

This discussion paper is/has been under review for the journal Atmospheric Measurement Techniques (AMT). Please refer to the corresponding final paper in AMT if available.

# Operational total and tropospheric NO<sub>2</sub> column retrieval for GOME-2

**P. Valks<sup>1</sup>, G. Pinardi<sup>2</sup>, A. Richter<sup>3</sup>, J.-C. Lambert<sup>2</sup>, N. Hao<sup>1</sup>, D. Loyola<sup>1</sup>,  
M. Van Roozendaal<sup>2</sup>, and S. Emmadi<sup>1</sup>**

<sup>1</sup>Remote Sensing Technology Institute, German Aerospace Center, Wessling, Germany

<sup>2</sup>Belgian Institute for Space Aeronomy, Brussels, Belgium

<sup>3</sup>Institute of Environmental Physics, University of Bremen, Germany

Received: 10 February 2011 – Accepted: 4 March 2011 – Published: 9 March 2011

Correspondence to: P. Valks (pieter.valks@dlr.de)

Published by Copernicus Publications on behalf of the European Geosciences Union.

**AMTD**

4, 1617–1676, 2011

## Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Abstract

This paper presents the algorithm for the operational near real time retrieval of total and tropospheric NO<sub>2</sub> columns from the Global Ozone Monitoring Experiment (GOME-2). The retrieval is performed with the GOME Data Processor (GDP) version 4.4 as used by the EUMETSAT Satellite Application Facility on Ozone and Atmospheric Chemistry Monitoring (O3M-SAF). The Differential Optical Absorption Spectroscopy (DOAS) method is used to determine NO<sub>2</sub> slant columns from GOME-2 (ir)radiance data in the 425–450 nm range. Initial total NO<sub>2</sub> columns are computed using stratospheric air mass factors, and GOME-2 derived cloud properties are used to calculate the air mass factors for scenarios in the presence of clouds. To obtain the stratospheric NO<sub>2</sub> component, a spatial filtering approach is used, which is shown to be an improvement on the Pacific reference sector method. Tropospheric air mass factors are computed using monthly averaged NO<sub>2</sub> profiles from the MOZART-2 chemistry transport model. An error assessment shows that the random error in the GOME-2 NO<sub>2</sub> slant columns is approximately  $0.45 \times 10^{15} \text{ molec cm}^{-2}$ . As a result of the improved quartz diffuser plate used in the GOME-2 instrument, the systematic error in the slant columns is strongly reduced compared to GOME/ERS-2. The estimated uncertainty in the GOME-2 tropospheric NO<sub>2</sub> column for polluted conditions ranges from 40 to 80%. An end-to-end ground-based validation approach for the GOME-2 NO<sub>2</sub> columns is illustrated based on MAX-DOAS measurements at the Observatoire de Haute Provence (OHP). The GOME-2 stratospheric NO<sub>2</sub> columns are found to be in good overall agreement with coincident ground-based measurements at OHP. A time series of the MAX-DOAS and the GOME-2 tropospheric NO<sub>2</sub> columns shows that pollution episodes at OHP are well captured by GOME-2. Monthly mean tropospheric columns are in very good agreement, with differences generally within  $0.5 \times 10^{15} \text{ molec cm}^{-2}$ .

AMTD

4, 1617–1676, 2011

## Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## 1 Introduction

Nitrogen dioxide ( $\text{NO}_2$ ) plays a key role in both stratospheric and tropospheric chemistry. In the stratosphere, it is involved in ozone destruction via a direct reaction with atomic oxygen and in the reaction cycles of halogen compounds (e.g. Solomon, 1999). In the troposphere,  $\text{NO}_2$  is an important air pollutant affecting human health and ecosystems and one of the most important ozone precursors. As a greenhouse gas,  $\text{NO}_2$  contributes significantly to radiative forcing locally over industrial and urban areas. Although the direct contribution of tropospheric  $\text{NO}_2$  to global warming is relatively small, emissions of nitrogen oxides ( $\text{NO}_x = \text{NO} + \text{NO}_2$ ) also have an indirect effect on the global climate by perturbing ozone and methane concentrations. The main anthropogenic sources of nitrogen oxides are combustion of fossil fuels and biomass burning, the most important natural sources are microbial production in soils, wildfires and lightning. The industrialisation and population growth in the 19th and 20th century have resulted in a strong increase of the anthropogenic  $\text{NO}_x$  emissions since pre-industrial times. Although recently emissions have decreased in several industrialized countries (e.g. in Europe and North America) as a result of pollution reduction measures, emissions in rapidly developing countries in Asia and elsewhere continue to increase. Studies using GOME and SCIAMACHY satellite measurements found especially strong increases in tropospheric  $\text{NO}_2$  over China during the last decades (Richter et al., 2005; van der A et al., 2006, 2008).

Complementary to ground-based measurements, which provide accurate information on the local  $\text{NO}_2$  concentrations, observations from space platforms offer the possibility to measure the distribution of  $\text{NO}_2$  globally, including remote places with few in-situ measurements, and to study its large scale temporal and spatial variability. The Global Ozone Monitoring Experiment (GOME), launched on ERS-2 in 1995, provides global  $\text{NO}_2$  column amounts on a daily basis (Burrows et al., 1999). GOME data has been used to study the variation of stratospheric  $\text{NO}_2$  and to monitor and investigate several important aspects of stratospheric chemistry and dynamics e.g. the Noxon-Cliff,

## AMTD

4, 1617–1676, 2011

### Operational $\text{NO}_2$ retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Operational NO<sub>2</sub>  
retrieval for GOME-2**

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



zonal symmetry, or interhemispheric differences (Wenig et al., 2004). Several papers have been published on the retrieval of tropospheric NO<sub>2</sub> from GOME (e.g. Leue et al., 2001; Velders et al., 2001; Martin et al., 2002; Richter and Burrows, 2002). Over NO<sub>2</sub> source regions, such as Europe, South East Asia and North America, the tropospheric component of the NO<sub>2</sub> column is comparable in magnitude to the stratospheric component. Major challenges are involved in quantifying this tropospheric NO<sub>2</sub> column from satellite measurements involving the subtraction of the estimated stratospheric NO<sub>2</sub>, and the conversion of the tropospheric residual into a vertical column using an accurate tropospheric air mass factor (Boersma et al., 2004).

A new generation of satellite instruments provides trace gases measurements with better spatial resolution that allow a detailed view of the NO<sub>2</sub> pollution patterns (Richter et al., 2005; Bucsela et al., 2006; Boersma et al., 2007). These are the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) (Bovensmann et al., 1999) on the Envisat platform, the Ozone Monitoring Instrument (OMI) (Levelt et al., 2006) on EOS-Aura, and GOME-2 aboard MetOp-A (Callies et al., 2005; Munro et al., 2006). GOME-2, the subject of the current study, observes about 4 times smaller ground pixels than its predecessor GOME on ERS-2 and provides an almost global coverage on a daily basis. With the launch of GOME-2 on MetOp-A, the foundation was laid for a continuous data set of at least 25 years of NO<sub>2</sub> measurements from space. Two more GOME-2 sensors on the MetOp-B and MetOp-C platforms will extend the GOME type data record until 2020. This unique data record will be further extended by the Sentinel-5 precursor to be launched by the middle of this decade, and the Sentinel-4 and Sentinel-5 sensors scheduled for the end of this decade.

In this paper, we describe the operational total and tropospheric NO<sub>2</sub> retrieval algorithms for GOME-2, as implemented in the GOME Data Processor (GDP) version 4.4 and developed within the framework of EUMETSAT's Satellite Application Facility on Ozone and Atmospheric Chemistry Monitoring (O3M-SAF). First, we give an overview of the GOME-2 satellite instrument, followed by a description of the near real time processing and data transport. Then the various steps in the total and tropospheric NO<sub>2</sub>



column retrieval algorithms are presented. In Sect. 3, the NO<sub>2</sub> slant column retrieval using the Differential Optical Absorption Spectroscopy (DOAS) method is described and in Sect. 4, the air mass factor and the total vertical column computation are discussed. Section 5 describes the tropospheric NO<sub>2</sub> column retrieval and some examples of GOME-2's NO<sub>2</sub> monitoring capabilities are given. An error assessment of the GOME-2 NO<sub>2</sub> columns is presented in Sect. 6. The final part of this paper is devoted to a ground-based validation study of the operational GOME-2 NO<sub>2</sub> data product. The validation methodology is described and the end-to-end validation is illustrated using measurement results obtained at the Observatoire de Haute Provence (44° N, 5.7° E).

## 2 The GOME-2 Instrument

The Second Global Ozone Monitoring Experiment (GOME-2) on the MetOp-A satellite, launched in October 2006, is part of the EUMETSAT Polar System (EPS). MetOp-A is flying on a sun-synchronous orbit with an equator crossing time of 09:30 local time (descending node) and a repeat cycle of 29 days. The GOME-2 instrument on MetOp continues the long-term monitoring of atmospheric trace gases, including O<sub>3</sub> and NO<sub>2</sub>, started by GOME (launched on ESA's ERS-2 platform in 1995) and continued with SCIAMACHY (launched on ESA's ENVISAT platform in 2002). GOME-2 is an improved version of the GOME instrument on the ERS-2 satellite (see Table 1 and Callies et al., 2000; Munro et al., 2006). It is a nadir-scanning UV-VIS spectrometer with four main optical channels, covering the spectral range between 240 and 790 nm with a spectral resolution between 0.26 nm and 0.51 nm (FWHM). Additionally, two polarisation components are measured with Polarisation Measurements Devices (PMDs) at 30 broad-band channels covering the full spectral range.

The default swath width of the GOME-2 scan is 1920 km, which enables global coverage in about 1.5 days. The along-track dimension of the instantaneous field of view is ~40 km, while the across-track dimension depends on the integration time used for each channel. For the 1920 km swath and the default integration time of 187.5 ms, the

## Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



ground pixel size is  $80 \times 40 \text{ km}^2$  (across-track  $\times$  along-track) in the forward scan. Owing to a non-linear movement of the scan mirror, the ground pixel size remains nearly constant over the full scan.

GOME-2 measures the back-scattered and reflected radiation from the earth-atmosphere system. In addition, a direct sun spectrum is recorded once per day via a diffuser plate. An important improvement of the GOME-2 instrument compared to GOME/ERS-2 is the use of a quartz quasi-volume diffuser for the direct sun measurements. The sun-angle dependent differential structures in the Bi-directional Scattering Distribution Function (BSDF) for this diffuser is strongly reduced compared the ground aluminium diffuser as used in GOME/ERS-2. The effect of the improved diffuser on the  $\text{NO}_2$  slant column retrieval with GOME-2 is discussed in Sect. 6.1.

## 2.1 Data transport and processing

The operational GOME-2 total column  $\text{NO}_2$  product is provided by the German Aerospace Center (DLR) in the framework of EUMETSAT's Satellite Application Facility on Ozone and Atmospheric Chemistry Monitoring (O3M-SAF). The focus of the O3M-SAF is to process, archive, validate and disseminate atmospheric data products of ozone,  $\text{NO}_2$  and various other trace gases, aerosols and surface ultraviolet radiation.

The first step in the processing chain is the production of calibrated and geolocated level 1 radiances (level 0-to-1 processing). MetOp data is transmitted once per orbit to the EUMETSAT ground station in Svalbard, Norway. Level 1 products are generated operationally in the Core Ground Segment (CGS) at EUMETSAT headquarters in Darmstadt, Germany and are broadcasted via the EUMETCast system in data chunks called PDUs, each containing 3 min of measurements.

The GOME-2 level 1 PDUs are received at the O3M-SAF processing facility in DLR, Germany, approximately 1 h and 45 min after sensing. The DLR multi-mission payload ground segment system (Heinen et al., 2009) controls the reception, processing, archiving, ordering and dissemination of the GOME-2 trace gas column products. The

## Operational $\text{NO}_2$ retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



GOME-2 level 1 PDUs are processed with the UPAS (Universal Processor for UV/VIS Atmospheric Spectrometers) system, a new generation system for the processing of operational trace gas and cloud property products in near-real time and off-line (Livschitz and Loyola, 2003; Valks et al., 2011). The resulting GOME-2 level 2 products are disseminated through EUMETCast, WMO/GTS and the Internet. The level 1 data reception, processing with UPAS and level 2 data dissemination takes less than 15 minutes in total. The end user receives the GOME-2 level 2 near-real-time total column products in less than two hours after sensing with a committed service of 24 h a day, 365 days a year. DLR provides also offline and reprocessed GOME-2 level 2 consolidated products on an orbital basis. These can be ordered via the EUMETSAT product navigator (<http://navigator.eumetsat.int>) or DLR EOWEB systems (<http://eoweb.dlr.de>).

### 3 DOAS slant column fitting

The first major algorithm component in the NO<sub>2</sub> column retrieval with the GOME Data Processor (GDP) is the Differential Optical Absorption Spectroscopy (DOAS) fitting (Platt, 1994; Platt and Stutz, 2008). This is a straightforward least-squares inversion to deliver the effective slant column of total NO<sub>2</sub>, plus a number of auxiliary fitted parameters and error diagnostics. In DOAS fitting for optically thin absorbers, such as NO<sub>2</sub> in the visible wavelength region, the basic model is the Beer-Lambert extinction law. A polynomial closure term accounts for broadband effects: molecular scattering, aerosol scattering and absorption and reflection from the Earth's surface. We also include an additive spectrum for Ring effect interference (Chance and Spurr, 1997). The fitting model is then:

$$Y(\lambda) \equiv \ln \left[ \frac{I(\lambda)}{I^0(\lambda)} \right] = - \sum_g S_g \sigma_g(\lambda) - \sum_{j=0}^3 \alpha_j (\lambda - \lambda^*)^j - \alpha_R R(\lambda) \quad (1)$$

Here,  $I(\lambda)$  is the earthshine spectrum at wavelength  $\lambda$ ,  $I^0(\lambda)$  the daily reference (solar) spectrum,  $S_g$  the slant column density of gas  $g$ , and  $\sigma_g(\lambda)$  is the associated trace gas

## Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Operational NO<sub>2</sub>  
retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



absorption cross section. The second term in Eq. (1) is the closure polynomial (a cubic polynomial has been assumed for the NO<sub>2</sub> retrieval), with  $\lambda^*$  a reference wavelength for this polynomial. The last term on the right hand side of Eq. (1) is the additive term for the Ring reference spectrum  $R(\lambda)$ . The fitting minimizes the weighted least squares difference between measurement based and simulated optical densities  $Y_{\text{meas}}(\lambda)$  (on the left side of Eq. (1)) and  $Y_{\text{sim}}(\lambda)$  (on the right hand side of Eq. (1)). The model in Eq. (1) is linear in the slant columns  $S_g$ , the polynomial coefficients  $\alpha_j$  and the Ring scaling parameter  $\alpha_R$ . The fitting window for NO<sub>2</sub> is 425–450 nm in GOME-2 Channel 3. In this wavelength region, the differential NO<sub>2</sub> absorption features are prominent, the interference by other species is small, and GOME-2 measurements have a relatively high signal-to-noise ratio. For the reference spectrum  $I^0(\lambda)$ , daily solar spectra measured with GOME-2 are used.

In the DOAS-fit, the absorption cross-section for NO<sub>2</sub> and O<sub>3</sub> from Gür et al. (2005) are used, which have been measured with the GOME-2 Flight Model (FM). The GOME-2 FM cross-sections were measured at five different temperatures (203, 223, 243, 273 and 293 K) under instrumental conditions of the in-flight operation aboard MetOp. An important advantage of these spectra is that the GOME-2 FM spectrometer is a well-characterized instrument including both spectral and radiometric calibration, and its instrumental line shape was accurately determined. DOAS analyses with GOME-2 data show that the Flight Model cross-sections provide consistent and stable fitting results in the NO<sub>2</sub> fitting window (Lambert et al., 2008).

The NO<sub>2</sub> absorption cross-section has a marked temperature dependence in this wavelength region, which has to be taken into account to improve the accuracy of the retrieved columns. In the DOAS fit, a single NO<sub>2</sub> cross-section reference spectrum at 243 K is used, and an a posteriori correction for the difference between the atmospheric temperature and the 243 K cross-sections temperature is performed on the AMF level (see Sect. 4).

Although the O<sub>3</sub> absorption in this part of the Chappuis band is weak (one reason for the fitting window choice), O<sub>3</sub> is included in the fit as interfering species (at 221 K). The

**Operational NO<sub>2</sub>  
retrieval for GOME-2**

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



other interfering species are O<sub>2</sub>–O<sub>2</sub> and H<sub>2</sub>O and their cross-sections are included in the fit as well. The inclusion of these interfering species in the fit reduces the uncertainty in the NO<sub>2</sub> slant column, especially for tropical areas (Lambert and Balis, 2004). Sources for the cross-section data are Greenblatt et al. (1990) for O<sub>2</sub>–O<sub>2</sub> (wavelength axis recalibrated) and HITRAN 2000 (Rothman et al., 2003) for H<sub>2</sub>O (the latter as input to line-by-line computations which are followed by convolution with the GOME-2 FM slit function).

Shift and squeeze parameters may be applied to cross-section wavelength grids to improve wavelength registration against level-1 spectra or compensate for inaccuracies in the wavelength calibration of the cross-section data. Experience with DOAS in the operational UPAS system has shown that fitting of such non-linear parameters on a pixel-by-pixel basis can sometimes lead to numerical instability, and an optimized pre-shift value needs to be applied (Lambert et al., 2002). For GOME-2, an optimized pre-shift value of –0.022 nm is applied to the Flight Model NO<sub>2</sub> cross-sections.

The Ring effect (filling-in of well-modulated solar and absorption features in earth-shine spectra) is due to inelastic rotational Raman scattering (RRS). In DOAS fitting, it is treated as an additional absorber, by means of an additive Ring reference spectrum and associated scaling parameter, as in Eq. (1) above. The “Fraunhofer” Ring spectrum is obtained by folding rotational Raman cross-sections at a fixed temperature with a high-resolution Fraunhofer spectrum taken from the Kitt Peak Observatory (Chance and Spurr, 1997). This does not include a telluric contribution (molecular Ring effect), but for NO<sub>2</sub> the error in the retrieved total column due to the molecular Ring effect is small (1–2%) as compared to the other error sources.

Intensity offset effects that may be induced by residual spectral stray-light, inelastic scattering in the atmosphere and the ocean or remaining calibration issues in the GOME-2 level-1 product are known to be possible sources of bias in DOAS retrievals of minor trace species; to correct for possible offset the inverse of the sun spectrum is fitted as another effective cross-section (Platt and Stutz, 2008).

In the GDP 4.4, the solar spectrum is used as the wavelength reference. Shift and squeeze parameters are fitted for each Earthshine spectrum to compensate for the Doppler shift and changing thermal stress. If necessary, the wavelength calibration of the GOME-2 level-1 spectra can be improved by applying window-dependent pre-shifts to parts of the solar spectrum before each orbit of data is processed. These pre-shifts are established by cross-correlation with a high-resolution solar spectrum (Chance and Spurr, 1997) over limited wavelength ranges covering the fitting window (i.e. 425–450 nm for NO<sub>2</sub> and 758–772 nm covering the oxygen A-band as used in the ROCINN cloud algorithm).

## 4 Air Mass Factor and initial total VCD computations

The second component in the retrieval is the conversion of the NO<sub>2</sub> slant column density into the vertical column density (VCD)  $V$ , using the air mass factor  $M$ :

$$M = \frac{S}{V} \quad (2)$$

The air mass factor depends on the vertical NO<sub>2</sub> profile and a set of forward model parameters  $\mathbf{b}$ , including the GOME-2 viewing geometry, surface albedo, clouds and aerosols. For optically thin absorbers, such as NO<sub>2</sub> in the visible wavelength region, the radiative transfer calculations can be decoupled from the trace gas profile shape (Palmer et al., 2001):

$$M = \frac{\sum_l m_l(\mathbf{b}) x_l c_l}{\sum_l x_l} \quad (3)$$

where  $m_l$  is the air mass factors for the individual layer  $l$  (independent of the NO<sub>2</sub> profile), and  $x_l$  the partial NO<sub>2</sub> column in layer  $l$ . The altitude-dependent air mass factors  $m_l$  are calculated with the LIDORT radiative transfer model (Spurr et al., 2001), as described in Sect. 4.1. The coefficients  $c_l$  are layer-specific correction factors that

account for the temperature dependence of the NO<sub>2</sub> absorption cross-section. This correction factor is a function of the atmospheric temperature in layer *l* and the fixed temperature (243 K) of the NO<sub>2</sub> absorption cross-sections assumed in the DOAS fit (Boersma et al., 2004; Nüß et al., 2006). In the GDP 4.4 algorithm, monthly mean temperatures taken from a run of the MOZART-2 model (see Sect. 5.2) are used to calculate the correction factors for the tropospheric layers.

The initial total VCD is computed under the assumption that the troposphere is not polluted. Therefore, the air mass factor is based on stratospheric NO<sub>2</sub> profiles only, whereas the tropospheric NO<sub>2</sub> amount is assumed to be negligible. This approach is valid over large parts of the Earth, but in areas with significant tropospheric NO<sub>2</sub>, the total column densities are underestimated and need to be corrected, as described in Sect. 5.2.

To incorporate the seasonal and latitudinal variation in stratospheric NO<sub>2</sub> in the air mass factor calculations, a harmonic climatology of stratospheric NO<sub>2</sub> profiles is used (Lambert et al., 2000). This harmonic climatology has been derived from satellite measurements by UARS/HALOE (Gordley et al., 1996) and SPOT-4/POAM-III (Randall et al., 1998) and complementary information from ground-based measurements from the Network for the Detection of Atmospheric Composition Change (NDACC). The stratospheric NO<sub>2</sub> profiles are time dependent and given for 16 latitude bands. Latitude and time of GOME-2 measurements are specified from level-1b geolocation information. In order to avoid jump artefacts associated with discrete latitude classifications, the climatological profiles are interpolated between latitude bands using a linear weighting scheme based on the cosine of the latitude. The resulting NO<sub>2</sub> concentration profile is then integrated to partial columns ( $x_l$ ) on the layer grid of the radiative transfer model. For stratospheric air mass factor calculations, the vertical resolution does not need to be very high, and it is sufficient to use a 13-layer grid based on Umkehr layers.

For GOME-2 scenarios in the presence of clouds, the air mass factor can be determined in conjunction with GOME-2 derived cloud information using a Lambertian

**Operational NO<sub>2</sub>  
retrieval for GOME-2**

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





reflecting boundary cloud model and the independent pixel approximation (IPA):

$$M = (1 - w)M_{\text{clear}} + w M_{\text{cloud}}, \quad (4)$$

where  $M_{\text{clear}}$  is the air mass factor for a completely cloud free pixel,  $M_{\text{cloud}}$  the air mass factor for a completely cloudy pixel, and  $w$  the cloud radiance fraction.  $M_{\text{clear}}$  and  $M_{\text{cloud}}$  are obtained with Eq. (3), with clouds treated as Lambertian Equivalent Reflectors and  $M_{\text{cloud}}$  calculated with  $m_1 = 0$  for all layers below the cloud-top pressure ( $p_c$ ).

The cloud radiance fraction  $w$  is defined as:

$$w = \frac{c_f I_{\text{cloud}}}{(1 - c_f) I_{\text{clear}} + c_f I_{\text{cloud}}}, \quad (5)$$

where  $c_f$  is the cloud fraction,  $I_{\text{clear}}$  and  $I_{\text{cloud}}$  are the backscattered radiances for cloud-free and cloud-covered scenes respectively.  $I_{\text{clear}}$  and  $I_{\text{cloud}}$  are calculated for the mid-point wavelength of the fitting window (437.5 nm) with the LIDORT radiative transfer model, and depend mainly on the surface and cloud albedos and on the GOME-2 viewing geometry.

#### 4.1 Radiative transfer calculations

LIDORT is a multiple scatter multi-layer discrete ordinate radiative transfer code (Spurr et al., 2001). In LIDORT, the atmosphere is assumed to be stratified into a number of optically uniform layers. The LIDORT code used here neglects light polarisation. For DOAS retrievals in the visible wavelength range, the polarisation signature is small and subsumed in the closure polynomial. We use the LIDORT Version 3.3, which possesses corrections for beam attenuation along curved line-of-sight paths, needed for the wide viewing angles of GOME-2 (scan angles in the range 40–50°) (Spurr, 2008).

For DOAS applications with optically thin absorbers, such as NO<sub>2</sub> in the visible wavelength region, the trace gas air mass factor wavelength dependence is weak and therefore it is sufficient to use the mid-point wavelength of the fitting window (437.5 nm).



**Operational NO<sub>2</sub>  
retrieval for GOME-2**

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



LIDORT is a scattering formalism, and requires as input the following optical properties in each layer: (1) total extinction optical thickness, (2) total single scattering albedo, and (3) total phase function scattering coefficients. LIDORT also requires knowledge of the surface reflection (assumed to be Lambertian). In the GDP 4.4, there is an “atmospheric/surface setup module” which deals with detailed radiative transfer physics of molecules, trace gases, aerosols, clouds and surface reflection as needed to create the necessary LIDORT inputs. This setup function is completely decoupled from LIDORT, and this gives the air mass factor computation great flexibility. It is straightforward to change input climatology and other reference atmospheric and surface datasets.

The climatology used for the surface albedo (including mean snow and ice cover) is derived from TOMS and GOME Lambert-equivalent reflectivity (LER) measurements at 380 and 440 nm, as described in Boersma et al. (2004). These monthly averaged surface albedo maps have a spatial resolution of  $1^\circ \times 1.25^\circ$  and represent climatological (monthly) mean situations. The surface albedo for each GOME-2 pixel is determined via area-weighted tessellation of the climatological surface albedo maps and linear interpolation in time to the measurement day.

Changes in surface albedo values will chiefly affect the clear-sky air mass factor  $M_{\text{clear}}$  and the intensity-weighted cloud fraction  $w$ . The effect on the total and tropospheric NO<sub>2</sub> column is largest for cloud-free and partly cloudy scenes; for completely cloud-covered scenes the effect is generally small, since  $M_{\text{clear}}$  plays no part in the total NO<sub>2</sub> column calculations (see Eq. (4) with  $w = 1$ ).

In addition to the albedo, the surface altitude is an important input for the air mass factor calculations, especially in the vicinity of mountainous terrain (Zhou et al., 2009). To determine an accurate pixel-average surface altitude, the high resolution ( $\sim 1 \times 1$  km) topography heights from the global digital elevation model GTOPO30 (<http://lpdaac.usgs.gov/gtopo30/gtopo30.asp>) are averaged over each GOME-2 pixel.

## 4.2 Cloud parameters

In the independent pixel approximation, clouds are regarded as reflecting Lambertian surfaces and cloud information is reduced to the specification of 3 parameters: cloud fraction, cloud-top albedo and cloud-top pressure. In the GDP 4.4, the OCRA and ROCINN algorithms (Loyola et al., 2007) are used for obtaining GOME-2 cloud information: OCRA provides the cloud fraction using the broad-band polarization measurements, and ROCINN provides cloud-top height and cloud-top albedo from measurements in and adjacent to the oxygen A-band around 760 nm. Note that being sensitive to light scattering by clouds, OCRA is also sensitive to scattering by aerosols present in a given GOME-2 scene, so that both effects are subsumed in the retrieved cloud fractions (see also Sect. 6.3)

With GOME-2, several improvements have been incorporated in the cloud algorithms. The initial ROCINN algorithm was based on transmittance-only calculations in the oxygen A-band. ROCINN version 2.0, as used for GOME-2, is based on radiative transfer simulations with Rayleigh scattering and polarization. Another important upgrade for GOME-2 is the ability to distinguish clouds in measurements affected by ocean surface sun-glint, a phenomenon that is common at the edges of the GOME-2 swath. OCRA discriminates clouds in the region affected by sun-glint by analysing the broad-band polarization measurements (Loyola et al., 2011).

## 5 Tropospheric NO<sub>2</sub> column algorithm

In this section, the retrieval of the tropospheric NO<sub>2</sub> column is presented. The tropospheric NO<sub>2</sub> column algorithm for GOME-2 consists of the following steps: (1) estimation of the stratospheric component of the NO<sub>2</sub> column using a spatial filtering approach and (2) the conversion of the residual tropospheric slant column into a tropospheric vertical column, using an accurate tropospheric air mass factor (including the effects

### Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



of clouds). In addition, the initial total NO<sub>2</sub> column is corrected for the tropospheric component under polluted conditions, to provide a more accurate total vertical column. A description of these algorithm steps is given below.

## 5.1 Stratospheric correction

5 The retrieval of the tropospheric NO<sub>2</sub> column starts with the estimation of the stratospheric component from the initial total VCD. Most methods for the stratosphere-troposphere separation reported in the literature are based on the observation that stratospheric NO<sub>2</sub> has a smooth spatial behaviour and that tropospheric contributions occur near source regions on smaller geographic scales. The “Pacific Reference Sector” method has been used in several studies (e.g. Richter and Burrows, 2002; Martin et al., 2002; Beirle et al., 2003) and rests on the assumption of a longitudinally homogeneous stratospheric NO<sub>2</sub> layer and negligible tropospheric NO<sub>2</sub> over the Pacific. The stratospheric NO<sub>2</sub> column can then be derived as the average of the total VCD for each latitude over the longitude band in the Pacific sector. The first assumption is reasonable at lower latitudes, since NO<sub>2</sub> in the stratosphere is mainly determined by day length (photolysis of reservoir species) and only to a lesser degree by transport, ozone concentrations, and temperature. However, longitudinal variations cannot be neglected at higher latitudes, because of the dynamical variability, especially in winter and spring. To reduce the uncertainties involved in the stratosphere-troposphere separation at mid and high-latitudes, a spatial filtering method is employed for GOME-2 in the GDP 4.4. Various spatial filtering methods have been developed that use data not only from the Pacific, but also from other relatively clean areas to determine the stratospheric NO<sub>2</sub> column (Leue et al., 2001; Wenig et al., 2004; Bucsela et al., 2006). The spatial filtering procedure used here works as follows. First a global map is constructed from the initial NO<sub>2</sub> columns by binning the last 24 h of GOME-2 data on a spatial grid of 2.5° latitude × 2.5° longitude. To minimize tropospheric biases in the stratospheric field, a global mask is applied to eliminate areas with potentially high amounts of tropospheric NO<sub>2</sub>. This pollution mask is derived from MOZART-2 model results: the areas in the model

## Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



with monthly mean tropospheric NO<sub>2</sub> columns larger than  $1.0 \times 10^{15}$  molec cm<sup>-2</sup> are masked as polluted (see next section).

After pollution masking, the stratospheric NO<sub>2</sub> column is determined by low-pass filtering the initial NO<sub>2</sub> columns in the zonal direction (30° boxcar filter). This is done in two steps, where first the unmasked measurements with initial total VCD exceeding the (preliminary) stratospheric NO<sub>2</sub> column by more than one standard deviation are identified and excluded from the final analysis. Unmasked polluted measurements can occur when pollution events are missed by the model, for instance during transient pollution events. Finally, the stratospheric NO<sub>2</sub> is interpolated between latitude bands in order to avoid jump artefacts associated with a discrete latitude grid.

A limitation of the spatial filtering approach used here is that it will also take up background NO<sub>2</sub> in the free troposphere with smooth spatial behaviour. In the GDP 4.4, a simple correction is applied for this effect: a fixed background NO<sub>2</sub> column ( $0.1 \times 10^{15}$  molec cm<sup>-2</sup>) is subtracted from the derived vertical stratospheric NO<sub>2</sub> column. This offset for the background NO<sub>2</sub> column has been derived from tropospheric NO<sub>2</sub> fields for unpolluted regions, as provided by the MOZART-2 model.

An example of the stratospheric NO<sub>2</sub> distribution obtained with the spatial filtering approach used in the GDP 4.4 is plotted in Fig. 1. This figure shows the initial total and stratospheric NO<sub>2</sub> columns from GOME-2 for the northern mid- and high-latitudes on 22 February 2008. Clearly visible in this figure are the longitudinal variations in stratospheric NO<sub>2</sub> as captured by the spatial filtering approach. The low values over the north polar area around Greenland and Eastern Canada indicate denoxified air masses inside the polar vortex. Due to dynamical variability, the location of these air masses can vary strongly within a time scale of a few days.

**Operational NO<sub>2</sub>  
retrieval for GOME-2**

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## 5.2 Tropospheric Air Mass Factor and VCD computation

After the stratosphere-troposphere separation, the tropospheric VCD can be determined via the relation:

$$V_t = \frac{S - M_s V_s}{M_t} \quad (6)$$

5 where  $S$  is the slant column density calculated in the DOAS fit and  $V_s$  is the stratospheric component, as calculated with the spatial filtering method described above.  $M_s$  is the stratospheric air mass factor, used for the calculation of the initial total VCD, as described in Sect. 4.  $M_t$  is a tropospheric air mass factor calculated with Eqs. (3) and (4), using an a priori tropospheric  $\text{NO}_2$  profile. The tropospheric air mass factor  
10 depends on the same forward model parameters as the stratospheric air mass factor (i.e. GOME-2 viewing geometry, surface albedo, clouds and aerosols). However, the dependence on the surface albedo, clouds and aerosols, as well as the a priori  $\text{NO}_2$  profile is much stronger for the tropospheric air mass factor. The variability in the tropospheric air mass factor is illustrated in Fig. 2. where a global map of the monthly  
15 averaged air mass factor is shown for March 2008. Small tropospheric air mass factors between 0.5 and 1.0 are found over polluted source regions, such as Europe, the Eastern US and Southeast Asia. Over unpolluted regions, like the oceans or the Sahara, and over snow covered areas the air mass factor is larger than 2.0.

After the calculation of the tropospheric column, a corrected total VCD  $V_c$  can be  
20 calculated via the relation:

$$V_c = V_s + V_t \quad (7)$$

In the GDP 4.4, a corrected total VCD is determined for all GOME-2 observations where the initial total VCD exceeds the estimated stratospheric component  $V_s$ .

25 The a priori  $\text{NO}_2$  profiles used in GDP 4.4 are obtained from a run of the global chemistry transport model MOZART version 2 (Horowitz et al., 2003). The model data has a horizontal resolution of  $1.875^\circ$  latitude by  $1.875^\circ$  longitude (T63), with 32

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



terrain-following hybrid layers extending from the surface up to  $\sim 3$  hPa. The number of layers in the troposphere varies from 10 to 16, depending on tropopause height, with about 4 layers in the boundary layer. For the tropospheric air mass factor computation in the GDP 4.4, monthly average profiles at the satellite overpass time have been determined, using MOZART-2 data from the year 1997 (Nüß et al., 2006). While this climatology will capture seasonal and spatial patterns and provides a good first guess of the atmospheric NO<sub>2</sub> profile, daily data from an online model run can capture short-term variability induced by meteorology and would therefore be preferable (Boersma et al., 2004). This option is currently under consideration for future versions of the GDP.

The calculation of the tropospheric VCD is complicated in case of (partly) cloudy conditions. For many measurements over cloudy scenes, the cloud-top is well above the NO<sub>2</sub> pollution in the boundary layer, and when the clouds are optical thick, the enhanced tropospheric NO<sub>2</sub> concentrations cannot be detected by GOME-2. Therefore, the tropospheric VCD is only calculated for GOME-2 observations with a cloud radiance fraction  $w < 50\%$ . Note that the “below cloud amount” (i.e. the amount of NO<sub>2</sub> that is inferred to be below the cloud top) for these partly cloudy conditions is implicitly accounted for via the cloudy air mass factor  $M_{\text{cloud}}$  (in which  $m_1 = 0$  for all layers below the cloud-top). As this procedure assumes knowledge of the vertical NO<sub>2</sub> profile (taken from the model) and neglects any possible differences of this profile in the cloudy and cloud-free part, cloudy scenes will have higher uncertainty than clear sky observations.

### 5.3 Examples of GOME-2 tropospheric NO<sub>2</sub>

Figure 3 (left) shows the yearly averaged tropospheric NO<sub>2</sub> column from GOME-2 for 2007 over Europe, as retrieved with the GDP 4.4 algorithm described above. For comparison, the GDP 4.4 algorithm has been applied to GOME/ERS-2 data as well, and the results for 2000 are shown in Fig. 3 (right) (due to limited spatial coverage of the GOME measurements after June 2003, the tropospheric NO<sub>2</sub> algorithm cannot be applied on more recent GOME measurements). Both panels show the high tropospheric NO<sub>2</sub> concentrations above large urban and industrial areas of Europe, such as the Po

Valley, the Benelux, South-East England and Germany's Ruhr area. However, the better spatial coverage and resolution of GOME-2 compared to GOME/ERS-2 results in more spatial detail in the tropospheric NO<sub>2</sub> field (for the region shown in Fig. 3, there are about  $6.5 \times 10^5$  GOME-2 measurements with a cloud radiance fraction <50% available, while the number of GOME/ERS-2 measurements is only  $\sim 5 \times 10^4$ ). The increase in spatial detail is clearly visible in, for example, the "city-size" polluted areas around Paris, Madrid and Moscow, which are much better resolved in the GOME-2 panel.

Differences between GOME and GOME-2 tropospheric NO<sub>2</sub> fields are expected for several reasons. Better spatial resolution leads to larger NO<sub>2</sub> columns over pollution hot-spots. Changes in NO<sub>x</sub> emissions are expected to lead to lower NO<sub>2</sub> values in some countries while increases have been reported for others (Konovalov et al., 2010). When comparing GOME-2 and GOME/ERS-2 measurements, the diurnal variation in tropospheric NO<sub>2</sub> should be taken into consideration as well. There is a one hour difference between the local overpass time of GOME-2 ( $\sim 09:30$  LT) and GOME/ERS-2 ( $\sim 10:30$  LT). For the polluted regions in the European area, 5–10% larger tropospheric NO<sub>2</sub> columns are expected at the earlier GOME-2 overpass time than at the GOME/ERS-2 overpass time (Boersma et al., 2008).

Figure 4 (left) shows the averaged tropospheric NO<sub>2</sub> columns from GOME-2 for the period 2007–2009 over East Asia. The world's largest area with high NO<sub>2</sub> pollution is found above east China, which is a result of China's spectacular economic growth during the last decade, accompanied by a strong increase in emissions of air pollutants. Another remarkable feature visible in the Fig. 4 is the enhanced tropospheric NO<sub>2</sub> along shipping lanes in the Bay of Bengal and the South China Sea (e.g. see Beirle et al., 2004; Franke et al., 2009; Richter et al., 2004, 2011). For comparison, the results for GOME/ERS-2 for the period 1997–1999 are also shown in Fig. 4 (right). The increased spatial detail of GOME-2 compared to GOME/ERS-2 is clearly visible. For example, the polluted areas around the large cities in Northern India, and around Bangkok are much better resolved in the GOME-2 panel. Also clearly visible is the increase in tropospheric NO<sub>2</sub> over Eastern China during the ten year period that lies between the two pictures,

**Operational NO<sub>2</sub>  
retrieval for GOME-2**

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





as a result of strong increased in NO<sub>2</sub> emissions (Richter et al., 2005; Van der A et al., 2008).

## 6 Error and sensitivity analyses

Referring to Eq. (6), the overall error on the tropospheric NO<sub>2</sub> column  $V_t$  (denoted as  $\sigma_{Vt}$ ) can be expressed as a function of the error on the slant column density  $S$ , the stratospheric column  $V_s$ , and the stratospheric and tropospheric air mass factors  $M_s$  and  $M_t$ :

$$\begin{aligned}\sigma_{Vt}^2 &= \left(\frac{\partial V_t}{\partial S}\right)^2 \sigma_S^2 + \left(\frac{\partial V_t}{\partial V_s}\right)^2 \sigma_{Vs}^2 + \left(\frac{\partial V_t}{\partial M_s}\right)^2 \sigma_{Ms}^2 + \left(\frac{\partial V_t}{\partial M_t}\right)^2 \sigma_{Mt}^2 \\ &= \left(\frac{1}{M_t}\right)^2 \sigma_S^2 + \left(\frac{M_s}{M_t}\right)^2 \sigma_{Vs}^2 + \left(\frac{V_s}{M_t}\right)^2 \sigma_{Ms}^2 + \left(\frac{S - M_s V_s}{M_t^2}\right)^2 \sigma_{Mt}^2\end{aligned}\quad (8)$$

This assumes that the errors are independent and random and therefore can be treated with standard error propagation. While this is not always the case and systematic errors also contribute to the overall uncertainty, this approach will provide a rough estimate of the uncertainties to be expected. In the following sections, the error components from the slant column density  $\sigma_S$ , the stratospheric column  $\sigma_{Vs}$ , and the air mass factors  $\sigma_{Ms}$  and  $\sigma_{Mt}$  are discussed in more detail.

### 6.1 Uncertainty in the slant column density

The precision of the NO<sub>2</sub> slant column densities is derived from a statistical analysis of the GOME-2 measurements in the clean tropical Pacific region (20° S–20° N; 160°–180° E). This region is divided into small boxes (2° × 2°), and from the variation of the NO<sub>2</sub> columns within each box, an estimate of the slant column precision can be made. (Note that the variability of the air mass factors within the boxes is small (<0.2%), and is taken into account by scaling the slant columns with an appropriate geometrical air mass factor). The analysis is based on the assumption that the variation in the total



Operational NO<sub>2</sub>  
retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



NO<sub>2</sub> columns in each box is a result of errors in the slant column only, originating from (random) instrument measurement noise. The deviation of each GOME-2 measurement from the corresponding box mean value is calculated on a daily basis. The slant column error is then derived from the distribution of the slant column deviations, as shown in Fig. 5 for April 2007. The distribution has a Gaussian shape and from the width of the Gaussian, an average slant column error  $\sigma_S$  of  $0.45 \times 10^{15}$  molec cm<sup>-2</sup> is estimated for GOME-2 ( $\sigma_S \approx 0.42 \times \text{FWHM}$ ). Since the instrument noise is mainly a result of the photoelectron shot noise, the slant column error depends significantly on the cloud fraction and surface albedo. For example, the average slant column error for GOME-2 measurements with cloud fraction <50% is  $0.56 \times 10^{15}$  molec cm<sup>-2</sup>, while for measurements with cloud fraction >50% the average slant column error is  $0.38 \times 10^{15}$  molec cm<sup>-2</sup>.

The GOME-2 slant column error is larger than the  $0.35 \times 10^{15}$  molec cm<sup>-2</sup> derived for GOME/ERS-2 using the same method. This is consistent with the higher signal-to-noise ratio and larger ground pixel size of the GOME/ERS-2 measurements. Boersma et al. (2007) report a slant column error of  $0.67 \times 10^{15}$  molec cm<sup>-2</sup> for the OMI NO<sub>2</sub> measurements, consistent with the smaller signal-to-noise ratio of the OMI instrument which has better spatial resolution.

The instrument degradation of the GOME-2 sensor in the visible wavelength range of Channel 3 (Lang et al., 2009; Dikty et al., 2010) has an impact on the derived slant column errors, as shown in Fig. 6. In the four years from the start of the operational GOME-2 measurements in January 2007, the GOME-2 slant column error for NO<sub>2</sub> has increase by about 35%. Here, it should be noted that during the more than 10 years of GOME/ERS-2 operations, no significant degradation has been detected for the visible wavelength region in Channel 3 (Coldewey-Egbers et al., 2008), and therefore the estimated slant column error for the GOME NO<sub>2</sub> measurements remains relatively constant ( $\sim 0.35 \times 10^{15}$  molec cm<sup>-2</sup>).

The spectral structures in the BSDF of the ground aluminium diffuser plate used for the direct sun measurements with GOME/ERS-2 result in spectral features in the

**Operational NO<sub>2</sub>  
retrieval for GOME-2**

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



differential spectra that correlate with the NO<sub>2</sub> absorption features (Richter and Wagner, 2001; Wenig et al., 2004). This severely affects the DOAS retrieval, resulting in a systematic error of up to 100% in the GOME NO<sub>2</sub> columns in the tropics. This error appears as a seasonal pattern in the NO<sub>2</sub> time-series, which is related to the annual cycle in the elevation and azimuth angles of the incoming solar radiation (see Fig. 7 (top)). The quartz quasi-volume diffuser used in the GOME-2 instrument has strongly reduced differential structures in the BSDF and therefore, the induced spectral features in the GOME-2 solar spectra are much smaller. This is clearly visible in the NO<sub>2</sub> time-series for the tropics, as shown in Fig. 7 (bottom). The seasonal patterns in the NO<sub>2</sub> time-series from GOME-2 are more than five times smaller than for GOME/ERS-2 (see also Richter et al., 2011).

Another source of systematic error in the NO<sub>2</sub> slant column is the uncertainty in the laboratory absorption cross-section of NO<sub>2</sub>. As described in Sect. 3, the GDP 4.4 algorithm uses the GOME-2 FM cross-sections. Comparisons of the GOME-2 FM cross-sections with the Vandaele et al. (1998) NO<sub>2</sub> cross-sections at 294K (convolved with the GOME-2 slit function) show a good agreement, with a mean deviation of ~2% (Gür et al., 2005; De Smedt, personal communication, 2009). This is consistent with earlier comparisons between various laboratory measurements of NO<sub>2</sub> cross section spectra by Vandaele et al. (1998), which showed mutual agreement of 2% as well. As discussed in Sects. 3 and 4, the temperature dependence of the NO<sub>2</sub> cross-sections is a potential source of systematic error in the NO<sub>2</sub> slant columns, and an a-posteriori correction for the difference between the atmospheric temperature and the cross-section temperature used in the DOAS-fit (243 K) is applied in the GDP 4.4 algorithm. Neglecting the atmospheric temperature variations and assuming a fixed NO<sub>2</sub> temperature of 243 K would result in large systematic errors up to 20% (Boersma et al., 2004; Nüß et al., 2006). Assuming a difference between model and real atmospheric temperature of 5K at the altitude of NO<sub>2</sub> absorption, an additional uncertainty of 2% in the tropospheric NO<sub>2</sub> columns results.

## 6.2 Uncertainties in the stratospheric air mass factor and stratospheric column

The stratospheric air mass factor, as used for the initial total VCD calculation, depends mainly on the viewing geometry. The variation in the stratospheric air mass factor due to the NO<sub>2</sub> profile shape, the albedo and cloud cover is ~2–3% for solar zenith angles smaller than ~80°. For higher solar zenith angles, the variation increases up to 5%. In the GDP 4.4, the dependence on the stratospheric NO<sub>2</sub> profile shape, the albedo and cloud cover is taken into account in the calculation of the stratospheric air mass factor (see Sect. 4). A conservative estimate of the uncertainty in the stratospheric air mass factor of 2% is assumed in this study.

In Boersma et al. (2004), the uncertainty in the stratospheric column calculation for GOME/ERS-2 has been estimated for various separation methods. For the reference sector and spatial filtering methods, they found uncertainties of  $0.2\text{--}0.45 \times 10^{15} \text{ molec cm}^{-2}$ . To analyse the uncertainty in the GOME-2 stratospheric column, the spatial filtering method used in the GDP 4.4 has been applied to one year (2004) of reanalyses model data from the IFS-MOZART assimilation system (Flemming et al., 2009) as provided by the Monitoring Atmospheric Composition and Climate (MACC) project (Hollingsworth et al., 2008). To that end, synthetic slant columns for the locations of all GOME-2 measurements were derived from vertical columns given by the IFS-MOZART model on a  $1.125^\circ \times 1.125^\circ$  latitude-longitude grid and processed with the GDP 4.4 algorithm. The differences between the retrieved and original stratospheric columns were then analysed to estimate the uncertainty in the GOME-2 stratospheric columns. Figure 8 shows an example of the stratospheric NO<sub>2</sub> field for 15 March, as derived with the GDP 4.4 using the synthetic slant columns, and the stratospheric NO<sub>2</sub> fields from the IFS-MOZART model for the same day. This figure shows a general good agreement between the GDP 4.4 and IFS-MOZART stratospheric columns with a typical stratospheric NO<sub>2</sub> distribution for March, i.e. relatively low stratospheric columns in the tropics ( $\sim 1.5 \times 10^{15} \text{ molec cm}^{-2}$ ), and higher columns at mid- and high-latitudes ( $2\text{--}3 \times 10^{15} \text{ molec cm}^{-2}$ ). The stratospheric columns retrieved with

### Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Operational NO<sub>2</sub>  
retrieval for GOME-2**

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



the GDP 4.4 also show most of the broad longitudinal variations visible in the IFS-MOZART model, for example in the tropical region with a minimum in the stratospheric columns over the Pacific ( $\sim 1.3 \times 10^{15}$  molec cm<sup>-2</sup>), and between 30–45° N with local enhancements over the Euro-Asia continent. However, the small-scale variations visible in the IFS-MOZART stratospheric NO<sub>2</sub> field cannot be captured with the GDP 4.4, which is an inherent limitation of the spatial filtering approach.

The uncertainty in the GOME-2 stratospheric NO<sub>2</sub> column has been estimated by calculating the mean absolute difference between the stratospheric columns retrieved with the GDP 4.4 and the original stratospheric columns from the IFS-MOZART model (using one year of model data). For the low-latitudes and southern hemisphere, the estimated uncertainty in the stratospheric column is  $\sim 0.15 \times 10^{15}$  molec cm<sup>-2</sup>. For the polluted northern latitudes (20–60° N), the estimated uncertainty is larger:  $\sim 0.26 \times 10^{15}$  molec cm<sup>-2</sup>. These values are consistent with previous estimates for GOME/ERS-2 as described above, and with estimates for the OMI NO<sub>2</sub> product ( $\sim 0.2 \times 10^{15}$  molec cm<sup>-2</sup>, see Bucsela et al., 2006).

### 6.3 Uncertainty in the tropospheric air mass factor

As described in Sect. 5.2, the tropospheric air mass factor depends mainly on the surface albedo, the cloud fraction and cloud-top pressure, as well as the a priori NO<sub>2</sub> profile shape. The uncertainty in the air mass factor due to the uncertainty in the surface albedo and cloud parameters was estimated in a sensitivity study on one year (2008) of GOME-2 data, by linearizing the air mass factor and varying the value for these forward model parameters around the optimal value used in the retrieval.

The uncertainty for the TOMS/GOME surface albedo used in the GDP 4.4 algorithm is assumed to be 0.02 (Boersma et al., 2004). This estimate is in agreement with recent comparisons between TOMS, GOME and OMI LER data, and MODIS black sky albedo, which show average differences smaller than 0.02 between the various surface albedo data-sets (Kleinpools et al., 2008). Using this value in the sensitivity study on one

year of GOME-2 data, the average uncertainty in the air mass factor due to the surface albedo uncertainty was found to be  $\sim 14\%$  for polluted conditions with maximum values of up to  $30\%$ . For clean and moderately polluted conditions, the derived uncertainty in the air mass factor is smaller ( $\sim 6\%$ ).

The uncertainty in the air mass factor due to the uncertainty in the cloud fraction and cloud-top pressure has been estimated using GOME-2 measurements with cloud radiance fraction  $< 50\%$  only. For small cloud fractions, the estimated uncertainty in the OCRA cloud fraction is  $\sim 0.06$  (Loyola et al., 2007). The effect of this uncertainty on the tropospheric columns depends mainly on the tropospheric  $\text{NO}_2$  load, the surface albedo and cloud-top pressure. The effect of uncertainties in the cloud-top pressure is largest at altitudes with the largest  $\text{NO}_2$  concentrations, which is usually within the boundary layer. For clouds in the lower troposphere (cloud top pressure  $> 700$  hPa), the uncertainty in the ROCINN cloud-top pressures is  $\sim 40$  hPa (Loyola et al., 2011).

From the sensitivity study on one year of GOME-2 data, the derived uncertainty in the tropospheric air mass factor due to the cloud fraction uncertainty is in the  $5\text{--}50\%$  range, with an average uncertainty of  $\sim 25\%$  for polluted regions. Figure 9 shows the average error in the tropospheric air mass factor for March 2008 due to uncertainty in the GOME-2 cloud fraction. Clearly visible are the relatively large uncertainties over continental source regions with high tropospheric  $\text{NO}_2$  columns. Since the ROCINN cloud-top pressures are usually above the  $\text{NO}_2$  pollution layer, the average uncertainty in the tropospheric air mass factor due to the cloud top pressure uncertainty is relatively small ( $\sim 3\%$ ). For individual GOME-2 measurements, the uncertainty can be  $\sim 30\%$  or larger, when the cloud is located inside the  $\text{NO}_2$  pollution layer.

The  $\text{NO}_2$  profile shape assumed for the airmass factor calculation also has a large effect on the airmass factor, in particular over dark surfaces where the measurement sensitivity decreases towards the surface. Boersma et al. (2004) evaluated this uncertainty by analysing the variability in the  $\text{NO}_2$  profiles from the TM3 chemistry-transport model that they used as a priori, and estimated an uncertainty of  $10\%$  on average but much larger values locally. In the GDP 4.4, monthly averages of MOZART profiles for

**Operational  $\text{NO}_2$   
retrieval for GOME-2**

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



one year (1997) are applied which introduces additional uncertainties as the effects of day-to-day variations in meteorology are not included. In order to quantify this source of uncertainty, daily and monthly airmass factors were calculated for two months of MOZART model data (January and July) (Nüß et al., 2006). The relative RMS of the daily values was between 5% and 10% for most locations with maxima of up to 20%. From this result, the uncertainty introduced by the use of monthly airmass factors is estimated to be 10%. The larger value was selected to account for the possibility of additional inter-annual changes, e.g. from changes in NO<sub>x</sub> emission strengths or transport patterns.

Table 2 summarizes the estimated uncertainties in the tropospheric air mass factor for polluted conditions ( $V_1 > 1.0 \times 10^{15}$  molec cm $^{-2}$ ). The total uncertainty in the tropospheric air mass factor is mostly in the 15–50% range, with an average uncertainty of ~33%. The total uncertainty in the tropospheric NO $_2$  column can be estimated with Eq. (8), and range from 40 to 80% for polluted conditions (see Table 3).

Aerosols are not included in the radiative transfer calculations performed in the GDP 4.4 retrieval. This is similar to the situation in most other retrievals (e.g. Bucsela et al., 2006; Boersma et al., 2007) and introduces uncertainties in the calculations of air mass factors and cloud properties. Leitão et al. (2010) have investigated the effect of aerosols on air mass factors in clear-sky scenarios for both idealised and more realistic scenarios. They found largely varying effects ranging from just a few percent up to a factor of 2 of reductions and enhancements of the air mass factors depending on AOD, single scattering albedo and relative vertical position of NO<sub>2</sub> and aerosol. However, the cloud treatment will provide some implicit correction of the aerosol effects as discussed in Boersma et al. (2004). In fact, in a recent study, Boersma et al. (2011) show that in very clear situations over the Eastern US, the OMI cloud fraction is well correlated to aerosol optical thickness retrieved from MODIS and that the implicit correction of the air mass factor is similar to that derived from an explicit radiative transfer calculation. However, it is not clear if this finding also holds for partly cloudy scenes and other regions of the world. A full treatment of aerosols in the radiative transfer will only be

possible if clouds and aerosols are represented as scattering layers (as e.g. in Martin et al., 2002) and if detailed information on aerosol optical properties is available.

## 7 Ground-based validation methodology

Ground-based Multi-Axis MAXDOAS instruments operated at many stations around the world provide tropospheric and stratospheric NO<sub>2</sub> columns using retrievals similar to those applied to satellite data. However, significant differences in sensitivity between satellite and ground-based NO<sub>2</sub> measurement systems exist and should be carefully taken into account when dealing with the validation of NO<sub>2</sub> columns derived from GOME-2. Atmospheric NO<sub>2</sub> exhibits large natural structures and cycles, among which are a vertical profile and a geographical distribution varying drastically with latitude and with the presence of emissions, and a diurnal cycle of photochemical origin. Differences in sampling and smoothing of these structures and cycles can hamper the comparison of GOME-2 and correlative observations if not taken into account properly. While the validation of stratospheric NO<sub>2</sub> columns derived from satellite can count on past experience based on an extended ground-based network of zenith sky light (ZSL) DOAS instruments covering different ranges of NO<sub>2</sub> values as part of the Network for the Detection of Atmospheric Composition Change (NDACC, Kurylo and Zander, 2000), the validation of tropospheric NO<sub>2</sub> data is still a matter of research. The development of appropriate instruments and validation methodologies remains an objective for field intercomparison campaigns, like the DANDELIONS and the CINDI campaigns (Brinksma et al., 2008; Roscoe et al., 2010).

As mentioned in Sect. 5, the retrieval of tropospheric NO<sub>2</sub> from satellite measurements is a complex process based on a chain of individual retrieval modules, each one relying on a set of assumptions. For this reason, a reliable validation of the final product should assign a validity indicator to each critical individual component of the level-1-to-2 retrieval chain. As a consequence, the validation of GOME-2 NO<sub>2</sub> columns in the O3M-SAF context has been set up as an end-to-end approach (Lambert et al., 2008),

### Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





consisting in the validation of each component of the retrieval, as recommended by Reference Protocols and Guidelines (CEOS 2004, Lambert et al., 2009). This approach is essential in that it allows hidden compensating errors to be unravelled. The end-to-end validation approach adopted for GOME-2 NO<sub>2</sub> data successively addresses the validity of: (a) DOAS analysis results, cloud properties retrievals and AMF calculations, (b) the stratospheric reference and (c) tropospheric NO<sub>2</sub> column data. An illustration of such a three level validation strategy is presented hereafter, based on measurement results obtained at the Observatoire de Haute Provence (OHP, 44° N, 5.7° E) station from March 2007 to July 2010. In a forthcoming paper, this kind of exercise will be extended including more ground-based measurements, covering different pollution levels.

## 8 End-to-end GOME-2 NO<sub>2</sub> validation at the OHP station

To illustrate the end-to-end validation of the operational GOME-2 NO<sub>2</sub> data product as retrieved with the GDP 4.4, a complete set of correlative observations available at the OHP station is used. Slant, total, stratospheric and tropospheric columns are separately assessed as to their “validity” in comparison to ground-based MAXDOAS observations as well as other satellite datasets, such as the operational NO<sub>2</sub> product from GOME/ERS-2 (Lambert and Balis, 2004) and the TEMIS NO<sub>2</sub> products from SCIAMACHY (Blond et al., 2007; Boersma et al., 2007).

### 8.1 Ground-based MAXDOAS observations

The Belgian Institute for Space Aeronomy (BIRA-IASB) has been operating a MAXDOAS instrument at the NDACC station OHP since 2005 and in the framework of the O3M-SAF, these data have been used to test and set up a method for the validation of the operational GOME-2 NO<sub>2</sub> product (Lambert et al., 2008). Although it is largely rural, OHP can occasionally be influenced by polluted air masses transported from neighbouring cities, hence providing interesting test cases for GOME-2 sensitivity to

## Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





tropospheric NO<sub>2</sub>. The MAXDOAS technique has been developed as an extension of the ZSL-DOAS technique, for the determination of vertically resolved abundances of atmospheric trace species in the lowermost troposphere (Hönninger et al., 2004; Wagner et al., 2004; Wittrock et al., 2004; Heckel et al., 2005). MAXDOAS instruments collect scattered sky light in a series of line-of-sight (LOS) angular directions extending from the horizon to the zenith. High sensitivity towards absorbers present near the surface is obtained for the smallest elevation angles, while measurements at higher elevation provide information on the rest of the column. In this way, a separation between near-surface concentration, tropospheric column and stratospheric column can be obtained. The MAXDOAS instrument at OHP is based on a grating spectrometer covering the 330–390 nm range and NO<sub>2</sub> differential slant column densities (DSCDs) are retrieved in the 364–384 nm wavelength interval range using the DOAS technique. More information on the instrument and the retrieval settings can be found in Lambert et al. (2008) and Pinardi et al. (2008).

## 8.2 Slant columns

Figure 10 shows time series and scatter plot of monthly averaged normalized slant columns measured from January 2007 until July 2010 within 300 km around OHP by different satellites: GOME (GDP 4.1), SCIAMACHY (TEMIS) and GOME-2 (GDP 4.4). The slant columns have been normalised by their geometrical air mass factor in order to consider the different angular dependencies of the three satellite instruments. Error bars represent the variability (one sigma standard deviation) in the measurements. Note the significant increase of the standard deviations for all instruments during the winter periods, which can be explained by a larger contribution from tropospheric pollution events and the time difference between the different satellite overpasses.

Considering the combined variability in the slant column data and the remaining instrumental differences (such as the different degradation of the three instruments and the effect of the diffuser plate anomaly which introduces time-dependent

## Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



pseudo-random variations in the measured columns, especially for GOME/ERS-2), one can conclude that all satellite data sets agree within their uncertainties.

### 8.3 Total and stratospheric columns

Figure 11 shows the initial total VCD (i.e. computed with stratospheric air mass factors, and thus “uncorrected” for tropospheric pollution) of GOME-2, and the corresponding stratospheric column  $V_s$  above OHP, for different cloud selections. All GOME-2 measurements within 300 km of OHP between March 2007 and July 2010 are selected and binned according to cloud fractions. The left panel corresponds to a selection of high clouds (cloud top pressure (CTP) smaller than 400hPa) while the right panel corresponds to lower clouds (CTP larger than 400hPa). As expected, the stratospheric content is similar in both plots, while the total (uncorrected) VCD strongly depends on the bulk altitude of the clouds. High clouds effectively mask the signal from surface  $\text{NO}_2$  while in case of low-lying clouds, the satellite observations remain sensitive to a part of the tropospheric  $\text{NO}_2$  column, even for fully cloudy pixels

For the validation of satellite  $\text{NO}_2$  columns, twilight sunrise zenith sky light (ZSL) data from SAOZ and DOAS-like instruments, mostly sensitive to stratospheric  $\text{NO}_2$ , have been used in past studies (Lambert et al., 2004; Ionov et al., 2008; Celarier et al., 2008). Stratospheric  $\text{NO}_2$  columns presented here are derived from zenith-sky measurements performed at sunrise between  $87^\circ$ – $91^\circ$  SZA by the BIRA MAXDOAS instrument at OHP. Zenith-sky AMFs are accurately determined using a-priori climatological stratospheric  $\text{NO}_2$  profiles (Lambert et al., 2000) similar to those used in the satellite evaluations. GOME-2 data are selected at the intersection with ZSL-DOAS air masses, according to the procedure described in Balis et al. (2007). This approach allows the reduction of horizontal smoothing uncertainties by matching the optical air masses extensions.  $\text{NO}_2$  cross-sections used for retrieving the columns are at same temperature (243 K) as used for the satellite data.

Figure 12 shows the comparison between time-series of  $\text{NO}_2$  stratospheric column data from GOME-2 (GDP 4.4), SCIAMACHY (TEMIS) and coincident ground-based

## Operational $\text{NO}_2$ retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Operational NO<sub>2</sub>  
retrieval for GOME-2**

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



ZSL measurements performed at OHP between March 2007 and July 2010. The GOME-2 stratospheric columns as derived with the GDP 4.4 are found to be in good overall agreement with the other datasets showing a good correlation and a similar seasonal variation. The mean differences are less than  $2.5 \times 10^{14}$  molec cm<sup>-2</sup> and less than  $6.2 \times 10^{13}$  molec cm<sup>-2</sup> respectively compared to the ground-based data and to the SCIAMACHY dataset. The difference between the two satellites datasets is mainly related to the different approach used to infer the stratospheric correction (spatial filtering versus assimilation) and is coherent with the estimated uncertainty for the stratospheric columns in Sect. 6.2. These results are in line with those reported in a previous study using the SAOZ network as a source of correlative data (Ionov et al., 2008).

**8.4 Tropospheric columns**

The GOME-2 tropospheric NO<sub>2</sub> column has been compared to ground-based MAXDOAS measurements performed at OHP. Here, tropospheric vertical columns are obtained from MAXDOAS differential SCD measurements considering a geometrical approximation for the tropospheric AMF, as described e.g. in Brinksma et al. (2008), Pinardi et al. (2008), and Lambert et al. (2008). Figure 13 shows a time-series of the ground-based MAXDOAS and the GOME-2 tropospheric NO<sub>2</sub> columns over OHP from June 2007 to March 2010. Comparison datasets are selected by taking the daily mean value of all GOME-2 measurements flagged “cloud-free” within 100 km around OHP, and by interpolating the ground-based data at the satellite overpass time. Alternatively ground-based data can be averaged within a prescribed time window around the satellite overpass time, and compared to the spatially closest satellite pixel. Both comparison approaches are found to give similar results with no significant statistical differences (Lambert et al., 2008).

One can see in Fig. 13 that pollution episodes are well captured by GOME-2, although the scatter of the individual comparison points is relatively large. These results are qualitatively similar to those obtained in previous validation exercises (e.g. Brinksma et al., 2008; Celarier et al., 2008; Irie et al., 2008). However, in our study

## Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



a much longer comparison dataset is available (2.5 years) and averaging can be performed, in order to limit the influence of temporal and spatial sensitivity differences on the comparison. As can be seen, monthly mean values are in very good agreement, the seasonal variations in tropospheric NO<sub>2</sub> column being similarly captured by both observation systems. The differences are generally within  $0.5 \times 10^{15}$  molec cm<sup>-2</sup>, with maximum differences around  $2 \times 10^{15}$  molec cm<sup>-2</sup>, as for December 2008 and January 2009. During the latter period, however, the comparison was hampered by a lack of ground-based measurement points due to an instrumental deficiency. The scatter plot of the monthly mean values is shown in Fig. 14, as well as the line of a linear regression fit. A correlation coefficient of 0.82 and a regression slope of 0.96 are derived when comparing GOME-2 to the MAXDOAS columns.

The GOME-2 tropospheric columns have also been compared to the SCIAMACHY satellite datasets in Fig. 15. In this case, the monthly means of all satellite measurements flagged “cloud-free” within 100 km around OHP are considered. No ground-based data is included in this comparison as for each satellite dataset a different selection of coincident ground-based data would have to be performed. GOME-2 tropospheric NO<sub>2</sub> columns are found to agree well with SCIAMACHY (TEMIS product), considering their combined uncertainties and the large tropospheric NO<sub>2</sub> variability. The temporal variations are similar, and the corresponding scatter plot presents a correlation of 0.78 and a slope of 0.72.

In conclusion, the operational GOME-2 NO<sub>2</sub> column product shows a high level of consistency with correlative observations available at the OHP station. The slant, stratospheric and tropospheric columns derived with the GDP 4.4 agree well with the other datasets. Note that validation results of the operational GOME-2 NO<sub>2</sub> product, addressing both the stratospheric and tropospheric columns are regularly updated within the O3M-SAF, confirming a good global agreement with ground-based correlative data sets (Lambert et al., 2008). More detailed comparisons will be addressed in a forthcoming NO<sub>2</sub> validation paper, including results from the whole NDACC network and more MAXDOAS stations.

## 9 Summary and conclusions

In this paper, we have described the operational total and tropospheric NO<sub>2</sub> retrieval algorithms for GOME-2, as implemented in the GOME Data Processor (GDP) version 4.4. The DOAS method is used to determine NO<sub>2</sub> slant column densities from calibrated GOME-2 (ir)radiance data in the 425–450 nm wavelength range. Initial total VCDs are computed using an air mass factor based on a stratospheric NO<sub>2</sub> profile climatology. In the GDP 4.4, a spatial filtering method is used to obtain the stratospheric NO<sub>2</sub> component from the initial total VCD. This method has been shown to be an improvement on the Pacific reference sector method, which rests on the assumption of a longitudinally homogeneous stratospheric NO<sub>2</sub> layer. For the tropospheric air mass factor computation, monthly average NO<sub>2</sub> profiles from the MOZART-2 chemistry transport model are used, determined for the satellite overpass time. GOME-2 derived cloud properties, determined with the OCRA and ROCINN algorithms are used to calculate the air mass factors for scenarios in the presence of clouds. Example applications of the GOME-2 tropospheric NO<sub>2</sub> columns show the increased spatial detail compared to its predecessor GOME/ERS-2, which is results of the better spatial coverage and resolution of the GOME-2 instrument.

A statistical approach has been used to estimate the uncertainty in the GOME-2 slant columns. We find that the random error in the slant column is approximately  $0.45 \times 10^{15} \text{ molec cm}^{-2}$ . In the four years from the start of GOME-2 measurements in 2007, the slant column error has increased by about 35% due to the instrument degradation of the GOME-2 sensor. As a result of the improved quartz diffuser plate used in the GOME-2 instrument, the systematic error in the slant columns is strongly reduced compared to GOME/ERS-2. The estimated uncertainty in the stratospheric NO<sub>2</sub> column determined with the spatial filtering method is in the  $0.15\text{--}0.3 \times 10^{15} \text{ molec cm}^{-2}$  range. The most important uncertainties associated with the computation of the tropospheric air mass factor are cloud fraction, surface albedo and the a priori NO<sub>2</sub> profile. The estimated uncertainty in the tropospheric NO<sub>2</sub> column for polluted conditions ranges from 40 to 80%.

AMTD

4, 1617–1676, 2011

### Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Operational NO<sub>2</sub>  
retrieval for GOME-2**

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



We have also presented an end-to end ground-based validation approach for the GOME-2 NO<sub>2</sub> product, involving the validation of each component of the retrieval. This end-to-end validation was illustrated for three-and-a-half years (January 2007–July 2010) of GOME-2 NO<sub>2</sub> measurements based on MAX-DOAS measurements at the Observatoire de Haute Provence (OHP; 44° N, 5.7° E). The stratospheric columns from GOME-2 and coincident ground-based measurements at OHP are found to be in good overall agreement. A time series of the MAX-DOAS and the GOME-2 tropospheric NO<sub>2</sub> columns shows that pollution episodes at OHP are well captured by GOME-2. Monthly mean tropospheric columns are in very good agreement, with differences generally within  $0.5 \times 10^{15}$  molec cm<sup>-2</sup>.

The GOME-2 total and tropospheric NO<sub>2</sub> products are generated operationally at the O3M-SAF processing facility in DLR, and cover the period from January 2007 onwards. The GDP 4.4 algorithm is shown to be robust in performance and more than capable of real-time data turnover in operational execution. Near-real-time (i.e. two hours after sensing), offline and re-processed products are freely available. The GOME-2 NO<sub>2</sub> products are broadcasted via EUMETCast and WMO/GTS, and are available online on a FTP server. GOME-2 products can also be ordered at the Help Desk of the O3M-SAF hosted by the Finnish Meteorological Institute (FMI) (o3msaf@fmi.fi). The GOME-2 Algorithm Theoretical Basis Document (ATBD), Product User Manual (PUM), Validation Reports, as well as quick-look images and links to related services are available from following DLR web page: <http://atmos.caf.dlr.de/gome2>.

**Acknowledgements.** Development of the GOME-2 NO<sub>2</sub> column products and their validation has been funded by the O3M-SAF project with EUMETSAT and national contributions. Part of this work was funded by ProDex and the Belgian Science Policy via SECPEA. The authors would like to thank F. Boersma (KNMI) for useful discussions. We thank W. Zimmer and L. Butenko for the development work on the UPAS system. S. Kiemle, J. Jaeger, K. H. Seitz, B. Huber and T. Ruppert are kindly acknowledged for the day-to-day operations of the O3M-SAF facility at DLR. We thank EUMETSAT for the ground-segment interfacing work with the O3M-SAF systems and for the provision of GOME-2 level 1 products.

## References

- Balis, D., Lambert, J.-C., Van Roozendael, M., Spurr, R., Loyola, D., Livschitz, Y., Valks, P., Amiridis, V., Gerard, P., Granville, J., and Zehner, C.: Ten years of GOME/ERS2 total ozone data—The new GOME data processor (GDP) version 4: 2. Ground-based validation and comparisons with TOMS V7/V8, *J. Geophys. Res.*, 112, D07307, doi:10.1029/2005JD006376, 2007.
- Beirle, S., Platt, U., Wenig, M., and Wagner, T.: Weekly cycle of NO<sub>2</sub> by GOME measurements: a signature of anthropogenic sources, *Atmos. Chem. Phys.*, 3, 2225–2232, doi:10.5194/acp-3-2225-2003, 2003.
- Beirle, S., Platt, U., von Glasow, R., Wenig, M., and Wagner, T.: Estimate of nitrogen oxide emissions from shipping by satellite remote sensing, *Geophys. Res. Lett.*, 31, L18102, doi:10.1029/2004GL020312, 2004.
- Blond, N., Boersma, K. F., Eskes, H. J., van der A, R. J., Van Roozendael, M., De Smedt, I., Bergametti, G., and Vautard, R.: Intercomparison of SCIAMACHY nitrogen dioxide observations, in-situ measurements and air quality modeling results over Western Europe, *J. Geophys. Res.*, 112, D10311, doi:10.1029/2006JD007277, 2007.
- Boersma K.F., Eskes, H. J., and Brinksma, E. J.: Error analysis for tropospheric NO<sub>2</sub> retrieval from space, *J. Geophys. Res.*, 109, D04311, doi:10.1029/2003JD003962, 2004.
- Boersma, K. F., Eskes, H. J., Veefkind, J. P., Brinksma, E. J., van der A, R. J., Sneep, M., van den Oord, G. H. J., Levelt, P. F., Stammes, P., Gleason, J. F., and Bucsela, E. J.: Near-real time retrieval of tropospheric NO<sub>2</sub> from OMI, *Atmos. Chem. Phys.*, 7, 2103–2118, doi:10.5194/acp-7-2103-2007, 2007.
- Boersma, K. F., Jacob, D. J., Eskes, H. J., Pinder, R. W., Wang, J., and van der A, R. J.: Intercomparison of SCIAMACHY and OMI tropospheric NO<sub>2</sub> columns: Observing the diurnal evolution of chemistry and emissions from space, *J. Geophys. Res.*, 113, doi:10.1029/2007JD008816, 2008.
- Boersma et al.: An improved retrieval of tropospheric NO<sub>2</sub> columns from the Ozone Monitoring Instrument, *Atmos. Meas. Tech.*, in preparation, 2011.
- Bovensmann, H., Burrows, J. P., Buchwitz, M., Frerick, J., Rozanov, V. V., Chance, K. V., and Goede, A. P. H.: SCIAMACHY: Mission objectives and measurement modes, *J. Atmos. Sci.*, 56, 127–150, doi:10.1175/1520-0469, 1999.
- Brinksma, E., Pinardi, G., Braak, R., Volten, H., Richter, A., Schonhardt, A., Van Roozendael,

AMTD

4, 1617–1676, 2011

## Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





## Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



- M., Fayt, C., Hermans, C., Dirksen, R., Vlemmix, T., Berkhout, A. J. C., Swart, D. P. J., Oetjen, H., Wittrock, F., Wagner, T., Ibrahim, O., de Leeuw, G., Moerman, M., Curier, L., Celarier, E. A., Knap, W. H., Veefkind, J. P., Eskes, H. J., Allaart, M., Rothe, R., PETERS, A. J. M., and Levelt, P.: The 2005 and 2006 DANDELIONS NO<sub>2</sub> and Aerosol Intercomparison Campaigns, *J. Geophys. Res.*, 113, D16S46, doi:10.1029/2007JD008808, 2008.
- Bucsela, E. J., Celarier, E. A., Wenig, M. O., Gleason, J. F., Veefkind, J. P., Boersma, K. F., and Brinksma, E.: Algorithm for NO<sub>2</sub> vertical column retrieval from the Ozone Monitoring Instrument, *IEEE Trans. Geosci. Remote Sens.*, 44(5), 1245–1258, doi:10.1109/TGRS.2005.863715, 2006.
- Burrows, J., Weber, M., Buchwitz, M., Rozanov, V., Ladstätter-Weißmayer, A., Richter, A., Debeek, R., Hoogen, R., Bramstedt, K., Eichmann, K.-U., and Eisinger, M.: The Global Ozone Monitoring Experiment (GOME): Mission concept and first scientific results, *J. Atmos. Sci.*, 56, 151–175, 1999.
- Callies, J., Corpaccioli, E., Eisinger, M., Hahne, A., Lefebvre, A.: GOME-2 – MetOp's Second Generation Sensor for Operational Ozone Monitoring, *ESA Bulletin*, No. 102, 2000.
- Celarier, E. A., Brinksma, E. J., Gleason, J. F., Veefkind, J. P., Cede, A., Herman, J. R., Ionov, D., Goutail, F., Pommereau, J.-P., Lambert, J.-C., van Roozendaal, M., Pinardi, G., Wittrock, F., Schönhardt, A., Richter, A., Ibrahim, O. W., Wagner, T., Bojkov, B., Mount, G., Spinei, E., Chen, C. M., Pongetti, T. J., Sander, S. P., Bucsela, E. J., Wenig, M. O., Swart, D. P. J., Volten, H., Kroon, M., and Levelt, P. F.: Validation of Ozone Monitoring Instrument Nitrogen Dioxide Columns, *J. Geophys. Res.*, 113, D15S15, doi:10.1029/2007JD008908, 2008.
- CEOS: Data Quality Guidelines for Satellite Sensor Observations Relevant to GEOSS – Calibration and Validation Issues, Recommendations by CEOS/ WGCV to the CEOS Task Force, 2004.
- Chance, K. V. and Spurr R. J. D.: Ring effect studies: Rayleigh scattering, including molecular parameters for rotational Raman scattering, and the Fraunhofer spectrum, *Applied Optics* LP, 36, Issue 21, 5224–5230, doi:10.1364/AO.36.005224, 1997.
- Coldewey-Egbers M., Slijkhuis S., Aberle B., and Loyola, D.: Long-term analysis of GOME in-flight calibration parameters and instrument degradation, *Appl. Optics*, 47(28), 4749–4761, 2008.
- Dikty, S., Richter, A., Bovensmann, H., Wittrock, F., Weber, M., Noël, S., Burrows, J. P., Munro, R., and Lang, R.: GOME-2 level 2 products at IUP Bremen and first results on the quantification of the effects of optical degradation, *EUMETSAT Meteorological Satellite Conference*,



Córdoba, Spain, 20–24 September 2010.

Flemming, J., Inness, A., Flentje, H., Huijnen, V., Moinat, P., Schultz, M. G., and Stein, O.: Coupling global chemistry transport models to ECMWF's integrated forecast system, *Geosci. Model Dev.*, 2, 253–265, 2009.

5 Franke, K., Richter, A., Bovensmann, H., Eyring, V., Jöckel, P., Hoor, P., and Burrows, J. P.: Ship emitted NO<sub>2</sub> in the Indian Ocean: comparison of model results with satellite data, *Atmos. Chem. Phys.*, 9, 7289–7301, doi:10.5194/acp-9-7289-2009, 2009.

Gordley, L. L., Russell III, J. M., Mickley, L. J., Frederick, J. E., Park, J. H., Stone, K. A., Beaver, G. M., McInemey, J. M., Deaver, L. E., Toon, G. C., Murcray, F. J., Blatherwick, R. D., Gunson, M. R., Abbatt, J. P. D., Mauldin III, R. L., Mount, G. H., Sen, B., and Blavier, J.-F.: Validation of Nitric Oxide and Nitrogen Dioxide Measurements Made by Halogen Occultation Experiment for UARS Platform, *J. Geophys. Res.* 101, p. 10241, 1996.

Greenblatt, G. D., Orlando, J. J., Burkholder, J. B., and Ravishankara, A. R.: Absorption measurements of oxygen between 330 and 1140 nm, *J. Geophys. Res.*, 95, 18577–18582, 1990

15 Gür, B., Spietz, P., Orphal, J., and Burrows, J.: Absorption Spectra Measurements with the GOME-2 FMs using the IUP/IFE-UB's Calibration Apparatus for Trace Gas Absorption Spectroscopy CATGAS, Final Report, IUP University of Bremen, October 2005.

Heckel, A., Richter, A., Tarsu, T., Wittrock, F., Hak, C., Pundt, I., Junkermann, W., and Burrows, J. P.: MAX-DOAS measurements of formaldehyde in the Po-Valley, *Atmos. Chem. Phys.*, 5, 909–918, doi:10.5194/acp-5-909-2005, 2005

20 Heinen, T., Kiemle, S., Buckl, B., Mikusch, E., and Loyola, D.: The Geospatial Service Infrastructure for DLR's National Remote Sensing Data Library, *IEEE J. Sel. Top. Appl.*, 2, 260–269, 2009.

Hollingsworth, A., Engelen, R. J., Textor, C., Benedetti, A., Boucher, O., Chevallier, F., Dethof, A., Elbern, H., Eskes, H., Flemming, J., Granier, C., Kaiser, J. W., Morcrette, J. -J., Rayner, P., Peuch, V.-H., Rouil, L., Schultz, M. G., Simmons, A. J., and the GEMS Consortium: Toward a monitoring and forecasting system for atmospheric composition: The Gems Project. *Bull. Amer. Meteor. Soc.*, 89, 1147–1164, doi:10.1175/2008BAMS2355.1, 2008.

Hönninger, G., von Friedeburg, C., and Platt, U.: Multi axis differential optical absorption spectroscopy (MAX-DOAS), *Atmos. Chem. Phys.*, 4, 231–254, doi:10.5194/acp-4-231-2004, 2004.

30 Horowitz, L., Walters, S., Mauzerall, D., Emmons, L., Rasch, P., Granier, C., Tie, X., Lamarque, J., Schultz, M., Tyndall, G., Orlando, J., and Brasseur, G.: A global simulation of tropospheric

AMTD

4, 1617–1676, 2011

## Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



- ozone and related tracers: description and evaluation of MOZART, version 2, J. Geophys. Res., 108(D24), 4784, doi:10.1029/2002JD002853, 2003.
- Irie, H., Kanaya, Y., Akimoto, H., Tanimoto, H., Wang, Z., Gleason, J. F., and Bucsela, E. J.: Validation of OMI tropospheric NO<sub>2</sub> column data using MAX-DOAS measurements deep inside the North China Plain in June 2006: Mount Tai Experiment 2006, Atmos. Chem. Phys., 8, 6577–6586, doi:10.5194/acp-8-6577-2008, 2008.
- Ionov, D. V., Timofeyev, Y. M., Sinyakov, V. P., Semenov, V. K., Goutail, F., Pommereau, J.-P., Bucsela, E. J., Celarier, E. A., and Kroon, M.: Ground-based validation of EOS-Aura OMI NO<sub>2</sub> vertical column data in the midlatitude mountain ranges of Tien Shan (Kyrgyzstan) and Alps (France), J. Geophys. Res., 113, D15S08, doi:10.1029/2007JD008659, 2008.
- Kleinpool, Q. L., Dobber, M. R., de Haan, J. F., and Levelt, P. F.: Earth surface reflectance climatology from 3 years of OMI data. J. Geoph. Res., 113, D18308, doi:10.1029/2008JD010290, 2008.
- Konovalov, I. B., Beekmann, M., Richter, A., Burrows, J. P., and Hilboll, A.: Multi-annual changes of NO<sub>x</sub> emissions in megacity regions: nonlinear trend analysis of satellite measurement based estimates, Atmos. Chem. Phys., 10, 8481–8498, doi:10.5194/acp-10-8481-2010, 2010.
- Kurylo, M. J. and Zander, R. J.: The NDSC – Its status after ten years of operation, in: Proceedings of the Quadrennial Ozone Symposium, edited by: Bojkov, R. D. and Kazuo, S., Sapporo, Japan, 2–8 July 2000, 137–138, 2000.
- Lambert, J.-C. and Balis, D.: Delta validation report for ERS-2 GOME Data Processor Upgrade to Version 4.0. ERSE-CLVL-EOPG-TN-04-0001, ESA/ESRIN, Frascati, Italy, 2004.
- Lambert, J. C., Granville J., Van Roozendael M., Sarkissian A., Goutail F., Müller J.-F., Pommereau J.-P., Russell III, J. M.: A climatology of NO<sub>2</sub> profile for improved air mass factors for ground-based vertical column measurements, Proceedings of the 5th European Symposium Strat. Ozone, Saint Jean de Luz, Sept 1999, Air pollution research report 73, European Commission, edited by: Harris, N. R. P., Guirlet, M., Amanatidis, G. T., p. 703, ISBN 92-827-5672-6, 2000.
- Lambert, J.-C., Granville, J., Soebijanta, V., and Van Roozendael, M.: Geophysical Validation of GOME GDP 3.0 Total NO<sub>2</sub>, in: ERS-2 GOME GDP 3.0 Implementation and Delta Validation, Validation Report for GOME Level 1-to-2 Data Processor Upgrade to Version GDP 3.0, edited by: Lambert, J.-C., ERSE-DTEX-EOD-TN-02-0006, ESA/ESRIN, Frascati, Italy, 2002.
- Lambert, J.-C., Blumenstock, T., Boersma, F., Bracher, A., De Mazière, M., Demoulin, P., De

## Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

## Abstract

## Introduction

## Conclusions

## References

## Tables

## Figures



[Back](#)

Close

Full Screen / Esc

[Printer-friendly Version](#)

## Interactive Discussion



- Smedt, I., Eskes, H., Gil, M., Goutail, F., Granville, J., Hendrick, F., Ionov, D. V., Johnston, P. V., Kostadinov, I., Kreher, K., Kyrö, E., Martin, R., Meier, A., Navarro-Comas, M., Petritoli, A., Pommereau, J-P., Richter, A., Roscoe, H. K., Sioris, C., Sussmann, R., Van Roozendael, M., Wagner, T., Wood, S., and Yela, M.: Geophysical Validation of SCIAMACHY NO<sub>2</sub> Vertical columns: Overview of Early 2004 Results. Proceedings of the Second Workshop on the Atmospheric Chemistry Validation of ENVISAT (ACVE-2), 3–7 May 2004, ESA-ESRIN, Frascati, Italy (ESA SP-562), edited by: Danesy, D., 6.1–6.13, 2004.
- Lambert, J.-C., Pinardi, G., Hao, N., and Valks, P.: GOME-2 GDP 4.2 total NO<sub>2</sub> (NTO/OTO) validation update and tropospheric NO<sub>2</sub> validation set-up, TN-IASB-GOME2-O3MSAF-NO2-02\_ORR-B\_1, 24 November, 2008.
- Lambert, J.-C., et al.: Atmospheric Monitoring Services – Stage 2 of the Earthwatch GMES Services Element, PROMOTE-2, C5 Service Validation Protocol, Version 3, October, 2009.
- Lang, R., Munro, R., Livschitz, Y., Dyer, R., and Lacan, A.: GOME-2 FM3 Long-Term In-Orbit Degradation – Basic Signatures After 2nd Throughput Test, EUMETSAT Technical report, EUM.OPS-EPS.DOC.09.0464, 2009.
- Leitão, J., Richter, A., Vrekoussis, M., Kokhanovsky, A., Zhang, Q. J., Beekmann, M., and Burrows, J. P.: On the improvement of NO<sub>2</sub> satellite retrievals – aerosol impact on the air mass factors, *Atmos. Meas. Tech.*, 3, 475–493, doi:10.5194/amt-3-475-2010, 2010.
- Leue, C., Wenig, M., Wagner, T., Klimm, O., Platt, U., and Jähne, B.: Quantitative analysis of NO<sub>x</sub> emissions from Global Ozone Monitoring Experiment satellite image sequences, *J. Geophys. Res.*, 106, 5493–5505, 2001.
- Levelt, P. F., van den Oord, G. H. J., Dobber, M. R., Mälkki, A., Visser, H., de Vries, J., Stammes, P., Lundell, J. O. V., and Saari, H.: The Ozone Monitoring Instrument, *IEEE Trans. on Geosci. Rem. Sens.*, 44(5), 1093–1101, doi:10.1109/TGRS.2006.872333, 2006.
- Livschitz, Y., and Loyola, D.: Design Document for the GOME-2 Universal Processor for Atmospheric Spectrometers, SAF/O3M/DLR/DD/001, Issue 2.0, October 2003.
- Loyola D., Thomas W., Livschitz Y., Ruppert T., Albert P., and Hollmann. R.: Cloud properties derived from GOME/ERS-2 backscatter data for trace gas retrieval, *IEEE T. Geosci. Remote*, 45(9), 2747–2758, 2007.
- Loyola, D., Koukouli, M. E., Valks, P., Balis, D. S., Hao, N., Van Roozendael, M., Spurr, R. J. D., Zimmer, W., Kiemle, S., Lerot, C., and Lambert, J.-C.: The GOME-2 Total Column Ozone Product: Retrieval Algorithm and Ground-Based Validation, *J. Geophys. Res.*, accepted, doi:10.1029/2010JD014675, 2011.

## Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



- Martin, R. V., Chance, K., Jacob, D. J., Kurosu, T. P., Spurr, R. J. D., Bucsela, E., Gleason, J. F., Palmer, P. I., Bey, I., Fiore, A. M., Li, Q., Yantosca, R. M., and Koelemeijer, R. B. A.: An improved retrieval of tropospheric nitrogen dioxide from GOME, *J. Geophys. Res.*, 107(D20), 4437, doi:10.1029/2001JD001027, 2002.
- 5 Munro, R., Eisinger, M., Anderson, C., Callies, J., Corpaccioli, E., Lang, R., Lefebvre, A., Livschitz, Y., and Perez Albinana, A.: GOME-2 on MetOp: From in-orbit verification to routine operations, in: *Proceedings of EUMETSAT Meteorological Satellite Conference*, Helsinki, Finland, 12–16 June 2006.
- 10 Nüß, H., Richter, A., Valks, P., and Burrows, J.: Improvement of the NO<sub>2</sub> total column retrieval for GOME-2, O3M-SAF Visiting Scientist Report, IUP University of Bremen, available at: <http://o3msaf.fmi.fi>, November 2006.
- Palmer, P. I., Jacob, D. J., Chance, K., Martin, R. V., Spurr, R. J. D., Kurosu, T. P., Bey, I., Yantosca, R., Fiore, A., and Li, Q.: Air-mass factor formulation for spectroscopic measurements from satellites: application to formaldehyde retrievals from GOME, *J. Geophys. Res.*, 106, 14539–14550, 2001.
- 15 Pinardi, G., Hendrick, F., Clémer, K., Lambert, J.C., Bai, J., and Van Roozendael, M.: On the use of the MAXDOAS technique for the validation of tropospheric NO<sub>2</sub> column measurements from satellite, proceeding of the EUMETSAT Meteorological Satellite Conference, 9–12 September 2008, Darmstadt, Germany, 2008.
- 20 Platt, U.: Differential optical absorption spectroscopy (DOAS), in *Air Monitoring by Spectroscopic Techniques*, Chem. Anal. Ser. 127, 27–84, John Wiley, New York, 27–84, 1994.
- Platt, U. and Stutz, J.: *Differential Optical Absorption Spectroscopy, Principles and Applications*, Springer, Physics of Earth and Space Environments, ISBN 978-3-540-21193-8, 2008.
- Randall, C. E., Rusch, D. W., Bevilacqua, R. M., Hoppel, K. W., and Lumpe, J. D.: Polar Ozone and Aerosol Measurements (POAM) II Stratospheric NO<sub>2</sub>, 1993–1996, *J. Geophys. Res.*, 25 103, 28361–28371, 1998.
- Richter, A. and Burrows, J.: Tropospheric NO<sub>2</sub> from GOME measurements, *Adv. Space Res.*, 29, 1673–1683, 2002.
- Richter, A. and Wagner, T.: Diffuser plate spectral structures and their influence on GOME slant columns. Technical note, available at: [www.iup.uni-bremen.de/gome/data/diffuser\\_gome.pdf](http://www.iup.uni-bremen.de/gome/data/diffuser_gome.pdf), 2001.
- 30 Richter, A., Eyring, V., Burrows, J., Bovensmann, H., Lauer, A., Sierk, B., and Crutzen, P.: Satellite measurements of NO<sub>2</sub> from international shipping emissions, *Geophys. Res. Lett.*,

Operational NO<sub>2</sub>  
retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



31, L23110, doi:10.1029/2004GL020822, 2004

Richter, A., Burrows, J. P., Nüß, H., Granier, C., and Niemeier, U.: Increase in tropospheric nitrogen dioxide over China observed from space, *Nature*, 437, 129–132, 2005.

Richter, A., Begoin, M., Hilboll, A., and Burrows, J. P.: An improved NO<sub>2</sub> retrieval for the GOME-2 satellite instrument, *Atmos. Meas. Tech. Discuss.*, 4, 213–246, doi:10.5194/amtd-4-213-2011, 2011.

Roscoe, H. K., Van Roozendael, M., Fayt, C., du Piesanie, A., Abuhassan, N., Adams, C., Akrami, M., Cede, A., Chong, J., Clémer, K., Friess, U., Gil Ojeda, M., Goutail, F., Graves, R., Griesfeller, A., Grossmann, K., Hemerijckx, G., Hendrick, F., Herman, J., Hermans, C., Irie, H., Johnston, P. V., Kanaya, Y., Kreher, K., Leigh, R., Merlaud, A., Mount, G. H., Navarro, M., Oetjen, H., Pazmino, A., Perez-Camacho, M., Peters, E., Pinardi, G., Puertedura, O., Richter, A., Schönhardt, A., Shaiganfar, R., Spinei, E., Strong, K., Takashima, H., Vlemmix, T., Vrekoussis, M., Wagner, T., Wittrock, F., Yela, M., Yilmaz, S., Boersma, F., Hains, J., Kroon, M., Piters, A., and Kim, Y. J.: Intercomparison of slant column measurements of NO<sub>2</sub> and O<sub>4</sub> by MAX-DOAS and zenith-sky UV and visible spectrometers, *Atmos. Meas. Tech.*, 3, 1629–1646, doi:10.5194/amt-3-1629-2010, 2010.

Rothman, L. S., Barbe, A., Chris Benner, D., Brown, L. R., Camy-Peyret, C., Carleer, M. R., Chance, K., Clerbaux, C., Dana, V., Devi, V. M., Fayt, A., Flaud, J. -M., Gamache, R. R., Goldman, A., Jacquemart, D., Jucks, K. W., Lafferty, W. J., Mandin, J. -Y., Massie, S. T., Nemtchinov, V., Newnham, D. A., Perrin, A., Rinsland, C. P., Schroeder, J., Smith, K. M., Smith, M. A. H., Tang, K., Toth, R. A., Vander Auwera, J., Varanasi, P., and Yoshino, K.: The HITRAN molecular spectroscopic database: edition of 2000 including updates through 2001, *J. Quant. Spectrosc. Rad. Transfer*, 82, 5–44, 2003.

Solomon, S.: Stratospheric ozone depletion: A review of concepts and history, *Rev. Geophys.*, 37(3), 275–316, 1999.

Spurr, R.: LIDORT and VLIDORT: Linearized pseudo-spherical scalar and vector discrete ordinate radiative transfer models for use in remote sensing retrieval problems. *Light Scattering Reviews*, 3, edited by: Kokhanovsky, A., Springer, 2008.

Spurr, R. J. D., Kurosu, T. P., and Chance, K. V.: A Linearized discrete Ordinate Radiative Transfer Model for Atmospheric Remote Sensing Retrieval, *J. Quant. Spectrosc. Radiat. Transfer*, 68, 689–735, 2001.

Valks, P., Loyola, D., Hao, N., Rix, M., and Slijkhuis, S.: Algorithm Theoretical Basis Document for GOME-2 Total Column Products of Ozone, Minor Trace Gases and Cloud Properties

Operational NO<sub>2</sub>  
retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



(GDP 4.4 for O3M-SAF OTO and NTO), DLR/GOME-2/ATBD/01, Iss./Rev.: 2/D, available at: <http://o3msaf.fmi.fi>, January 2011.

Vandaele A. C., Hermans, C., Simon, P. C., Carleer, M., Colin, R., Fally, S., Merienne, M. F., Jenouvrier, A., and Coquart, B.: Measurements of the NO<sub>2</sub> absorption cross-section from 42 000 cm<sup>-1</sup> to 10 000 cm<sup>-1</sup> (238–1000 nm) at 220 K and 294 K, *J. Quant. Spectrosc. Radiat. Transfer*, 59, 171–184, 1998

Van der A, R. J., Peters, D. H. M. U., Eskes, H., Boersma, K. F., Van Roozendael, M., De Smedt, I., and Kelder, H. M.: Detection of the trend and seasonal variation in tropospheric NO<sub>2</sub> over China, *J. Geophys. Res.*, 111, D12317, doi:10.1029/2005JD006594, 2006.

Van der A, R. J., Eskes, H. J., Boersma, K. F., van Noije, T. P. C., Van Roozendael, M., De Smedt, I., Peters, D. H. M. U., and Meijer, E. W.: Trends, seasonal variability and dominant NO<sub>x</sub> source derived from a ten year record of NO<sub>2</sub> measured from space, *J. Geophys. Res.*, 113, D04302, doi:10.1029/2007JD009021, 2008.

Velders, G. J. M., Granier, C., Portmann, R. W., Pfeilsticker, K., Wenig, M., Wagner, T., Platt, U., Richter, A., and Burrows, J. P.: Global tropospheric NO<sub>2</sub> column distributions: Comparing three-dimensional model calculations with GOME measurements, *J. Geophys. Res.*, 106, 12643–12660, 2001.

Wagner, T., Dix, B., Friedburg, C. v., Frieß, U., Sanghavi, S., Sinreich, R., and Platt, U.: MAX-DOAS O<sub>4</sub> measurements: A new technique to derive information on atmospheric aerosols- Principles and information content, *J. Geophys. Res.*, 109, D22205, doi:10.1029/2004JD004904, 2004.

Wenig, M., Köhl, S., Beirle, S., Bucsela, E., Jähne, B., Platt, U., Gleason, J., and Wagner, T.: Retrieval and analysis of stratospheric NO<sub>2</sub> from the Global Ozone Monitoring Experiment, *J. Geophys. Res.*, 109, D04315, doi:10.1029/2003JD003652, 2004.

Wittrock, F., Oetjen, H., Richter, A., Fietkau, S., Medeke, T., Rozanov, A., and Burrows, J. P.: MAX-DOAS measurements of atmospheric trace gases in Ny-Ålesund – Radiative transfer studies and their application, *Atmos. Chem. Phys.*, 4, 955–966, doi:10.5194/acp-4-955-2004, 2004.

Zhou, Y., Brunner, D., Boersma, K. F., Dirksen, R., and Wang, P.: An improved tropospheric NO<sub>2</sub> retrieval for OMI observations in the vicinity of mountainous terrain, *Atmos. Meas. Tech.*, 2, 401–416, doi:10.5194/amt-2-401-2009, 2009.

**Operational NO<sub>2</sub>  
retrieval for GOME-2**

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Table 1.** Summary of GOME-2 instrument characteristics (Munro et al., 2006). The main improvements as compared to GOME/ERS-2 are shown in *italic*.

Principle	Nadir-scanning UV/VIS grating spectrometer
Wavelength range	240–790 nm in 4 channels <i>300–800 nm in 2 polarisation channels (s/p) with 15 bands</i>
Spectral sampling	0.12–0.21 nm (main channels)
Spectral resolution	<i>0.26–0.51 nm (FWHM)</i>
Swath width	<i>1920 km (default)</i>
Swath type	<i>Earth-curvature compensating</i>
Integration time	<i>187.5 ms (default)</i>
Spatial resolution	<i>80 × 40 km<sup>2</sup> (default)</i>
Internal calibration	<i>Spectral lamp (PtCrNeAr), White lamp, LED</i>
Sun diffuser	<i>Quartz quasi-volume</i>



# Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

**Table 2.** Estimated mean uncertainty in the tropospheric air mass factor  $M_t$  for polluted conditions ( $V_t > 1.0 \times 10^{15} \text{ molec cm}^{-2}$ ) due to errors in the surface albedo, cloud parameters, and the a priori NO<sub>2</sub> profile shape.

Error source	Uncertainty	Uncertainty in $M_t$
Surface albedo	0.02	14%
Cloud fraction	0.06	25%
Cloud-top pressure	40 hPa	3%
NO <sub>2</sub> profile shape	see Sect. 6.3	15%
Total uncertainty*		33%

\* The uncertainty due to the effect of aerosols is not included, see Sect. 6.3.

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[I◀](#)
[▶I](#)
[◀](#)
[▶](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)


**Operational NO<sub>2</sub>  
retrieval for GOME-2**

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

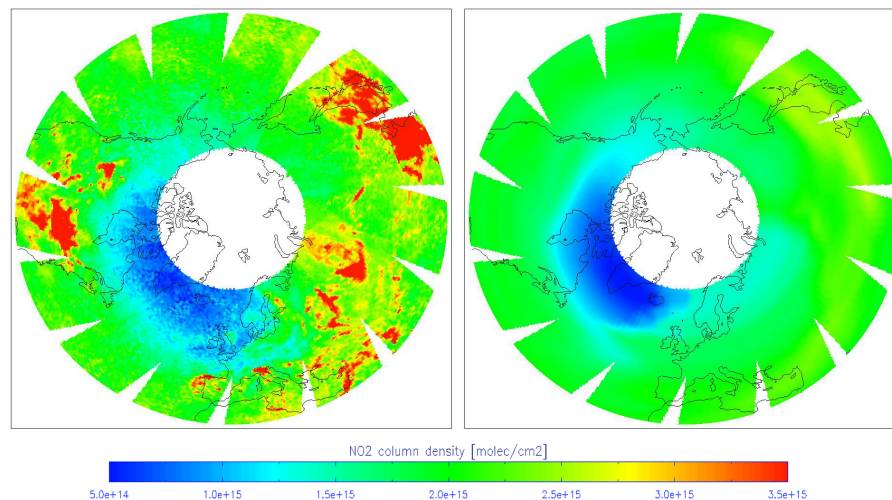


**Table 3.** Contributions to the overall uncertainty in the GOME-2 tropospheric NO<sub>2</sub> column retrieved with the GDP 4.4 for polluted conditions ( $V_t > 1.0 \times 10^{15}$  molec cm<sup>-2</sup>).

Error source	Uncertainty
Slant column	$0.45 \times 10^{15}$ molec cm <sup>-2</sup>
Stratospheric column	$0.15\text{--}0.30 \times 10^{15}$ molec cm <sup>-2</sup>
Tropospheric AMF	15–50%
Tropospheric column	40–80%

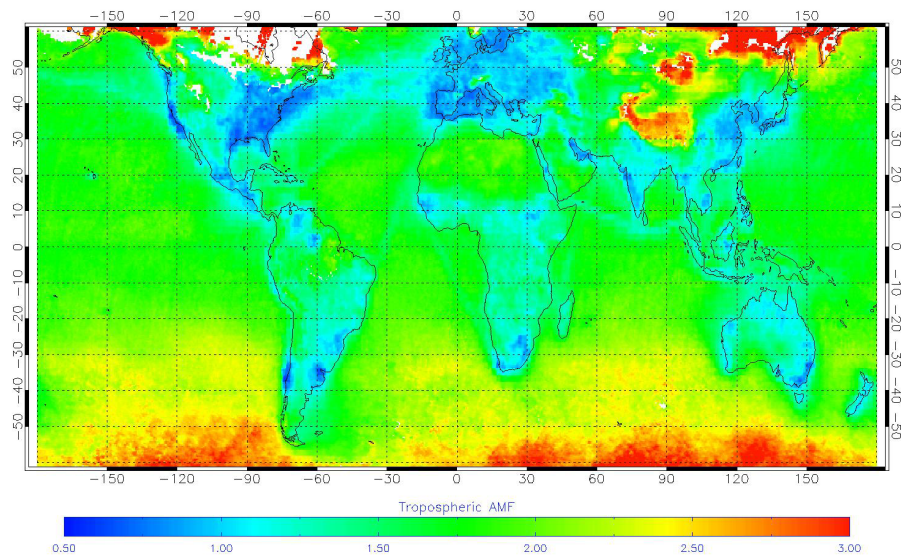
**Operational NO<sub>2</sub>  
retrieval for GOME-2**

P. Valks et al.



**Fig. 1.** Total NO<sub>2</sub> distribution from GOME-2 for 22 February 2008 (left) and the corresponding stratospheric NO<sub>2</sub> distribution as obtained with the spatial filtering approach (right).

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I◀](#)[▶I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)



**Fig. 2.** Monthly averaged tropospheric air mass factors for March 2008, as calculated with the GDP 4.4.

# Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

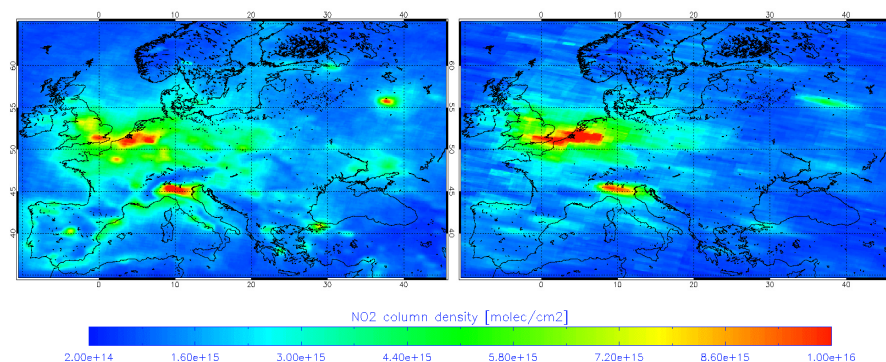
Printer-friendly Version

Interactive Discussion



**Operational NO<sub>2</sub>  
retrieval for GOME-2**

P. Valks et al.

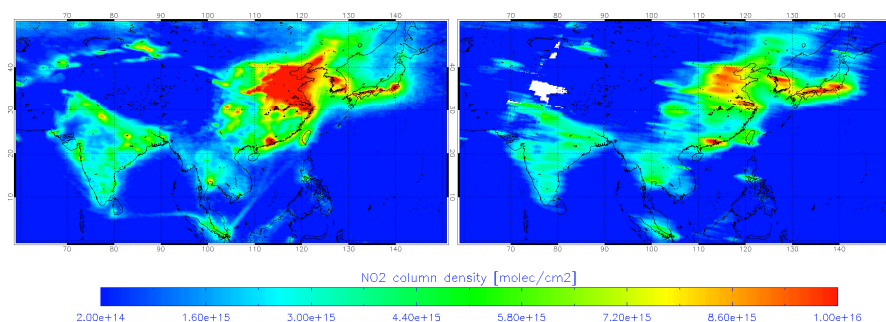


**Fig. 3.** Yearly average tropospheric NO<sub>2</sub> columns over Europe measured by GOME-2 for 2007 (left) and by GOME/ERS-2 for 2000 (right).

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

**Operational NO<sub>2</sub>  
retrieval for GOME-2**

P. Valks et al.



**Fig. 4.** Average tropospheric NO<sub>2</sub> columns over East Asia measured by GOME-2 for 2007–2009 (left) and by GOME/ERS-2 for 1997–1999 (right).

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

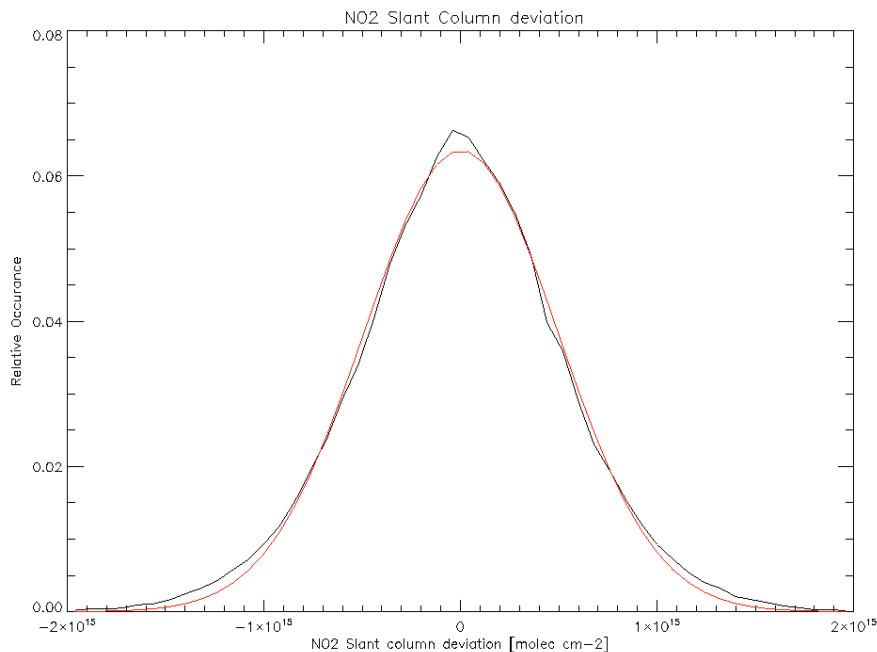
Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





**Fig. 5.** Distribution of the deviations of GOME-2 NO<sub>2</sub> slant columns from corresponding (2° × 2°) box mean values in the tropical Pacific region (20° S–20° N; 160°–180° E) for April 2007. The red line shows the Gaussian function fitted to the measured distribution. The width of the Gaussian corresponds to a slant column error of  $0.45 \times 10^{15} \text{ molec cm}^{-2}$ .

## Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

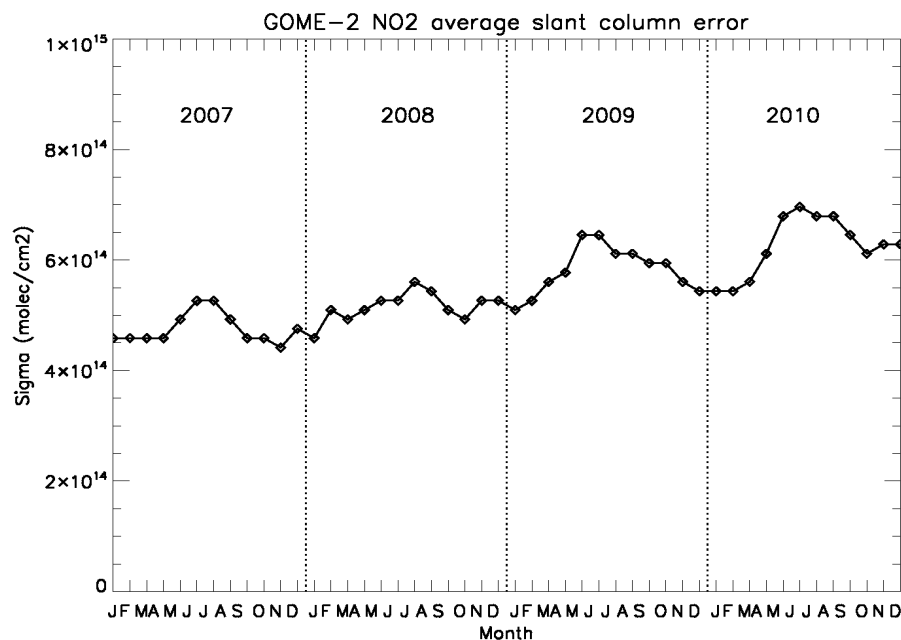
Interactive Discussion





Operational NO<sub>2</sub>  
retrieval for GOME-2

P. Valks et al.



**Fig. 6.** Estimated NO<sub>2</sub> slant column error for the GOME-2 instrument for the period January 2007–December 2010.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

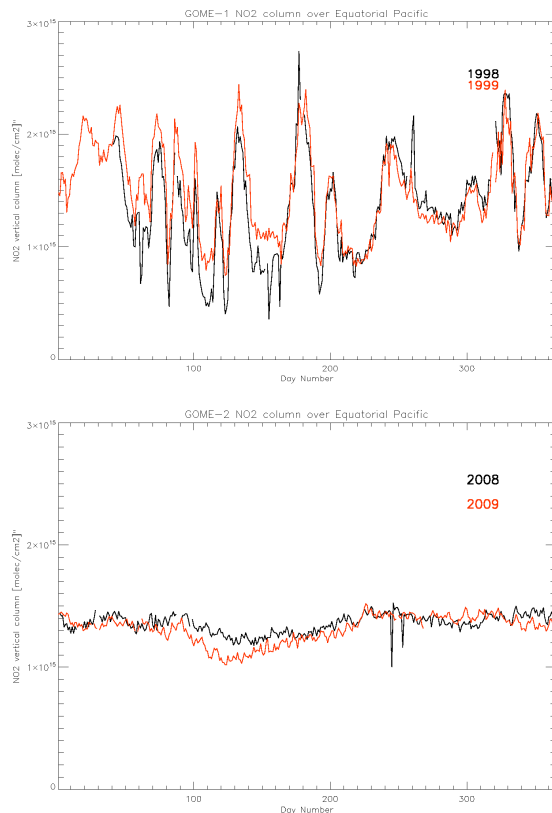
Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





**Fig. 7.** NO<sub>2</sub> vertical column density from GOME/ERS-2 for the Equatorial Pacific for 1998 and 1999 (Top), and from GOME-2/MetOp for 2008 and 2009 (Bottom). The GOME NO<sub>2</sub> columns show large and systematic seasonal variations induced by the diffuser plate. The seasonal structures in the GOME-2 NO<sub>2</sub> columns are much smaller as a result of the use of a quartz quasi-volume Diffuser in the GOME-2 instrument.

**Operational NO<sub>2</sub>  
retrieval for GOME-2**

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

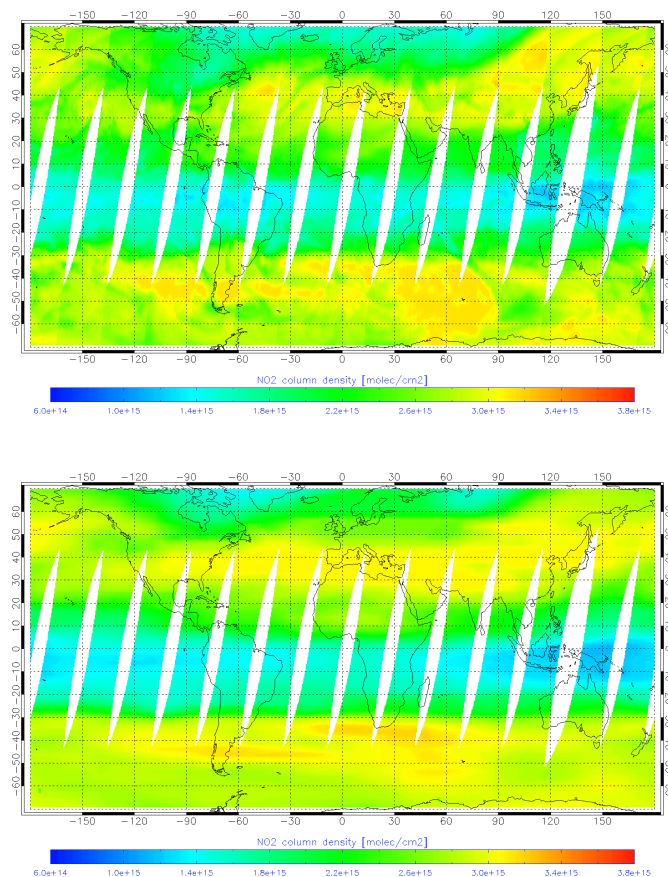
Back

Close

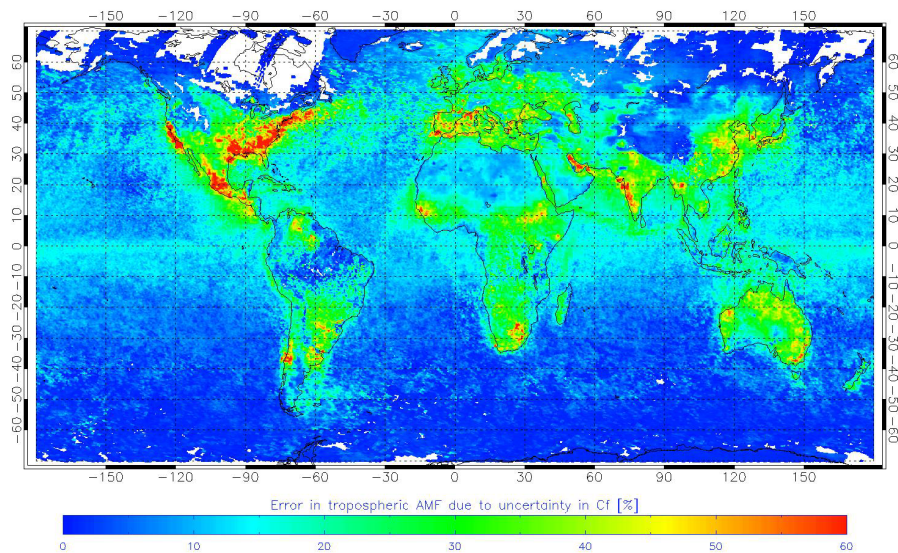
Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Fig. 8.** Stratospheric NO<sub>2</sub> columns from the IFS-MOZART reanalysis model for 15 March 2004 (Top) and those retrieved with the spatial filtering method (as used in the GDP 4.4), using synthetic slant column derived from IFS-MOZART model data as input (Bottom).



**Fig. 9.** Uncertainty in the tropospheric air mass factor for March 2008 due to the uncertainty in the GOME-2 cloud fraction. Only measurements with a cloud radiance fraction <50% were used.

# Operational NO<sub>2</sub> retrieval for GOME-2

P. Valks et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

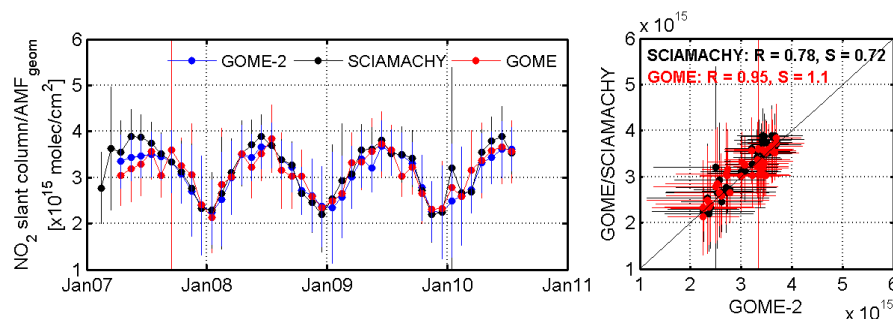
Printer-friendly Version

Interactive Discussion



Operational NO<sub>2</sub>  
retrieval for GOME-2

P. Valks et al.



**Fig. 10.** Comparison of normalized NO<sub>2</sub> slant column densities as measured by GOME (GDP 4.1), SCIAMACHY (TEMIS) and GOME-2 (GDP 4.4) at the Observatoire de Haute Provence (OHP, 44° N, 5.7° E) between January 2007 and July 2010. Dots represent the monthly average of all satellite measurements within a radius of 300 km around OHP and the error bars the one sigma variability. The left panel shows the time series of the three datasets, while the right panel presents the scatter plot of the GOME and SCIAMACHY datasets with respect to the GOME-2 data. The correlation coefficient  $R$  and the slope  $S$  of the linear regression line are given as insert.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

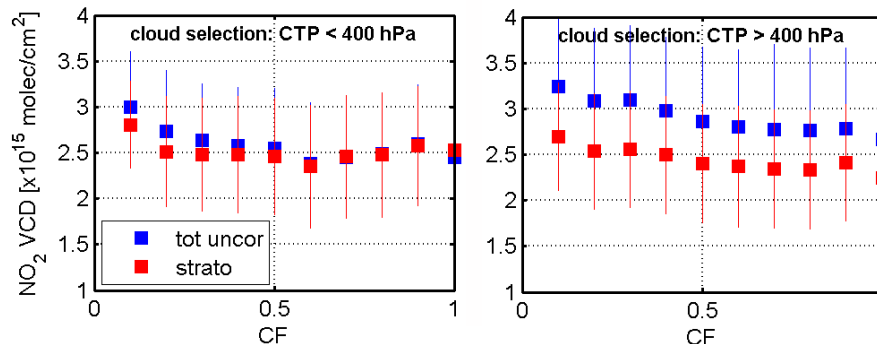
Printer-friendly Version

Interactive Discussion



Operational NO<sub>2</sub>  
retrieval for GOME-2

P. Valks et al.



**Fig. 11.** Comparison of initial total NO<sub>2</sub> columns (i.e. uncorrected for tropospheric pollution) and stratospheric NO<sub>2</sub> columns from GOME-2 around OHP. The squares correspond to NO<sub>2</sub> column averages in bins of 0.1 cloud fraction unit and the error bars to the variability of all pixels in time and space. On the left hand side, GOME-2 measurements with a cloud top pressure (CTP) smaller than 400 hPa are selected, while on the right hand side the selection applies to measurements with a CTP higher than 400 hPa.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

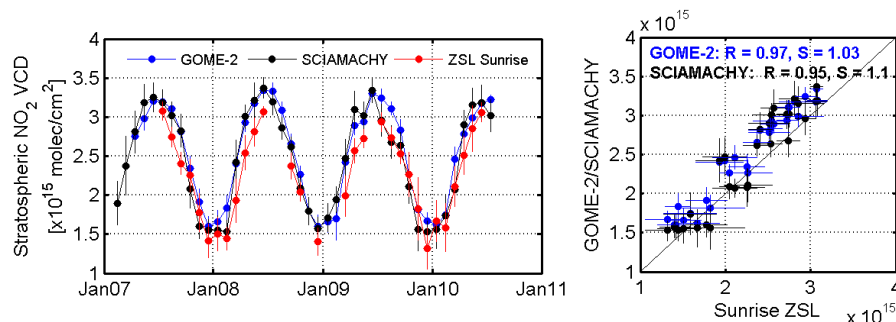
Printer-friendly Version

Interactive Discussion



**Operational NO<sub>2</sub>  
retrieval for GOME-2**

P. Valks et al.



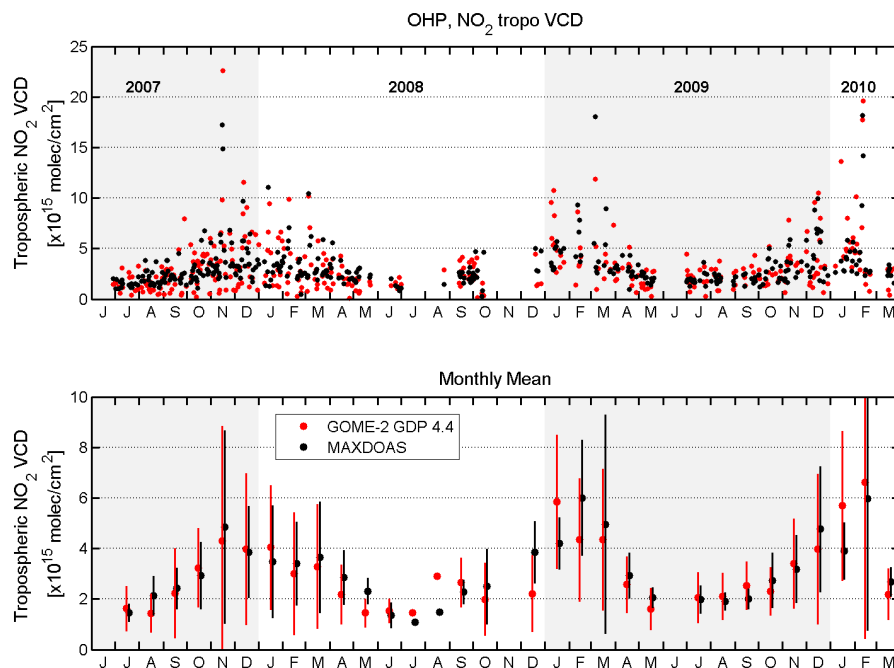
**Fig. 12.** Comparison of stratospheric NO<sub>2</sub> column data measured by GOME-2 (GDP 4.4), SCIAMACHY (TEMIS) and ground-based ZSL sunrise measurements from the MAXDOAS instrument at OHP, between January 2007 and July 2010. Left panel: time series of NO<sub>2</sub> monthly means around OHP; right panel: scatter plot between the satellite and the ground-based ZSL measurements. The coefficient  $R$  and the slope  $S$  of the linear regression line are given as insert.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)



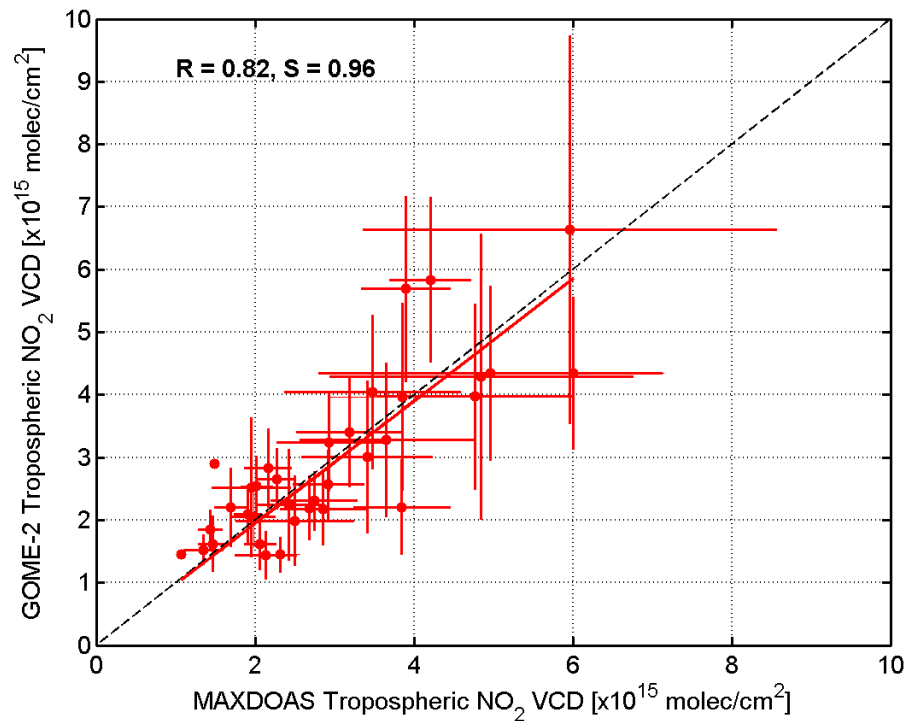
Operational NO<sub>2</sub>  
retrieval for GOME-2

P. Valks et al.



**Fig. 13.** Comparison of MAXDOAS and GOME-2 tropospheric NO<sub>2</sub> columns (mean value of all the pixels within 100 km around OHP, after flag selection) from June 2007 to March 2010. In the first subplot daily values (only days with both successful measurements) are represented while the second subplot displays monthly averaged values and corresponding one sigma standard deviations (if there was only one coincident MAXDOAS measurement in a particular month, the error bar is omitted).

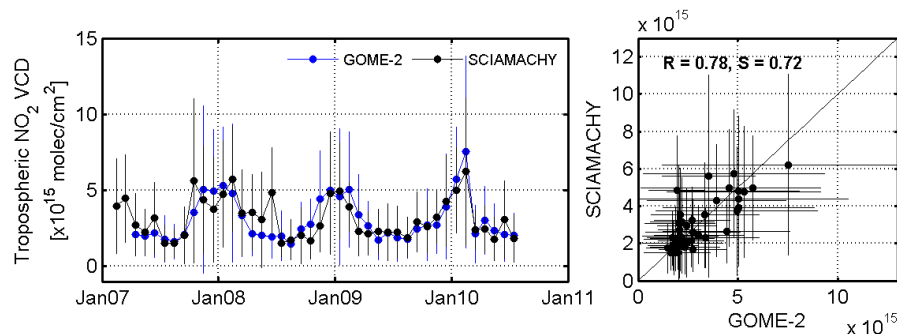
[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I◀](#)[▶I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)



**Fig. 14.** Scatter plot of the monthly averaged MAXDOAS and GOME-2 tropospheric NO<sub>2</sub> columns at OHP for the period displayed in Fig. 13. The correlation coefficient  $R$  and the slope  $S$  of the linear regression line are given in the legend.

**Operational NO<sub>2</sub>  
retrieval for GOME-2**

P. Valks et al.



**Fig. 15.** Comparison of monthly mean tropospheric NO<sub>2</sub> columns measured by GOME-2 (GDP 4.4) and SCIAMACHY (TEMIS), around OHP between January 2007 and July 2010. Only satellite measurements with a valid tropospheric flag and within 100 km of OHP are used for the comparison.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

