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from OMI

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# Near-real time retrieval of tropospheric NO<sub>2</sub> from OMI

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## Abstract

We present a new algorithm for the near-real time retrieval – within 3 h of the actual satellite measurement – of tropospheric NO<sub>2</sub> columns from the Ozone Monitoring Instrument (OMI). The retrieval system is based on the combined retrieval-assimilation-modelling approach developed at KNMI for off-line tropospheric NO<sub>2</sub> from the GOME and SCIAMACHY satellite instruments. We have adapted the off-line system such that the required a priori information – profile shapes and stratospheric background NO<sub>2</sub> – is now immediately available upon arrival of the OMI NO<sub>2</sub> slant columns and cloud data at KNMI. Slant column NO<sub>2</sub> and cloud information arrives at KNMI typically within 80 min of actual OMI observations. Slant columns for NO<sub>2</sub> are retrieved using differential optical absorption spectroscopy (DOAS) in the 405–465 nm range. Cloud fraction and cloud pressure are provided by a new cloud retrieval algorithm that uses the absorption of the O<sub>2</sub>–O<sub>2</sub> collision complex near 477 nm. On-line availability of stratospheric slant columns and NO<sub>2</sub> profiles is achieved by running the TM4 chemistry transport model (CTM) forward in time based on forecast ECMWF meteorological and assimilated NO<sub>2</sub> information from all previously observed orbits. OMI NO<sub>2</sub> slant columns, after correction for spurious across-track variability, show a random error for individual pixels of approximately  $0.7 \times 10^{15}$  molec.cm<sup>-2</sup>. As NO<sub>2</sub> retrievals are very sensitive to clouds, we evaluated the consistency of cloud fraction and cloud pressure from the new O<sub>2</sub>–O<sub>2</sub> (OMI) algorithm and from the Fast Retrieval Scheme for Cloud Observables (FRESCO). Cloud parameters from the O<sub>2</sub>–O<sub>2</sub> (OMI) algorithm have similar frequency distributions as cloud parameters retrieved from FRESCO (SCIAMACHY) for August 2006. On average, OMI cloud fractions are higher by 0.011, and OMI cloud pressures exceed FRESCO cloud pressures by 60 hPa. As a consistency check, we intercompared OMI near-real time NO<sub>2</sub> columns measured at 13:45 h local time to SCIAMACHY off-line NO<sub>2</sub> columns measured at 10:00 h local time. In August 2006, both instruments observe very similar spatial patterns of tropospheric NO<sub>2</sub> columns, and small differences for most locations on Earth where tropospheric NO<sub>2</sub> columns

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are small. For regions that are strongly polluted, SCIAMACHY observes higher tropospheric NO<sub>2</sub> columns than OMI.

## 1 Introduction

The daily global coverage and the pixel size of 24×13 km<sup>2</sup> make the Ozone Monitoring Instrument (OMI) on the Earth Observing System (EOS) Aura satellite well suited to observe the sources of air pollution with an unprecedented spatial and temporal coverage. Recently, satellite-based observations of tropospheric NO<sub>2</sub> have been proven useful in estimating anthropogenic emissions of nitrogen oxides (Leue et al., 2001; Martin et al., 2003, 2006; Beirle et al., 2003; Richter et al., 2005; van der A et al., 2006), in observing emissions by soils (Jaeglé et al., 2004), and in putting constraints on NO<sub>x</sub> production by lightning (Beirle et al., 2004, 2005; Boersma et al., 2005). Tropospheric NO<sub>2</sub> columns derived from the Global Ozone Monitoring Experiment (GOME) have been compared with outputs from various-scale models (Velders et al., 2001; Lauer et al., 2002; Savage et al., 2004; Ma et al., 2006). The results of the regional-scale chemistry-transport model CHIMERE have been evaluated against GOME and SCIAMACHY-derived tropospheric NO<sub>2</sub> columns (Konovalov et al., 2004; Blond et al., 2006<sup>1</sup>). These comparisons have clearly demonstrated the potential of satellite NO<sub>2</sub> data sets for model evaluation and emission estimates. On the other hand, sometimes large and often systematic differences persist between retrievals by different groups (van Noije et al., 2006), calling into question the quality of space-based constraints on NO<sub>x</sub> sources. However, validation efforts for various retrievals show acceptable accuracy (Heland et al., 2002; Petritoli et al., 2004; Martin et al., 2004, 2006; Cede et al., 2006; Ordóñez et al., 2006; Schaub et al., 2006) for GOME and SCIAMACHY NO<sub>2</sub>.

<sup>1</sup>Blond, N., Boersma, K. F., Eskes, H. J., van der A, R., Van Roozendaal, J. M., De Smedt, I., Bergametti, G., and Vautard, R.: Intercomparison of SCIAMACHY nitrogen dioxide observations, in-situ measurements and air quality modeling results over Western Europe, submitted, J. Geophys. Res., 2006.

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The unique characteristics of OMI –the small pixel size and daily global coverage– allow for an important contribution to air quality monitoring and modelling. GOME has a resolution of  $320 \times 40 \text{ km}^2$ , too coarse to resolve the areas with high emissions that are relevant in regional air quality modelling, e.g. medium-sized cities. SCIAMACHY's horizontal resolution is  $60 \times 30 \text{ km}^2$  but needs six days to achieve global coverage. Despite the fact that interesting regional-scale daily variability has been observed with SCIAMACHY by Blond et al. (2006)<sup>1</sup>, it is not well suited for a day-to-day monitoring of air quality. The daily coverage of OMI has been an important motivation to set up the near-real time NO<sub>2</sub> retrieval system described in this paper. An additional motivation originates from the data set of tropospheric NO<sub>2</sub> columns retrieved from the GOME and SCIAMACHY instruments that now spans more than 10 years (1996–2006) and is publicly available through <http://www.temis.nl>. NO<sub>2</sub> data sets from GOME and SCIAMACHY have been retrieved with one and the same retrieval-assimilation-modelling approach described in Boersma et al. (2004) and show excellent mutual consistency (van der A et al., 2006). The OMI NO<sub>2</sub>-retrievals described here are expected to add considerable value to the GOME and SCIAMACHY dataset. Health regulations concerning air quality require a routine monitoring, typically on an hourly basis, of surface concentrations of several species including NO<sub>2</sub>. Clearly, this requirement cannot be directly fulfilled by satellite instruments in general, nor by OMI in particular. Nevertheless we anticipate that instruments like OMI will make essential contributions to air quality monitoring and modelling:

- Daily maps of NO<sub>2</sub> columns provided by OMI show extensive transport features that are changing from day to day, and that are politically interesting as they directly show air pollution being transported across national borders. These changeable distributions can be directly compared with model output, and they constitute strong tests for the description of horizontal and vertical transport processes, as well as NO<sub>x</sub> removal processes.
- A direct relationship exists between columns of NO<sub>2</sub> and surface emissions of

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NO<sub>x</sub>. OMI data can thus be combined with regional-scale models through inverse modeling or data assimilation to adjust or improve emission estimates in the model and to detect unknown sources.

- Incidental releases, such as from major fires, can be monitored and quantified, and subsequent plumes can be tracked from day to day.
- A routine assimilation of satellite data may improve air quality “nowcasting” and forecasting capabilities, and may thereby contribute to the monitoring of emission and health regulations.

All these applications are new and largely untested. Despite this, there exists a considerable interest in the community to establish atmospheric chemistry data assimilation systems that will exploit the available satellite data sets of atmospheric composition and air pollution. One example is the European GEMS project (Global and regional Earth-system (Atmosphere) Monitoring using Satellite and in-situ data; (<http://www.ecmwf.int/research/EU/projects/GEMS/>) which is scheduled to deliver an operational atmospheric composition assimilation system by 2009.

This paper presents a new retrieval algorithm designed for near-real time retrieval of tropospheric NO<sub>2</sub> from OMI. This algorithm differs from the standard, off-line OMI NO<sub>2</sub> retrieval-procedure that is a joint NASA/KNMI effort and has been described by Bucsela et al. (2006). The differences are shortly discussed in Sect. 3.1. In Sect. 2 we shortly discuss Ozone Monitoring Instrument characteristics, and describe the fast transport of measurement data from the satellite to the computer system where the retrievals take place. The retrieval itself is discussed in Sect. 3, with a focus on the innovations with respect to previous NO<sub>2</sub> column retrieval work at KNMI. Section 4 is devoted to errors in the NO<sub>2</sub> slant columns that occur through calibration errors in current the level 1 data, and through the propagation of measurement noise in the spectral fitting. The stratospheric correction and computation of the air mass factor (AMF) are described in Sect. 5. As SCIAMACHY and OMI cloud retrieval use different spectral features, we also discuss in Sect. 5 the consistency of the OMI O<sub>2</sub>–O<sub>2</sub> cloud

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product with the SCIAMACHY FRESCO cloud retrievals. As a first order quality-check of OMI near-real time NO<sub>2</sub> retrievals, in Sect. 6 we compare SCIAMACHY and OMI tropospheric NO<sub>2</sub> columns for the month August 2006 and show some examples of OMI's capabilities. This is followed by conclusions in Sect. 7.

## 2 OMI overview

### 2.1 Ozone Monitoring Instrument

The Dutch-Finnish Ozone Monitoring Instrument (OMI) on NASA's EOS Aura satellite is a nadir-viewing imaging spectrograph measuring direct and atmosphere-backscattered sunlight in the ultraviolet-visible (UV-VIS) range from 270 nm to 500 nm (Levelt et al., 2006a). EOS Aura was launched on 15 July 2004 and traces a Sun-synchronous, polar orbit at approximately 705 km altitude with a period of 100 min and a local equator crossing time between 13.40 h and 13.50 h. In contrast to its predecessors GOME and SCIAMACHY, instruments operating with scanning mirrors and one-dimensional photo diode array detectors, OMI has been equipped with two two-dimensional CCD detectors. The CCDs record the complete 270–500 nm spectrum in one direction, and observe the Earth's atmosphere with a 114° field of view, distributed over 60 discrete viewing angles, perpendicular to the flight direction. OMI's wide field of view corresponds to a 2600 km wide spatial swath on the Earth's surface for one orbit, large enough to achieve complete global coverage in one day. The exposure time of the CCD-camera measures 2 s, corresponding to a spatial sampling of 13 km along track (2 s × 6.5 km/s, the latter being the orbital velocity projected onto the Earth's surface). Across track the size of an OMI pixel varies with viewing zenith angle from 24 km in the nadir to approximately 60 km for the extreme viewing angles of 57° at the edges of the swath. OMI has three spectral channels; UV1 (270–310 nm) and UV2 (310–365 nm) are covered by CCD1. CCD2 covers the VIS-channel from 365–500 nm with a spectral sampling of 0.21 nm and a spectral resolution of 0.63 nm. It is in this channel that the

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spectral features of NO<sub>2</sub> are most prominent. The spectral resolution and sampling of OMI have been designed to resolve trace gas absorption signatures of chemically important atmospheric species. The spectral sampling rate (resolution/sampling) is ~3, large enough to avoid spectral undersampling or aliasing difficulties in the spectral fitting process. A polarization scrambler makes the instrument insensitive to the polarization state of the reflected Earth radiance to less than 0.5%. During nominal operations OMI takes one measurement of the solar irradiance per day. Radiometric calibration is accomplished in-flight by a series of special on-board measurements that involve a.o. monitoring detector degradation with a white-light source, and dark signal measurements when OMI is at the dark side of the Earth. Spectral calibration is achieved by a cross-correlation of Fraunhofer lines in theoretical and observed in-flight irradiance spectra. For more details on the instrument and calibration procedures, the reader is referred to [Dobber et al. \(2005, 2006\)](#).

Retrievals of ozone column and vertical distribution (as well as BrO and OCIO) are meant to address the first EOS Aura science question whether the ozone layer is recovering. Of no less importance are the retrievals of trace gases related to air pollution and the production of photochemical smog, i.e. NO<sub>2</sub>, HCHO, and SO<sub>2</sub>. In addition, cloud and aerosol parameters, and UV-B surface flux are derived from OMI. For an overview of EOS-AURA and OMI targets, see [Levelt et al. \(2006b\)](#). In this paper we focus on the fast delivery of OMI NO<sub>2</sub> data relevant for the purposes of air quality monitoring and forecasting.

## 2.2 Data transport

OMI science data is stored on-board on a solid-state recorder. These unprocessed data are down-linked once every orbit (100 min) to one of the Polar Ground Stations in Alaska or Svalbard (Spitsbergen) or to the Wallops Ground Station in Virginia (USA). As soon as a spacecraft contact session has finished, the OMI data are sent to the EOS Data Operations System (EDOS) at NASA GSFC in Maryland (USA). At GSFC, the raw OMI data are processed in three different ways, resulting in Rate-Buffered

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Data, Expedited Data, and Production Data. Near-real time processing is based on Rate-Buffered Data (RBD). RBD data are made available with the highest priority, at the expense of data integrity. The other data types are scheduled for off-line level 2 retrievals.

5 EDOS subsequently forwards the RBD data to the OMI Science Investigator-led Processing System (SIPS), where production starts as soon as all engineering, ancillary and science data have been received. A preprocessor run improves data integrity, removes duplicate packages, and time-orders the data. Then, the resulting level 0 data sets (in Analog to Digital Units) are processed into level 1b data, i.e. estimated radiances and irradiances in units of  $W/m^2/nm(/sr)$  (van den Oord et al., 2006). The only  
10 difference with standard production at this time is that the near-real time processing uses the predicted altitude and ephemeris data received from the spacecraft rather than definitive altitude and ephemeris data. Using predicted orbital parameters may lead to errors in geolocation parameters (latitude, longitude, solar, viewing, and azimuth angles) that influence the retrievals, but in practice, these errors are small. Once  
15 OMI level 1b data has been generated at the SIPS, the  $O_2-O_2$  cloud level 1–2 algorithm (Acarreta et al., 2004) is run, followed by the DOAS ozone (Veefkind and De Haan, 2002) and  $NO_2$  slant column spectral fitting retrieval algorithms. As soon as the ozone and  $NO_2$  slant column files are available, they are picked up by the OMI  
20 Dutch Processing system (ODPS) and forwarded to the processing system at KNMI developed within the DOMINO project (see acknowledgment). Subsequent steps are intrinsic parts of the DOMINO retrieval algorithm and are described in the next section.

25 Typical data volumes per orbit are 450 MB level 0 data, 400 MB level 1b data, and 17 MB  $NO_2$  slant column data (including cloud retrievals). Processed orbital data arrives at KNMI on average within three hours after the start of an orbit. An orbit takes 100 min (indicated as the red part of Fig. 1) and the process described above (downlink, transfer to EDOS, transfer to SIPS, level 1b and 2 processing, and transfer to KNMI) takes 80 min (the orange part in Fig. 1). Final processing from  $NO_2$  slant columns to tropospheric vertical columns at KNMI is typically faster than 2 min on a linux worksta-

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tion. Including the generation of images and web publishing, the processing step takes less than 45 min (the green part of Fig. 1), so that data and images are available for the public at approximately 16:00 h local time.

### 3 Algorithm description

#### 3.1 Heritage: the retrieval-assimilation-modelling approach

The near-real time NO<sub>2</sub> retrieval algorithm (DOMINO version: TM4NO2A-OMI v0.8, February 2006) in this work is based on the retrieval-assimilation-modelling approach (hereafter: RAM) described in [Boersma et al. \(2004\)](#). The first DOAS slant column fitting part is identical to the standard, off-line OMI NO<sub>2</sub> product discussed by [Bucsela et al. \(2006\)](#). The RAM-approach has been applied at KNMI to generate a tropospheric NO<sub>2</sub> data base from GOME and SCIAMACHY measurements. The off-line RAM approach consists of a three-step procedure:

1. a slant column density is determined from a spectral fit to the Earth reflectance spectrum with the so-called DOAS approach (differential absorption spectroscopy, e.g. [Platt, 1994](#); [Boersma et al., 2002](#); [Bucsela et al., 2006](#)),
2. the stratospheric contribution to the slant column is estimated from assimilating slant columns into a CTM including stratospheric chemistry and wind fields, and
3. the residual tropospheric slant column is converted into a vertical column by application of a tropospheric air mass factor (AMF).

The standard, off-line NO<sub>2</sub> product and the near-real time retrieval have step 1 in common. This step is discussed in detail in Sects. 3.2 and 3.3. Step 2 and 3 are different between the OMI off-line and NRT algorithms, for details the reader is referred to [Bucsela et al. \(2006\)](#); [Boersma et al. \(2004\)](#) and this work.

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Until now, a large NO<sub>2</sub> data set has been generated with the RAM-approach for the GOME and SCIAMACHY instruments. The data sets cover the period April 1996–June 2003 for GOME and from January 2003 onwards for SCIAMACHY. The two data sets contain tropospheric NO<sub>2</sub> columns along with error estimates and averaging kernels (Eskes and Boersma, 2003) for every individual pixel and they are publicly available through <http://www.temis.nl>. NO<sub>2</sub> retrieved from GOME with the RAM approach has been used to estimate the production of NO<sub>x</sub> by lightning in the tropics (Boersma et al., 2005). Furthermore, Schaub et al. (2006) showed that GOME tropospheric NO<sub>2</sub> over Northern Switzerland in the period 1996–2003 compares favourably to NO<sub>2</sub> profiles observed with in-situ techniques. Blond et al. (2006)<sup>1</sup> reported considerable consistency between SCIAMACHY tropospheric NO<sub>2</sub> columns and both surface observations as well as simulations from the regional air-quality model CHIMERE over Europe, especially over moderately polluted rural areas. Merged GOME and SCIAMACHY observations showed a distinct increase in NO<sub>2</sub> columns from 1996 to 2003 over China, consistent with a strong growth of NO<sub>x</sub> emissions in that area (van der A et al., 2006). Moreover, this paper showed an almost seamless continuity from GOME to SCIAMACHY NO<sub>2</sub> values retrieved with the same RAM-approach. This finding provides confidence in the consistency of the two data sets and their retrieval method, even though they originated from two different satellite instruments (with similar overpass times of 10:30 h (GOME) and 10:00 h (SCIAMACHY)).

Tropospheric NO<sub>2</sub> columns are generally retrieved as follows:

$$V_{tr} = \frac{S - S_{st}}{M_{tr}(\mathbf{x}_{a,tr}, \mathbf{b})}, \quad (1)$$

with  $S$  the slant column density from step 1,  $S_{st}$  the stratospheric slant column obtained from step 2, and  $M_{tr}$  the tropospheric AMF that depends on the a priori NO<sub>2</sub> profile  $\mathbf{x}_{a,tr}$  and the set of forward model parameters  $\mathbf{b}$  including cloud fraction, cloud pressure, surface albedo, and viewing geometry.

For the RAM-approach as well as the NRT-retrieval described here, AMFs and averaging kernels are computed with a pseudo-spherical version of the DAK radiative

transfer model (Stammes, 2001). Given the best estimate of the forward model parameters the DAK forward model describes the scattering and absorbing processes that define the average optical path of photons from the Sun through the atmosphere to the OMI.

5 Also similar as in our RAM-approach for off-line retrievals, we obtain here the a priori NO<sub>2</sub> profile shapes ( $x_{a, tr}$ ) from the global chemistry-transport model TM4 at a resolution of 2° latitude by 3° longitude and 35 vertical levels extending up to 0.38 hPa. Given the very few available in situ NO<sub>2</sub> measurements, a global 3-D model of tropo-  
10 spheric chemistry is the best source of information for the vertical distribution of NO<sub>2</sub> at the time and location of the OMI measurement. The TM4 model is driven by forecast and analysed six hourly meteorological fields from the European Centre for Medium Range Weather Forecast (ECMWF) operational data. These fields include global distributions for horizontal wind, surface pressure, temperature, humidity, liquid and ice water content, cloud cover and precipitation. A mass conserving preprocessing of the  
15 meteorological input is applied according to Bregman et al. (2003). Key processes included are mass conserved tracer advection, convective tracer transport, boundary layer diffusion, photolysis, dry and wet deposition as well as tropospheric chemistry including non-methane hydrocarbons to account for chemical loss by reaction with OH (Houweling et al., 1998). In TM4, anthropogenic and natural emissions of NO<sub>x</sub> are  
20 based on results from the EU POET-project (Precursors of Ozone and their Effects on the Troposphere) for the year 1997 (Olivier et al., 2003). Including free tropospheric emissions from air traffic (0.8 Tg N/yr) and lightning (5 Tg N/yr), total NO<sub>x</sub> emissions for 1997 amount to 46 Tg N/yr.

25 As in the RAM-approach for GOME and SCIAMACHY retrievals, cloud fraction and cloud pressure are determined from the actual satellite measurements. Because OMI does not detect the O<sub>2</sub> A band at 760 nm, we use cloud parameters retrieved from the VIS-channel using the O<sub>2</sub>-O<sub>2</sub> absorption feature at 477 nm (Acarreta et al., 2004). The cloud retrieval is based on the same set of assumptions (i.e. clouds are modelled as Lambertian reflectors with albedo 0.8) as the FRESKO-algorithm (Koelemeijer et al.,

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2001). Before launch, the precision of the  $O_2-O_2$  cloud fraction and cloud pressure was discussed in [Acarreta et al. \(2004\)](#) with encouraging results. In Sect. 5.2.1 we test the accuracy of the  $O_2-O_2$  cloud parameters by comparing them to FRESCO cloud parameters retrieved by SCIAMACHY on the same days and same locations.

5 As for GOME and SCIAMACHY retrievals, we use a surface albedo database derived from TOMS and GOME Lambert-equivalent reflectivity (LER) measurements at 380 nm and 440 nm as described in [Boersma et al. \(2004\)](#). These monthly average surface albedo maps have a spatial resolution of  $1^\circ \times 1.25^\circ$  and represent climatological (monthly) mean situations. The uncertainty in the surface albedo is estimated to be approximately 0.01 ([Koelemeijer et al., 2003](#); [Boersma et al., 2004](#)).

10 In summary, the near-real time algorithm is based on the RAM-approach used for GOME and SCIAMACHY retrievals at KNMI. The main differences with RAM are: (1) near-real time requirement, (2) different spectral fitting method, and (3) cloud inputs derived from (similar but) different algorithms. The near-real time algorithm differs from the off-line standard NASA/KNMI  $NO_2$  algorithm in the method to estimate the stratospheric  $NO_2$  content, in the choice of radiative transfer model, and in the a priori  $NO_2$  profiles and different surface albedo data sets for AMF computations

### 3.2 Near-real time retrieval

20 The most important difference between the RAM-approach and the NRT-retrieval is related to the required near-real time delivery of the retrieval product. In the RAM-approach, the estimated stratospheric  $NO_2$  column (step 2) and the modelled profile shape (required for step 3), are provided by off-line assimilation and modelling based on analysed ECMWF meteorological data. In contrast, for the NRT-retrieval, the assimilation and modelling steps are operational, based on daily ECMWF meteorological analyses and forecasts.

25 The NRT-retrieval consists of two distinct subsystems. The first is the TM forecast subsystem shown in Fig. 2. This forecast system is run once per day, as soon meteorological data becomes available. The second subsystem is activated each and every

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time that new OMI data becomes available, and incorporates the model information provided by subsystem 1.

In the forecast subsystem (1), the actual chemical state of the atmosphere is based on the analysis and forecast run starting from the analysis of the previous day. The update consists of running the chemistry-transport model forward in time with the forecast ECMWF meteorological data and the assimilation of all available OMI NO<sub>2</sub> slant columns measurements. The updated analysis, the new actual state, is then stored as input for a subsequent time step. The outputs are the necessary inputs to the near-real time subsystem (2); the stratospheric NO<sub>2</sub> column and the NO<sub>2</sub> and temperature profiles (needed in AMF computations).

The NRT-subsystem is illustrated in Fig. 2. As soon as an orbit of observed NO<sub>2</sub> slant columns arrives at KNMI, the forecast TM stratospheric slant column, prepared by the TM forecast subsystem, is ready and is subtracted. Subsequently, the residual tropospheric slant column is converted into a vertical column by the tropospheric AMF. The AMF is computed as described as in Sect. 3.1. The averaging kernel is also calculated for output and furthermore serves as the observation operator required in the assimilation part of the TM forecast subsystem.

### 3.3 Slant column retrieval

A second difference between the OMI near-real time and SCIAMACHY off-line implementation is the wide spectral window that is used in the DOAS retrieval. For GOME and SCIAMACHY, it has been conventional wisdom that a 425–450 nm window yields the most precise and stable fitting results. For OMI a much wider fit window, 405–465 nm, has been proposed by Boersma et al. (2002) in order to compensate for OMI's lower signal-to-noise ratio (approximately 1400 under normal mid-latitude conditions) compared to GOME and SCIAMACHY (approximately 2000, Bovensmann et al., 1999) in this wavelength region. Pre-flight testing showed that a least squares fitting of reference spectra from NO<sub>2</sub>, O<sub>3</sub>, a theoretical Ring function, and a 3rd order polynomial to the reflectance spectrum yields results that are stable for multiple viewing geometries

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with a better than 10% NO<sub>2</sub> slant column precision (Boersma et al., 2002). The NO<sub>2</sub> absorption cross section spectrum is taken from Vandaele et al. (1998), who tabulated the cross section at different temperatures. To account for the temperature sensitivity of the NO<sub>2</sub> cross section spectrum – determined from the data by Vandaele et al. (1998) – an effective atmospheric temperature is calculated for the NO<sub>2</sub> along the average photon path. Subsequently an *a posteriori* correction for the difference between the computed effective temperature and the 220 K cross section spectrum used in the fitting procedure is applied (Boersma et al., 2002). The ozone absorption cross section spectrum is taken from WMO (1975) and the theoretical Ring spectrum from De Haan (personal communication, 2006) based on irradiance data by Voors et al. (2006). All reference spectra have been convolved with the OMI instrument transfer function (Dobber et al., 2005). OMI NO<sub>2</sub> slant column retrieval with synthetic and flight model data (Dobber et al., 2005) yields results that fulfill the OMI science requirement of better than 10% slant column precision (Boersma et al., 2002; Bucsele et al., 2006). However, upon first inspection of actual flight-data, systematic enhancements in the OMI NO<sub>2</sub> slant columns show up at specific satellite viewing angles. This across-track variability is has also been reported by Kurosu et al. (2005) for HCHO-retrievals. Below we discuss a procedure to reduce across-track variability and investigate the error budget of the OMI NO<sub>2</sub> slant columns.

## 4 Slant column density errors

### 4.1 Across-track variability

Calibration errors in the level 1b OMI irradiance measurements used here are likely responsible for across-track variability observed in the NO<sub>2</sub> slant columns. This variability will likely be significantly reduced in future level 1b releases with improved calibration data (expected in Spring 2007), using daily dark current corrections. Across-track variability appears as constant offsets for specific satellite viewing angles along an orbit,

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allowing for an a posteriori correction that consists of four steps:

1. Determine the orbital segment (50 along track by 60 across track pixels in size) with the minimum variance in NO<sub>2</sub> columns.
2. Within this window, average the 50 NO<sub>2</sub> columns with the same viewing angle. This gives 60 average across-track NO<sub>2</sub> columns.
3. Separate low and high frequencies of the across-track columns with a Fourier analysis.
4. Perform the correction by subtracting the (high-frequency) across-track variability for all across-track rows along the orbit.

In this scheme, the selection of the minimum variance window avoids areas with large anthropogenic emissions. The high-frequencies are then interpreted as the across-track variability due to calibration errors in the OMI level 1b data. Similarly, the low frequencies describe any (weak, stratospheric) natural variability. The low frequencies are described by the first three Fourier terms. Subsequently we subtract the high-frequency signal obtained for the minimum variance window for all rows along the orbit. If it turns out to be not possible to determine a correction for a particular orbit, the across-track variability correction from the previous orbit is taken.

Figure 4 shows corrections for across-track variability computed from 10 consecutive orbits measured on the same day (22 September 2004). Although the corrections have been computed from independent data, they are remarkably similar. This justifies taking the correction determined from the previous orbit if for some reason a correction cannot be determined from the actual orbit. Furthermore, Fig. 4 provides further evidence that irradiance data are most likely at the basis of the across-track variability. All orbits shown have been retrieved with the same irradiance measurement. However, when the correction was plotted for the first orbit retrieved with a new irradiance measurement, a distinctly different correction pattern was seen (not shown).

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## 4.2 Slant column precision

In this section we estimate the DOAS fitting error of individual NO<sub>2</sub> slant columns. We do so by a statistical analysis of OMI NO<sub>2</sub> columns observed over separate 2°×2° boxes in the meridional band between 178° and 180° W. The basic assumption is that the OMI pixels within a 2°×2° box observe the same total vertical column over this clean part of the Pacific Ocean. Any variability in the observed total vertical columns is assumed to originate from errors in the slant columns if the ensemble of AMFs within a box has little variability. Boxes with appreciable AMF variability were rejected if the box-mean vertical column computed by averaging the ensemble of individual slant columns ratioed by a box-mean AMF ( $\overline{V'}$ ) differed by more than 0.1% with the box-mean vertical column computed by averaging the ensemble of individual slant columns ratioed by the original AMFs ( $\overline{V}$ ). We find that for most of the 2°×2° boxes,  $\overline{V}$  and  $\overline{V'}$  indeed do not differ by more than 0.1%, and thus the slant columns have been observed under almost identical viewing geometries. For these boxes, we take the standard deviation of the ensemble of slant-column as a reasonable estimate for the precision of the slant columns.

For 7 August 2006, we computed estimates for the slant column errors for every box between 60° S and 60° N. The results are shown in Fig. 5a for boxes with a relative difference between  $\overline{V}$  and  $\overline{V'}$  of less than 0.1%. The figure shows that there is no appreciable change of fitting error with latitude. Averaged over all latitudes, the fitting error is close to  $0.75 \times 10^{15}$  molec.cm<sup>-2</sup> (dashed line). For the vertical column error, the contribution from the fitting error is smaller than  $0.4 \times 10^{15}$  molec.cm<sup>-2</sup> (solid line, computed as  $\sigma_N/M_{tr}$ ). We also made a distribution of all member deviations from box means and collected these deviations in a histogram ( $n=2893$ ). This is shown in Fig. 5b. The distribution closely follows a Gaussian shape that is fitted to the histogram data. The fact that the distribution follows a Gaussian distribution is consistent with our assumption that the variability within each box is dominated by random errors in the slant columns, originating from measurement noise. The corresponding width of

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the Gaussian for this day is  $0.67 \times 10^{15} \text{ molec.cm}^{-2}$ , and we interpret this as a reasonable estimate for the average slant column error of all 2893 pixels we investigated on 7 August 2006. We also looked into a few other days and other longitude bands with different across-track variability corrections, and found very similar numbers. The slant column error for an individual OMI pixel is somewhat larger than the better-than-10% ( $\approx 0.3 \times 10^{15} \text{ molec.cm}^{-2}$ ) number quoted in Boersma et al. (2002). This number was computed from synthetic spectra (i.e. without across-track variability) under the assumption that 4 OMI pixels would be binned (increasing signal-to-noise by a factor of 2). For individual pixels, the OMI fitting error is larger than the  $0.45 \times 10^{15} \text{ molec.cm}^{-2}$  found for GOME (Boersma et al., 2004) and SCIAMACHY (I. DeSmedt, personal communication), consistent with the higher signal-to-noise ratios for individual GOME and SCIAMACHY pixels than for OMI pixels.

## 5 Stratospheric contribution and tropospheric air mass factor

### 5.1 Stratospheric slant column

In order to determine the tropospheric  $\text{NO}_2$  column accurately, the stratospheric component of the slant column density needs to be predicted. We attain this by data-assimilation of OMI slant columns in TM4:

- Modelled  $\text{NO}_2$  profiles are convolved with the appropriate averaging kernel to give model-predicted slant column densities. The differences between observed and modelled columns (the model innovations) are used to force the modelled columns to generate an analysed state that is based on the modelled  $\text{NO}_2$  distribution and the OMI observations.
- The forcing depends on weights (from observation representativeness and model errors) attributed to model and observation columns. Observed columns are attributed a low weight in regions and times with large tropospheric model columns.

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This reflects the uncertainty in the averaging kernel, and minimizes the influence of slant columns with an estimated large tropospheric contribution.

- The forcing equation (based on the Kalman filter technique) is solved with the statistical interpolation method. This involves a covariance matrix that describes the forecast error and spatial correlations. The most important characteristics of this forecast covariance matrix are: (1) the conservation of modelled profile shapes. The altitude dependence of the forecast error is set to be proportional to the local NO<sub>2</sub> profile shape. (2) The horizontal correlation model function is assumed to follow a Gaussian shape with a  $1/e$  correlation length of 600 km. This length was derived from the assimilation of ozone in the stratosphere (Eskes et al., 2003) and should be a reasonable guess for stratospheric NO<sub>2</sub> as well. (3) In addition we introduce a correlation scaling to reduce the correlation with increasing differences in concentration (Riishøjgaard, 1998). This avoids problems with negative concentrations near sharp gradients that occur for instance at the polar vortex edge. (4) The vertical distribution of the assimilation increments is determined by the covariance matrix and the averaging kernel profile. The kernel peaks in most cases in the stratosphere, which is an additional reason why the adjustment caused by the assimilation is mainly taking place in the stratosphere.
- The adjustments made by the assimilation are therefore mainly occurring in the stratosphere at places where the tropospheric concentrations are low. The stratospheric information inserted by the assimilation is transported to the stratosphere above more polluted areas by the advection in the model.
- The model NO<sub>x</sub> species (NO, NO<sub>2</sub>, NO<sub>3</sub>, N<sub>2</sub>O<sub>5</sub>, HNO<sub>4</sub>) are assumed to be fully correlated and are all scaled in the same way as NO<sub>2</sub>.
- Based on the most recent analysed state, a forecast run of the model predicts the stratospheric field. This is used by the near-real time retrieval branch as shown in Fig. 2.

For further reading on the assimilation method we refer to [Eskes et al. \(2003\)](#). The advantage of the approach is that slant column variations due to stratospheric dynamics are now accounted for in the retrieval. The purpose is to improve the detection limit for tropospheric NO<sub>2</sub> columns that can be retrieved. An additional advantage is that the assimilation scheme provides a statistical estimate of the uncertainty in the stratospheric slant column. Generally, this statistical estimate is on the order of 0.1–0.2×10<sup>15</sup> molec.cm<sup>-2</sup>, much smaller than the slant column uncertainty reported on in the previous section. Hence, the detection limit in our method is mainly determined by the random slant column error ( $\sigma_S$ ) that is easily averaged out by taking large numbers of observations. Not accounting for stratospheric dynamics however, would lead to systematic errors in the estimate of the stratospheric column ([Boersma et al., 2004](#)) that cannot be easily averaged out and would raise the detection threshold.

## 5.2 Tropospheric air mass factor

We convert the tropospheric slant column into a vertical column by using a tropospheric AMF (Eq. 1). For OMI NRT we follow as much as possible the same approach (same look-up tables, computational methods) as for the GOME and SCIAMACHY data set described in Sect. 3.1. But the forward model input parameters cloud fraction and cloud pressure differ between the OMI NRT and the GOME and SCIAMACHY RAM retrievals. This, along with the much finer spatial resolution of OMI compared to GOME, is expected to lead to different error budgets for OMI tropospheric NO<sub>2</sub> than for GOME and SCIAMACHY.

### 5.2.1 Cloud parameters

DOAS-type retrievals are very sensitive to errors in cloud parameters. Boersma et al. (2004) showed that errors in FRESCO cloud fractions of ±0.05 lead to retrieval errors of up to 30% for situations with strong NO<sub>2</sub> pollution. Errors in the cloud pressure may also affect retrievals, especially in situations where the retrieved cloud top is lo-

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cated within a polluted layer. In such situations, errors in the cloud pressure of 50 hPa lead to retrieval errors of up to 25%.

Here we evaluate any differences between cloud parameters retrieved by SCIAMACHY and OMI. Cloud retrievals using FRESKO have been the subject of intensive validation. FRESKO applied on GOME measurements compared favourably to AVHRR (Koelemeijer et al., 2001) and ISCCP (Koelemeijer et al., 2002) cloud fractions (differences <0.02) and cloud pressure (differences <50–80 hPa). Of more relevance to our work is the application of FRESKO on SCIAMACHY measurements, as reported on by Fournier et al. (2006). They showed that FRESKO cloud parameters are in good agreement with other SCIAMACHY cloud algorithms, and estimate that the accuracy of the effective cloud fraction from FRESKO is better than 0.05 for all surfaces apart from snow and ice.

The retrieval methods for SCIAMACHY (FRESKO) and OMI ( $O_2-O_2$ ) are based on the same principles, i.e. they both retrieve an effective cloud fraction, that holds for a cloud albedo of 0.8, and to do so, they both use the continuum top-of-atmosphere reflectance as a measure for the brightness, or cloudiness, of a scene. Furthermore they both use the depth of an oxygen band as a measure for the length of the average photon path from the Sun, through the atmosphere back to the satellite instrument. The length of this light path is converted to cloud pressure. On the other hand, there are some distinct differences between the cloud parameter retrievals from SCIAMACHY and OMI:

1. FRESKO uses reflectances measured inside and outside of the strong oxygen A band (758–766 nm), whereas the OMI cloud retrieval uses the weakly absorbing  $O_2-O_2$  band at 477 nm.
2. FRESKO and  $O_2-O_2$  have different sensitivities to cloud pressure. This originates from the use of absorption by a single molecule (FRESKO,  $O_2$  A band), scaled with oxygen number density, versus the use of absorption by a collision complex ( $O_2-O_2$ ), scaled with oxygen number density squared. This is expected to lead

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to higher cloud pressures for the  $O_2-O_2$  cloud algorithm.

3. SCIAMACHY observes clouds at 10:00 h local time, and OMI at 13:45 h local time.

Because SCIAMACHY and OMI observe the Earth at different times, FRESCO and  $O_2-O_2$  cloud parameters cannot be compared directly. However, since temporal variation in global cloud fraction and cloud pressure between 10:00 h and 13:30 h is small (Bergman and Salby, 1996), frequency distributions of cloud parameters may be compared as a first evaluation of consistency between the two cloud retrievals. Here we compare SCIAMACHY FRESCO cloud retrievals version SC-v4 (August 2006, with improved desert surface albedos) to OMI  $O_2-O_2$  retrievals v1.0.1.1 (available since 7 October 2005, orbit 6541). We compare the cloud retrievals for locations between  $60^\circ$  N and  $60^\circ$  S to avoid situations with ice and snow, where cloud retrieval traditionally is difficult. We gridded FRESCO and  $O_2-O_2$  cloud parameters to a common  $0.5^\circ \times 0.5^\circ$  grid and selected only grid cells filled with successfully retrieved cloud values for both FRESCO and  $O_2-O_2$ . Doing so, we obtain SCIAMACHY and OMI cloud parameter distributions on a spatial grid comparable to the SCIAMACHY grid ( $30 \times 60 \text{ km}^2$ ) that have been sampled on the same days and locations.

Figure 6a shows the cloud fraction distribution as observed by FRESCO (dashed line) and  $O_2-O_2$  averaged over the period 5–11 August 2006. The distributions show a high degree of similarity, with the smallest effective cloud fractions observed most often. The differences between the two distributions are most appreciable for small effective cloud fractions, with OMI more frequently observing cloud fractions smaller than 0.05, and SCIAMACHY more frequently observing cloud fractions in the 0.05–0.20 range. These differences are likely related to different surface albedo data bases used in the cloud retrievals, and the way effective cloud fractions outside the 0.0–1.0 range are treated. The  $O_2-O_2$  retrieval uses the TOMS/GOME surface albedo datasets at 463 and 494.5 nm, consistent with albedos used for  $NO_2$  AMF computations. But for FRESCO, the GOME albedo dataset at 760 nm is used. Since OMI's horizontal resolution is much finer than the  $1^\circ \times 1^\circ$  surface albedo datasets, this will lead

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to cloud fraction errors, especially for small cloud fractions. On average for 5–11 August 2006, FRESCO observes a mean effective cloud fraction of 0.311, and  $O_2-O_2$  observes 0.300. The small mean difference between FRESCO and  $O_2-O_2$  of 0.011 is encouraging and gives a first order indication of the accuracy of the  $O_2-O_2$  cloud fraction retrieval.

Because the FRESCO version SC-v4 (August 2006) does not account for Rayleigh scattering (reflectances at the top-of-atmosphere are modelled by a purely  $O_2$ -absorbing atmosphere), FRESCO cloud pressures are likely to be underestimated for small cloud fractions. This effect has been discussed by Wang et al. (2006) for GOME and they showed that taking into account Rayleigh scattering in the GOME FRESCO retrieval on average increases cloud pressures by 60 hPa (for cloud fractions larger than 0.1). To avoid the effects of neglecting Rayleigh scattering in our comparison of FRESCO (that does not account for Rayleigh scattering) and  $O_2-O_2$  (that does account for Rayleigh scattering), we have restricted ourselves to situations with cloud fractions higher or equal to 0.05, where the signal from Rayleigh scattering is outshined by the signal from the cloudy part of the scene. Figure 6b shows the distributions of cloud pressures observed by FRESCO (dashed line) and  $O_2-O_2$  averaged over the period 5–11 August 2006. The figure shows that  $O_2-O_2$  more frequently retrieves high cloud pressures than FRESCO. On average,  $O_2-O_2$  cloud pressures are 58 hPa higher than FRESCO cloud pressures. As discussed above,  $O_2-O_2$  retrievals are more sensitive to levels deeper in the cloud, as the absorber slant column scales with the oxygen number density (or pressure) squared profile rather than with the single oxygen molecule number density profile as in the FRESCO retrieval. It is important for the  $NO_2$  retrieval to use the most appropriate cloud pressure in the context of the AMF computation. The “best” cloud pressure is the pressure level that, within the concept of the Lambertian reflector, indicates the effective scattering height for photons (in the 405–465 nm range). Dedicated validation campaigns with simultaneous observations of  $NO_2$  profiles and cloud parameters that coincide with OMI observations will help address these issues.

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## 5.2.2 Profile shape and representativity issues

In previous work, we estimated that errors in the a priori profiles obtained from a 3-D CTM give rise to approximately 10% error in the retrieved NO<sub>2</sub> columns (Boersma et al., 2004). Since then, Martin et al. (2004) and Schaub et al. (2006) showed that vertical distributions of NO<sub>2</sub> over southern U.S. and over northern Switzerland calculated with a CTM (GEOS-Chem and TM4 respectively) are consistent with observed NO<sub>2</sub> profiles in these regions, increasing confidence in the model-derived a priori profile shapes.

For OMI (as well as for reprocessed GOME and SCIAMACHY data), a priori NO<sub>2</sub> profile shapes are obtained from TM4, the successor of TM3. In earlier retrievals, the a priori profile shape was sampled at the TM3 spatial resolution of 5°×3.75°. TM4 vertical distributions are now sampled at 3°×2°. This resolution is still too coarse to properly resolve vertical distributions at OMI-scale resolution (roughly 0.1°×0.1°). Thus a certain degree of spatial smearing or smoothing may be expected. This also holds for the surface albedo (Koelemeijer et al., 2003; Herman and Celarier, 1997), stored on a 1.0°×1.25°-grid. Sub-grid variation in albedo, for instance as a result of land-sea or land-snow transitions, will lead to smearing errors in the final retrieval product. One other source of error that may lead to significant retrieval errors for high spatial resolution instruments such as SCIAMACHY and OMI is surface pressure (or altitude). Retrieval algorithms that use surface pressures from a coarse-resolution CTM-run typically undersample surface pressure over regions with marked topography. For instance over northern Switzerland this leads to significant retrieval errors (Schaub et al., 2006b), suggesting that future retrievals need to use surface pressure fields at a resolution compatible with the satellite observations.

## 5.2.3 Error budget

Table 1 compares contributions to AMF errors for polluted situations as presented in Boersma et al. (2004) for GOME to our best estimates for OMI in this work. We estimate that the uncertainty in the O<sub>2</sub>–O<sub>2</sub> cloud fractions is ±0.05. Although the cloud

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pressure uncertainty for OMI is comparable to that for GOME, there is a stronger sensitivity to cloud pressure errors for low clouds (OMI cloud pressures are on average 58 hPa higher than FRESKO cloud pressures) within the polluted NO<sub>2</sub> layer (see Fig. 5b in Boersma et al., 2004). For GOME, with pixel sizes comparable to the grid size of the forward model input parameters (albedo, profile shape, surface pressure), undersampling errors could be discarded. But for OMI with its much smaller pixel size this can no longer be done. However, undersampling effects are relevant for some regions and times only. For instance, coarse-gridded surface pressures and albedos over a flat, rather homogeneous area like northern Germany will have little effect on fine-scale OMI retrievals. Over that same area though, spatial gradients in NO<sub>2</sub> profiles are not resolved by the 3° by 2° TM4 model, and this may lead to smearing errors. Since we have little quantitative information about undersampling errors, and because their effect is limited to certain locations and times (and highly variable), we indicate them in our table as  $\epsilon_U$  (unknown undersampling error). Table 1 shows that the AMF error budget for individual retrievals from well quantified error sources is similar: 29% for GOME and 31% for OMI. However, previously discarded undersampling, or representativity errors, may contribute considerably to especially OMI AMF errors for specific locations and times. Again, these issues may be addressed through dedicated validation efforts and retrieval improvements including the use of high(er)-resolution surface pressure and albedo data bases, and improved spatial resolution chemistry-transport models.

Table 2 summarizes the contribution of various errors to the overall error budget for individual retrievals in cloud-free situations. Pixels are defined as cloud-free when the cloud radiance fraction does not exceed 50%, which corresponds to effective cloud fractions smaller than 15–20%. For comparison, in Table 2 we also included error estimates for GOME and SCIAMACHY retrievals computed with the RAM-approach. The uncertainty in the AMF is determined by errors in the forward model parameters as shown in Table 1. These error contributions are quite similar as in Boersma et al. (2004), although we expect that in certain locations and times smearing effects are larger for OMI than for SCIAMACHY and especially GOME. We estimate that a rough

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1-sigma uncertainty for an individual OMI retrieval can be expressed as a base component (from spectral fitting uncertainty divided by AMF value, see Eq. 5 in Boersma et al., 2004) plus a relative uncertainty in the 10%–40% range due to AMF uncertainty. The base component is in the  $0.5\text{--}1.5 \times 10^{15}$  molec.cm<sup>-2</sup> range ( $0.5 \times 10^{15}$  molec.cm<sup>-2</sup> for situations with high tropospheric AMF values,  $1.5 \times 10^{15}$  molec.cm<sup>-2</sup> for situations with low tropospheric AMF values).

## 6 Intercomparison of OMI and SCIAMACHY tropospheric NO<sub>2</sub>

We evaluate the consistency of OMI and SCIAMACHY tropospheric NO<sub>2</sub> retrievals for August 2006. The SCIAMACHY set has been retrieved with the RAM-approach, and has been described and validated in Blond et al. (2006)<sup>1</sup> and van der A et al. (2006). For all days on which both SCIAMACHY and OMI reported a cloud-free observation in the same 0.5°×0.5° grid cell, we averaged the data. Doing so, we avoid artificial differences that would otherwise show up as a consequence of the differences in spatial and day-to-day sampling between the two satellite instruments. As in the cloud parameter comparison, we have averaged the satellite data on a 0.5°×0.5° grid, as the size of these grid cells is in the same range as the size of an individual SCIAMACHY pixel (30×60 km<sup>2</sup>). One important sampling difference remains as SCIAMACHY observations have been taken at 10:00 hr local time whereas OMI observations have been taken at 13:45 h local time.

Figure 7 shows the monthly mean tropospheric NO<sub>2</sub> field derived from SCIAMACHY and OMI for August 2006. For grey grid cells, the requirement that both SCIAMACHY and OMI have observed at least one, cloud-free scene in August 2006 has not been fulfilled. The spatial distributions tropospheric NO<sub>2</sub> observed by SCIAMACHY and OMI look consistent. Both satellite instruments observe the highest values over the industrial regions of North America, Europe, and China, and moderate levels of pollution in the biomass burning regions of South America, Southern Africa, and Northern Australia. Also, weak plumes of NO<sub>2</sub> over tropical oceans originating from especially

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biomass burning and lightning in Africa, South America, and Australia, are observed by both SCIAMACHY and OMI.

The OMI mean tropospheric NO<sub>2</sub> field is much smoother than the SCIAMACHY field. This can be understood from the fact that 10× more pixels have been used in the OMI composite than in the SCIAMACHY composite, leading to much better statistics for the OMI average. Figure 7b also shows that after averaging the data over a month, features related to across-track variability may still be visible.

SCIAMACHY and OMI tropospheric NO<sub>2</sub> show appreciable consistency ( $r=0.77$ ,  $n=1.9\times 10^5$ ). For clean, unpolluted regions that make up the bulk of the grid cells, Fig. 7 shows quantitative agreement between OMI and SCIAMACHY. However, for strongly polluted regions, SCIAMACHY observes up to 30% higher values than OMI. We expect that these differences are to considerable extent due to differences in emissions, meteorological (boundary layer mixing, transport) and (photo) chemical regimes between 10:00 h and 13:45 h. Furthermore, we observe a strong land-sea contrast in tropospheric NO<sub>2</sub>, especially over the Mediterranean. A manuscript with a detailed investigation of these differences is in preparation.

As a further illustration of OMI's capabilities, we show monthly averaged tropospheric NO<sub>2</sub> columns over Europe. Other than in Fig. 7, here we did not sample SCIAMACHY and OMI identically in space and time. We computed monthly averages on a 0.1°×0.1° grid to emphasize OMI's spatial resolution and sampled whenever a cloud-free observation was available. Figure 8 shows monthly mean tropospheric NO<sub>2</sub> columns from OMI (left panel) and from SCIAMACHY (right panel) in August 2006, a month with almost permanent cloud-cover over north-western Europe. The left panel shows that it was still possible to compute a monthly mean based on cloud-free OMI measurements. But SCIAMACHY – with less spatial and temporal coverage – did not record any cloud-free measurements over large parts of Europe, so that many grid cells show up grey in Fig. 8b. Apart from the better spatial and temporal coverage, the OMI average also shows much more spatial detail in the tropospheric NO<sub>2</sub> field. For instance, individual large cities including Madrid, Paris, and Moscow can be tracked

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down easily on the OMI map. Industrial regions such as the Po Valley, the Ruhr Area, and large parts of the UK also stand out.

## 7 Conclusions

The DOMINO near-real time algorithm retrieves tropospheric NO<sub>2</sub> columns from OMI within 3 h after measurement. This is possible with a new technique that is based on the assimilation of recently observed OMI NO<sub>2</sub> slant columns in the TM4 CTM. After the most recent observations have been digested by the assimilation scheme, TM4 is run forward in time with forecast ECMWF meteorological fields to predict the required retrieval inputs. Because of this, these inputs (stratospheric slant column, NO<sub>2</sub> and temperature profile) are available when a newly processed orbit of NO<sub>2</sub> and cloud data arrives at KNMI, and retrieval of tropospheric NO<sub>2</sub> columns is completed within a few minutes upon arrival of the data. We introduced a correction method for across-track variability associated with calibration errors in the OMI level 1b data and show that our procedure removes most of the spurious across-track variability. A simple statistical approach has been used to estimate the uncertainty in the slant columns. We find that the random error in the slant column is approximately  $0.70 \times 10^{15} \text{ molec. cm}^{-2}$  for a single OMI observation.

From a comparison of SCIAMACHY cloud parameters retrieved by FRESKO from the O<sub>2</sub> A band, and OMI cloud parameters retrieved from the O<sub>2</sub>–O<sub>2</sub> absorption band at 477 nm, we find similar distributions of cloud fractions and cloud pressures. On average, SCIAMACHY cloud fractions are higher by 0.011 than OMI cloud fractions. OMI cloud pressures are approximately 60 hPa higher than FRESKO cloud pressures, consistent with different sensitivities of the two algorithms. The consistency between the SCIAMACHY and OMI cloud parameters, and the similar design and inputs for the NO<sub>2</sub> algorithms provide confidence in the OMI retrieval approach.

As a first-order test of OMI NRT NO<sub>2</sub> retrievals, we compared SCIAMACHY and OMI tropospheric NO<sub>2</sub> columns – identically sampled – for the month August 2006.

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After regridding to a common  $0.5^\circ \times 0.5^\circ$  grid, the only sampling difference is due to the 10:00 h SCIAMACHY and the OMI 13:45 h equator crossing time. The comparison shows that the two instruments observe the same spatial  $\text{NO}_2$  patterns over the globe ( $r=0.77$ ,  $n=1.9 \times 10^5$ ). The quantitative agreement between SCIAMACHY and OMI is appreciable over areas with little  $\text{NO}_2$  pollution. Over polluted areas, OMI observes up to 30% smaller tropospheric  $\text{NO}_2$  amounts than SCIAMACHY, related to differences in emissions, meteorology (boundary layer mixing, transport), and (photo) chemistry between 10:00 h and 13:45 h. This issue will be further investigated in a future study. Monthly mean maps of Europe, the U.S., and eastern Asia show the distribution of  $\text{NO}_2$  with a large amount of detail.

We expect that for  $\text{NO}_2$  from OMI, in contrast with the previous error analyses for GOME, a new type of retrieval error becomes increasingly relevant. Forward model parameters, including a priori profile shapes, surface albedo's, and surface pressures, are currently obtained from data bases with spatial resolutions much coarser than the actual spatial resolution of the retrieval. This is expected to lead to significant retrieval errors for some locations and times. We recommend detailed studies into the extent of these errors, and furthermore strongly encourage validation activities. Higher spatial resolution a priori information and models are needed for the full exploitation of the high resolution OMI data.

*Acknowledgements.* The near-real time retrieval was developed within the DOMINO project, "Derivation of Ozone Monitoring Instrument tropospheric  $\text{NO}_2$  in near-real time", funded by the NIVR and the Dutch User Support Programme, project code USP 4.2 DE-31. The authors would like to thank D. Schaub (EMPA) and J. de Haan for useful discussions. Thanks to the NASA SIPS for processing the slant column near-real time  $\text{NO}_2$  and cloud data, and delivery to the KNMI system. I. DeSmedt and M. Van Roozendaal (BIRA) are kindly acknowledged for their retrievals of SCIAMACHY  $\text{NO}_2$  slant columns.

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**Table 1.** Overview of forward model error contributions to the (relative) AMF uncertainty for GOME and OMI.  $\epsilon_{u_a}$  is the unknown error due to albedo undersampling,  $\epsilon_{u_p}$  due to profile shape undersampling, and  $\epsilon_U$  the overall undersampling AMF error that is highly variable in space and time.

Error source	GOME (Boersma et al., 2004)		OMI (this work)	
	Uncertainty	Uncertainty $M_{tr}$	Uncertainty	Uncertainty $M_{tr}$
Surface albedo	0.02	15%	$0.02 + \epsilon_{u_a}$	$15\% + \epsilon_{u_a}$
Cloud fraction	0.05	30%	0.05	30%
Cloud pressure	50–80 hPa	2%	60 hPa	15%
Profile shape	N.A.	9%	N.A.	$9\% + \epsilon_{u_p}$
Total AMF error		29%		$31\% + \epsilon_U$

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**Table 2.** Contributions to the overall OMI tropospheric NO<sub>2</sub> retrieval error for individual, cloud-free pixels retrieved with the KNMI retrieval-assimilation-modelling approach (with the exception of the OMI standard product).

\*It is anticipated that improved lv1 calibration (due Spring 2007) and standard-product correction for spurious across-track variability will reduce the OMI standard product NO<sub>2</sub> slant column uncertainty.

\*\*For “mostly clear” conditions, Bucsela (personal communication, 2006) estimates 15–30% AMF uncertainty range.

Instrument	Reference	$\sigma_S$	$\sigma_{S_{sl}}$	$\sigma_{M_{tr}}$
OMI near-real time product	This work	$0.70 \times 10^{15}$ molec.cm <sup>-2</sup>	$0.15 \times 10^{15}$ molec.cm <sup>-2</sup>	10%–40%
OMI standard product	<a href="#">Bucsela et al. (2006)</a>	$1.10 \times 10^{15*}$ molec.cm <sup>-2</sup>	$0.20 \times 10^{15}$ molec.cm <sup>-2</sup>	not given**
SCIAMACHY	Blond et al. (2006) <sup>1</sup> , DeSmedt (2006)	$0.47 \times 10^{15}$ molec.cm <sup>-2</sup>	$0.25 \times 10^{15}$ molec.cm <sup>-2</sup>	10%–40%
GOME	<a href="#">Boersma et al. (2004)</a>	$0.45 \times 10^{15}$ molec.cm <sup>-2</sup>	$0.25 \times 10^{15}$ molec.cm <sup>-2</sup>	15%–50%

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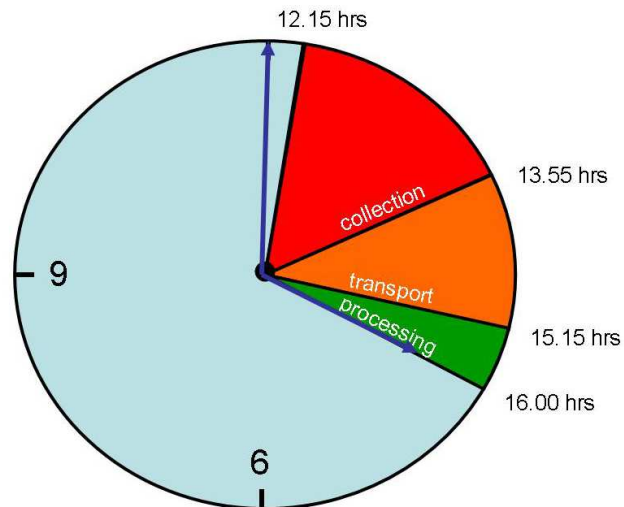
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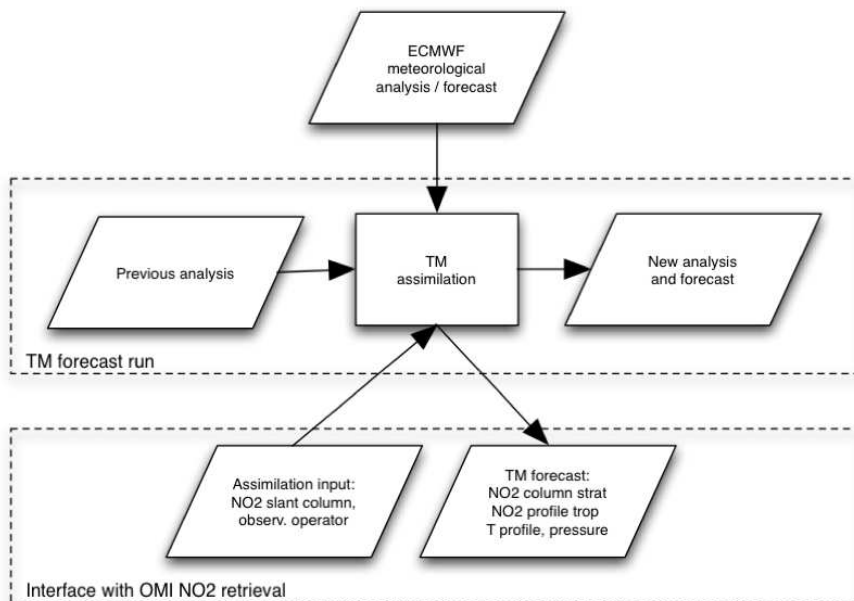
**Fig. 1.** OMI science data is linked down to ground stations once per orbit, resulting in a time delay between the OMI observations and reception at the ground station of at most 100 min. The collection, time-ordering, consecutive transfers, and level 1b and 2 processing, takes approximately 80 min. Final processing, image generation and web publishing generally occurs within 45 min.

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**Fig. 2.** Flowchart for the DOMINO forecast/assimilation subsystem. The lowest part shows the input-output interface with the NRT-subsystem.

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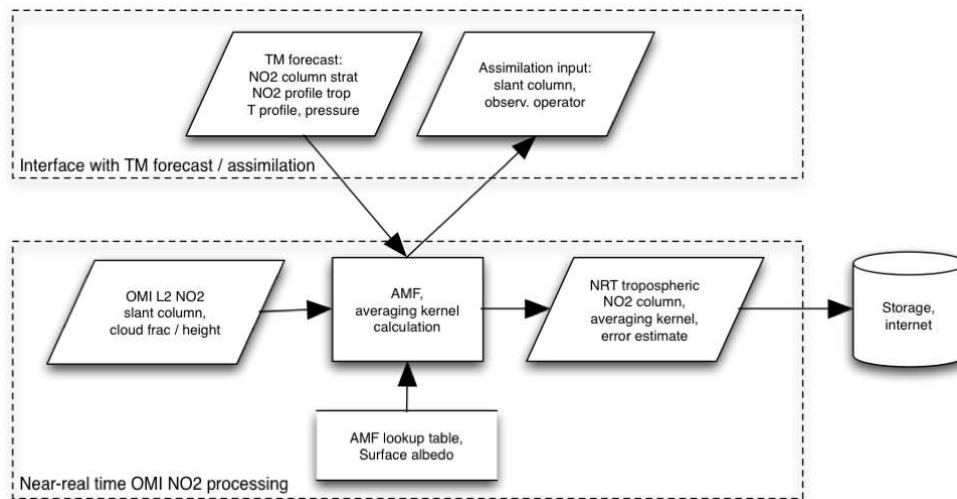
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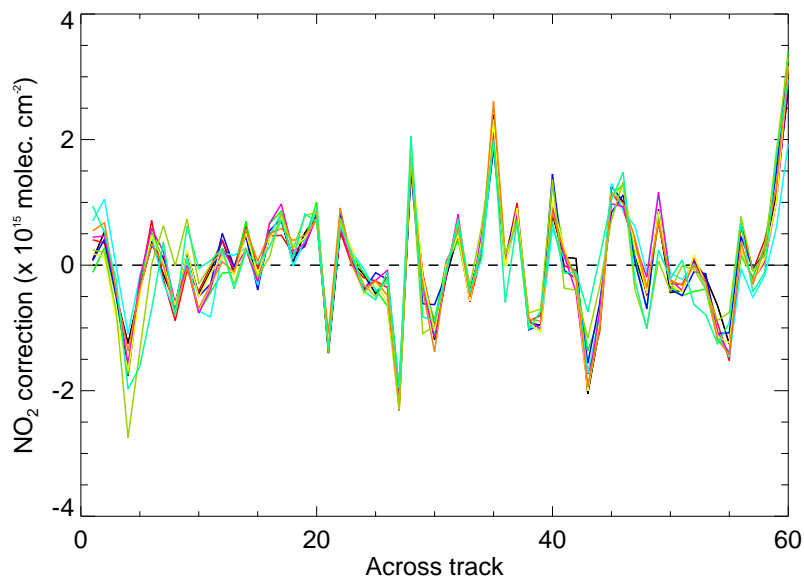


**Fig. 3.** Flowchart for the DOMINO NRT-subsystem. The upper part shows the input-output interface with the forecast/assimilation-subsystem (the lower part of Fig. 2).



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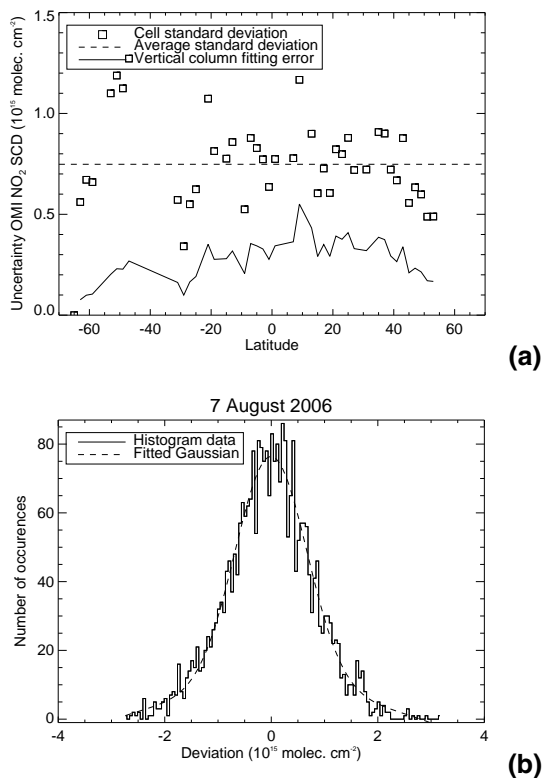
**Fig. 4.** Across-track variability corrections, computed with the method described in Sect. 4.1, for 10 consecutive orbits on 22 September 2004. Every orbital correction is shown with a unique colour.

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**Fig. 5.** (a) Left panel: standard deviation of slant columns within a  $2^\circ \times 2^\circ$  (longitudes between  $178^\circ$  and  $180^\circ$  W) box as a function of latitude. The dashed line shows the meridional average of the standard deviations for all the boxes. The solid line shows the contribution of the (slant column) fitting error to the vertical column error (i.e.  $S/M_{tr}$ ). (b) Right panel: distribution of member deviations from box means for 2893 pixels. The dashed line shows a Gaussian function fitted to the histogram data. The width of the Gaussian corresponds to a slant column error of  $0.67 \times 10^{15}$  molec.cm<sup>-2</sup>.

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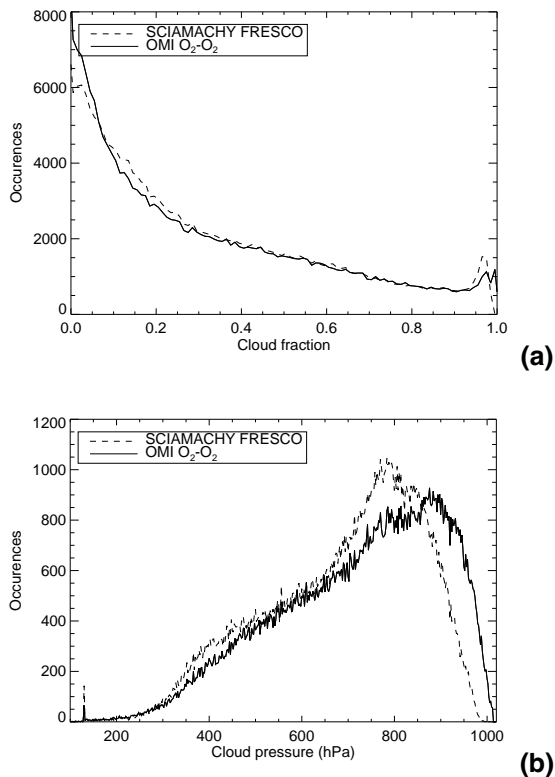
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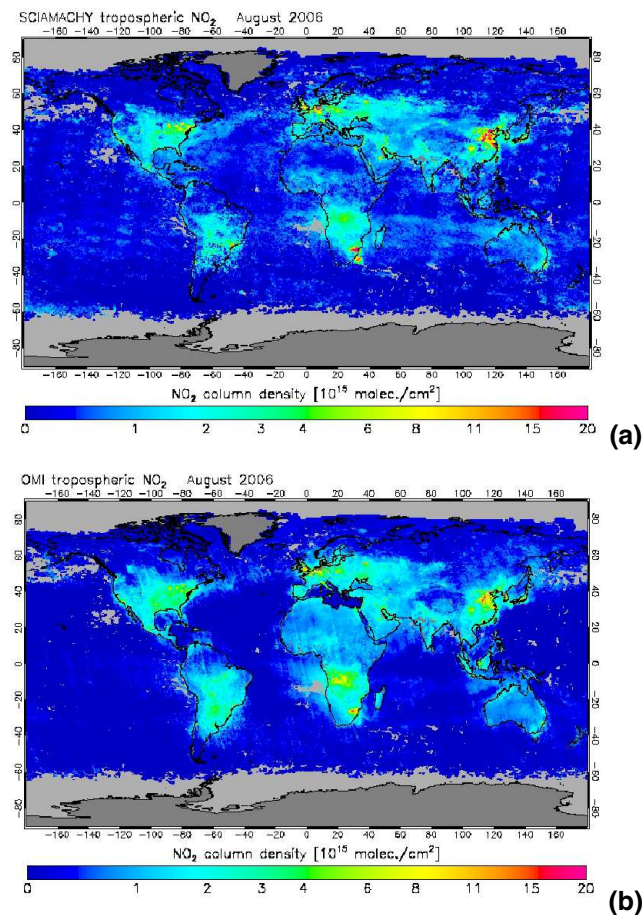
**Fig. 6.** (a) Left panel: histogram of  $0.5^\circ \times 0.5^\circ$  gridded effective cloud fractions from SCIAMACHY FRESCO (dashed line) and OMI  $\text{O}_2\text{-O}_2$  for seven consecutive days (5–11 August 2006). (b) Right panel: histogram of  $0.5^\circ \times 0.5^\circ$  gridded effective cloud pressures from SCIAMACHY FRESCO (dashed line) and OMI  $\text{O}_2\text{-O}_2$  for seven consecutive days (5–11 August 2006). Only observations with a cloud fraction higher or equal to 0.05 are shown.

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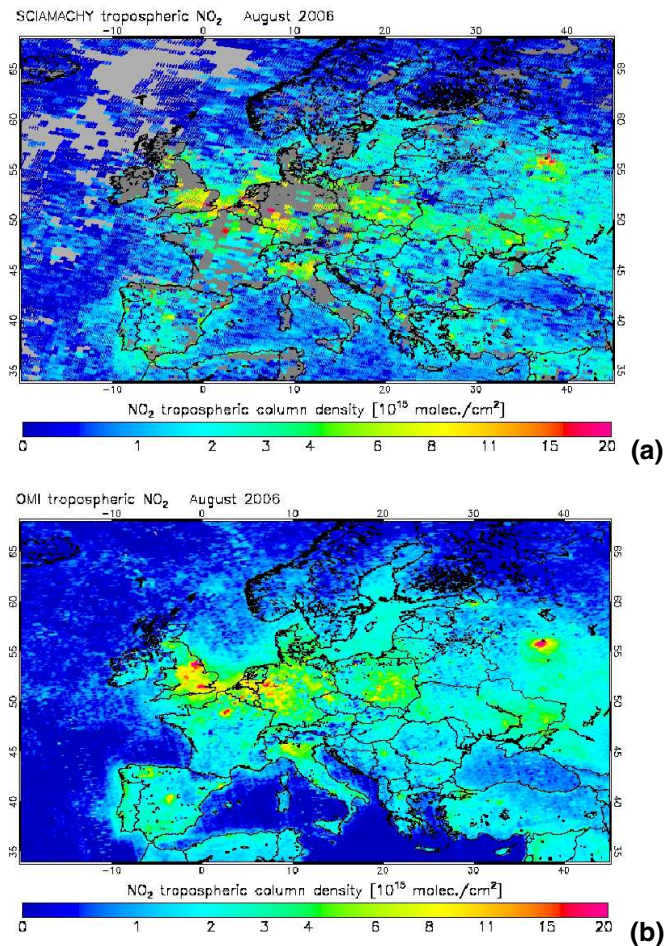


**Fig. 7. (a)** Monthly mean tropospheric NO<sub>2</sub> column from SCIAMACHY for situations when both SCIAMACHY and OMI observed a cloud-free scene. Grey grid cells have not been observed or had persistent cloud cover over August 2006. **(b)** Same as (a), but now for OMI.

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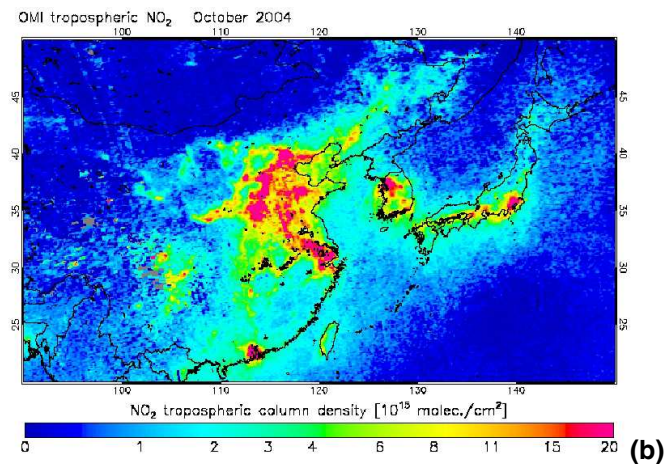
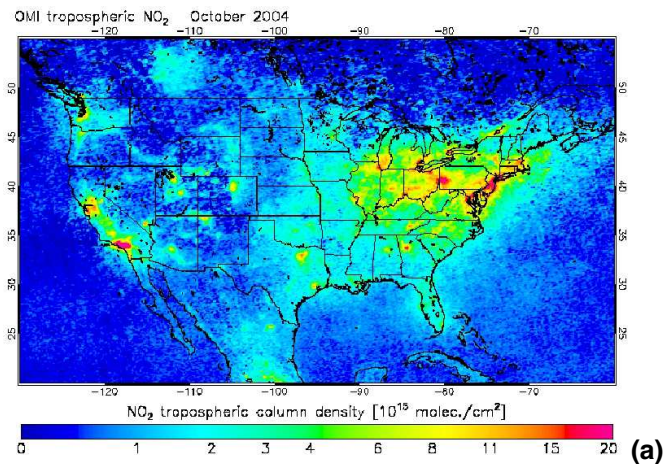


**Fig. 8.** (a) Monthly mean tropospheric NO<sub>2</sub> column from SCIAMACHY (left panel) for cloud-free situations. Grey grid cells have not been observed or had persistent cloud cover over August 2006. (b) Same as (a), but now for OMI.

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**Fig. 9.** (a) Monthly mean tropospheric NO<sub>2</sub> column from OMI over the U.S. for cloud-free situations. Grey grid cells have not been observed or had persistent cloud cover over October 2004. (b) Same as (a), but now over eastern Asia.

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