climatology of NO <sub>2</sub> AMFs | A generic climatology of NO <sub>2</sub> AMFs has been generated at IASB-BIRA for NDACC. This table is constructed using the harmonic climatology of stratospheric NO <sub>2</sub> profile of Lambert et al. (1999; 2000), complemented at low altitudes (below 17 km) by a climatology derived from SAOZ balloon soundings. It has been designed to be applicable at any NDACC station (see section 3 of this note). Note that the twilight photochemistry of NO <sub>2</sub> is not taken into account, which may introduce biases on the resulting vertical columns of about 5-10% (similar to the error introduced by the simplified Ring correction approach). |
| <b>Determination of residual amount in reference spectrum</b>    | Modified Langley plot                     | If possible, modified Langley plots are the preferred approach to determine the residual NO <sub>2</sub> amount in the reference spectrum. Alternative more empirical approaches such as the minimization of the diurnal variation of the NO <sub>2</sub> VCD are also suitable.  |
| <b>SZA range used for twilight averaging of vertical columns</b> | 86° – 91° SZA                             | Best compromise between accuracy and precision is achieved in the 86-91° SZA range. The effective SZA of the reported NO <sub>2</sub> average should be explicitly mentioned in the data product.   |

### 3. NO<sub>2</sub> air mass factors climatology

As reported in the existing literature (e.g. *Sarkissian et al., 1995, Van Roozendaal et al., 1994*), differences in the radiative transfer model, pressure and temperature profiles, NO<sub>2</sub> profile shape, and wavelength can have a significant impact (up to 10-15%) on the resulting NO<sub>2</sub> AMF values. For the sake of homogeneity, it is desirable to improve the level of standardization of the UV-Vis data evaluation process. We describe a new multi-entry data base of NO<sub>2</sub> AMFs applicable at the global scale.

The proposed data base of NO<sub>2</sub> AMFs is based on the harmonic climatology of stratospheric NO<sub>2</sub> 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<sub>2</sub> profile data records. It has been used for the retrieval of global total ozone fields from recent European UV-VIS nadir sounders (GOME and SCIAMACHY). For the NO<sub>2</sub> AMF calculation, the NO<sub>2</sub> concentration between 20 and 60 km altitude is taken from the Lambert et al.'s climatology. Between 12 and 17 km, the NO<sub>2</sub> profiles are complemented by a climatology derived from SAOZ balloon observations (F. Goutail, personal communication). The NO<sub>2</sub> concentration is set to zero below 12 km altitude.

The parameters considered in building the NO<sub>2</sub> AMF climatology are: wavelength, ground albedo, altitude of the station, and SZA. Table 1 summarizes these different parameters and their corresponding values.

| Parameter  | Values   |
|--|--|
| Lambert et al.'s NO <sub>2</sub> profile climatology | - Latitude: 85°S to 85°N step 10°<br>- Month: 1 (Jan) to 12 (Dec) step 1 |

|  |  |
|--|--|
|  | <ul style="list-style-type: none"> <li>- Sunrise and sunset</li> <li>- Altitude range : 20-60 km</li> </ul>  |
| SAOZ balloon NO <sub>2</sub> profile climatology | <ul style="list-style-type: none"> <li>- Latitude: tropics, mid- and high-latitudes</li> <li>- Resolved in seasons (spring, summer, fall, winter)</li> <li>- Altitude range: 12-20 km</li> </ul> |
| Wavelength                                       | 350 to 550 nm step 40 nm   |
| Surface albedo                                   | 0 and 1  |
| Altitude of the station                          | 0 and 4 km   |
| SZA  | 10, 30, 50, 70, 80, 82.5, 85, 86, 87, 88, 89, 90, 91, and 92°  |

Table 1: Parameters of the NO<sub>2</sub> AMF climatology and their corresponding values.

NO<sub>2</sub> AMFs have been calculated using the UVSPEC/DISORT radiative transfer (RT) model which includes a treatment of the multiple scattering in a pseudo-spherical geometry. This model has been validated through several intercomparison exercises (e.g. *Hendrick et al., 2006*; *Wagner et al., 2007*). An aerosol extinction profile corresponding to background conditions has been used for the AMF calculation. It has been constructed from the aerosol model of *Shettle* (1989) included in UVSPEC/DISORT, and is therefore not suitable for volcanic conditions. The O<sub>3</sub> and temperature profiles are from US Standard Atmosphere and the TOMS V8 climatology, respectively.

The NO<sub>2</sub> AMF climatology consists of 2 look-up tables (size: 416 KB each), one for sunrise and one for sunset conditions. An interpolation routine has been developed to extract appropriately parameterized NO<sub>2</sub> AMFs for the different NDACC stations. The interpolation routine is written in FORTRAN 77 and a DOS executable has been created. The source code is also available for compilation on LINUX machines. In addition, a global monthly climatology of the surface albedo at two wavelengths (380 and 494 nm) is coupled to the interpolation routine so that realistic albedo values can be obtained in a transparent way. This albedo climatology is extracted from the GOME surface albedo database developed by *Koelemeijer et al. (2003)*. It consists of 24 (12x2) look-up tables, one for each month of the year and each wavelength. Albedo values are given for grid-cells of 1° x 1° (latitude: -89.5° to 89.5°; longitude: -179.5° to 179.5°).

#### 4. How to use the AMF climatology?

The zip file contains 31 files: 2 NO<sub>2</sub> AMF look-up tables, 24 surface albedo look-up tables (size: 1.1 MB each), a DOS executable ('no2\_amf\_interpolation\_dos'), the source code in FORTRAN 77 ('no2\_amf\_interpolation.for'), two input files for selecting parameter values, and 1 output file. All the files should be located in the same directory for a proper use of the climatology. In the file 'input\_no2\_amf.dat', the user can enter a value for wavelength, latitude, longitude, surface albedo, and altitude of the station. Regarding the albedo, the user has to give a value to a flag in order to determine whether he wants to use the albedo climatology (flag=1) or not (flag=2). The user has also to define the name of the file containing lines with year, decimal day numbers (decimals being decimal UT time), and SZAs at which AMF should be extracted (here this file is called 'DAY\_SZA.dat'; maximum number of lines in this file: 500000). The last flag is for the display of the interpolation results on the screen (1: display; 2: no display). The resulting NO<sub>2</sub> AMFs are stored in a file called 'no2\_amf\_output.dat'.

## 5. Points for discussion

The following points have been discussed at the NORS/NDACC UV-VIS Working Group meeting held on 3-4 July 2012 at BIRA in Brussels:

- *Should we introduce a more refined (post-processing) correction for variations of the stratospheric temperature (around 220°K) ?*

Decision: It is recommended to apply a temperature correction using a simple monthly zonal climatology of temperature profiles. In the future, this correction could be coupled to the NO<sub>2</sub> AMF climatology.

- *How to filter data contaminated by tropospheric NO<sub>2</sub> ?*

Decision: The Langley-plot method should be applied (data points not lying on 'smooth' lines indicate a tropospheric contamination). There was also a consensus about the fact that even with a fixed reference spectra, a photochemically modified Langley plot is needed to check that the instrument is stable on long time periods.

- *How to filter data for clouds – is it necessary?*

Decision: Clouds have a small effect on stratospheric NO<sub>2</sub>. However, all groups agreed to eliminate measurements with large enhancements of O<sub>4</sub> and/or H<sub>2</sub>O.

- *Should we explicitly correct for the impact of the Ring effect on the NO<sub>2</sub> SCDs ?*

Decision: The post-processing option is chosen, based on simple SZA parameterization.

- *Should we take into account the NO<sub>2</sub> photochemistry and its impact on the AMF ?*

Decision: The NO<sub>2</sub> photochemistry at twilight will not be taken into account in the standard AMF calculation since it is rather complex to implement at a global scale. However, sensitivity tests are needed in order to estimate the impact of the NO<sub>2</sub> photochemistry on the AMF. This could be included in the error budget.

- *How to derive best estimates of our random and systematic uncertainties? (both need to be reported according to GEOMS)*

Decision: Random uncertainty should be derived from fitting residuals and systematic uncertainty has to be estimated off-line based on sensitivity studies. The error budget should be estimated independently for each station.

- *For historical long data series going back to early 1990 or before: Is there a need to introduce an AMF correction for the period affected by the Pinatubo aerosols?*

Decision: An AMF correction for Pinatubo aerosols will not be included in the standard recommendations. However, sensitivity tests are needed to estimate the impact of the Mount Pinatubo eruption on the NO<sub>2</sub> AMFs.

- *How to proceed with UV instruments which can not follow the DOAS recommendation of 425-490nm ?*

Decision: Visible instruments are preferable than UV ones for NO<sub>2</sub> but a good UV instrument is better than a bad visible one. It is recommended to derive an error estimate representative of the instrument.

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## 7. Contact

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