

# An improved tropospheric NO<sub>2</sub> column retrieval algorithm for the Ozone Monitoring Instrument

K. F. Boersma<sup>1,2</sup>, H. J. Eskes<sup>1</sup>, R. J. Dirksen<sup>1</sup>, R. J. van der A<sup>1</sup>, J. P. Veefkind<sup>1</sup>, P. Stammes<sup>1</sup>, V. Huijnen<sup>1</sup>, Q. L. Kleipool<sup>1</sup>, M. Sneep<sup>1</sup>, J. Claas<sup>1</sup>, J. Leitão<sup>3</sup>, A. Richter<sup>3</sup>, Y. Zhou<sup>4</sup>, and D. Brunner<sup>4</sup>

<sup>1</sup>Royal Netherlands Meteorological Institute, De Bilt, The Netherlands

<sup>2</sup>Eindhoven University of Technology, Fluid Dynamics Lab, The Netherlands

<sup>3</sup>Institute for Environmental Physics and Institute of Remote Sensing, University of Bremen, Germany

<sup>4</sup>EMPA, Swiss Federal Laboratories for Materials Science and Technology, Dübendorf, Switzerland

Received: 15 March 2011 – Published in Atmos. Meas. Tech. Discuss.: 26 April 2011

Revised: 26 July 2011 – Accepted: 1 September 2011 – Published: 16 September 2011

**Abstract.** We present an improved tropospheric nitrogen dioxide column retrieval algorithm (DOMINO v2.0) for OMI based on better air mass factors (AMFs) and a correction for across-track stripes resulting from calibration errors in the OMI backscattered reflectances. Since October 2004, NO<sub>2</sub> retrievals from the Ozone Monitoring Instrument (OMI), a UV/Vis nadir spectrometer onboard NASA's EOS-Aura satellite, have been used with success in several scientific studies focusing on air quality monitoring, detection of trends, and NO<sub>x</sub> emission estimates. Dedicated evaluations of previous DOMINO tropospheric NO<sub>2</sub> retrievals indicated their good quality, but also suggested that the tropospheric columns were susceptible to high biases (by 0–40%), probably because of errors in the air mass factor calculations. Here we update the DOMINO air mass factor approach. We calculate a new look-up table (LUT) for altitude-dependent AMFs based on more realistic atmospheric profile parameters, and include more surface albedo and surface pressure reference points than before. We improve the sampling of the TM4 model, resulting in a priori NO<sub>2</sub> profiles that are better mixed throughout the boundary layer. We evaluate the NO<sub>2</sub> profiles simulated with the improved TM4 sampling as used in the AMF calculations and show that they are highly consistent with in situ NO<sub>2</sub> measurements from aircraft during the INTEX-A and INTEX-B campaigns in 2004 and 2006. Our air mass factor calculations are fur-

ther updated by the implementation of a high-resolution terrain height and a high-resolution surface albedo climatology based on OMI measurements. Together with a correction for across-track stripes, the overall impact of the improved terrain height and albedo descriptions is modest (<5%) on average over large polluted areas, but still causes significant changes locally. The main changes in the DOMINO v2.0 algorithm follow from the new LUT and the improved TM4 sampling that results in more NO<sub>2</sub> simulated aloft, where sensitivity to NO<sub>2</sub> is higher, and amount to reductions in tropospheric NO<sub>2</sub> columns of up to 20% in winter, and 10% in summer over extended polluted areas. We investigate the impact of aerosols on the NO<sub>2</sub> retrieval, and based on a comparison of concurrent retrievals of clouds from OMI and aerosols from MODIS Aqua, we find empirical evidence that OMI cloud retrievals are sensitive to the presence of scattering aerosols. It follows that an implicit correction for the effects of aerosols occurs through the aerosol-induced cloud parameters in DOMINO, and we show that such an empirical correction amounts to a 20% AMF reduction in summer and ±10% changes in winter over the eastern United States.

## 1 Introduction

Nitrogen oxides (NO<sub>x</sub> = NO + NO<sub>2</sub>) are released into the atmosphere by anthropogenic and natural sources. These species largely control the production of ozone in the global troposphere (Jacob et al., 1996), and also affect OH



Correspondence to: Folkert Boersma  
(boersma@knmi.nl)

concentrations, thereby modifying the residence time of greenhouse gases and other pollutants (e.g. Shindell et al., 2009). The chemical decay product of  $\text{NO}_2$ ,  $\text{HNO}_3$ , contributes to nitrate aerosol formation (Basset and Seinfeld, 1983) and fertilization of soils and surface waters (Holland and Lamarque, 1997; Michaels et al., 1996). Global mapping of atmospheric  $\text{NO}_2$  concentrations can provide important information on  $\text{NO}_x$  emissions, on the formation of secondary pollutants, as well as on the transport and chemistry of tropospheric nitrogen oxides.

Since the mid-nineties, satellite remote sensing using spectral fitting techniques has been used to derive tropospheric  $\text{NO}_2$  concentrations on global, regional, and near-urban scales. Measurements from the Global Ozone Monitoring Experiment (GOME, Burrows et al., 1999) revealed hotspots of air pollution throughout the world (e.g. Leue et al., 2001; Martin et al., 2002), and showed episodes with long-range transport of nitrogen oxides from wildfires (e.g. Spichtinger et al., 2001). Since 2002, retrievals from the Scanning Imaging Spectrometer for Atmospheric Cartography (SCIAMACHY, Bovensmann et al., 1999) have mapped the  $\text{NO}_x$  pollution in finer spatial detail, and allowed the detection of significant trends in  $\text{NO}_2$  concentrations (Richter et al., 2005; van der A et al., 2008). With the launch of the Ozone Monitoring Instrument (OMI, Levelt et al., 2006) in 2004, the spatial detail (up to  $13 \times 24 \text{ km}^2$ ) improved to near-urban scales (Wang et al., 2007; Boersma et al., 2009a). In combination with mid-morning overpasses from SCIAMACHY and GOME-2 (Callies et al., 2000), the 13:40 h overpasses from OMI provide critical information on the timing of  $\text{NO}_x$  emissions and test our understanding of diurnal  $\text{NO}_x$ -chemistry (Boersma et al., 2008a, 2009a). In spite of these and many other successful applications of  $\text{NO}_2$  satellite measurements, a number of scientific questions about the accuracy of the retrievals remain. As concluded by the community workshop “Tropospheric  $\text{NO}_2$  measured by satellites” (September 2007), retrievals should be improved by reducing errors in the air mass factor (AMF), which is the dominant source of error in tropospheric  $\text{NO}_2$  retrievals over areas with enhanced  $\text{NO}_2$  (Boersma et al., 2004). The AMF defines the relationship between the  $\text{NO}_2$  abundance along the average photon path from the Sun through the atmosphere to the satellite (slant column) and the vertical column amount above a certain ground pixel. AMF calculations require external information on atmospheric scattering by air molecules, aerosols, and clouds, the shape of the  $\text{NO}_2$  vertical distribution, and on the surface albedo. The AMFs of the current OMI retrievals are based on external datasets which have coarse resolution compared to the small OMI pixels (Bucsela et al., 2006; Boersma et al., 2007). This spatial undersampling of the forward model parameters is a source of additional retrieval uncertainties beyond the ‘classical’ error budget as discussed in Boersma et al. (2004).

Here we describe the first major update of the Dutch OMI  $\text{NO}_2$  (DOMINO) retrieval algorithm version 1.02 originally

described in Boersma et al. (2007, 2009b). Data processed with DOMINO v1.02 were released in 2008. Since then, dedicated validation activities and model comparisons brought a number of DOMINO retrieval weaknesses to light. Table 1 summarizes the outcome of the comparisons, bearing in mind that the independent data used for the comparisons was imperfect. The independent data listed in Table 1 struggle with measurement errors and rely (amongst others) on assumptions on boundary layer thickness, vertical mixing, and spatial representativity. Even so, the table shows that DOMINO v1.02 generally agrees well with independent data, but appears to be biased high by 0–40%. The only study that suggested a possible seasonally dependent bias in the OMI retrievals was by Huijnen et al. (2010), who found good agreement between tropospheric  $\text{NO}_2$  from DOMINO v1.02 and regional chemistry transport models in winter, but reported that DOMINO tropospheric  $\text{NO}_2$  was much higher than model simulations in summer. In contrast, Lamsal et al. (2010) found that DOMINO v1.02 captures the seasonal variation in tropospheric  $\text{NO}_2$  over the United States well. Most studies cited in Table 1 made recommendations on how to improve the AMF calculations. In the DOMINO upgrade described here, we improve AMFs by better radiative transfer modelling, more accurate, higher-resolution descriptions of surface albedo and surface pressure, and using more realistic a priori vertical distributions of  $\text{NO}_2$ . Following on our earlier work (Boersma et al., 2004), we investigate the effect of aerosols in cloud-free scenes on the OMI  $\text{O}_2\text{-O}_2$  retrievals. This is important because  $\text{O}_2\text{-O}_2$ -retrieved cloud parameters are used to correct for cloud effects in the DOMINO  $\text{NO}_2$  retrieval.

It is unlikely that the separation of stratospheric and tropospheric  $\text{NO}_2$  slant columns leads to systematic, large-scale biases in the DOMINO tropospheric  $\text{NO}_2$  columns. A recent evaluation of the DOMINO stratosphere-troposphere separation scheme indicated that OMI stratospheric  $\text{NO}_2$  columns agree well with independent, ground-based measurements (Dirksen et al., 2011). However, DOMINO version 1.02 tropospheric  $\text{NO}_2$  columns do show a persistent, stripe-like pattern that indicates an across-track bias. This bias is caused by small jumps in the solar irradiance spectra due to measurement noise and wavelength calibration that are different from one OMI viewing angle to the other. In this study we will also describe and implement a simple method to correct for the across-track bias, but our main concern will be the improved calculation of the OMI  $\text{NO}_2$  AMF.

## 2 OMI tropospheric $\text{NO}_2$ retrieval

### 2.1 Ozone Monitoring Instrument

OMI is the Dutch-Finnish UV-Vis spectrometer on NASA’s EOS-Aura satellite. Aura was launched on 15 July 2004 into a Sun-synchronous orbit with a local equator crossing

**Table 1.** Summary of validation studies with DOMINO v1.02 tropospheric NO<sub>2</sub> data.

Study	Region	Period	Independent data	Result	Recommendation
Boersma et al. (2009)	Israeli cities (6 stations)	2006	NO <sub>2</sub> columns based on surface air NO <sub>2</sub> obs. <sup>1</sup>	Moderate agreement ( $r = 0.6$ ), insignificant bias.	
Hains et al. (2009)	Netherlands, Gulf of Mexico	09/2006 03/2006	Ground-based and aircraft NO <sub>2</sub> profiles	Good agreement ( $r = 0.8 - 0.9$ ), OMI biased high by 0–40 %	Implement OMI surface albedo database
Huijnen et al. (2009)	Northwestern Europe	2008–2009	Regional air quality models, surface air NO <sub>2</sub> measurements	Good agreement ( $r = 0.8$ ), OMI biased high by 0–40 %	Evaluate TM4 NO <sub>2</sub> profiles
Lamsal et al. (2009)	Southeastern United States	2005–2006	Surface air NO <sub>2</sub> measurements	Good agreement, OMI biased high by 0–40 %	Evaluate TM4 NO <sub>2</sub> profiles and destriping
Zhou et al. (2009)	Switzerland and Po Valley (35 stations)	2006–2007	NO <sub>2</sub> columns based on surface air NO <sub>2</sub> obs. <sup>2</sup>	Good agreement ( $r = 0.6 - 0.8$ ), OMI biased high by 0–60 %	Implement high resolution terrain height database
Zhou et al. (2010)	Europe	2006–2007	Not available	Choice of albedo set more important than accounting for BRDF effects	Implement OMI surface albedo dataset
Zyrichidou et al. (2009)	Southeastern Europe (6 stations)	2004–2008	Surface air NO <sub>2</sub> observations	Moderate agreement ( $r = 0.6$ )	

<sup>1</sup> NO<sub>2</sub> columns based on extrapolating observed surface air NO<sub>2</sub> concentration throughout the depth of the boundary layer (from a local climatology). <sup>2</sup> NO<sub>2</sub> columns based on extrapolating observed surface air NO<sub>2</sub> concentration throughout the depth of the boundary layer (from TM4).

time of approximately 13:40 h. Within the UV-Vis window that ranges from 270–500 nm, OMI detects direct and atmospheric backscattered sunlight. Two-dimensional CCD detectors are used to simultaneously record (ir)radiance spectra for 60 individual satellite viewing angles (rows). The dimensions of the ground pixels are 13–26 km along track and 24–128 km across track depending on the satellite viewing angle. Current scientific data products from OMI are based on Collection 3 level 1b data. Collection 3 data has been better calibrated than previous level 1 data versions (Dobber et al., 2008), although retrievals of minor trace gases based on Collection 3 spectra have shown spurious across-track variability, or stripes, since launch. This situation has deteriorated on 25 June 2007. From that date onwards, OMI has been affected by a number of so-called row anomalies that appear as signal suppressions in the level 1 radiance spectra for particular satellite viewing angles over the complete illuminated orbit. On 1 January 2011, 29 of the 60 rows were (partially) affected. The origin of

these anomalies is currently unknown. Until a satisfying correction for these row anomalies has been implemented, data from rows affected by the anomalies should not be used (see e.g. <http://www.knmi.nl/omi/research/calibration/instrument.status.v3/index.html>). On the other hand, OMI's radiometric stability is very good for a UV-Vis spectrometer. The optical degradation in the visible channel is less than 2 % over the period 2004–2010, and together with the good instrument performance, the OMI science data are generally considered to be of high quality for the full 6 yr period of the mission thusfar.

## 2.2 DOMINO retrieval algorithm

The Dutch OMI NO<sub>2</sub> retrieval algorithm consists of three steps: (1) using Differential Optical Absorption Spectroscopy (DOAS) to obtain NO<sub>2</sub> slant columns from the OMI reflectance spectra (Boersma et al., 2002, 2007; Bucsela et al., 2006), (2) separating the stratospheric and tropospheric

contribution to the slant column (Boersma et al., 2007; Dirksen et al., 2011), and (3) converting the tropospheric slant column to a vertical column with the tropospheric air mass factor (AMF). These principles are the same in the previous v1.02 algorithm and in the improved retrieval presented here, and they have been described in more detail elsewhere (Boersma et al., 2007, 2009b).

We chose a wide fitting window of 405–465 nm for the spectral fitting of NO<sub>2</sub> in order to compensate for OMIs moderate signal-to-noise ratio compared to other sensors. The absorption cross sections of NO<sub>2</sub>, ozone, H<sub>2</sub>O, as well as a synthetic Ring spectrum, are fitted to the satellite reflectance spectra. A fifth order polynomial is included in the fit to account for scattering effects. Annual average irradiance measurements for the year 2005 are used as reference for the reflectance spectra. The uncertainty in individual retrievals due to spectral fitting is  $0.7 \times 10^{15}$  molecules cm<sup>-2</sup> and dominates the overall retrieval error over the oceans and remote areas (Boersma et al., 2007). The NASA Standard Product retrieval (Bucsela et al., 2006) uses the same OMI NO<sub>2</sub> slant columns as the DOMINO retrievals discussed here.

In the second retrieval step, the stratospheric contribution is separated from the total slant column. The stratospheric NO<sub>2</sub> slant column is estimated by assimilating OMI total slant columns in the TM4 chemistry-transport model (Dentener et al., 2003; Boersma et al., 2007). The average (annual) differences between stratospheric NO<sub>2</sub> columns from DOMINO and independent, ground-based techniques were recently shown to be smaller than  $0.3 \times 10^{15}$  molecules cm<sup>-2</sup> (Dirksen et al., 2011).

The third retrieval step is the focus of this paper. We apply the tropospheric AMF formulation of Palmer et al. (2001) and Boersma et al. (2004) to convert the resulting tropospheric slant columns into vertical columns. Because the slant optical thickness of NO<sub>2</sub> is generally <0.005, the AMF can be written as the linear sum of atmospheric layer contributions to the slant column ratioed by the vertical column:

$$M = \frac{\sum_l m_l(\hat{\mathbf{b}}) x_{a,l}}{\sum_l x_{a,l}} \quad (1)$$

with  $m_l$  the altitude-dependent AMFs that describe the vertically resolved sensitivity to NO<sub>2</sub>, and  $x_{a,l}$  the layer specific subcolumns from the a priori profile  $\mathbf{x}_a$  for atmospheric layer  $l$ . The altitude-dependent air mass factors are calculated by adding a finite amount of NO<sub>2</sub> to layer  $l$  and subsequently ratioing the NO<sub>2</sub> slant column (simulated with a radiative transfer model) to the vertical column added to that layer ( $m_l = \partial N_s / \partial x_l$ , see Eskes and Boersma, 2003 for more detail). As radiative transfer model we use the Doubling Adding KNMI (DAK) model (Stammes, 2001) version 3.0. The altitude-dependent air mass factors are stored in a look-up table as a function of forward model parameters  $\hat{\mathbf{b}}$ , including the satellite viewing geometry, surface pressure and albedo. Pixel-specific altitude-dependent air mass

factors are obtained by using the best estimates for forward model parameters  $\hat{\mathbf{b}}$ , and an interpolation scheme. The previous DOMINO dataset (v1.02) uses a priori NO<sub>2</sub> profiles based on the TM4 assimilation run at a resolution of  $2^\circ \times 3^\circ$  (lat  $\times$  lon) and 35 vertical levels up to 0.38 hPa and spatially interpolated to the OMI pixel center. Similarly, DOMINO v1.02 uses interpolated surface pressures from the  $2^\circ \times 3^\circ$  TM4 model, which is driven by operational meteorological fields from ECMWF. Surface albedo information in v1.02 is from the combined  $1^\circ \times 1.25^\circ$  TOMS/GOME climatologies (Boersma et al., 2004) for the period October 2004 to February 2009. From 17 February 2009 onwards, the DOMINO v1.02 retrievals were based on the surface albedo from the  $0.5^\circ \times 0.5^\circ$  OMI climatology (Kleipool et al., 2008), but the impact of this update on AMFs has not yet been studied. Any changes in the AMF are important to data users because such changes affect the altitude-dependent air mass factors ( $m_l$ ), and these, combined with the a priori NO<sub>2</sub> profile, determine the averaging kernel (Eskes and Boersma, 2003), which is provided in the data product.

The AMF formulation accounts for cloud-contaminated pixels. Following Martin et al. (2002) and Boersma et al. (2002), we use the independent pixel approximation to express the AMF as a linear combination of a clear-sky AMF and a cloudy AMF:

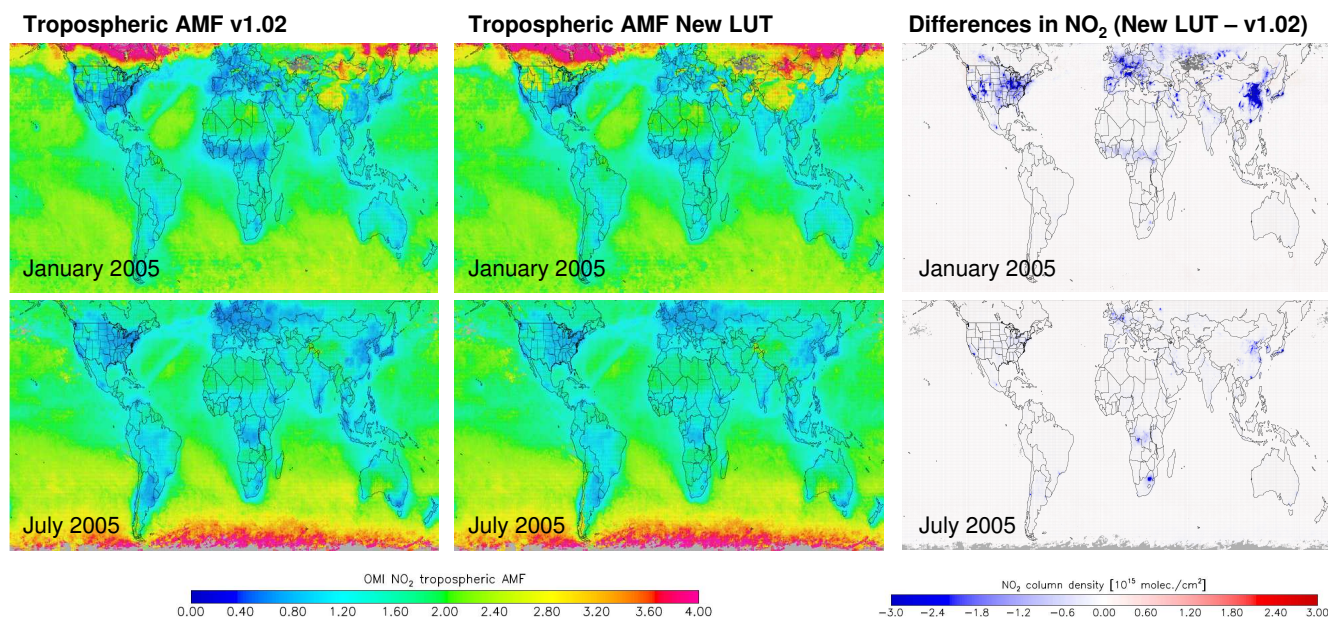
$$M = w M_{cl} + (1 - w) M_{cr} \quad (2)$$

with  $w$  the cloud radiance fraction that depends on the effective cloud fraction ( $w = f_{cl} I_{cl} / R$  with  $I_{cl}$  the radiance from the cloud part of the pixel, and  $R$  the total scene radiance), and  $M_{cl}$  and  $M_{cr}$  signifying the cloudy-sky and clear-sky AMFs, respectively. Our AMF calculation uses the effective cloud fraction and cloud pressure from the OMI O<sub>2</sub>-O<sub>2</sub> retrieval (OMCLDO2, Acarreta et al., 2004). The O<sub>2</sub>-O<sub>2</sub> retrieval is consistent with the NO<sub>2</sub> retrieval in the sense that both use the independent pixel approximation which represents clouds as opaque Lambertian surfaces of albedo 0.8.

AMFs are very sensitive to assumed surface albedo. Clear-sky AMFs ( $M_{cr}$ ) increase with increasing surface albedo (Boersma et al., 2004). Cloudy-sky AMFs ( $M_{cl}$ ) depend on the assumed albedo of 0.8 for the Lambertian reflector, but they are independent of the assumed surface albedo. Because the radiance-weighted cloud fractions  $w$  decrease with increasing surface albedo, the magnitude of the effect of changes in surface albedo on the AMF is difficult to predict. We will investigate this issue further in Sect. 3.3 where we discuss the direct (clear-sky) and indirect (partly cloudy) impact of improved OMI surface albedos (Kleipool et al., 2008) on the DOMINO retrievals.

### 3 Improved DOMINO AMF calculations for v2.0

Here we focus on improving four different aspects of the DOMINO air mass factors: the improved (1) radiative



**Fig. 1.** Tropospheric air mass factors (AMF) calculated for v1.02 (left panels) and calculated with the new AMF lookup table (middle panels). The right panels indicate the differences between tropospheric  $\text{NO}_2$  columns as calculated with the new AMF LUT minus tropospheric  $\text{NO}_2$  columns calculated with the v1.02 AMF LUT. Values are monthly means for January 2005 (upper row) and July 2005 (bottom row) based on clear-sky (cloud radiance < 50 %) observations.

transfer calculations, (2) terrain heights, (3) surface albedo, and (4) sampling of the TM4 model. All these are important aspects by themselves, and we will discuss the effects of the improvements on the retrieval separately. Section 5 discusses the combined effect of all improvements on v2.0 together.

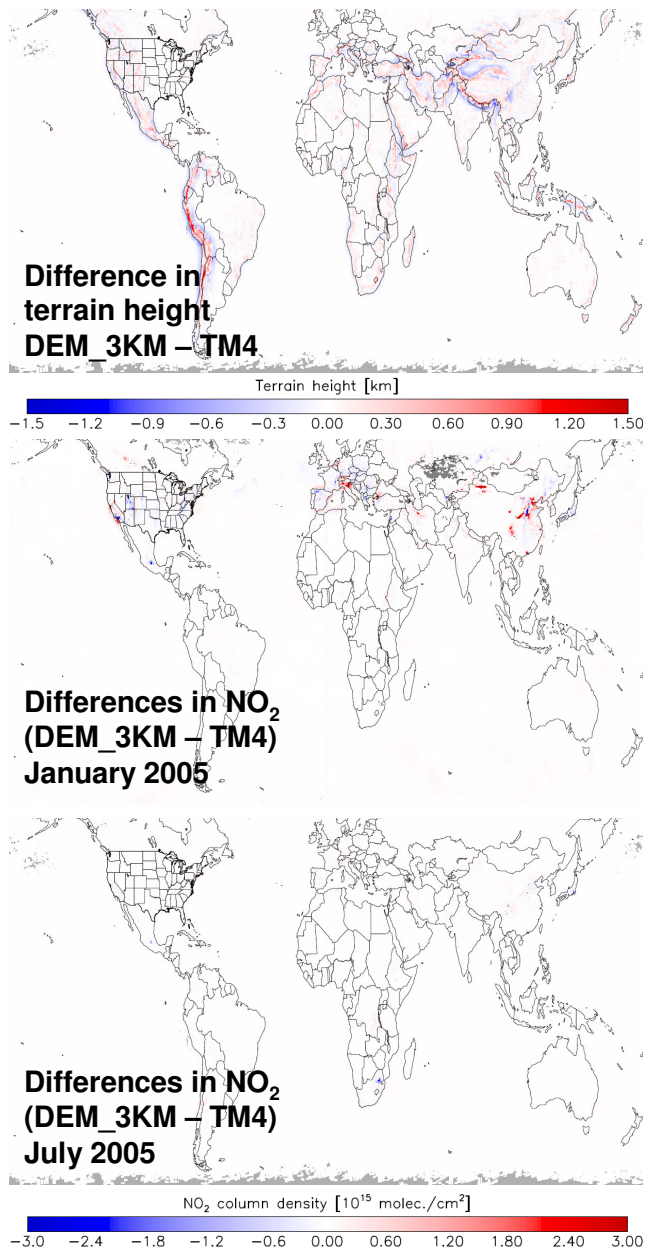
### 3.1 Altitude-dependent AMFs

Retrieving tropospheric  $\text{NO}_2$  columns from solar backscatter observations requires information about the vertical sensitivity to  $\text{NO}_2$ , or the averaging kernel. These sensitivities, expressed as altitude-dependent AMFs for both clear and cloudy parts of the pixel, are calculated with the DAK radiative transfer model. The DAK model atmosphere consists of a Lambertian surface albedo, and 24 atmospheric layers. Atmospheric data are from the standard AFGL midlatitude summer profile. We calculate the AMF at 439 nm, in the middle of the spectral fitting window for the corresponding OMI  $\text{NO}_2$  slant column retrievals. Using a midlatitude winter atmosphere profile ( $p$ ,  $T$ ) instead of a summer profile would change the tropospheric AMFs by 1%. Our calculations have been done with a plane parallel version of DAK that accounts for polarization. This is consistent with DAK settings in retrievals of cloud properties and surface albedo from OMI (Acarreta et al., 2004; Kleipool et al., 2008). Although polarization is accounted for, its impact is small, and reduces tropospheric AMFs by <0.5 % and affects stratospheric retrievals by <0.1 %.

The altitude-dependent AMFs are stored in a look-up table (LUT) as a function of solar zenith angle, viewing zenith angle, relative azimuth angle, Lambertian surface albedo, surface pressure, and (midlevel) atmospheric pressure. Compared to earlier versions of our AMF LUT used in OMI  $\text{NO}_2$  retrievals, we extend here the set of surface albedo (from 10 to 16) and surface pressure (from 10 to 15) reference points in the LUT, and include a more realistic pressure and temperature profile. In addition, we eliminate an interpolation error in calculating the altitude-dependent AMF from the LUT for the lowest layer, as suggested by Zhou et al. (2009). For any (TM4) pressure level, the altitude-dependent AMF is found by interpolating between the LUT-values at the two adjacent reference pressure levels. For reference pressure levels exceeding the actual (TM4) surface pressure, the altitude-dependent AMF was assigned a value of zero (assuming that the sensitivity to below-surface  $\text{NO}_2$  is zero). This led to too low (interpolated) altitude-dependent AMFs for the lowest TM4 model layer. By extrapolating the higher-layer AMF curve downward, we now avoid interpolation and obtain more realistic altitude-dependent AMFs for the lowest model layer.

Figure 1 shows the global distribution of the monthly mean January and July 2005 tropospheric AMF for OMI  $\text{NO}_2$  calculated with the old (v1.02, left panels) and new AMF LUT (middle panels) for clear-sky situations (cloud radiance fraction < 50 %). The AMFs are smallest over polluted regions indicating reduced sensitivity to  $\text{NO}_2$  in the boundary layer.





**Fig. 2.** Difference in terrain height calculated with the Global 3km Digital Elevation Model data (DEM\_3km, upper panel). The middle panel indicates the differences in tropospheric NO<sub>2</sub> columns for January 2005 as calculated with the DEM\_3km terrain heights and with the coarse TM4-derived heights as in v1.02. The lower panel indicates the differences for July 2005. Values are monthly means based on clear-sky (cloud radiance < 50 %) observations.

AMFs are relatively high over areas with high surface albedo (e.g. the Alps and much of Mongolia in January 2005) and over the oceans where the larger fraction of NO<sub>2</sub> is in the free troposphere. Over polluted regions, tropospheric AMFs with the new LUT are larger by 20–30 % (January) and 10–15 %

(July), leading to reductions in tropospheric NO<sub>2</sub> columns of 20 % (January) and 10 % (July) over North America, Europe, and eastern Asia. Over snow-covered terrain, AMFs increase by up to 100 %, but the absolute reduction in NO<sub>2</sub> columns is marginal over these mostly clean regions. The AMF increases are smaller in July than in January because of the increase in mixing and depth of the boundary layer, reducing the relative importance of downward extrapolation for the lowest layers.

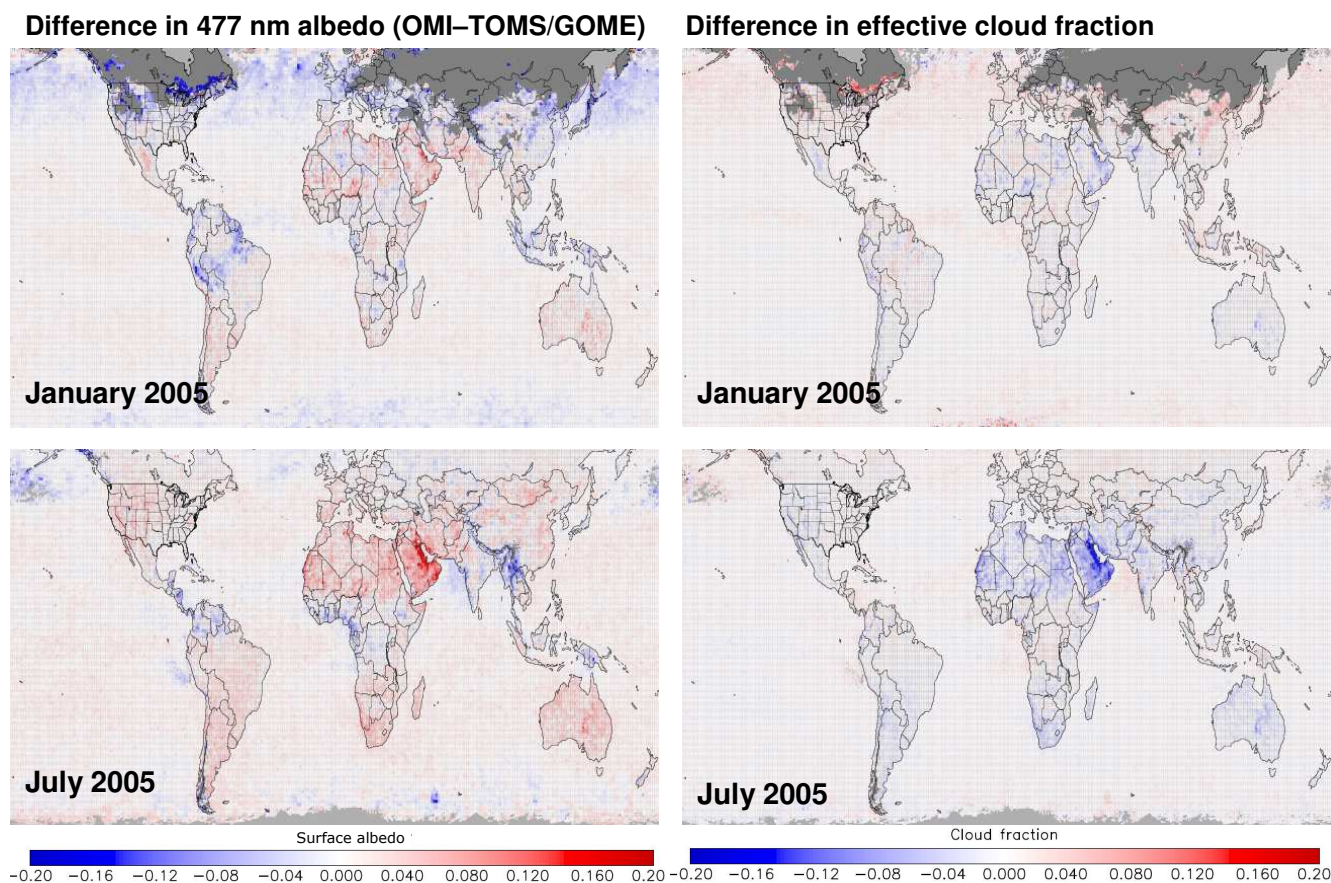
### 3.2 Terrain height

The AMF calculation also requires local information about the surface pressure. In earlier retrievals this information was obtained from the coarse-resolution (2° × 3°) TM4 model, driven by ECMWF meteorological data. Here we follow the approach presented by Zhou et al. (2009), and take more accurate surface pressures based on Global 3km Digital Elevation Model data (DEM\_3km), an Earth Science Data type routinely provided with EOS-Aura data. We convert the coarse-resolution TM4 surface pressure by applying the hypsometric equation and the assumption that temperature changes linearly with height, which is often used for reducing measured surface pressures to sea level (Wallace and Hobbs, 1977):

$$p_{\text{DEM}} = p_{\text{TM4}} \left( \frac{T_{\text{surf}}}{(T_{\text{surf}} + \Gamma(h_{\text{TM4}} - h_{\text{DEM}}))} \right)^{-g/R\Gamma} \quad (3)$$

where  $p_{\text{TM4}}$ ,  $T_{\text{surf}}$  are the TM4 surface pressure and temperature,  $\Gamma = 6.5 \text{ K km}^{-1}$  the lapse rate,  $h_{\text{TM4}}$  the TM4 terrain height, and  $h_{\text{DEM}}$  the 3km-resolution terrain height according to the DEM\_3km database.  $R = 287 \text{ J kg}^{-1} \text{ K}^{-1}$  is the gas constant for dry air, and  $g = 9.8 \text{ m s}^{-2}$  the gravitational constant. For the improved retrieval, the pressure levels for the a priori NO<sub>2</sub> profiles are based on the improved surface pressure level  $p_{\text{DEM}}$  (with  $p_i = a_i + b_i * p_{\text{DEM}}$  and  $a_i, b_i$  ECMWF constants that effectively define the vertical coordinate), so that mixing ratios are conserved.

Figure 2 (upper panel) shows the absolute difference between the new high-resolution terrain heights and the coarse TM4 (v1.02) terrain heights for July 2005. Over regions with pronounced orography, the smooth TM4 terrain heights underestimate the elevation of the highest mountains and overestimate the elevation of adjacent valleys, illustrated by marked transitions from red to blue. The improved terrain heights have marginal effect on the retrieved clear-sky (cloud radiance < 50 %) NO<sub>2</sub> columns for regions with little pollution or small differences in effective terrain heights as shown in the middle (January 2005) and bottom (July 2005) panels of Fig. 2. Averaged over the polluted regions extended over North America, Europe, and eastern Asia, the change in tropospheric NO<sub>2</sub> columns is within 1 % for January and July 2005 (Table 2). Locally the effects can be much stronger. Correcting for overestimated terrain heights leads



**Fig. 3.** Left column: difference in surface albedo between OMI (477 nm) and TOMS-GOME (479.5 nm) for monthly mean climatologies for January and July 2005. Right column: (OMI–v1.02) difference in retrieved effective cloud fractions from the  $O_2-O_2$  algorithm calculated with different surface albedo climatologies (v1.02: TOMS-GOME, improved algorithm: Kleipool et al., 2008). Scenes with snow or ice have been excluded.

to increased  $NO_2$  columns (e.g. Po Valley, Tehran), by reducing the – previously too high – AMF. Correcting for underestimated terrain height leads to increased AMFs (sensitivity for  $NO_2$  at higher altitude is higher) and reduced  $NO_2$  columns (e.g. Mexico City, Highveld area). A net increase in terrain height by 30 m for the polluted Los Angeles area reduces  $NO_2$  columns by 4 % during January and July 2005. Net decreases in terrain height by approximately 450 m over the Po Valley and Beijing areas lead to columns that are higher by 12–14 % in January and by 3 % in July 2005 (Table 3). The impact of improved terrain heights is stronger in winter than in summer, mainly reflecting the stronger sensitivity to errors in the surface pressure for situations with shallow boundary layers when most  $NO_2$  is concentrated near the surface.

### 3.3 Surface albedo

AMFs are affected by the surface albedo both directly and via the cloud retrieval. Both these aspects need to be considered

when implementing an improved surface albedo climatology in the  $NO_2$  retrieval. In previous OMI retrievals at KNMI, both cloud retrievals (Stammes et al., 2008) and  $NO_2$  AMF calculations used surface albedos from Lambert Equivalent Reflectance, or LER, data sets based on TOMS and GOME measurements. These estimates are limited by the spatial resolution of the TOMS ( $1^\circ \times 1.25^\circ$ ) and GOME ( $1^\circ \times 1^\circ$ ) climatologies. Kleipool et al. (2008) recently developed a surface albedo climatology using three years of OMI data with improved spatial resolution ( $0.5^\circ \times 0.5^\circ$ ). The higher spatial resolution of the OMI reflectance dataset reduces cloud contamination in the surface albedo compared to TOMS/GOME (that caused high-biased values in the dataset). Another major advantage of the Kleipool et al. (2008) climatology is that either the mode (most frequently observed value) or the 1 % cumulative probability threshold of the LER distribution has been used instead of the minimum LER as in the TOMS and GOME climatologies. Because of transient effects, like ground and cloud shading, and darkening by incidental precipitation, the minimum LER is likely to

**Table 2.** Summary of individual changes in regional tropospheric NO<sub>2</sub> retrievals relative to DOMINO v1.02. v2.0 refers to the combined effect of all individual changes.

Region <sup>1</sup>	Improved LUT	Improved terrain height	OMI surface albedo	Profile shape	Across-track variability	v2.0
North America (Jan. 2005)	−20 %	−0.6 %	+3 %	−1 %	+0.1 %	−18.9 %
Europe	−20 %	+0.7 %	+9 %	−2 %	−0.1 %	−11.3 %
Eastern Asia	−17 %	+0.9 %	−5 %	−4 %	−0.1 %	−13.9 %
Pacific	+0.02 × 10 <sup>15</sup> molec cm <sup>−2</sup>	n.a.	−0.02 × 10 <sup>15</sup> molec cm <sup>−2</sup>	n.a.	n.a.	−0.03 × 10 <sup>15</sup> molec cm <sup>−2</sup>
North America (Jul. 2005)	−7 %	−0.8 %	−2 %	−6 %	−0.3 %	−12.4 %
Europe	−8 %	−0.2 %	−0.3 %	−6 %	−0.1 %	−8.2 %
Eastern Asia	−10 %	−0.4 %	+3 %	−4 %	−0.4 %	−8.1 %
Pacific	−0.02 × 10 <sup>15</sup> molec cm <sup>−2</sup>	n.a.	n.a.	n.a.	n.a.	−0.02 × 10 <sup>15</sup> molec cm <sup>−2</sup>

<sup>1</sup> North America defined as region between 35°–45° N, and 100°–75° W. Europe: 40°–55° N, 5° W–20° E, eastern Asia: 30°–45° N, 110° E–140° E, Pacific: 40°–0° S, 160°–110° W.

**Table 3.** Summary of individual changes in local (1° × 1°) tropospheric NO<sub>2</sub> retrievals relative to DOMINO v1.02. v2.0 refers to the combined effect of all individual changes.

Region <sup>1</sup>	Improved LUT	Improved terrain height	OMI surface albedo	Profile shape	Across-track variability	v2.0
Los Angeles (Jan. 2005)	−40 %	−4 %	−14 %	−1 %	+0.5 %	−38.7 %
Po Valley	−29 %	+12 %	+10 %	−3 %	<0.1 %	+8.9 %
Beijing	−26 %	+14 %	−20 %	−5 %	+0.1 %	−2.7 %
Los Angeles (Jul. 2005)	−19 %	−4 %	−15 %	−4 %	+0.3 %	−30.1 %
Po Valley	−9 %	+3 %	−6 %	−8 %	+1.3 %	−14.1 %
Beijing	−15 %	+2 %	+7 %	−7 %	−0.3 %	−5.3 %

<sup>1</sup> Los Angeles defined as 1° × 1° box centered at 34° N, and 118° W. Po Valley: 45° N, 9° E, Beijing: 40° N, 116.5° E.

underestimate the overall albedo of a scene. Furthermore, the Kleipool et al. (2008) climatology represents the surface albedo at approximately 13:40 local time, consistent with the OMI observations. Using the OMI albedo climatology for OMI NO<sub>2</sub> retrievals also reduces errors arising from unaddressed instrumental effects and long term trends associated with TOMS/GOME but not with OMI. Furthermore it ensures that the optimal LER is derived under the same illumination conditions (solar zenith angle) so that Bidirectional Reflectance Distribution Function (BRDF) effects are reduced.

### 3.3.1 Effect of surface albedo changes on OMI cloud parameters

Figure 3 (left panels) shows the monthly mean difference in surface albedo between the OMI (479.5 nm) and the TOMS/GOME (477 nm) climatologies for January and July 2005. Scenes flagged as covered by snow or ice in the OMI data have been excluded. Large negative differences

occur over regions where the GOME climatology most likely suffered from contamination by residual cloud or snow/ice. This is observed, for instance, over tropical rain forests in the case of clouds (January and July), and over the northern parts of North America, in the Rhine Valley, and interior China in case of snow (January). For these regions, the TOMS/GOME data set is more likely to report contaminated and therefore higher surface albedos, because of the larger GOME pixels and smaller data volume analysed than in the case of OMI. Surface albedos for other regions over land are mostly higher in the OMI LER climatology due to the selection of the mode rather than the minimum LER. On average, the albedo differences between OMI and TOMS/GOME (477 nm) in the 60° S to 60° N region are within 0.001 with a standard deviation smaller than 0.012 (see Table 4).

Retrieved cloud parameters are sensitive to changes in the assumed surface albedo. Here we compare cloud parameters from the O<sub>2</sub>-O<sub>2</sub> algorithm retrieved with surface albedo data from Kleipool et al. (2008), to cloud parameters based on the



**Table 4.** Comparison of OMI and TOMS/GOME surface albedo climatologies.

	Mean difference (440 nm) (OMI-TOMS/GOME)	Standard deviation (440 nm)	Mean difference (477 nm) (OMI-TOMS/GOME)	Standard deviation (477 nm)
January 2005	+0.0002	0.012	−0.001	0.010
July 2005	−0.002	0.013	+0.001	0.012

TOMS/GOME surface albedo, as in DOMINO v1.02. The reprocessing of the O<sub>2</sub>-O<sub>2</sub> algorithm for the October 2004 – February 2009 timeframe ensures that we now have one consistent cloud dataset at our disposal for the complete duration of the OMI mission. The topography used in the O<sub>2</sub>-O<sub>2</sub> retrieval is DEM\_3KM, i.e. identical to that used in DOMINO v2.0. For more information on the O<sub>2</sub>-O<sub>2</sub> algorithm, we refer to the readme-file, available on [http://disc.sci.gsfc.nasa.gov/Aura/data-holdings/OMI/omcldo2\\_v003.shtml](http://disc.sci.gsfc.nasa.gov/Aura/data-holdings/OMI/omcldo2_v003.shtml). The right panels of Fig. 3 illustrate that for regions with higher surface albedo (e.g. Sahara, Arabian Peninsula), O<sub>2</sub>-O<sub>2</sub>-retrieved effective cloud fractions are reduced by up to 20%, and vice versa, for regions with lower surface reflectivity (e.g. South America). For the days 1 January and 1 July 2005 (and averaging over all retrieval scenes) the change in effective cloud fraction  $\Delta f_{cl}$  depends on the change in the (477 nm) surface albedo approximately as  $\Delta f_{cl} = -0.6\Delta a_{sf}$  (not shown), consistent with theoretical predictions by Koelemeijer et al. (2001). Using the OMI surface albedo climatology leads to a smaller global mean effective cloud fraction (0.303 instead of 0.309), and increases the number of pixels with cloud radiance fractions below 0.5 by 0.5%.

O<sub>2</sub>-O<sub>2</sub> cloud pressures are also sensitive to changes in the surface albedo, in particular when the effective cloud fractions are small. For retrievals with low cloud fractions (cloud radiance fractions <0.5, of most interest for our retrievals), we found that using the OMI climatology results, on average, in cloud pressures that are lower (by less than −5 hPa) when compared to those resulting from using the TOMS/GOME climatology. The small decrease in average cloud pressures is consistent with the small decrease in average effective cloud fractions (less than −0.006 for both months), because the same strength of the observed spectral O<sub>2</sub>-O<sub>2</sub> features needs to be explained by the cloud algorithm (e.g. Acarreta et al., 2004). We find that for situations with low cloud fractions (between 0.05 and 0.20), the change in effective cloud pressure approximately depends on the change in surface albedo as  $\Delta p_{cl} = -1400 \Delta a_{sf}$  hPa (an albedo increase of +0.01 leads to cloud pressures reduced by 14 hPa).

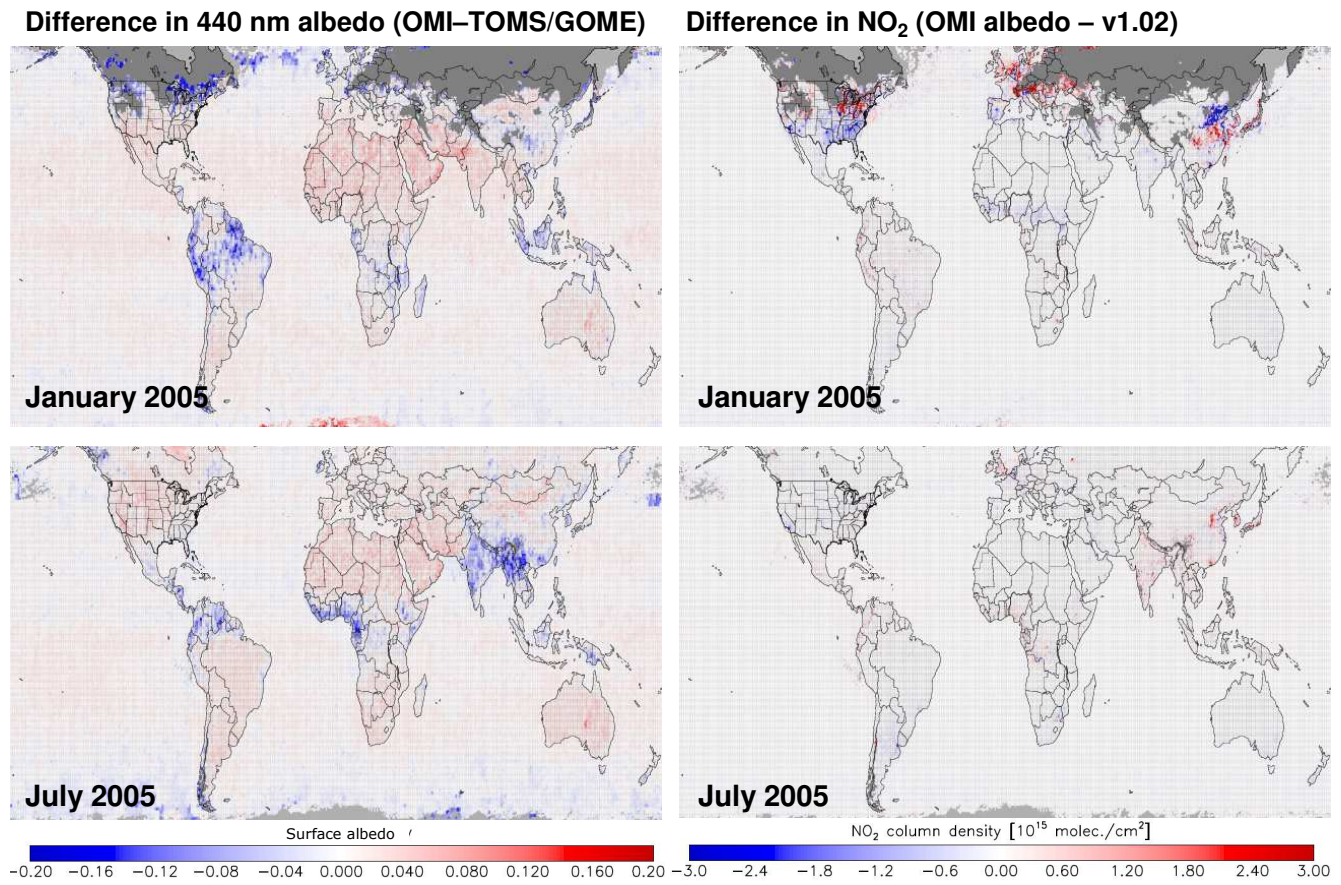
### 3.3.2 Effect of surface albedo changes on NO<sub>2</sub> retrievals

The left panels in Fig. 4 show the absolute differences between the OMI and TOMS/GOME (v1.02) surface albedo climatologies (440 nm) used in the clear-sky AMF calculations for January and July 2005. Scenes flagged as covered

by snow or ice in the OMI data have been excluded. The differences between the 440 nm albedos are broadly consistent with those at 479 nm shown in Fig. 3. One exception is the larger difference for desert areas where the increase at 479 nm is stronger than at 440 nm, which can be explained by the significant increase in albedo as a function of wavelength over deserts (e.g. Kleipool et al., 2008). Similar to 479 nm, we see large negative differences over areas that were contaminated by clouds or snow/ice in the TOMS/GOME climatology. Over land (with the exception of rainforests), the OMI climatology generally reports somewhat higher values than the TOMS/GOME set. On average, the differences between OMI and TOMS/GOME (440 nm) between 60° S and 60° N are within 0.002 with a standard deviation smaller than 0.013 (see Table 4).

From the right panels in Fig. 4 we see that the change in albedo has a negligible effect on the retrieved NO<sub>2</sub> columns for regions with little pollution. The effects of the improved albedo are best seen over the polluted areas of the Northern Hemisphere in winter, when the NO<sub>2</sub> lifetime is longest. Reductions in the 440 nm albedo generally lead to increases in retrieved NO<sub>2</sub> (northeastern United States, Europe). Vice versa, over polluted areas with increased albedo, we see a distinct reduction in NO<sub>2</sub> (southern United States, Spain, sub-Saharan Africa). Over northeastern China, the strong reduction in retrieved NO<sub>2</sub> is not accompanied by an increase in the 440 nm albedo. There, the reduction reflects the considerable increase in effective cloud fraction (+0.02) together with a significant increase (+23 hPa) in cloud pressures that are already high (~900 hPa) over this area. These relatively high cloud fractions and pressures may be indicative of large amounts of aerosols in the polluted boundary layer, as we will see in Sect. 6. Higher cloud fractions and higher cloud pressures imply increased sensitivity to NO<sub>2</sub> when the effective cloud pressure is situated within the polluted layer (e.g. Fig. 5a and b in Boersma et al., 2004). Averaged over the polluted regions of the Northern Hemisphere, the change in tropospheric NO<sub>2</sub> columns due to the improved surface albedo is within 10% in January 2005 and within 3% in July 2005 (see Table 2). The choice for albedo matters most in winter, because in that season most NO<sub>2</sub> is confined in a thin layer near the surface where the satellite vertical sensitivity depends strongest on albedo.

For the polluted hotspots Los Angeles (LA), the Po Valley, and Beijing, albedo changes are stronger than albedo changes averaged over larger polluted areas. Consequently,



**Fig. 4.** Left column: difference in surface albedo between OMI and TOMS-GOME (440 nm) for January and July 2005. Right column: difference (OMI – v1.02) in retrieved tropospheric NO<sub>2</sub> columns from the DOMINO algorithm calculated with different surface albedo climatologies (v1.02: TOMS-GOME, improved algorithm: OMI). Only scenes with cloud radiance fractions smaller than 0.5 have been selected, and scenes with snow or ice have been excluded.

the effects on retrieved NO<sub>2</sub> are stronger (see Table 3): significant increases in albedo over LA (January: +0.023, July: +0.027) lead to 15 % reductions in retrieved NO<sub>2</sub>, mostly reflecting increased clear-sky AMFs. In contrast, the reduced albedo over the Po Valley in January (–0.032) leads to a 10 % increase in NO<sub>2</sub>. Similar to the situation over eastern China, the reduction in NO<sub>2</sub> over Beijing in winter is largely attributed to a significant increase in cloud fraction (from 0.04 to 0.08) and in cloud pressure (from 928 to 950 hPa).

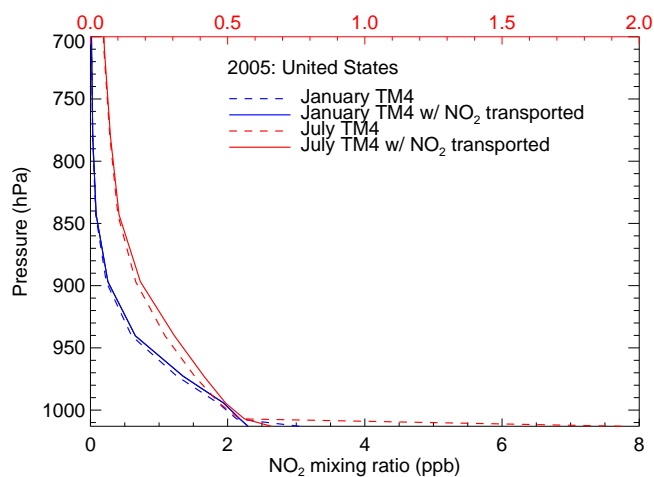
### 3.4 A priori vertical profiles

Previous studies (Hains et al., 2010; Huijnen et al., 2010) suggested that TM4 NO<sub>2</sub> profiles in polluted regions possibly suffer from too weak mixing in the lowest few model layers. A critical review of the TM4 output module pointed out that upon sampling the TM4 tracer field at 13:30 h local time, emissions, chemistry, deposition and vertical transport all have operated on NO<sub>x</sub>, but only the first two have operated

on NO<sub>2</sub>. In the TM4 model, the chemistry step computes NO<sub>2</sub> from NO<sub>x</sub>, but prior to deposition and vertical transport. The resulting 30-min “lag” in the vertical redistribution of NO<sub>2</sub> leads to profiles with artificially strong NO<sub>2</sub> concentrations in the lowest model layers and, consequently, with too little vertical NO<sub>2</sub> transport. By explicitly propagating NO<sub>2</sub> (as well as NO, NO<sub>3</sub>, N<sub>2</sub>O<sub>5</sub>, and HNO<sub>4</sub>) as tracers in TM4, we now ensure appropriate sampling of the NO<sub>2</sub> field, i.e. after NO<sub>2</sub> has been subject to deposition and convection.

Figure 5 shows a comparison between TM4 vertical NO<sub>2</sub> profiles simulated for January 2005 (blue) and July 2005 (red) over the United States. We see that the high surface layer concentrations in the original simulations (v1.02, dashed lines) are significantly reduced by the simulations with NO<sub>2</sub> as transported tracer in the model. These improvements are most prominent in Summer, when vertical transport is stronger than in winter, resulting in better mixed vertical distributions of NO<sub>2</sub> in that season.

We evaluate the improved TM4 vertical profiles of NO<sub>2</sub> at 13:30 h local time used as a priori in the AMF calculation.



**Fig. 5.** Vertical profiles of  $\text{NO}_2$  simulated with TM4 in January and July 2005. The  $\text{NO}_2$  profiles have been sampled at 13:30 h local time and averaged over the United States ( $100.5^\circ \text{W}$ – $76.5^\circ \text{W}$ ,  $31^\circ \text{N}$ – $51^\circ \text{N}$ ). The dashed lines represent the original simulations as used in DOMINO v1.02 retrievals. The solid lines represent the improved simulations with  $\text{NO}_2$  as a transported tracer in the model. The blue and red lines indicate the simulations for January and July 2005, respectively.

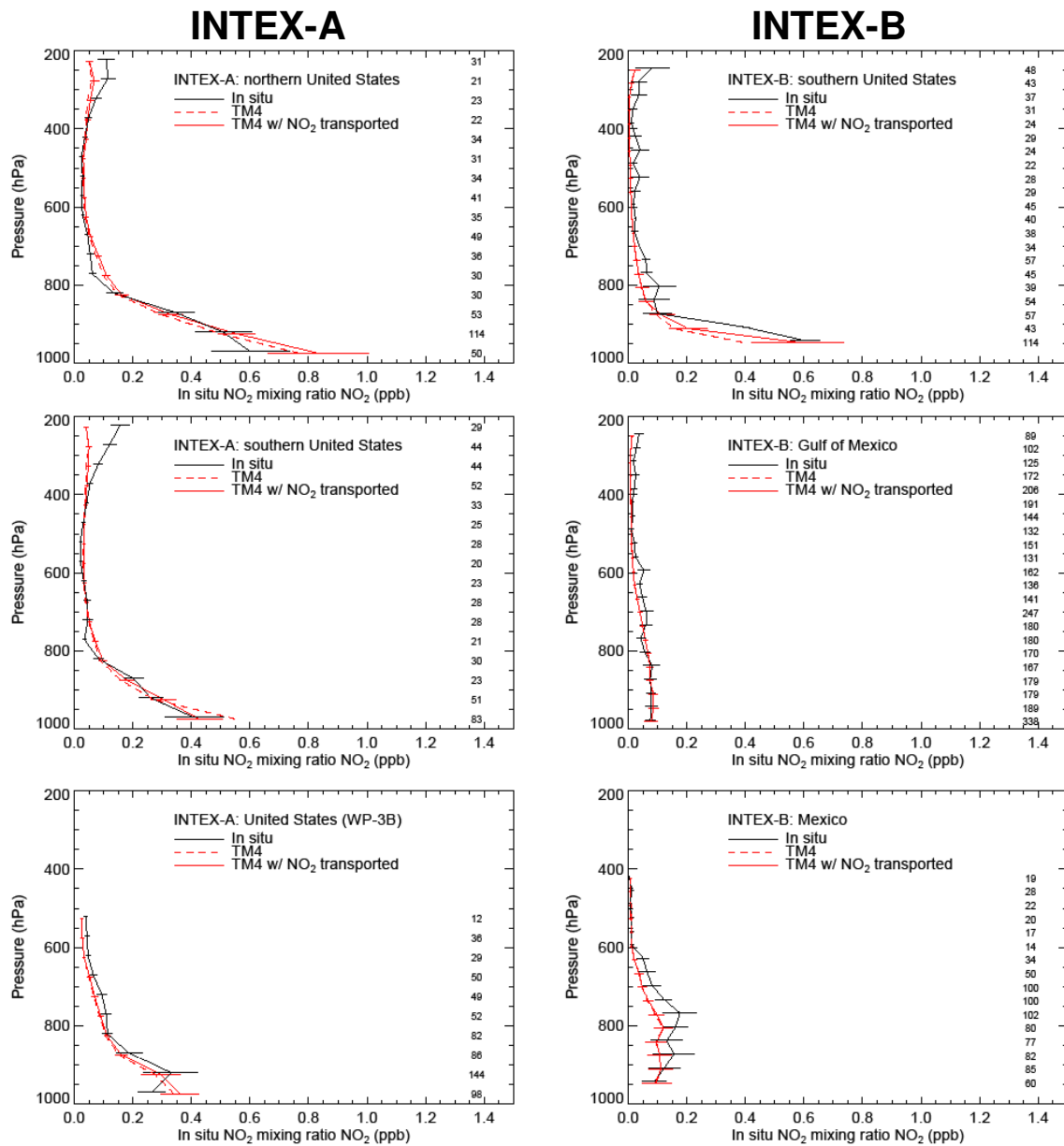
We compare to vertical  $\text{NO}_2$  profiles observed from NASA's DC-8 aircraft during the INTEX-A (July–August 2004) and INTEX-B (March 2006) field campaigns. The flight conditions during these campaigns ranged from remote marine to highly polluted. During INTEX-A, the DC-8 sampled tropospheric air over the eastern United States and the North Atlantic Ocean. NOAA's WP-3D also flew over the polluted east coast of North America and over the North Atlantic Ocean (ICARTT campaign). During INTEX-B, the DC-8 sampled the troposphere over the southern United States, and (the Gulf of) Mexico.  $\text{NO}_2$  was measured from the DC-8 by laser induced fluorescence (Thornton et al., 2000) with 0.02–0.05 ppb precision (for 1-s measurements), and from the WP-3D by chemiluminescence (Ryerson et al., 2000) during ICARTT with a precision of 0.025 ppbv for the 1-Hz data at low values of  $\text{NO}_2$ .

Figure 6 compares  $\text{NO}_2$  profile shapes simulated by TM4 to those observed from aircraft during the INTEX-A/ICARTT and INTEX-B campaigns. The TM4 simulations are sampled at 13:30 hours local time, nearly coinciding with the OMI overpass time, and consistent with our AMF calculation. All aircraft observations within three hours of the model sampling time have been selected. Aircraft measurements and model simulations for the INTEX-A/ICARTT campaign are generally highly consistent throughout the lower troposphere, but the model simulations underestimate  $\text{NO}_2$  in the upper troposphere by 0.1 ppb. This is likely due to an underestimate in mid-latitude lightning  $\text{NO}_x$  production reported earlier by Martin et al. (2006). During INTEX-B,

we find reasonable agreement between TM4 and the DC-8 observations for the southern United States and Mexico, and very good agreement over the Gulf of Mexico.

The TM4 simulation with  $\text{NO}_2$  propagated is in somewhat better agreement with the observations than the original simulation, especially for the southern United States, although the largest improvements are found below the aircraft bottom altitude of 150–300 m above surface (see Fig. 5). The underestimation by TM4 of  $\text{NO}_2$  over Mexico (0.1 ppb), reflects too low  $\text{NO}_x$  emissions from the POET inventory (1997) for that area. POET  $\text{NO}_x$  emissions are consistent with GEIA emissions (1999) that underestimate recently updated estimates from the Mexico National Emissions Inventory by a factor 1.6–1.8 (Boersma et al., 2008a). The AMFs calculated from the observed profiles are generally within 15 % of the AMFs calculated from simulated profiles (Hains et al., 2010). Our comparison suggests that boundary layer mixing in TM4, simulated with a non-local scheme, is vigorous enough to appropriately simulate vertical distributions of  $\text{NO}_2$ . The non-local scheme used in TM4 (Holtslag and Boville, 1993) has recently also been implemented in the GEOS-Chem CTM, which resulted in significantly improved simulations of vertical distributions for  $\text{NO}_2$  and  $\text{O}_3$  in that model (Lin and McElroy, 2010), and further supports using “non-local” mixing schemes for vertical tracer transport as in TM4.

Figure 7 shows the effect of the improved tropospheric air mass factors on OMI  $\text{NO}_2$  retrievals for January and July 2005. We see that, in both seasons, the improved simulations of  $\text{NO}_2$  profiles lead to reductions of more than  $1.0 \times 10^{15} \text{ molec cm}^{-2}$  over regions with strong  $\text{NO}_x$  emissions. These reductions are due to increases in the tropospheric AMFs that reflect generally higher  $\text{NO}_2$  concentrations aloft in our improved TM4 simulations. The impact of the improved TM4 profiles is strongest in summer, when vertical transport is stronger than in winter. This is reflected by Fig. 5 showing a larger difference between the original and improved TM4 simulation in July than in January 2005. In winter, we see small increases ( $< 0.5 \times 10^{15} \text{ molec cm}^{-2}$ ) over regions such as rural France and the Midwest of the United States, reflecting more efficient outflow towards these regions from the adjacent pollution hotspots in TM4. In summer, this effect is negligible because the  $\text{NO}_2$  lifetime is too short for  $\text{NO}_x$  to be transported in significant amounts to these regions. Averaged over the polluted regions of the Northern Hemisphere, the change in tropospheric  $\text{NO}_2$  columns due to the improved TM4  $\text{NO}_2$  profiles is within  $-4\%$  in January 2005 and within  $-6\%$  in July 2005 (see Table 2). For the polluted hotspots Los Angeles, the Po Valley, and Beijing, the effects of the improved TM4 sampling are somewhat stronger than averaged over the larger polluted areas (approximately  $-5\%$  and  $-8\%$  in January and July 2005, respectively). The changes in TM4  $\text{NO}_2$  profiles directly affect the retrieved  $\text{NO}_2$  columns, but model-retrieval comparisons using the averaging kernel will not be



**Fig. 6.** Vertical profiles of  $\text{NO}_2$  averaged over the INTEX-A/ICARTT (July–August 2004) and INTEX-B (March 2006) periods over the northern United States ( $100^\circ\text{W}$ – $87^\circ$ ,  $34^\circ\text{N}$ – $46^\circ\text{N}$ ), the southern United States ( $100^\circ\text{W}$ – $87^\circ$ ,  $28^\circ\text{N}$ – $34^\circ\text{N}$ ), the Gulf of Mexico ( $97^\circ\text{W}$ – $86^\circ$ ,  $20^\circ\text{N}$ – $27^\circ\text{N}$ ), and Mexico ( $100^\circ\text{W}$ – $97^\circ$ ,  $18^\circ\text{N}$ – $26^\circ\text{N}$ ) averaged over the INTEX-B period. The comparisons exclude strongly localized and recent pollution as diagnosed by  $\text{NO}_x/\text{NO}_y > 0.4$  or  $\text{NO}_2 < 4$  ppb. The black and red lines represent the average profiles from the in situ measurements and the TM4 model, respectively. The error bars represent the standard deviations divided by  $\sqrt{n}$ .

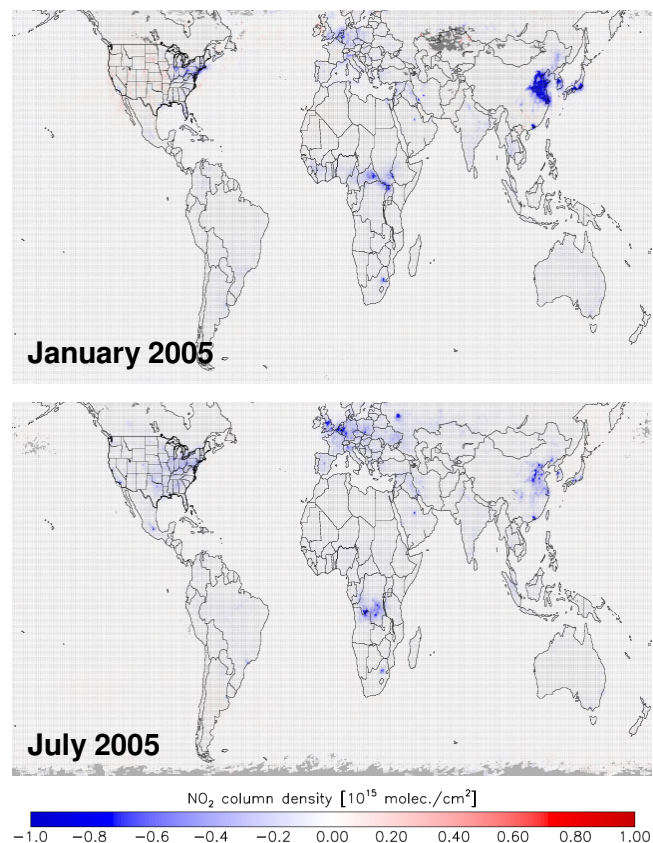
affected by changes in the TM4  $\text{NO}_2$  profiles, because in such comparisons, the dependence on the a priori (TM4)  $\text{NO}_2$  profile cancels (Eskes and Boersma, 2003; Boersma et al., 2004).

#### 4 Destriping

Since its launch in 2004, OMI retrievals have been suffering from spurious across-track variability, or stripes. Recent improvements in the calibration and data processing of the level 1b fields (Collection 3, Dobber et al., 2008) have suppressed striping, and further reductions have been achieved by using the annual average solar irradiance spectrum (2005) instead



### Difference in NO<sub>2</sub> (improved TM4 – v1.02)



**Fig. 7.** Differences in retrieved OMI tropospheric NO<sub>2</sub> columns as calculated with the improved TM4 simulations minus v1.02. Values are monthly means for January 2005 (upper row) and July 2005 (bottom row).

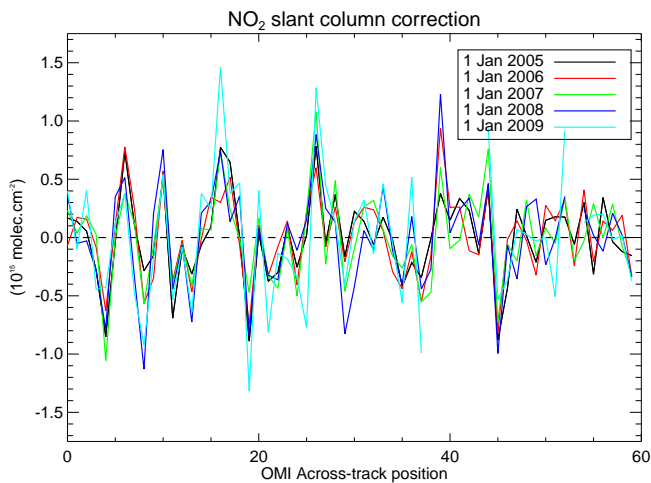
of individual, daily irradiance measurements for level 1 to 2 retrievals. But even after these improvements, OMI NO<sub>2</sub> retrievals still show some degree of striping. Here we use a simple method to remove the across-track bias in the OMI NO<sub>2</sub> slant columns.

OMI NO<sub>2</sub> slant columns are derived from fitting modeled spectra to observed reflectance spectra in the 405–465 nm window. The reflectance spectra are defined as the ratio of OMI radiance to the annual average irradiance spectra. For OMI, the spectral fit uses 60 independently observed irradiance spectra as the reference for the along-track radiance measurements. But even after averaging over a complete year, the OMI irradiance measurements for each of the 60 satellite viewing angles still have slightly different levels of noise. In combination with small differences in wavelength calibration for each of the 60 angles, this causes small, but systematic jumps in the top-of-atmosphere reflectance spectra (even if the radiance spectra would happen to be identical). Because the DOAS spectral fit is very sensitive to such

jumps from one viewing angle to the other, we observe spurious jumps in the NO<sub>2</sub> columns, with columns retrieved at some of the 60 viewing angles persistently higher or lower than the values at adjacent viewing angles. In principle, this situation is not different for scanning spectrometers that also contend with irradiance measurement noise and wavelength calibration errors, but since these instruments only have one single detector compared to OMI's 60 viewing angle specific CCD-detectors, such errors do not appear as stripes, but rather as constant, unknown offsets. This means that striping is inherent to hyperspectral imagers with 2-D detectors. Taken together, the jumps in NO<sub>2</sub> columns retrieved from OMI resemble a stripe-like pattern that does not represent "true" geophysical variability in NO<sub>2</sub>.

Over relatively clean regions, away from sources of pollution and at low and mid-latitudes, NO<sub>2</sub> columns are not expected to vary significantly across-track. The difference in local time between the far left (west) and far right (east) side of the OMI track is on the order of an hour (depending on latitude), too small to result in an appreciable chemistry-induced decrease between the left (early) and right (later) parts of the orbit (see e.g. Boersma et al., 2008b and Dirksen et al., 2011). We propose here to use a simple box-car averaging method to obtain a better, destriped measure of the true NO<sub>2</sub> column than the original value. For OMI observations between 50° S and the equator, we average total NO<sub>2</sub> columns from 15 adjacent viewing angles for every along-track array of the 60 viewing angles. Because there is no decisive a priori information guiding us to prefer particular viewing angles over others, columns retrieved for different viewing angles are attributed the same weight. Any 60-element array with more than 17.5 % variability is discarded so as to avoid interpreting real geophysical variability (for instance from biomass burning) as spurious across-track variability. We then calculate the average difference between the smoothed and original NO<sub>2</sub> columns between 50° S and the equator as a function of across-track position, map these back to slant column corrections, and subsequently store these as de-striping correction along the complete orbit on a daily basis.

Figure 8 shows corrections for across-track variability computed for days in five different years (2005–2009). Although these corrections have been computed from independent data, they are still similar, reflecting the systematic character of the striping error. The corrections in Fig. 8 are well within  $1.5 \times 10^{15}$  molec cm<sup>-2</sup>, a factor 3 smaller than our earlier corrections for retrievals based on Collection 2 data (Boersma et al., 2007). That de-striping corrections for Collection 3 (improved irradiance spectrum calibration) are much smaller than corrections for Collection 2 confirms that the stripes are dominated by across-track jumps in the solar reference spectrum. This is further supported by corrections for July 2005 being highly consistent with those for January 2005. Since 25 June 2007, a number of so-called row anomalies have occurred. These anomalies



**Fig. 8.** OMI NO<sub>2</sub> slant column destriping corrections computed for 1 January 2005–2009. The corrections represent the daily averages of 14 consecutive orbital corrections.

have affected the quality of level 1B and level 2 data products for particular viewing angles (rows, see also <http://www.knmi.nl/omi/research/product/index.php#row>). Our correction method discards rows (from the boxcar averaging) that have been affected by row anomalies occurring along the complete orbit, but works the same otherwise. From the corrections we obtain for 1 January 2008 (when row 53 was discarded) and 1 January 2009 (when rows 38–43 and 53 were discarded), we see that the method still results in similar but somewhat stronger corrections for the later years. The root mean square correction (per row) has increased from  $3.6 \times 10^{14}$  for 1 January 2005 to  $5.3 \times 10^{14}$  molec cm<sup>-2</sup> for 1 January 2009, reflecting the degradation in OMI and the fact that the 2005 average irradiance measurements are less appropriate as reference spectra for later years. Nevertheless, the monthly mean tropospheric NO<sub>2</sub> column fields shown on [www.temis.nl](http://www.temis.nl) suggest that the destriping remains effective, also for later years.

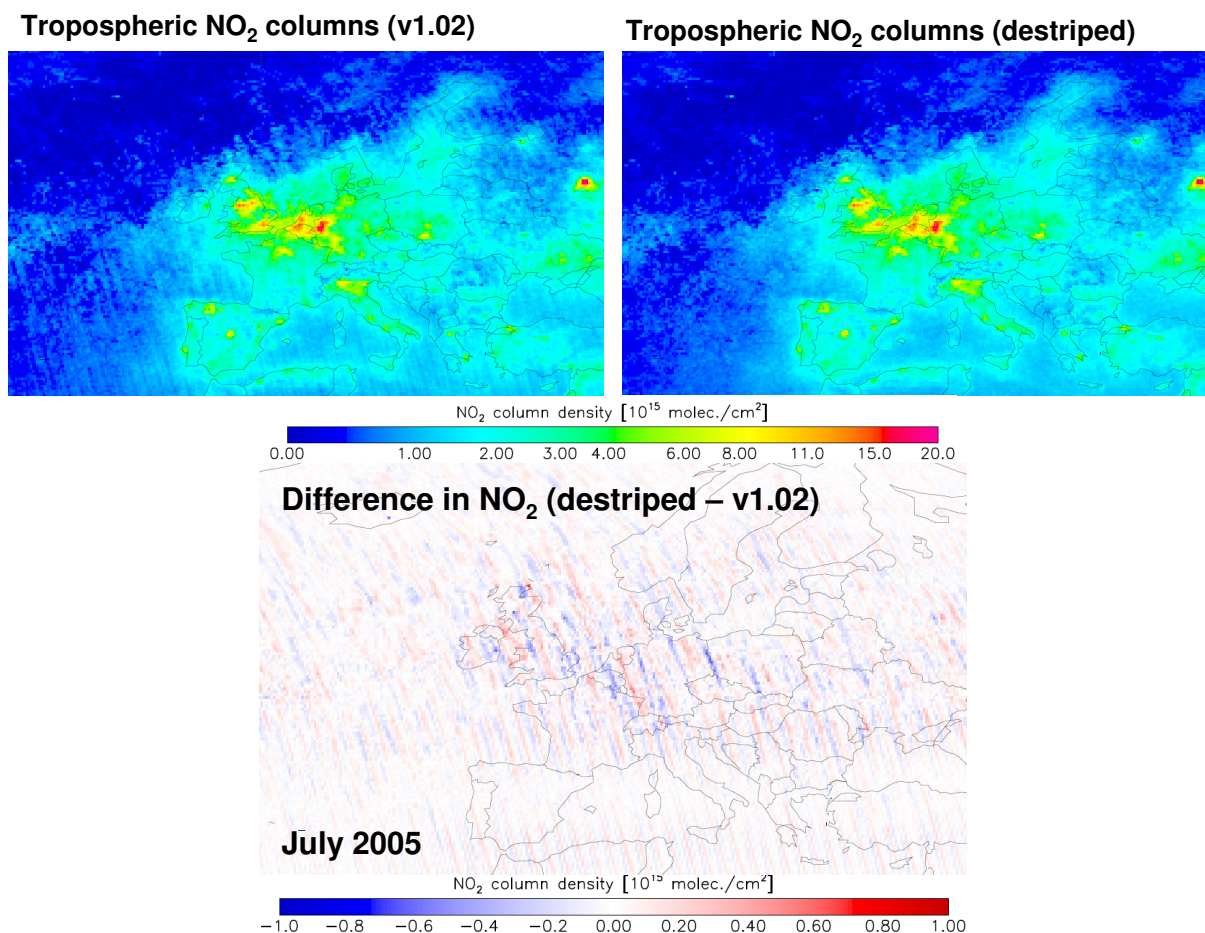
Figure 9 illustrates the effect of de-striping on monthly mean OMI tropospheric NO<sub>2</sub> columns over Europe in July 2005. We see that the de-striping corrections largely remove the artificial across-track pattern in v1.02. The lower panel of Fig. 9 shows that the local effects of de-striping on the tropospheric columns are strongest over the polluted region of northwestern Europe (up to  $0.5 \times 10^{15}$  molec cm<sup>-2</sup>), because of the amplifying effect of low AMF values there (see Fig. 1). Averaged over the large regions of the United States, Europe, and China, our destriped NO<sub>2</sub> columns are always within  $0.01 \times 10^{15}$  molec cm<sup>-2</sup> of the original retrievals (Table 2). We conclude that our destriping has little effect on regional levels of NO<sub>2</sub>, but are of importance for local studies, including validation.

## 5 Impact of the combined algorithm changes

After having discussed individual algorithm improvements, we now combine all the changes to evaluate the overall effect on the DOMINO NO<sub>2</sub> retrievals. Figure 10 and Tables 2 and 3 summarize the impact of the v2.0 algorithm on the DOMINO datasets. The tables illustrate that the sum of the individual changes is not necessarily the same as the overall impact, because some changes counteract others, and because of feedbacks (for instance between albedo and cloud effects, see Boersma et al., 2004). The left panels of Fig. 10 show the absolute differences between DOMINO-derived tropospheric NO<sub>2</sub> columns for the v2.0 and the v1.02 algorithm for January and July 2005. In winter, the changes are strongest (−10% to −20%) over the polluted regions of the world, reflecting the combined effect of individual improvements. The generally reduced NO<sub>2</sub> columns over polluted areas are due to higher AMFs following mainly from our improved altitude-dependent AMFs (look-up table) and also from improved TM4 sampling that results in more NO<sub>2</sub> simulated aloft, where sensitivity to NO<sub>2</sub> is higher. In January 2005, reductions in surface albedo counteract the decreases over the Rhine and Po Valley in Europe, near the Great Lakes in North America, and in interior China. The strong increases in tropospheric NO<sub>2</sub> over these regions result from the lower albedo in the Kleipool et al. (2008) dataset compared to the TOMS/GOME set that suffered from residual snow/ice over these areas (see upper panels of Fig. 4). The right panels in Fig. 10 show the differences between monthly mean v2.0 and v1.02 as a function of the original v1.02 retrieval. The slope of the black triangles in the right panels of Fig. 10 is somewhat larger in January than in 2005, in line with somewhat stronger relative differences between v2.0 and v1.02 in wintertime.

Summertime changes in retrieved NO<sub>2</sub> (−10%) are smaller than in winter because improved altitude-dependent AMFs for the lowest atmospheric layers have less effect in summer, with relatively more NO<sub>2</sub> aloft, than in winter. Together with the improved sampling of a priori NO<sub>2</sub> profiles from TM4, the improved altitude-dependent AMFs are driving the reductions in tropospheric NO<sub>2</sub>, but albedo reductions (Fig. 4) over the United Kingdom, parts of China and South Korea lead to the small increases in NO<sub>2</sub> over those areas that can be seen in lower left panel of Fig. 10.

The changes between v1.02 and v2.0 mostly -but not exclusively- reflect the impact of the improved forward model parameters on the retrieval. The changes in the forward model parameters also propagate through the observation operator (averaging kernel) in the data assimilation used to calculate the stratospheric columns (see Boersma et al., 2007; Dirksen et al., 2011). This effect is generally small, as indicated by tropospheric NO<sub>2</sub> columns differences on the order of  $10^{13}$  molec cm<sup>-2</sup> over the Pacific in Table 2, with the exception of the northern Pacific and the northwestern United States (January 2005) where tropospheric NO<sub>2</sub>



**Fig. 9.** Monthly mean OMI tropospheric NO<sub>2</sub> columns (July 2005) for v1.02 (left panel), after correction for the stripes (middle panel), and differences between destriped and original (v1.02) monthly mean OMI tropospheric columns (right panel). Cloudy scenes (cloud radiance fractions > 0.5) have been omitted in calculating the monthly mean.

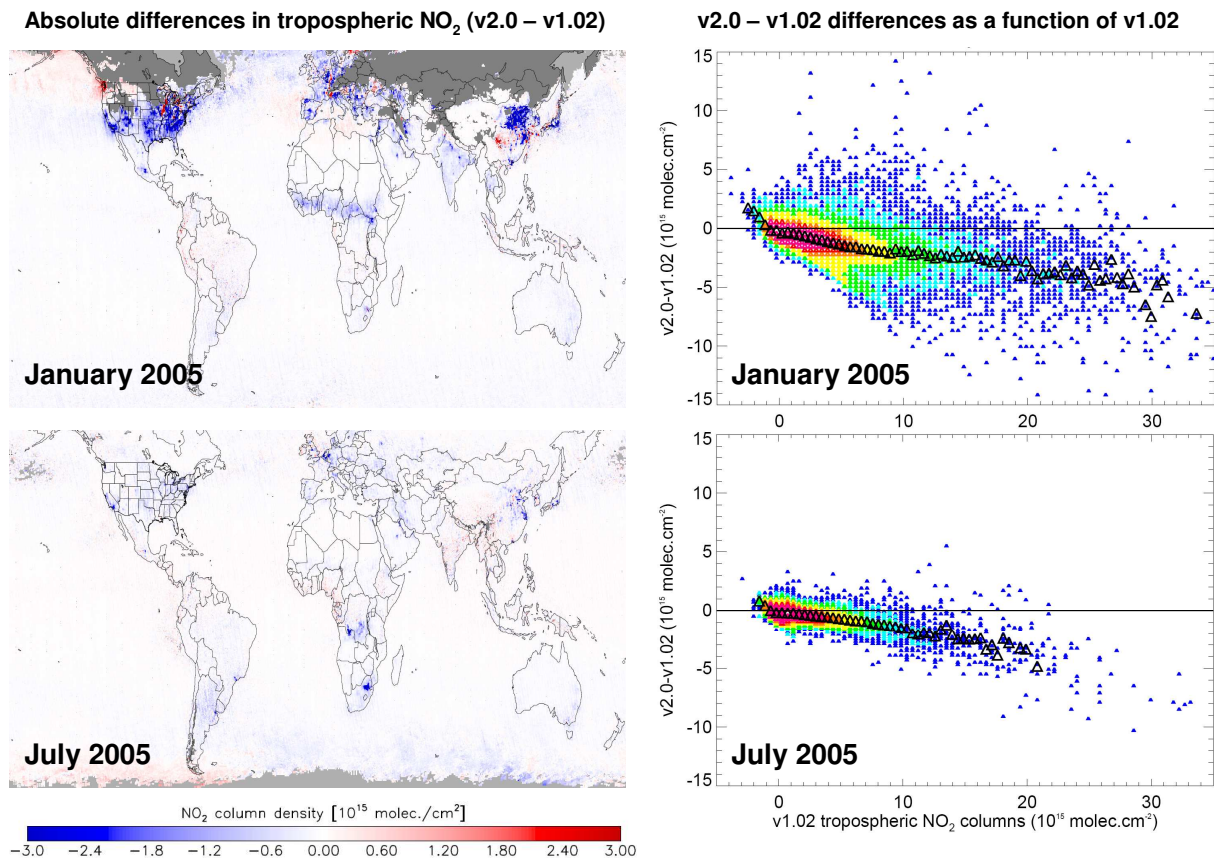
columns have increased because of reduced stratospheric NO<sub>2</sub> in v2.0. Users of the DOMINO data who apply the averaging kernel can expect the impact of the v2.0 algorithm to be somewhat smaller than indicated in Tables 2 and 3. When using the averaging kernel, the changes in a priori NO<sub>2</sub> profile shape are no longer relevant, and the reductions in NO<sub>2</sub> columns going from v1.02 to v2.0 will be smaller by a few percent.

The uncertainty in DOMINO v2.0 tropospheric NO<sub>2</sub> columns has probably decreased relative to v1.02. The improved spatial resolution and quality of the surface pressure and surface albedo data are likely to decrease the contribution of these parameters to the overall uncertainty. Because the errors from cloud parameters and a priori profile shapes propagate unabated, we cautiously estimate that the stated uncertainty for individual tropospheric NO<sub>2</sub> columns of  $1.0 \times 10^{15} \text{ molec cm}^{-2} + 30\%$  for v1.02 (Boersma et al., 2007) improves to  $1.0 \times 10^{15} \text{ molec cm}^{-2} + 25\%$  for v2.0 retrievals.

## 6 Implicit aerosol correction in DOMINO v2.0

Scattering and absorption by aerosols influence the top-of-atmosphere radiances measured by satellite instruments and can have significant effects on retrievals of atmospheric trace gases and clouds. The sensitivity of the satellite measurements for a particular trace gas can be increased (albedo effect) or decreased (screening), depending on the amount and optical properties of the aerosols, and its vertical distribution relative to that of the trace gas (e.g. Palmer et al., 2001; Martin et al., 2003; Fu et al., 2007; Leitão et al., 2010). In an earlier study with radiative transfer simulations (Boersma et al., 2004), we found that cloud retrievals are also sensitive to aerosols: higher cloud fractions and lower cloud pressure levels are retrieved in the presence of aerosols compared to a pure molecular scattering atmosphere. The good agreement between explicit corrections for aerosols and the actual correction through the AMF formalism (see Eq. (2)) in that study, showed that an implicit correction for aerosols occurs





**Fig. 10.** Monthly average difference between v2.0 and v1.02 DOMINO tropospheric NO<sub>2</sub> column retrievals for January 2005 and July 2005 (left panels). Cloudy scenes (cloud radiance fractions > 0.5) have been omitted in calculating the monthly mean. The right panels show the difference between v2.0 and v1.02 as a function of the original (binned) monthly mean v1.02 values. Blue symbols indicate bins filled with 1–3 values, light blue 4–9, green 10–29, yellow 30–99, orange 100–199, red 200–399, and purple more than 400 values. The black triangles indicate the mean difference for a particular bin.

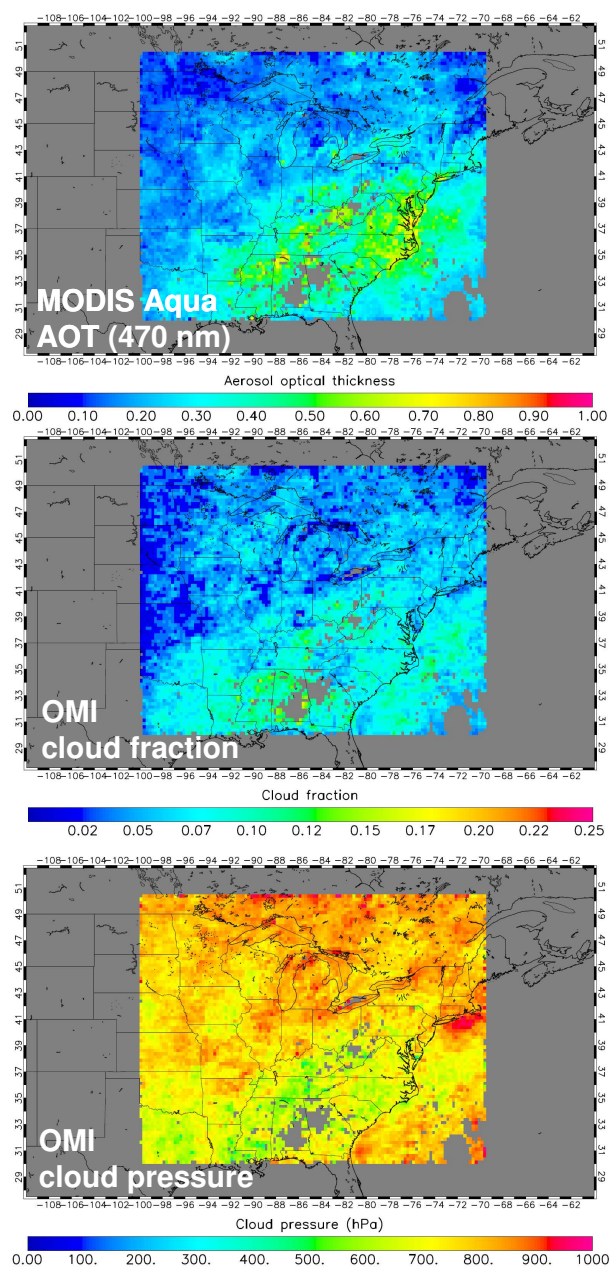
partially through the modified cloud fraction and pressure, at least in theory.

Here we investigate the impact of aerosols on O<sub>2</sub>-O<sub>2</sub> and NO<sub>2</sub> retrievals by taking advantage of near-simultaneous satellite measurements of aerosols and clouds from the A-Train (Stephens et al., 2002). We focus on clear-sky situations. MODIS onboard EOS-Aqua observes the Earth's atmosphere approximately 15 min prior to OMI onboard EOS-Aura. We use MODIS-Aqua aerosol optical thickness (AOT) data (Remer et al., 2008) that is retrieved exclusively for cloud-free situations. We subsequently correlate the MODIS observations to any collocated OMI O<sub>2</sub>-O<sub>2</sub> cloud parameters whenever the MODIS and OMI measurements were taken within 15 minutes of another, and store the observations onto a common  $0.25^\circ \times 0.25^\circ$  grid. Selecting OMI retrievals that are collocated in space and time with available MODIS AOT retrievals implies that the OMI measurements have been taken under (nearly) cloud-free conditions. In such situations, retrievals of non-zero O<sub>2</sub>-O<sub>2</sub> cloud frac-

tions are thus due to aerosol scattering leading to increased top-of-atmosphere radiances. This is in line with the use of a Lambertian reflector with fixed albedo (0.8) as the simplified model in the O<sub>2</sub>-O<sub>2</sub> retrieval. The fractional coverage of the Lambertian reflector that yields a top-of-atmosphere reflectance that best agrees with the observed reflectance, is interpreted as the radiometrically equivalent, or effective, cloud fraction (Stammes et al., 2008). The O<sub>2</sub>-O<sub>2</sub> retrieval does not distinguish between clouds or aerosols to explain the observed reflectances.

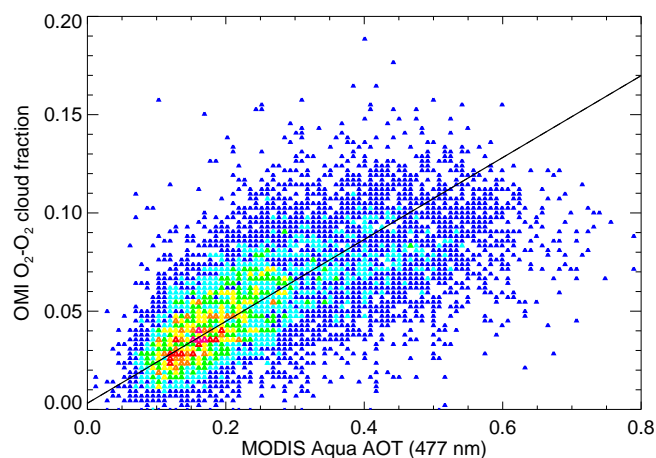
The upper panel of Fig. 11 shows the monthly mean AOT from MODIS-Aqua (470 nm) for July 2005 over the eastern United States. The middle panel shows the corresponding monthly mean of O<sub>2</sub>-O<sub>2</sub> effective cloud fractions from OMI sampled at the same locations and same time as the MODIS observations. We see that the AOT values are highest (up to 0.7) over the southeastern United States, and that this is reflected by effective cloud fractions with values up to 0.15. The MODIS Ångström exponent over the eastern United





**Fig. 11.** Monthly average aerosol optical thickness observed from MODIS Aqua at 470 nm (upper panel), and corresponding OMI O<sub>2</sub>-O<sub>2</sub> effective cloud fraction (middle panel) and effective cloud pressure (lower panel) for July 2005. Cloud fractions have been selected only for those days and locations that had a successful, cloud-free, MODIS AOT retrieval 15 min prior to the OMI observations. Grey areas indicate less than 3 successful coincidences in July 2005.

States is approximately 1.75, corresponding to small particles (e.g. Dubovik et al., 2002; Russell et al., 2010). Upon inspection of the OMI Absorbing Aerosol Index (Torres et al., 1998) during July 2005, we did not find any evidence for a significant contribution from absorbing particles to the



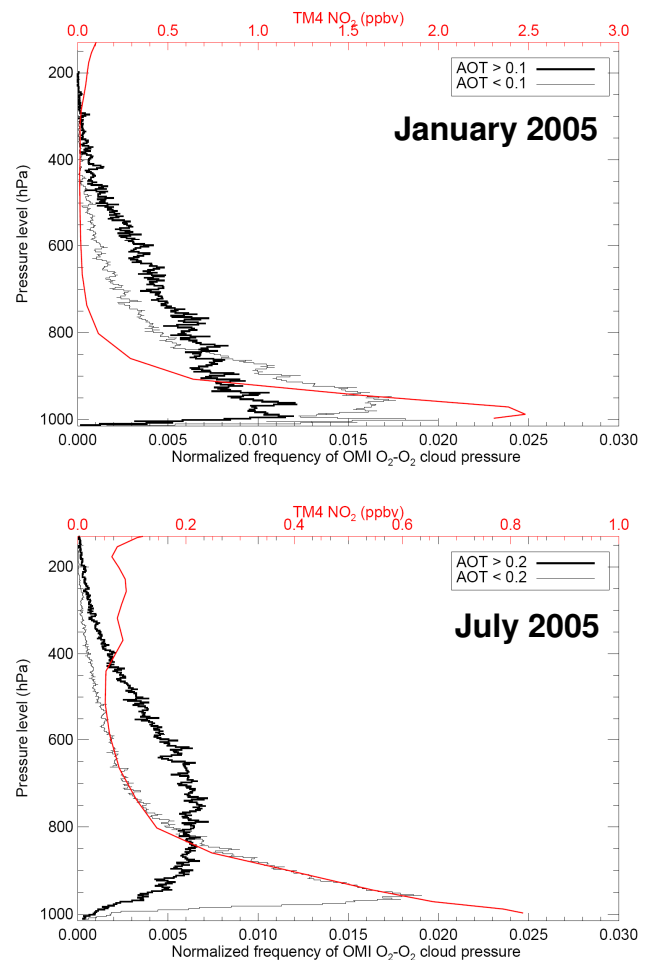
**Fig. 12.** Scatterplot for MODIS Aqua AOT at 470 nm (x-axis) and coinciding OMI O<sub>2</sub>-O<sub>2</sub> effective cloud fractions (y-axis) observed in July 2005 (13:30 h local time). The colours indicate the number of times a particular grid cell has been filled, where dark blue corresponds to 1 time and red to more than 6 times. OMI effective cloud fractions can be expressed as  $0.21 \times \text{AOT}$  (reduced major axis regression). Cloud fractions have been selected only for those days and locations that had a successful, cloud-free, MODIS AOT retrieval 15 min prior to OMI.

AOT. Instead, there is strong evidence that the high aerosol loadings originate from local emissions of volatile organic compounds (e.g. Goldstein et al., 2009; Jiang et al., 2010; Veefkind et al., 2011), and these aerosols are known to be highly reflective (Penning de Vries et al., 2009). Figure 12 shows significant positive correlation ( $r = 0.66$ ,  $n = 9685$ ) between OMI effective cloud fractions and MODIS AOT over the eastern United States. A reduced major axis regression suggests that the OMI effective cloud fractions can be expressed as  $f_{\text{cl}} = 0.21 \times \tau$ , with  $\tau$  the MODIS AOT. We conclude that OMI cloud retrievals are sensitive to the presence of scattering aerosols, particularly in situations with predominantly scattering aerosols, such as over the southeastern United States in July 2005.

The lower panel of Fig. 11 shows the monthly mean O<sub>2</sub>-O<sub>2</sub> effective cloud pressures over the eastern United States in July 2005. In the absence of enhanced effective cloud fractions, the O<sub>2</sub>-O<sub>2</sub> retrieval essentially returns values close to surface pressure (Acarreta et al., 2004) such as for the northern part of the domain. For high AOT (increased effective cloud fractions) in the southeastern United States, O<sub>2</sub>-O<sub>2</sub> pressures are lowest, indicative of elevated aerosol layers. The median O<sub>2</sub>-O<sub>2</sub> pressure corresponds to 720 hPa for hazy situations with MODIS AOT > 0.2. These elevations are somewhat higher than the significant amounts of aerosols well above the boundary layer in the summertime (south)eastern United States as observed with ground-based, airborne, and space-based lidars (Turner et al., 2001; Lewis et al., 2010; Yu et al., 2010). Because of differences

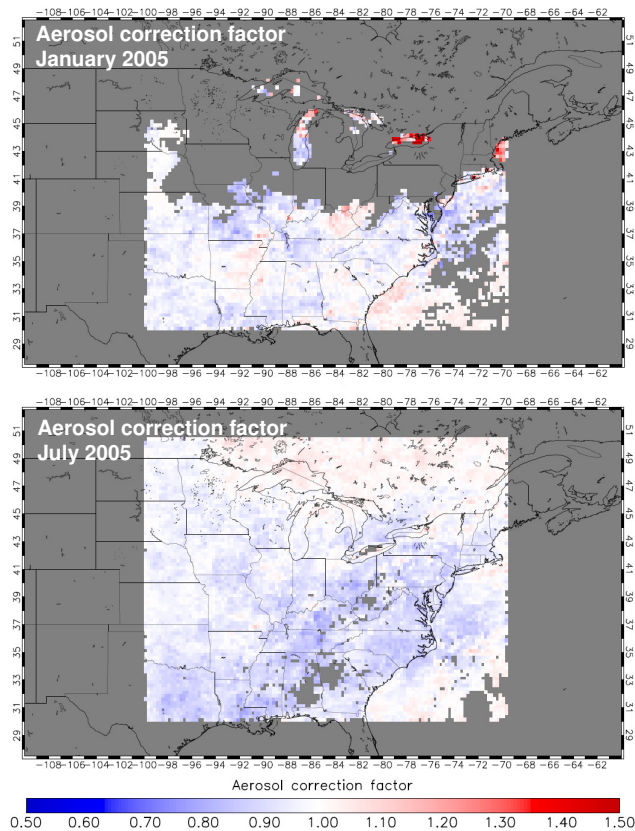
between the mid latitude standard atmosphere temperature profile used in the  $O_2-O_2$  retrieval and the actual, higher temperatures over the eastern United States in July 2005, it is likely that the number density of the  $O_2-O_2$  collision complexes is underestimated, leading to lower cloud pressures. Additional simulations suggest that the bias in cloud pressure is  $<50$  hPa for a surface temperature increase of 20 K relative to the mid latitude standard temperature for effective cloud fractions  $>0.2$ . However, for cloud fractions  $<0.2$ , the retrieved cloud pressures may be too low by up to 50–200 hPa. We therefore estimate that for a temperature increase of 10 K, which may be expected in this season over the eastern United States, the retrieved cloud pressures are too low by 25–100 hPa. Bearing these biases in mind, we examine the vertical differences between the cloud pressures and the a priori  $NO_2$  profile, by comparing the probability distribution of cloud pressures to the average  $NO_2$  vertical distribution simulated by TM4 under “hazy” conditions ( $AOT > 0.2$ ). Figure 13 (lower panel) shows that the scattering levels (due to aerosol scattering) mostly reside above the bulk of the  $NO_2$  in July 2005. A layer of scattering aerosols above an  $NO_2$  layer is most likely to have a screening effect, i.e. reduce the AMF compared to situations without aerosols. This is not different from the effect of clouds above a polluted layer that also tend to decrease the AMF (see for example Fig. 5a–b in Boersma et al., 2004).

We investigate whether an implicit AMF aerosol correction occurs through Eq. (2) with modified cloud parameters, now that we have shown that OMI cloud retrievals are obviously sensitive to scattering aerosols. Figure 14 shows the ratio of OMI  $NO_2$  AMFs calculated in our standard retrieval with the small, positive effective cloud fractions resulting from aerosol scattering ( $M$ ) to the clear-sky AMFs ( $M_{cr}$ ) with zero cloud fraction (consistent with successful MODIS AOT retrievals in the absence of clouds). This implicit aerosol correction factor, defined as  $M/M_{cr}$ , is 0.8–1.0 in the “hazy” region where  $AOT > 0.2$ , and close to 1.0 in the northern part of the domain with low AOT and small cloud fractions. Indeed, AMFs influenced by aerosols through the modified  $O_2-O_2$  cloud parameters are smaller than clear-sky AMFs, consistent with the expected screening effect, that may well be too strong given the indications for a low bias in the  $O_2-O_2$  pressures. To evaluate our implicit corrections, we also calculated correction factors from simulations with a separate, independent radiative transfer model (SCIATRAN 2.2, Rozanov et al., 2005). SCIATRAN AMFs were calculated at 440 nm for a typical summertime southern United States scenario with the average “hazy” TM4  $NO_2$  profile shown in Fig. 13, with and without aerosols. For the simulations with aerosols, we used an AOT (440 nm) of 0.5, in line with the MODIS observations in Fig. 11. We used aerosol profiles with significant extinction up to 3–4 km (box profile, and profiles observed by CALIOP and simulated by GO-CART over the eastern United States from Yu et al., 2010). We assumed a surface albedo of 0.05 (0.054 for summer,



**Fig. 13.** Probability density functions for OMI  $O_2-O_2$  cloud pressure levels in January 2005 (upper panel), and July 2005 (lower panel). In both panels, the pdf for the “hazy” southern United States (January: MODIS  $> 0.1$ , July: MODIS  $> 0.2$ ) is indicated by the thick black line, and the pdf for the clean part of the domain (January: MODIS  $< 0.1$ , July: MODIS  $< 0.2$ ) has been indicated by the thin line. The red lines in both panels indicate the average vertical distribution of  $NO_2$  (layer columns) simulated by TM4 for the “hazy” region. The upper x-axis units are in ppbv.

0.049 for winter), consistent with the Kleipool et al. (2008) albedo climatology, and the aerosols to consist mostly of small particles with phase functions consistent with long-term AERONET observations summarized by Dubovik et al. (2002) (single scattering albedo 0.97 in summer, 0.95 in winter). Table 5 shows that the SCIATRAN simulations result in aerosol correction factors of 0.97–1.11, which is higher than our satellite-inferred implicit correction factors. The SCIATRAN simulations that agree most with our implicit correction factors, with values  $<1.0$ , are the ones with the average “hazy” TM4  $NO_2$  profile (Fig. 13) replaced by a 1 km  $NO_2$  “box profile”. Apparently, for screening by a haze layer to



**Fig. 14.** Monthly average aerosol correction factor for January 2005 (upper panel) and July 2005 (lower panel). The aerosol correction factor was calculated as the ratio of the tropospheric AMF as used in the DOMINO v1.02 retrieval to the clear-sky AMF (i.e.  $M/M_{cr}$ , see Eq. (2)). The aerosol correction factor was calculated only for those days and locations that had a successful, cloud-free, MODIS AOT retrieval 15 minutes prior to OMI. Grey areas indicate less than 3 successful coincidences.

occur, most of the  $\text{NO}_2$  needs to be well below the bulk of the aerosol particles. We conclude that our satellite-inferred implicit correction factors are 10–20 % lower than the explicit correction factors simulated with the SCIATRAN radiative transfer model. This discrepancy likely reflects the low bias in the  $\text{O}_2\text{-O}_2$  pressures. To investigate this further, we recommend to repeat this type of analysis for situations where most of the aerosol and  $\text{NO}_2$  characteristics are known, such as during dedicated measurement campaigns such as DANDELIONS (Hains et al., 2010) or CINDI (Piters et al., 2011).

Compared to summertime retrievals, the impact of aerosols on wintertime retrievals is modest. Figure 15 shows monthly mean MODIS AOT values for January 2005 up to 0.3, much smaller than in summer. The MODIS Ångström exponent over areas with high AOT is 0.7, indicating that particles are coarser in winter than in summer and that these coarse particles dominate over the ocean (Dubovik et al., 2002). Previous studies showed that lower AOT and coarse

particles affect AMFs less than high AOT and fine particles (Boersma et al., 2004; Leitão et al., 2010). Figure 15 also shows that both AOT values and cloud fractions are enhanced over the western Atlantic, and both are low over the southern United States. There is positive correlation between AOT and cloud fractions over the domain ( $r = 0.42$ ,  $n = 4465$ ) in January, but it is less significant than in July 2005. The lower panel of Fig. 15 shows that aerosol-induced cloud pressures are high in January compared to July 2005, indicating that the scattering aerosols reside closer to the surface in winter. Indeed, the median  $\text{O}_2\text{-O}_2$  pressure corresponds to 830 hPa when MODIS AOT > 0.1. Figure 13 indicates that in winter, aerosols are more likely to be mixed with  $\text{NO}_2$  than in summer, but also that aerosols can still be elevated relative to the  $\text{NO}_2$  layer. We can thus expect AMFs to be enhanced in some circumstances (when the aerosols are mixed with the  $\text{NO}_2$  layer) and reduced in others (when aerosols are well above the  $\text{NO}_2$  layer, as in summer). Indeed, Fig. 14 (upper panel) shows most (implicit) aerosol correction factors in the range 0.9–1.1. SCIATRAN correction factors for a number of plausible wintertime scenarios with and without aerosols over the eastern United States are similar with values in the 0.95–1.10 range. Table 6 confirms that correction factors are lowest for the scenario with the most elevated aerosol (2 km box profile).

## 7 Conclusions

We have improved the Dutch OMI  $\text{NO}_2$  (DOMINO) algorithm for global retrievals of tropospheric  $\text{NO}_2$  columns from the Ozone Monitoring Instrument. DOMINO was first described in Boersma et al. (2007), and has been used successfully in many scientific applications since then. Various validation exercises suggest that the previous version (v1.02) of DOMINO retrievals was of good quality, but biased high by 0–40 %, mostly because of air mass factor errors.

Here we focused especially on improving the  $\text{NO}_2$  air mass factors in order to generate a new DOMINO tropospheric  $\text{NO}_2$  dataset, named version 2 (v2.0). The improvements concern a better description of the radiative transfer for the lowest atmospheric layers, surface albedo, terrain height, clouds, and a priori vertical  $\text{NO}_2$  profiles. We have calculated a new altitude-dependent air mass factor look-up table (LUT) based on a more realistic atmospheric profile, and with an increased number of reference vertical layers and surface albedos, which reduces interpolation errors. The new LUT also mends another interpolation error in the computation of the altitude-dependent air mass factors for the lowest atmospheric layer. We showed that the new LUT alone increases air mass factors by 20–30 %, thereby reducing retrieved columns by up to 20 %.

We implemented a recently developed surface albedo climatology based on measurements from OMI in both the  $\text{NO}_2$  and cloud retrieval. The OMI surface albedo database with



**Table 5.** Summertime aerosol correction factors, defined as the ratio of the air mass factor with and without aerosols (fine particles,  $SSA = 0.97$ ), calculated with the SCIATRAN radiative transfer model for a typical summertime scenario over the eastern United States. The AOT was 0.5 (at 440 nm), and the solar zenith angle  $30^\circ$ . The aerosol vertical profiles refer to a constant extinction from the surface up to 3 km, and to average observed (CALIPSO) and simulated (GOCART) profiles over the eastern United States (see Yu et al., 2010). The assumed  $NO_2$  profiles refer to the TM4 average shown in Fig. 13 (lower panel) for situations when MODIS AOT  $> 0.2$ , and a profile with the same vertically integrated  $NO_2$  amount, but now confined to a 1-km boundary layer.

$NO_2$ profile	3 km box profile	CALIPSO profile	GOCART profile
TM4 average	1.10	1.11	1.09
1 km box profile	0.99	0.99	0.97

**Table 6.** Wintertime aerosol correction factors, defined as the ratio of the AMF with and without aerosols, calculated with the SCIATRAN radiative transfer model for typical wintertime scenarios over the eastern United States. The AOT was 0.15 (at 470 nm), and the solar zenith angle  $50^\circ$ . The vertical profiles refer to a constant extinction from the surface up to 0.5 km, and to extinction values that decrease exponentially with altitude. The assumed  $NO_2$  profiles refer to the TM4 average shown in Fig. 13 (upper panel) for situations when MODIS AOT  $> 0.1$ .

Aerosol type	0.5 km box profile	0.5 km exp. profile	2 km box profile
Fine ( $SSA = 0.95$ )	1.13	1.11	1.01
Coarse ( $SSA = 0.95$ )	1.05	1.04	0.99

improved spatial resolution ( $0.5^\circ \times 0.5^\circ$ ) reduces cloud and snow/ice contamination in the surface albedo, and leads to lower cloud fractions compared to TOMS/GOME. The overall impact of the OMI albedo is generally modest in summer but can be strong (up to +10 %) in winter over polluted regions that previously suffered from residual snow or ice in the TOMS/GOME database. We improved the sampling of the TM4 model, resulting in a priori  $NO_2$  profiles that are better mixed throughout the boundary layer. The TM4 profiles were also compared with in situ measurements from the INTEX-A/ICARTT and INTEX-B aircraft campaigns, showing good consistency throughout the lower troposphere. The improved sampling of  $NO_2$  from TM4 leads to more  $NO_2$  aloft and higher air mass factors over polluted regions. The strongest impact on  $NO_2$  retrievals is in summer (up to -8 %), when vertical transport is stronger than in winter. We reduced topography-related errors by replacing the  $2^\circ \times 3^\circ$  TM4 surface pressures by more accurate values based on the high-resolution DEM\_3km database, and scaling the a priori TM4  $NO_2$  accordingly. We found small changes (<1 %) in tropospheric  $NO_2$  columns over extended polluted regions, but significant enhancements (up to 14 %) in winter over polluted areas that previously had too high terrain heights such as the Po Valley and Beijing, and we found similar decreases for polluted highlands such as Mexico City and the Highveld area in South Africa that had too low terrain heights.

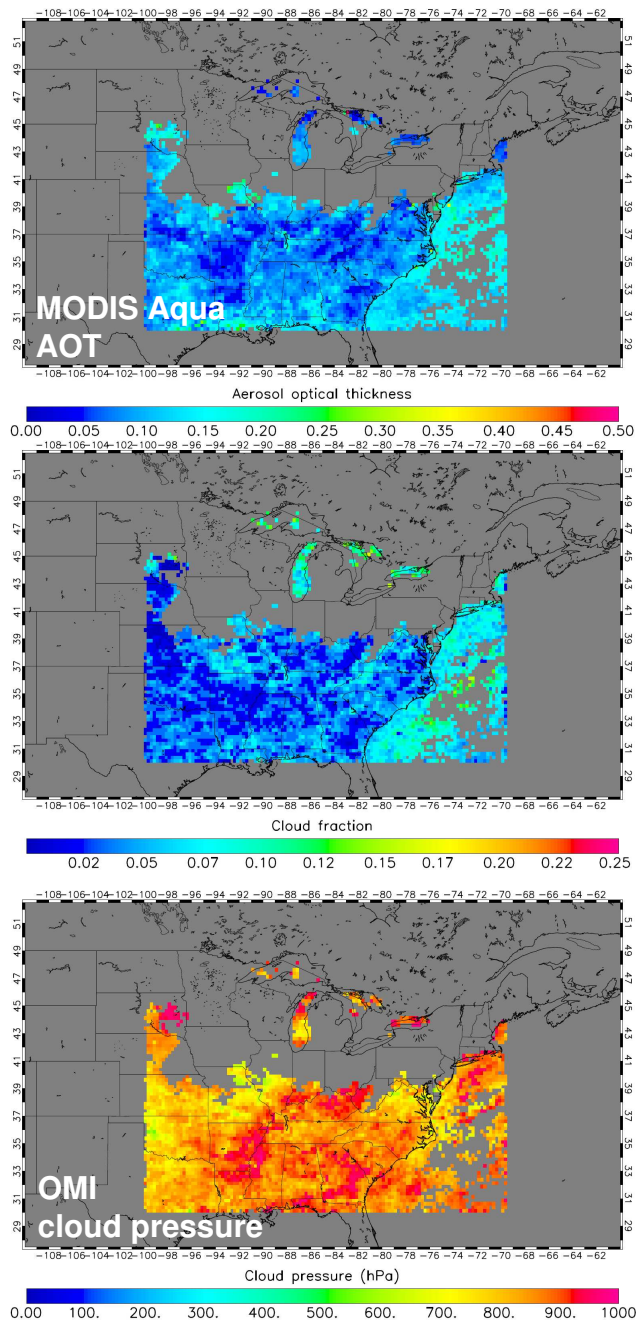
Apart from the air mass factor improvements, we introduced an a posteriori correction for stripes that are still apparent in the v1.02 tropospheric  $NO_2$  columns based on Collection 3 level 1 data. Our correction is based on the assumption that the jumps in  $NO_2$  slant columns that occur from one OMI viewing angle to the other are unrealistic, and that

a simple low-pass filter effectively eliminates them. Averaged over polluted areas, the effect of the stripe correction is marginal (<1 %), but locally the correction of tropospheric  $NO_2$  columns is on the order of  $0.5 \times 10^{15}$  molec  $cm^{-2}$ .

The picture that emerges from the individual improvements is that our new LUT and the improved sampling of TM4 have a significant effect on the  $NO_2$  retrievals for the polluted regions in the world. For users taking the averaging kernel into account – such as data assimilation groups – the improved sampling of  $NO_2$  is not relevant, and the changes between v1.02 and v2.0 will be somewhat smaller. The overall impact of the OMI surface albedo dataset, the improved description of the terrain height, and the stripe correction, is modest on average (changes are typically a few percent) over large polluted areas, but still causes significant changes on the local scale. These improvements are still important, since our satellite measurements are often used for studies with a distinct regional (air quality) or even local character (trend and validation studies).

Our new DOMINO v2.0 retrievals of tropospheric  $NO_2$  columns are reduced by 20 % in winter, and by 10 % during summer compared to v1.02 over extended polluted regions. These reductions are mainly driven by the improved air mass factor look-up table, and the better sampling of a priori  $NO_2$  profiles from TM4. On smaller spatial scales, the differences between v2.0 and v1.02 can be larger, reflecting the sometimes considerable changes in local surface albedo, clouds, and terrain height, and the effects of destriping. These reductions would bring DOMINO tropospheric columns in better agreement with independent measurements and model simulations. We cautiously estimate that the stated uncertainty for individual retrievals





**Fig. 15.** Monthly average aerosol optical thickness observed from MODIS Aqua (upper panel), and corresponding OMI O<sub>2</sub>-O<sub>2</sub> effective cloud fraction (middle panel) and effective cloud pressure (lower panel) for January 2005. Cloud fractions have been selected only for those days and locations that had a successful, cloud-free, MODIS AOT retrieval 15 min prior to OMI. Grey areas indicate less than 3 successful coincidences in January 2005.

of  $1.0 \times 10^{15}$  molec cm<sup>-2</sup> + 30 % for v1.02 (Boersma et al., 2007) improves to  $1.0 \times 10^{15}$  molec cm<sup>-2</sup> + 25 % for v2.0.

The conclusions of most published studies based on previous DOMINO v1.02 retrievals remain intact when they would be repeated with the improved v2.0 set, often because those studies made use of the averaging kernels, or because they were mainly concerned with relative changes in the NO<sub>2</sub> data. Absolute values for NO<sub>x</sub> emissions directly estimated from our previous DOMINO retrievals (e.g. Boersma et al., 2008b; Zhao and Wang, 2009) were probably too high by 10–20 %, but generally still well within the stated limits of uncertainty.

Following up on our earlier work that predicted OMI cloud retrievals to be sensitive to the presence of scattering aerosols, we examined the relationship between concurrent OMI O<sub>2</sub>-O<sub>2</sub> cloud observations and MODIS-Aqua aerosol optical thickness (AOT) over the eastern United States. We found that high aerosol loadings tend to increase retrieved O<sub>2</sub>-O<sub>2</sub> effective cloud fractions with reduced cloud pressures, especially over the southeastern United States which is covered by a layer of elevated, small (scattering) particles during Summer. The combined effect of the enhanced summertime cloud parameters on the NO<sub>2</sub> retrieval corresponds to screening: OMI is treated as being less sensitive to NO<sub>2</sub> because of the elevated aerosol layer is treated as residing above the NO<sub>2</sub> layer. In the DOMINO retrieval, the aerosol correction proceeds implicitly through the air mass factor formulation. The aerosol-induced cloud parameters reduce air mass factors over the southeastern United States by up to 20 % in summertime. In winter, aerosol loadings are lower, particles are coarser, and air mass factors are much less affected. Radiative transfer calculations with crudely estimated aerosol and NO<sub>2</sub> parameters reproduced the range of implicit aerosol correction factors for the winter case, but were higher by 0–20 % in summer. Radiative transfer calculations with observed rather than guessed NO<sub>2</sub> and aerosol parameters are required to further investigate this discrepancy, and provide more insight into the exact influence that aerosols exert on cloud and trace gas retrievals.

We focused here on NO<sub>2</sub> retrievals from OMI, but the described improvements are of similar importance for NO<sub>2</sub> retrievals from the GOME, SCIAMACHY, and GOME-2 series provided by KNMI on [www.temis.nl](http://www.temis.nl). The new altitude-dependent air mass factor look-up table, a more realistic surface albedo dataset (from MERIS, see Popp et al., 2011), the improved description of terrain height, and better sampling of TM4 profiles are currently being implemented in the GOME(-2) and SCIAMACHY NO<sub>2</sub> retrievals and preliminary results indicate similar effects on those retrievals as found here for OMI. New versions of the GOME(-2) and SCIAMACHY NO<sub>2</sub> data products can be expected in 2011. The new v2.0 DOMINO NO<sub>2</sub> retrievals are currently being made available (data for 2004–2007 is already online) on [www.temis.nl](http://www.temis.nl).

Tropospheric NO<sub>2</sub> retrievals for existing platforms can be further improved by better knowledge of the state of the atmosphere for individual pixels. The largest gains will likely be obtained by higher-resolution a priori profile shapes, and improved surface albedo databases. For the DOMINO v2 algorithm, an improved description of stratospheric chemistry in TM5 (Dirksen et al., 2011) is expected to improve our data assimilation for situations when the denoxified air masses make excursions into polluted continental regions. We anticipate that the next major update (v3) will consist of coupling our retrieval to TM5 (with 1° × 1° instead of 3° × 2°) capability and improved stratospheric chemistry.

**Acknowledgements.** We wish to thank Randall Martin (Dalhousie University) for encouraging discussions and Johan de Haan (KNMI) for additional simulations. NO<sub>2</sub> measurements from the NASA DC-8 aircraft were kindly made available by R. C. Cohen and co-workers (University of California, Berkeley), and from the NOAA's WP-3D aircraft by Tom Ryerson (NOAA). We acknowledge the free use of MODIS Aqua aerosol optical thickness data, obtained from <http://ladsweb.nascom.nasa.gov>, and publicly available information derived from the AERONET database. Comments from two anonymous reviewers helped to improve this paper. This research was funded by the Netherlands Organisation for Scientific Research, NWO Vidi grant 864.09.001.

Edited by: I. Aben

## References

- Acarreta, J. R., de Haan, J. F., and Stammes, P.: Cloud pressure retrieval using the O<sub>2</sub>-O<sub>2</sub> absorption band at 477 nm, *J. Geophys. Res.*, 109, D05204, doi:10.1029/2003JD003915, 2004.
- Basset, M. and Seinfeld, J. H.: Atmospheric Equilibrium Model of Sulfate and Nitrate Aerosols, *Atmos. Environ.*, 17, 2237–2252, 1983.
- Boersma, K. F., Bucsel, E. J., Brinksma, E. J., and Gleason, J. F.: NO<sub>2</sub>, in: OMI Algorithm Theoretical Basis Document, vol. 4, OMI Trace Gas Algorithms, ATB-OMI-04, Version 2.0, edited by: Chance, K., 13–36, NASA Distrib. Active Archive Cent., Greenbelt, Md., August, 2002.
- 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 Bucsel, 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., and Wang, 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, D16S26, doi:10.1029/2007JD008816, 2008a.
- Boersma, K. F., Jacob, D. J., Bucsel, E. J., Perring, A. E., Dirksen, R., van der A, R. J., Yantosca, R. M., Park, R. J., Wenig, M. O., Bertram, T. H., and Cohen, R. C.: Validation of OMI tropospheric NO<sub>2</sub> observations during INTEX-B and application to constrain NO<sub>x</sub> emissions over the eastern United States and Mexico, *Atmos. Environ.*, 42, 4480–4497, doi:10.1016/j.atmosenv.2008.02.004, 2008b.
- Boersma, K. F., Jacob, D. J., Trainic, M., Rudich, Y., DeSmedt, I., Dirksen, R., and Eskes, H. J.: Validation of urban NO<sub>2</sub> concentrations and their diurnal and seasonal variations observed from the SCIAMACHY and OMI sensors using in situ surface measurements in Israeli cities, *Atmos. Chem. Phys.*, 9, 3867–3879, doi:10.5194/acp-9-3867-2009, 2009a.
- Boersma, K. F., Dirksen, R. J., Veefkind, J. P., Eskes, H. J., and Van der A, R. J.: Dutch OMI NO<sub>2</sub> (DOMINO) data product HE5 data file user manual, TEMIS website, available at: <http://www.temis.nl/airpollution/no2.html>, 2009b.
- Bucsel, E. J., Celarier, E. A., Wenig, M. O., Gleason, J. F., Veefkind, J. P., Boersma, K. F., and Brinksma, E. J.: Algorithm for NO<sub>2</sub> vertical column retrieval from the Ozone Monitoring Instrument, *IEEE T. Geosci. Remote Sens.*, 44, doi:10.1109/TGRS.2005.863715, 2006.
- Bovensmann, H., Burrows, J. P., Buchwitz, M., Frerick, J., Noël, S., 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)056<0127:SMOAMM>2.0.CO;2, 1999.
- Burrows, J. P., Weber, M., Buchwitz, M., Rozanov, V., Ladstätter-Weissenmayer, 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., and Lefebvre, A.: GOME-2 – Metop's Second-Generation Sensor for Operational Ozone Monitoring, *ESA Bulletin*, 102, 28–36, 2000.
- Dentener, F., van Weele, M., Krol, M., Houweling, S., and van Velthoven, P.: Trends and inter-annual variability of methane emissions derived from 1979–1993 global CTM simulations, *Atmos. Chem. Phys.*, 3, 73–88, doi:10.5194/acp-3-73-2003, 2003.
- Dirksen, R. J., Boersma, K. F., Eskes, H. J., Ionov, D. V., Bucsel, E. J., Levelt, P. F., and Kelder, H. M.: Evaluation of stratospheric NO<sub>2</sub> retrieved from the Ozone Monitoring Instrument: intercomparison, diurnal cycle and trending, *J. Geophys. Res.*, 116, D08305, doi:10.1029/2010JD014943, 2011.
- Dobber, M., Kleipool, Q., Dirksen, R., Levelt, P. F., Jaross, G., Taylor, S., Kelly, T., Flynn, L., Leppelmeier, G., and Rozemeijer, N.: Validation of Ozone Monitoring Instrument level 1b data products, *J. Geophys. Res.*, 113, D15S06, doi:10.1029/2007JD008665, 2008.
- Dubovik, O., Holben, B., Eck, T. F., Smirnov, A., Kaufman, Y. J., King, M. D., Tanré, D., and Slutsker, I.: Variability of absorption and Optical Properties of Keu Aerosol Types Observed in Worldwide Locations, *J. Atmos. Sci.*, 59, 590–608, 2002.
- Eskes, H. J. and Boersma, K. F.: Averaging kernels for DOAS total-column satellite retrievals, *Atmos. Chem. Phys.*, 3, 1285–1291, doi:10.5194/acp-3-1285-2003, 2003.
- Fu, T.-M., Jacob, D. J., Palmer, P. I., Chance, K., Wang, Y. X., Barletta, B., Blake, D. R., Stanton, J. C., and Pilling, M. J.: Space-based formaldehyde measurements as constraints on volatile organic compound emissions in east and south Asia and implications for ozone, *J. Geophys. Res.*, 112, D06312, doi:10.1029/2006JD007853, 2007.
- Goldstein, A., Koven, C. D., Heald, C. L., and Fung, I. Y., Bio-

- genic carbon and anthropogenic pollutants combine to form a cooling haze over the southeastern United States, *Proc. Natl. Acad. Sci.*, 106, 8835–8840, available at: [www.pnas.org/cgi/doi/10.1073/pnas.0904128106](http://www.pnas.org/cgi/doi/10.1073/pnas.0904128106), 2009.
- Hains, J. C., Boersma, K. F., Kroon, M., Dirksen, R. J., Cohen, R. C., Perring, A. E., Bucsele, E. J., Volten, H., Swart, D. P. J., Richter, A., Wittrock, F., Schoenhardt, A., Wagner, T., Ibrahim, O. W., Van Roozendaal, M., Pinardi, G., Gleason, J. F., Veefkind, J. P., and Levelt, P. F.: Testing and improving OMI DOMINO tropospheric NO<sub>2</sub> using observations from the DANDELIONS and INTExJb validation campaigns, *J. Geophys. Res.*, 115, D05301, doi:10.1029/2009JD012399, 2010.
- Holland, E. A. and Lamarque, J.-F.: Modelling bio-atmospheric coupling of the nitrogen cycle through NO<sub>x</sub> emissions and NO<sub>y</sub> deposition, *Nutr. Cycl. Agroecosyst.*, 48, 7–24, 1997.
- Holtzlag, A. A. and Boville, B. A.: Local versus nonlocal boundary layer diffusion in a global climate model, *J. Climate*, 10, 1825–1842, 1993.
- Huijnen, V., Eskes, H. J., Poupkou, A., Elbern, H., Boersma, K. F., Foret, G., Sofiev, M., Valdebenito, A., Flemming, J., Stein, O., Gross, A., Robertson, L., D'Isidoro, M., Kioutsioukis, I., Friese, E., Amstrup, B., Bergstrom, R., Strunk, A., Vira, J., Zyryanov, D., Maurizi, A., Melas, D., Peuch, V.-H., and Zerefos, C.: Comparison of OMI NO<sub>2</sub> tropospheric columns with an ensemble of global and European regional air quality models, *Atmos. Chem. Phys.*, 10, 3273–3296, doi:10.5194/acp-10-3273-2010, 2010.
- Jacob, D. J., Heikes, B. G., Fan, S.-M., Logan, J. A., Mauzerall, D. L., Bradshaw, J. D., Singh, H. B., Gregory, G. L., Talbot, R. W., Blake, D. R., Sachse, G. W., Origin of ozone and NO<sub>x</sub> in the tropical troposphere: A photochemical analysis of aircraft observations over the South Atlantic basin, *J. Geophys. Res.*, 101, 24235–24250, 1996.
- Jiang, X., Yang, Z.-L., Liao, H., and Wiedinmyer, C.: Sensitivity of biogenic secondary organic aerosols to future climate change at regional scales: an online coupled simulation, *Atmos. Environ.*, 44, 4891–4907, 2010.
- Kleipool, Q. L., Dobber, M. R., de Haan, J. F., and Levelt, P. F.: Earth surface reflectance climatology from 3 years of OMI data, *J. Geophys. Res.*, 113, D18308, doi:10.1029/2008JD010290, 2008.
- Koelemeijer, R. B. A., Stammes, P., Hovenier, J. W., and de Haan, J. F.: A fast method for retrieval of cloud parameters using oxygen A band measurements from the Global Ozone Monitoring Experiment, *J. Geophys. Res.*, 106, 3475–3490, doi:10.1029/2000JD900657, 2001.
- Lamsal, L. N., Martin, R. V., van Donkelaar, A., Celarier, E. A., Bucsele, E. J., Boersma, K. F., Dirksen, R., Luo, C., and Wang, Y.: Indirect validation of tropospheric nitrogen dioxide retrieved from the Ozone Monitoring Instrument: Insight into the seasonal variation of nitrogen oxides at northern midlatitudes, *J. Geophys. Res.*, 115, D05301, doi:10.1029/2009JD012399, 2010.
- 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 airmass 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, doi:10.1029/2000JD900572, 2001.
- Levelt, P. F., van den Oord, G. H. J., Dobber, M. R., Malkki, A., Visser, H., de Vries, J., Stammes, P., Lundell, J., and Saari, H.: The Ozone Monitoring Instrument, *IEEE T. Geosci. Remote Sens.*, 44, 1093–1101, 2006.
- Lewis, J., De Young, R., and Chu, D. A.: A Study of Air Quality in the Southeastern Hampton-Norfolk-Virginia Beach Region with Airborne Lidar Measurements and MODIS Aerosol Optical Depth Retrievals, *J. Appl. Meteorol. Clim.*, 49, 3–19, doi:10.1175/2009JAMC2119.1, 2010.
- Lin, J.-T. and McElroy, M.: Impacts of boundary layer mixing on pollutant vertical profiles in the lower troposphere: Implications to satellite remote sensing, *Atmos. Environ.*, 44, 1726–1739, doi:10.1016/j.atmosenv.2010.02.009, 2010.
- Martin, R. V., Jacob, D. J., Chance, K., Kurosu, T. P., Palmer, P. I., and Evans, M. J.: An improved retrieval of tropospheric nitrogen dioxide from GOME, *J. Geophys. Res.*, 107, 4437, doi:10.1029/2001JD001027, 2002.
- Martin, R. V., Jacob, D. J., Chance, K., Kurosu, T. P., Palmer, P. I., and Evans, M. J.: Global inventory of nitrogen oxide emissions constrained by space-based observations, *J. Geophys. Res.*, 108, 4537, doi:10.1029/2003JD003453, 2003.
- Martin, R. V., Sioris, C. E., Chance, K., Ryerson, T. B., Bertram, T. H., Wooldridge, P. J., Cohen, R. C., Neuman, J. A., Swanson, A., and Flocke, F. M.: Evaluation of space-based constraints on global nitrogen oxide emissions with regional aircraft measurements over and downwind of eastern North America, *J. Geophys. Res.*, 111, D15308, doi:10.1029/2005JD006680, 2006.
- Michaels, A. F., Olson, D., Sarmiento, J. L., Ammerman, J. W., Fanning, K., Jahnke, R., Knap, A. H., Lipschultz, F., and Prospero, J. M.: Inputs, losses, and transformations of nitrogen and phosphorus in the pelagic North Atlantic Ocean, *Biogeochemistry*, 35, 181–226, 1996.
- 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 the Global Ozone Monitoring Experiment, *J. Geophys. Res.*, 106, 14539–14550, 2001.
- Penning de Vries, M. J. M., Beirle, S., and Wagner, T.: UV Aerosol Indices from SCIAMACHY: introducing the SCattering Index (SCI), *Atmos. Chem. Phys.*, 9, 9555–9567, doi:10.5194/acp-9-9555-2009, 2009.
- Peters, A. J. M., Boersma, K. F., Kroon, M., Hains, J. C., Van Roozendaal, M., Wittrock, F., Abuhassan, N., Adams, C., Akrami, M., Allaart, M. A. F., Apituley, A., Bergwerff, J. B., Berkhout, A. J. C., Brunner, D., Cede, A., Chong, J., Clémer, K., Fayt, C., Frieß, U., Gast, L. F. L., Gil-Ojeda, M., Goutail, F., Graves, R., Griesfeller, A., Großmann, K., Hemerijckx, G., Hendrick, F., Henzing, B., Herman, J., Hermans, C., Hoexum, M., van der Hoff, G. R., Irie, H., Johnston, P. V., Kanaya, Y., Kim, Y. J., Klein Baltink, H., Kreher, K., de Leeuw, G., Leigh, R., Merlaud, A., Moerman, M. M., Monks, P. S., Mount, G. H., Navarro-Comas, M., Oetjen, H. A., Perez-Camacho, M., Peters, E., du Piesanie, A., Pinardi, G., Pazmino, O., Puentedura, Richter, A., Roscoe, H. K., Schönhardt, A., Schwarzenbach, B., Shaiganfar, R., Sluis, W., Spinei, E., Stolk, A. P., Strong, K., Swart, D. P. J., Takashima, H., Vlemmix, T., Vrekoussis, M., Wagner, T., Whyte, C., Wilson, K. M., Yela, M., Yilmaz, S., Zieger, P., and

- Zhou, Y.: The Cabauw Intercomparison campaign for Nitrogen Dioxide Measuring Instruments (CINDI): Design, execution, and first results, in press, *Atmos. Meas. Tech. Discuss.*, 2011.
- Popp, C., Wang, P., Brunner, D., Stammes, P., Zhou, Y., and Grzegorski, M.: MERIS albedo climatology for FRESCO+O<sub>2</sub> A-band cloud retrieval, *Atmos. Meas. Tech.*, 4, 463–483, doi:10.5194/amt-4-463-2011, 2011.
- Remer, L. A., Kleidman, R. G., Levy, R. C., Kaufman, Y. J., Tanre, D., Mattoo, S., Vanderlei Martins, J., Ichoku, C., Koren, I., Yu, H., and Holben, B. N.: Global aerosol climatology from the MODIS satellite sensors, *J. Geophys. Res.*, 113, D14S07, doi:10.1029/2007JD009661, 2008.
- 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.
- Rozanov, A., Rozanov, V., Buchwitz, M., Kokhanovsky, A., and Burrows, J. P.: SCIATRAN 2.0 - A new radiative transfer model for geophysical applications in the 175–2400 nm spectral region, *Adv. Space Res.*, 36, 1015–1019, doi:10.1016/j.asr.2005.03.012, 2005.
- Russell, P. B., Bergstrom, R. W., Shinozuka, Y., Clarke, A. D., Decarlo, P. F., Jimenez, J. L., Livingston, J. M., Redemann, J., Dubovik, O., and Strawa, A.: Absorption Angstrom Exponent in AERONET and related data as an indicator of aerosol composition, *Atmos. Chem. Phys.*, 10, 1155–1169, doi:10.5194/acp-10-1155-2010, 2010.
- Ryerson, T. B., Williams, E. J. and Fehsenfeld, F. C.: An efficient photolysis system for fast-response NO<sub>2</sub> measurements, *J. Geophys. Res.*, 105, 26447–26461, 2000.
- Shindell, D. T., Faluvegi, G., Koch, D. M., Schmidt, G. A., Unger, N., and Bauer, S. E.: Improved Attribution of Climate Forcing to Emissions, *Science*, 326, 716–718, doi:10.1126/science.1174760, 2009.
- Spichtinger N., Wenig, M., James, P., Wagner, T., Platt, U., and Stohl, A.: Satellite detection of a continental-scale plume of nitrogen oxides from boreal fires, *Geophys. Res. Lett.*, 29, 4579–4583, 2001.
- Stammes, P., Sneep, M., de Haan, J. F., Veefkind, J. P., Wang, P., and Levelt, P. F.: Effective cloud fractions from the Ozone Monitoring Instrument: Theoretical framework and validation, *J. Geophys. Res.*, 113, D16S38, doi:10.1029/2007JD008820, 2008.
- Stephens, G. L., Vane, D. G., Boain, R. J., Mace, G. G., Sassen, K., Wang, Z., Illingworth, A. J., O'Connor, E. J., Rossow, W. B., Durden, S. L., Miller, S. D., Austin, R. T., Benedetti, A., Mitrescu, C., and the CloudSat Science Team: The CloudSat Mission and the A-Train, *B. Am. Meteorol. Soc.*, 83, 1771–1790, 2002.
- Thornton, J. A., Wooldridge, P. J., and Cohen, R. C.: Atmospheric NO<sub>2</sub>: In situ laser-induced fluorescence detection at parts per trillion mixing ratios, *Anal. Chem.*, 72, 528–539, 2000.
- Torres, O., Bhartia, P. K., Herman, J. R., Ahmad, Z., and Gleason, J.: Derivation of aerosol properties from a satellite measurement of backscattered ultraviolet radiation: Theoretical basis, *J. Geophys. Res.*, 103, 17099–17110, 1998.
- Turner, D. D., Ferrare, R. R., and Brasseur, L. A.: Average Aerosol Extinction and Water Vapor Profiles Over the Southern Great Plains, *Geophys. Res. Lett.*, 28, 4441–4444, 2001.
- van der A, R. J., Eskes, H. J., Boersma, K. F., van Noije, T. P. C., Van Roozendaal, 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.
- Veefkind, J. P., Boersma, K. F., Wang, J., Kurosu, T. P., Krotkov, N., Chance, K., and Levelt, P. F.: Global satellite analysis of the relation between aerosols and short-lived trace gases, *Atmos. Chem. Phys.*, 11, 1255–1267, doi:10.5194/acp-11-1255-2011, 2011.
- Wang, Y., McElroy, M. B., Boersma, K. F., Eskes, H. J., and Veefkind, J. P.: Traffic restrictions associated with the Sino-African summit: Reductions of NO<sub>x</sub> detected from space, *Geophys. Res. Lett.*, 34, L08814, doi:10.1029/2007GL029326, 2007.
- Yu, H., Chin, M., Winker, D. M., Omar, H., Liu, Z., Kittaka, C., and Diehl, T.: Global view of aerosol vertical distributions from CALIPSO lidar measurements and GOCART simulations: Regional and seasonal variations, *J. Geophys. Res.*, 115, D00H30, doi:10.1029/2009JD013364, 2010.
- Zhao, C. and Wang, Y.: Assimilated inversion of NO<sub>x</sub> emissions over east Asia using OMI NO<sub>2</sub> column measurements, *Geophys. Res. Lett.*, 36, L06805, doi:10.1029/2008GL037123, 2009.
- 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.
- Zhou, Y., Brunner, D., Spurr, R. J. D., Boersma, K. F., Sneep, M., Popp, C., and Buchmann, B.: Accounting for surface reflectance anisotropy in satellite retrievals of tropospheric NO<sub>2</sub>, *Atmos. Meas. Tech.*, 3, 1185–1203, doi:10.5194/amt-3-1185-2010, 2010.