



Development of a custom OMI NO₂ data product for evaluating biases in a regional chemistry transport model

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Abstract. In this paper, we present the custom Hong Kong NO₂ retrieval (HKOMI) for the Ozone Monitoring Instrument (OMI) on board the Aura satellite which was used to evaluate a high-resolution chemistry transport model (CTM) (3 km × 3 km spatial resolution). The atmospheric chemistry transport was modelled in the Pearl River Delta (PRD) region in southern China by the Models-3 Community Multiscale Air Quality (CMAQ) modelling system from October 2006 to January 2007. In the HKOMI NO₂ retrieval, tropospheric air mass factors (AMFs) were recalculated using high-resolution ancillary parameters of surface reflectance, a priori NO₂ and aerosol profiles, of which the latter two were taken from the CMAQ simulation. We tested the influence of the ancillary parameters on the data product using four different aerosol parametrizations. Ground-level measurements by the PRD Regional Air Quality Monitoring (RAQM) network were used as additional independent measurements.

The HKOMI retrieval increases estimated tropospheric NO₂ vertical column densities (VCD) by (+31 ± 38)%, when compared to NASA's standard product (OMNO2-SP), and improves the normalized mean bias (NMB) between satellite and ground observations by 26 percentage points from -41 to -15%. The individual influences of the parameters are (+11.4 ± 13.4)% for NO₂ profiles, (+11.0 ± 20.9)% for surface reflectance and (+6.0 ± 8.4)% for the best aerosol parametrization. The correlation coef-

ficient r is low between ground and satellite observations ($r = 0.35$). The low r and the remaining NMB can be explained by the low model performance and the expected differences when comparing point measurements with area-averaged satellite observations.

The correlation between CMAQ and the RAQM network is low ($r \approx 0.3$) and the model underestimates the NO₂ concentrations in the northwestern model domain (Foshan and Guangzhou). We compared the CMAQ NO₂ time series of the two main plumes with our best OMI NO₂ data set (HKOMI-4). The model overestimates the NO₂ VCDs by about 15% in Hong Kong and Shenzhen, while the correlation coefficient is satisfactory ($r = 0.56$). In Foshan and Guangzhou, the correlation is low ($r = 0.37$) and the model underestimates the VCDs strongly (NMB = -40%). In addition, we estimated that the OMI VCDs are also underestimated by about 10 to 20% in Foshan and Guangzhou because of the influence of the model parameters on the AMFs.

In this study, we demonstrate that the HKOMI NO₂ retrieval reduces the bias of the satellite observations and how the data set can be used to study the magnitude of NO₂ concentrations in a regional model at high spatial resolution of 3 × 3 km². The low bias was achieved with recalculated AMFs using updated surface reflectance, aerosol profiles and NO₂ profiles. Since unbiased concentrations are important, for example, in air pollution studies, the results of this paper can be very helpful in future model evaluation studies.

1 Introduction

Nitrogen oxides (NO_x = NO + NO₂) play an important role in atmospheric chemistry. As precursors of ozone and aerosols, they are vital in the formation of photochemical smog (Haagen-Smit, 1952) and acid rain (Driscoll et al., 2001). In the troposphere, NO₂ concentrations have a high spatial and temporal variability due to their short lifetime and the variety of sources and sinks. This spatiotemporal variability has been studied with chemistry transport models (CTM), air quality monitoring networks and satellite instruments (e.g. Beirle et al., 2003; Huijnen et al., 2010; Han et al., 2011, 2015; Chan et al., 2012; Mueller et al., 2015).

The first satellite instrument able to detect tropospheric NO₂ was the Global Ozone Monitoring Experiment (GOME) which was launched on board the second European Remote Sensing satellite (ERS-2) in 1995 (Burrows et al., 1999). Successor instruments to GOME (GOME-2) are payload on the MetOp satellites used for operational meteorology (Callies et al., 2000). In 2006, the Scanning Imaging Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY) was launched on board the ENVIRONMENTAL SATellite (ENVISAT) (Bovensmann et al., 1999) and in 2004, the Ozone Monitoring Instrument (OMI) was launched on board the Aura satellite (Levelt et al., 2006). Since the launch of GOME, the spatial resolution of the instruments has increased rapidly. While GOME had a smallest ground pixel size of 40 km × 320 km, which was suitable for coarse global analyses, OMI has a smallest ground pixel size of 13 km × 24 km. The higher spatial resolution makes OMI applicable for the study of NO₂ in large metropolitan areas.

The OMI NO₂ standard products are the NASA standard product (OMNO2, Version 2.1) (Bucsela et al., 2006, 2013) and the Derivation of OMI tropospheric NO₂ product (DOMINO, Version 2.0) (Boersma et al., 2007, 2011). The NO₂ retrieval algorithms use ancillary parameters, such as surface reflectance, a priori NO₂ profiles, and aerosol and cloud information, to calculate air mass factors (AMFs). The AMFs convert the retrieved NO₂ slant column densities (SCDs) to vertical column densities (VCDs). In the standard products, a priori NO₂ profiles are taken from global CTMs with a spatial resolution of 2° × 2.5° (OMNO2) and 2° × 3° (DOMINO). Surface reflectances are taken from an OMI climatology with a spatial resolution of 0.5° × 0.5° (Kleipool et al., 2008).

Since NO₂ profile and surface reflectance have a large impact on the NO₂ VCD, the standard products are not directly suitable to study VCDs on a scale below the resolution of these ancillary parameters. However, when more accurate NO₂ profiles are available, for example in model evaluation studies, the VCDs can be corrected using scattering weights (SWs) or averaging kernels (AKs). The correction removes the dependency on the a priori profile and can be applied either to the OMI NO₂ VCDs or the model profiles (for details see Eskes and Boersma, 2003; Boersma

et al., 2005). SWs and AKs are closely related to vertically resolved AMFs and are provided with the standard products. AKs have been applied, for instance, by Herron-Thorpe et al. (2010), who validated the regional-scale air quality model AIRPACT-3 (12 km × 12 km spatial resolution) over the Pacific Northwest, and Zyrichidou et al. (2013), who evaluated the Comprehensive Air Quality Model (CAMx) (10 km × 10 km) over southeastern Europe. Applying AKs has a large influence on the magnitude of tropospheric NO₂ VCDs (Herron-Thorpe et al., 2010).

In the standard products, AKs and SWs depend on the OMI surface reflectance climatology. The spatial resolution of this climatology is coarse compared to the OMI ground pixel size. Therefore, customized NO₂ products have been developed which recalculate AKs, and thus AMFs, using high-resolution surface reflectance products (Zhou et al., 2010; Russell et al., 2011; McLinden et al., 2014). Furthermore, AKs and SWs are affected by aerosols directly due to additional scattering and absorption (Leitão et al., 2010) as well as indirectly due to their impact on the retrieval of surface reflectance and cloud properties (Boersma et al., 2011; Lin et al., 2014). Since the ancillary parameters have a large impact on the magnitude of the retrieved NO₂ VCDs, it is important to consider their influence, for example, in air pollution studies where unbiased pollutant concentrations are important.

For this paper, we developed a customized OMI NO₂ retrieval and applied it to the Pearl River Delta (PRD) region in southern China. The Hong Kong OMI (HKOMI) retrieval recalculates tropospheric AMFs and uses them with OMNO2 SCDs to calculate VCDs. The HKOMI product was used to evaluate the Models-3 Community Multiscale Air Quality (CMAQ) modelling system (3 km × 3 km spatial resolution) (Byun and Schere, 2006). The study period is October 2006 to January 2007. Three automatic weather stations and 16 ground-level stations of the PRD Regional Air Quality Monitoring (RAQM) network were used to validate model and retrieval. The objective is to estimate the influence of the ancillary parameters on the evaluation and to demonstrate some possibilities and limitations for using satellite-based NO₂ observations.

This paper is organized as follows: OMI and the retrieval of tropospheric NO₂ including the AMF calculations using the SCIATRAN radiative transfer model are described in Sect. 2. The RAQM network, the CMAQ model run and the HKOMI NO₂ retrieval are described in Sect. 3. The results are presented in Sect. 4 and discussed in Sect. 5. Finally, Sect. 6 concludes this paper.

2 Background

2.1 Ozone Monitoring Instrument (OMI)

OMI is a nadir-viewing imaging spectrometer measuring Earth's reflectance spectra in the near-ultraviolet and visible wavelength range with two charge-coupled device (CCD) arrays. It was launched aboard NASA's EOS Aura satellite on 15 July 2004 (Levelt et al., 2006). The instrument provides near-daily global coverage at an overpass time of $13:45 \pm 15$ min local time (LT). Earth reflectance spectra are measured during the sunlit part of about 14.5 sun-synchronous orbits per day. Trace gases, such as ozone (O₃), sulfur dioxide (SO₂) and nitrogen dioxide (NO₂), are retrieved from the reflectance spectra as well as cloud and aerosol properties. The measurement principle is an along-track (push-broom) scanner with a swath width of 2600 km which is divided into 60 pixels. The ground pixel size varies between 13 km × 24 km at nadir and 40 km × 160 km at the swath edge (Levelt et al., 2006). Since June 2007, OMI is affected by a row anomaly which reduces the number of valid measurements (see <http://www.knmi.nl/omi/research/product/rowanomaly-background.php> for details).

2.2 Standard retrieval of tropospheric NO₂ column densities

The OMNO2 product is the basis for the HKOMI retrieval. Therefore, we give a brief introduction to their algorithm. The OMNO2 retrieval algorithm has three major steps:

1. The total slant column densities S are obtained from the reflectance spectra using the differential optical absorption spectroscopy (DOAS) technique (Platt and Stutz, 2008).
2. The stratospheric slant column densities S_{strat} are subtracted from the total column S using a stratosphere–troposphere separation (STS) algorithm (Bucsela et al., 2013).
3. The tropospheric slant column densities S_{trop} are converted to vertical column density V_{trop} using a tropospheric AMFs A_{trop} (Palmer et al., 2001).

The AMF is defined as the ratio of slant and vertical column density (Solomon et al., 1987). Thus the tropospheric column density V_{trop} is calculated by

$$V_{\text{trop}} = \frac{S - S_{\text{strat}}}{A_{\text{trop}}} = \frac{S_{\text{trop}}}{A_{\text{trop}}}. \quad (1)$$

The tropospheric AMF depends on parameters such as sun position, instrument viewing direction, surface reflectance, atmospheric scattering properties due to air molecules, aerosols and clouds, and the a priori NO₂ profile. It is related to the vertical sensitivity of the satellite instrument and

can be computed for N vertical layers by

$$A_{\text{trop}} = \frac{\sum_{k=1}^N \alpha_k m_k V_k}{\sum_{k=1}^N V_k} \quad (2)$$

with:

- α_k : an empirical temperature correction coefficient accounting for the temperature dependency of the NO₂ absorption cross-section,
- m_k : the differential or box air mass factor which describes the instrument sensitivity for layer k ,
- V_k : the partial NO₂ VCD of layer k .

The products $\alpha_k m_k$ are the scattering weights provided with the standard product. The correction coefficient α_k can be computed by the empirical formula

$$\alpha_k = 1 - 0.003 \text{ K}^{-1} (T_k - T_{\text{ref}}), \quad (3)$$

where T_k is the temperature in layer k and T_{ref} is reference temperature of the NO₂ absorption cross-section ($T_{\text{ref}} = 220 \text{ K}$) (Bucsela et al., 2013).

The box air mass factors m_k can be computed with a radiative transfer model. The OMNO2 algorithm uses the TOMRAD radiative transfer model (Dave, 1965). The AMF formulation used in OMNO2 is based on a AMF formulation by Palmer et al. (2001). In partly cloudy scenes, OMNO2 computes the box AMFs using the independent pixel approximation (IPA). The approximation calculates AMFs as weighted sums of a cloudy m_k^{cloudy} and a clear m_k^{clear} component:

$$m_k = w \cdot m_k^{\text{cloudy}} + (1 - w) \cdot m_k^{\text{clear}}, \quad (4)$$

where w is the aerosol/cloud radiance fraction (CRF), that is the fraction of measured radiation that results from clouds and aerosols (Acarreta et al., 2004).

In OMNO2, the partial VCD V_k are taken from the Global Modeling Initiative (GMI) CTM (Duncan et al., 2007; Strahan et al., 2007) which combines stratospheric chemistry described by Douglass et al. (2004) and tropospheric O₃–NO_x–hydrocarbon chemistry from the GEOS-Chem model (Bey et al., 2001). It should be noted that, for an optical thin absorber, only the relative shape of the profile $n_k = V_k/V_{\text{trop}}$ is required for the AMF calculation.

Bucsela et al. (2013) estimated tropospheric NO₂ VCD uncertainties to be $1 \times 10^{15} \text{ cm}^{-2}$ for clear skies and up to $3 \times 10^{15} \text{ cm}^{-2}$ for large cloud radiance fractions. In polluted regions, the main uncertainties are the DOAS fit (10% relative error) and the tropospheric AMFs (20–80%).

2.3 SCIATRAN radiative transfer model

SCIATRAN is a one-dimensional RTM which can be used for the calculation of box AMFs (Rozanov et al., 2005). The model is designed as a forward model for the retrieval of atmospheric constituents from measurements of scattered light by satellite, ground or airborne instruments. The wavelength range goes from 175 to 2380 nm which includes the ultraviolet, visible and near-infrared part of the spectrum.

SCIATRAN solves the integro-differential radiative transfer equation using the discrete-ordinates method (DOM) in the plane-parallel or pseudo-spherical mode to calculate box AMF profiles. Box AMFs m_k are derived from weighting functions which describe the sensitivity of the reflectance spectrum R to a perturbation of a model parameter Δn in layer z_k . In SCIATRAN, weighting functions are computed by a quasi-analytic approach (Rozanov et al., 1998). If Δn is a perturbation of the NO₂ number density n_k , the box AMF m_k is the negative weighting function (Rozanov and Rozanov, 2010).

3 Methodology

In this section, we describe the RAQM network, the CMAQ CTM and the HKOMI retrieval. Our study period is from October 2006 to January 2007 which has been chosen because cloud fractions in the PRD region are lowest in this season and OMI measurements in later years are affected by the aforementioned row anomaly.

3.1 Ground networks

3.1.1 Meteorological observations

Meteorological observations were used for the evaluation of the simulated meteorological fields which were used to drive the CTM. The data were measured by three automatic weather stations in Hong Kong. The stations are located at the Hong Kong Observatory (HKO) headquarters, the Hong Kong International Airport (HKIA) and on Waglan Island (WGL) (see Fig. 1). HKO is in the city centre of Kowloon and surrounded by high buildings. HKIA is located on an artificial island near Tung Chung to the north of mountainous Lantau Island. Waglan Island is a small island located to the east of Hong Kong Island. The island is too small to be resolved by the model grid. The WGL station is located 56 m above sea level and used as background station by the Hong Kong Observatory. The used meteorological parameters are hourly measurements of temperature (T), humidity (q), sea surface pressure (p) and wind (v). Temperature and humidity have been measured at 2 m above ground level (a.g.l.).

3.1.2 Pearl River Delta regional Air Quality Monitoring network

Ground-level NO₂ mixing ratios were provided by the RAQM network. The network was established by the governments of the Guangdong province and Hong Kong to monitor the air quality in the PRD region and has been in operation since 30 November 2005. It consists of 16 automatic air quality monitoring stations (see Fig. 1, round markers). The network measures NO₂, SO₂, O₃ and PM₁₀ hourly. The monitoring network was used to validate the NO₂ mixing ratios of the OMI NO₂ products and the CMAQ simulation. In the network, NO₂ is measured by chemiluminescence and DOAS technique with an accuracy and precision of about 10 % (GDEMC and HKEPD, 2006).

3.2 CMAQ model simulation

Atmospheric chemistry was simulated with the CMAQ modelling system version 4.7.1 (Byun and Schere, 2006). Three model domains were defined using a Lambert conformal conic projection (Fig. 1). The coarse domain (D1) covers East Asia with a spatial resolution of 27 km × 27 km. The nested domains have grid resolutions of 9 km × 9 km (D2) and 3 km × 3 km (D3), respectively.

Meteorological fields were provided by the Weather Research and Forecasting (WRF) modelling system (Skamarock et al., 2008) driven by NCEP Final Analysis (FNL) data (NCEP, 1997). Horizontal advection was modelled with the mass-conserving YAMO scheme (Yamartino, 1993). The default vertical advection scheme was replaced by the new advection scheme implemented in CMAQ version 5.

The gas-phase chemistry was modelled with the Euler backward iterative solver optimized for the Carbon Bond-05 mechanism with chlorine (Yarwood et al., 2005). Aerosol chemistry was modelled with the fifth-generation CMAQ aerosol model (aero5) while the impact of clouds on deposition, mixing, photolysis, and aqueous chemistry was set by the Asymmetrical Convective Model (ACM) cloud processor (Pleim and Chang, 1992). Three-dimensional extinction coefficients were computed by the empirical IMPROVE formula (Malm et al., 1994).

The emission inventory used in this simulation was compiled by Du (2008). The inventory combines monthly anthropogenic emission from INTEX-B (Zhang et al., 2009) with biogenic emissions from GEIA (Global Emissions Inventory Activity, Guenther et al., 1995), and biomass burning and ship emissions from TRACE-P (Streets et al., 2003). The INTEX-B emissions were updated with regional emissions for Hong Kong provided by the Hong Kong Environmental Protection Department.

Model outputs used were hourly ground-level values and three-dimensional fields of NO₂ mixing ratios, aerosols and meteorological parameters averaged from 13:00 to 15:00 LT (OMI overpass time). In addition, at each ground station,

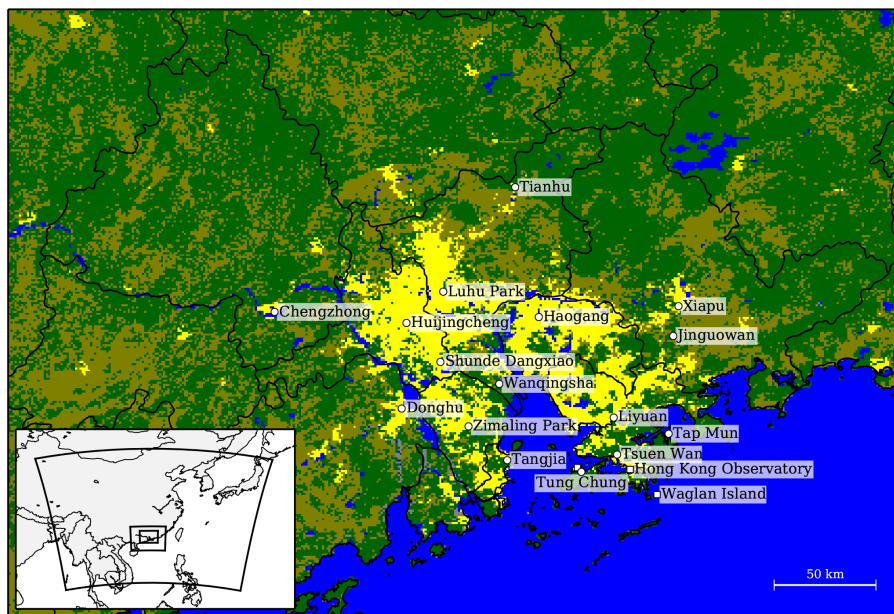


Figure 1. The CMAQ model domain (D3) with MODIS land categories in the Pearl River Delta (PRD) region grouped into the following categories: forest (dark green), crop lands (olive), bare land (grey), urban areas (yellow) and water (blue). The stations of the PRD Regional Air Quality Monitoring (RAQM) network and HKO automatic weather stations are marked by circles and squares, respectively. The inset shows the three CMAQ model domains which are D1, D2 and D3 from the largest to smallest.

time series of hourly meteorological parameters and NO₂ mixing ratios were extracted from the model output. The surface pressure in the model was converted to sea level pressure using the simulated temperature. The wind vector in the model was taken at the height of the measurement stations to account for the elevation of the stations above averaged surface height.

3.3 The Hong Kong OMI (HKOMI) NO₂ retrieval

For the HKOMI NO₂ retrieval, we recalculated tropospheric AMFs with Eq. (2) with new ancillary parameters. The new AMFs were used to compute the tropospheric VCDs with Eq. (1).

For the AMF calculation, a set of ancillary parameters was compiled for each OMI ground pixel. The parameters were taken mainly from the WRF/CMAQ simulation. Thus, the retrieval does not depend on any other CTM model which makes the model evaluation easier. The parameters are surface elevations, temperature, pressure and NO₂ profiles, as well as aerosol extinction coefficients. Further ancillary parameters are cloud height and CRF, which were taken from the OMI O₂–O₂ cloud product (Acarreta et al., 2004), and surface reflectance, which was taken from the MODIS MCD43C2 product (Wanner et al., 1997; Lucht et al., 2000).

All ancillary parameters were projected to a 0.01° × 0.01° (about 1 km × 1 km) longitude–latitude grid and then averaged to each OMI ground pixel. The grid points were weighted based on the instrument's spatial sensitivity within

the pixel boundaries (Dobber et al., 2006; Kurosu and Celarier, 2010), in contrast to other custom retrievals, where each grid point was given equal weight.

We recalculated temperature correction coefficients $\alpha_k(T_k)$, box AMFs m_k and partial VCDs V_k . The temperature correction coefficients $\alpha_k(T_k)$ were calculated by Eq. (3) from the WRF/CMAQ output. The box AMFs were computed with the SCIATRAN radiative transfer model. The partial VCDs were also calculated from the WRF/CMAQ output. As an example, two NO₂ profiles are shown in Fig. 2.

3.3.1 Surface reflectance

The surface reflectance was calculated from the MODIS MCD43C2 product. The product is available every 8 days compiled from 16 days of data. The spatial resolution is 0.05° × 0.05° (Wanner et al., 1997; Lucht et al., 2000). We calculated the black-sky albedo (BSA) from the polynomial representation of the bidirectional reflectance distribution function (BRDF) using solar zenith angle (SZA) and model parameters for MODIS Band 3 (Lucht et al., 2000). MODIS Band 3 has a wavelength range from 459 to 479 nm and a centre wavelength of 470 nm. This band is closest to the DOAS fitting window used in the NO₂ retrieval (405–465 nm). Systematic errors due to the wavelength inconsistency are expected to be small. The MODIS surface reflectance has been used in other custom OMI NO₂ products (Zhou et al., 2010; Russell et al., 2011; Lin et al., 2014; McLinden et al., 2014).

Since the BSA model parameters have missing values due to cloud contamination, Zhou et al. (2010) filled the data gaps by applying a series of spatial and temporal interpolations. They also reduced measurement noise by applying a smoothing filter. In this work, we combined their steps by using normalized convolution which is a useful algorithm for filling missing values (Knutsson and Westin, 1993). We used a three-dimensional, uniform kernel of size 5 to fill the gaps in the model parameters. The BSA was calculated from the model parameters for each OMI ground pixel.

3.3.2 Aerosols and clouds

Scattering by aerosols and clouds affects the AMF (Leitão et al., 2010). Aerosol scattering at or below an NO₂ layer increases the AMF, but scattering above the NO₂ layer will reduce the AMF. In addition, aerosol optics affects the retrieval of cloud properties.

Clouds are typically handled by the independent pixel approximation (Eq. 4), while aerosols are only treated implicitly in the standard and most custom NO₂ products. In OMNO2, cloud pressure and CRF are taken from the OMI O₂–O₂ cloud product which is sensitive to weakly absorbing aerosols (Boersma et al., 2011). Aerosols are also included in the OMI surface climatology which includes ground haze and persistent cloud features (Kleipool et al., 2008). Lin et al. (2014) included aerosols from a regional GEOS-Chem simulation. They used aerosol optical thickness (AOT) and assumed different profiles of aerosol extinction and absorption coefficients. They also recalculated the OMI cloud product to remove the aerosol component. Boersma et al. (2011) derived an empirical relationship between MODIS AOT and OMI CRF:

$$\text{CRF} = 0.21 \cdot \text{AOT}. \quad (5)$$

The formula was derived from observations over North America which were cloud free according to the MODIS AOT retrieval but had non-zero CRF according to the OMI retrieval.

In the HKOMI retrieval, we treated clouds as in OMNO2 using the independent pixel approximation. In SCIATRAN, clouds were implemented as an opaque surface at the height of the OMI cloud pressure product with a surface reflectance of 0.8 and the box AMF was calculated with Eq. (4).

Aerosol extinction coefficients can be calculated from the CMAQ output using the empirical IMPROVE formula (Malm et al., 1994). In the standard output, CMAQ calculates only ground-level extinction coefficients. In our custom retrieval, we implemented four different aerosol parametrizations:

Case 1: No explicit aerosol treatment, i.e. aerosols were only included implicitly through the OMI cloud product.

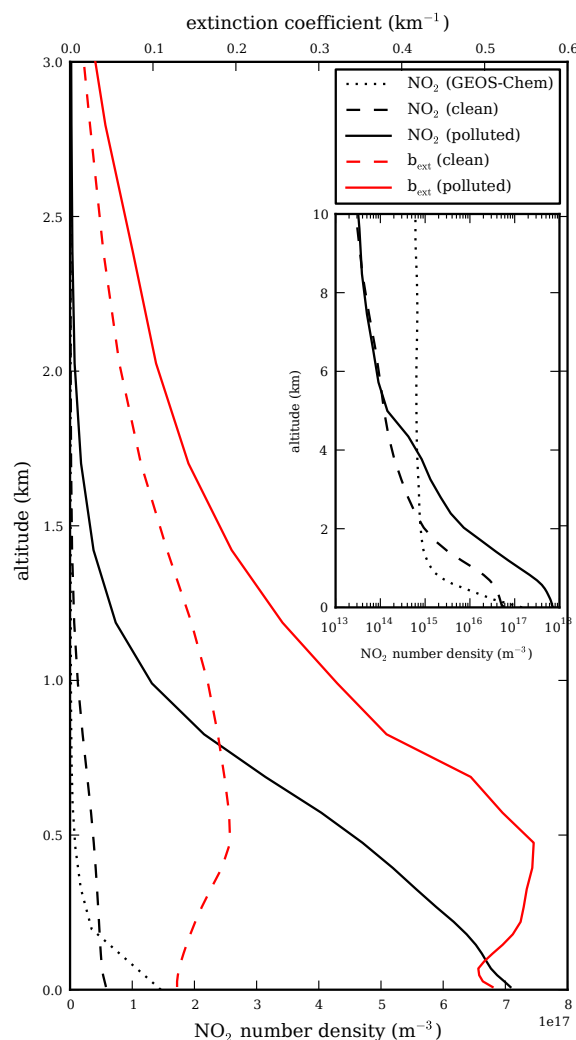


Figure 2. Averaged CMAQ NO₂ and b_{ext} profiles under “clean” and “polluted” conditions. A NO₂ profiles was categorized as polluted, if the ground number density was larger than $4.8 \times 10^{17} \text{ m}^{-3}$ (about 20 ppbv). A b_{ext} profile was categorized as polluted if the ground extinction coefficient was larger than 0.4. The clean b_{ext} profile has an AOT of 0.3 while the polluted has an AOT of 0.6. In addition, an annual GEOS-Chem NO₂ profile is shown for Hong Kong ($2^\circ \times 2.5^\circ$ spatial resolution).

Case 2: Aerosols were described by the LOWTRAN aerosol parametrization which requires only very limited information about season, aerosols type, visibility and relative humidity at four different layers: planet boundary layer (PBL) (0–2 km), troposphere (2–10 km), stratosphere and mesosphere. We set the season to autumn/winter and the aerosol type to urban. The PBL visibility was calculated from the CMAQ ground extinction coefficients β using the definition of the meteorological optical range (MOR) (World Meteorological Organization, 2008):

$$\text{MOR} = -\frac{\log 0.05}{\beta}. \quad (6)$$

The visibility in the free troposphere was set to 23 km. Furthermore, we assumed that no volcanic aerosols were in the stratosphere or mesosphere. The relative humidity in the PBL and the free troposphere were taken from WRF. Visibility and relative humidity were set to the nearest predefined value in the LOWTRAN parametrization.

Case 3: Vertical profiles of extinction coefficients β were computed from CMAQ output using the IMPROVE formula. The formula includes a constant Rayleigh extinction coefficient of 0.01 which is subtracted to obtain the aerosol extinction coefficient β_{ext} . Since the IMPROVE formula calculates β_{ext} at 550 nm, an Ångström exponent α for urban aerosols is used to calculate extinction coefficients at 435 nm (Hess et al., 1998). Furthermore, a single scattering albedo ω_0 of 0.82 (urban aerosol) in PBL (below 2 km) and 0.93 in free troposphere is used to calculate aerosol absorption coefficients β_{abs} . The phase function is modelled by Henyey–Greenstein parametrization with an asymmetry factor g of 0.689 (Hess et al., 1998). As examples, two β_{ext} profiles are shown in Fig. 2.

Case 4: Since these three parametrizations include aerosols implicitly through the cloud product, aerosol might be counted twice. Therefore, the CRF was corrected by Eq. (5) using the AOT in CMAQ. This was done mainly because recalculation of the OMI cloud product was outside the scope of our study. Since this formula was derived from cloud free observations over North America, generalization to cloudy pixels in other regions should be considered with great caution. Otherwise, aerosols were treated as in Case 3.

3.3.3 OMI NO₂ data sets for the PRD region

For our study, we created six OMI NO₂ data sets. The first data set uses the OMNO2 standard product (OMNO2-SP). For the second data set (OMNO2-SW), AMFs were recalculated using OMNO2 scattering weights and CMAQ NO₂ profiles. The remaining four data sets were created with our HKOMI retrieval for each aerosol case (HKOMI-1, -2, -3 and -4).

The number of OMI NO₂ VCDs, which allows for the study of the NO₂ distribution, is limited due to two factors. First, the ground pixel size at the end of the swath is very large and thus not suitable for studying the local spatial distribution. Therefore, only the inner 50 out of 60 rows were

used in this study. Second, the presence of clouds increases the retrieval uncertainty. Therefore, only ground pixels were used with CRFs smaller than 50 %. Since the CRF is also sensitive to aerosols, this filter criterion is likely to remove heavily polluted days as well. We further removed all orbits that did not have valid pixels in the urbanized area of the model domain (see Fig. 1).

3.4 Data processing

The OMI NO₂ data sets have been compiled in the instrument's frame of reference (level 2). For comparison and visualization, the data sets need to be projected to the model grid (level 3) using a suitable gridding algorithm. This is not a trivial task, because the algorithm should conserve the NO₂ VCDs within the OMI ground pixels which are overlapping in along-track direction. In this study, we use a new gridding algorithm which reconstructs the spatial distribution using a parabolic spline surface (Kuhlmann et al., 2014, the source code was downloaded at <https://github.com/gkuhl/omi>). Each orbit was projected to a 0.01° × 0.01° longitude–latitude grid. It would be more plausible to project all data to the CMAQ model grid, but the gridding algorithm code currently only supports longitude–latitude grids.

Since OMI has a lower spatial resolution than the CMAQ simulation, the satellite instrument cannot resolve small features in the simulated NO₂ distribution. In addition, OMI's spatial resolution depends on the current satellite orbit and thus changes from day to day. Therefore, we create a processed CMAQ data set (CMAQ processed) from the model output (CMAQ raw). The raw CMAQ VCDs were averaged for each OMI ground pixel in an orbit. Then, the CMAQ data were projected back onto a 0.01° × 0.01° longitude–latitude grid using the gridding algorithm (Kuhlmann et al., 2014). In this step, missing OMI pixels were also removed from the CMAQ data. Using this approach, the NO₂ horizontal distributions of OMI and CMAQ are directly comparable.

For each RAQM ground station location, time series of tropospheric NO₂ VCDs were computed from the six OMI data sets. To study the additive and proportional differences, the OMI VCDs were converted to mixing ratios using the CMAQ NO₂ profile shapes. Since many stations are located on top of buildings, the mixing ratios were calculated at the station height using nearest-neighbour interpolation. In addition, these time series were also computed for the processed CMAQ data set.

3.5 Evaluation study

For our evaluation study, we have six OMI data sets, two CMAQ data sets as well as time series of these data sets at each RAQM station and the measurements of the RAQM stations. Furthermore, we have simulated and measured meteorological parameters at three ground stations. In this paper, we performed the following analyses with these data sets:

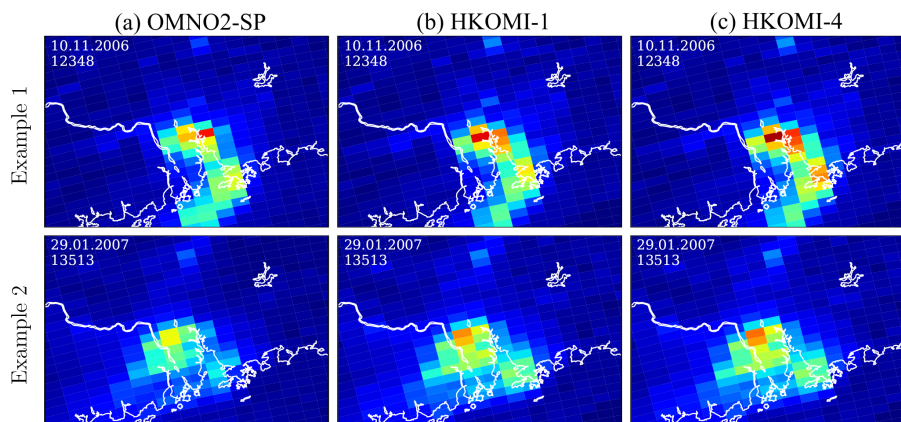


Figure 3. Two example orbits of OMI NO₂ distributions for OMNO2-SP, HKOMI-1 and HKOMI-4. The overall spatial distribution is similar but different in details. The HKOMI-1 and HKOMI-4 data sets have larger NO₂ column densities than OMNO2-SP.

- First, we compared the six OMI data sets (level 2) with each other to identify the impact of the ancillary parameters on the tropospheric NO₂ VCDs. We calculated normalized mean bias (NMB) and coefficient of variance (CV) for all valid OMI VCDs in the data sets. Since high NO₂ values are particularly important for air pollution studies, we also calculated NMB and CV for the 10 % highest VCDs based on HKOMI-3. Furthermore, we used a standard set of typical ancillary parameters (see Table 1) to obtain a better understanding of the differences between the data sets.
- Second, we compared the WRF/CMAQ time series with the ground network measurements using index of agreement (IOA), Pearson's correlation coefficient r , root mean square error (RMSE), mean bias (MB), CV and NMB (Table 2). In our analysis, we concentrated on two large urban areas which have the highest NO₂ mixing ratios. The first area consists of the Hong Kong area and the Shenzhen prefecture (HKSZ) and the second area consists of the Foshan and Guangzhou prefectures (FSGZ). HKSZ includes the RAQM stations Liyuan, Tap Mun, Tung Chung and Tsuen Wan. FSGZ includes the RAQM stations Huijingcheng, Luhu Park, Shunde Dangxiaoxia and Wanqingsha.
- Third, we validated the OMI data sets (level 3) with ground measurements. The validation with point measurements is challenging, because OMI measures the mean value within the ground pixels ($\geq 13 \text{ km} \times 24 \text{ km}$). Since NO₂ has a high spatial variability, area average and point measurement may not agree well – in particular in an urban area with complex NO₂ sources and sinks. In order to get an idea of the expected deviations, we compared the time series of the processed CMAQ data set, which have been averaged to the OMI ground pixels, with the raw CMAQ data set at the 16 stations. Furthermore, if the VCDs are con-

Table 1. Standard set of ancillary parameters used in the AMF sensitivity study.

Ancillary parameter	Value
solar zenith angle (SZA)	48°
viewing zenith angle (VZA)	0° (nadir)
relative azimuth angle (RAA)	180°
terrain height	0.0 km
surface reflectance	0.05
aerosol/cloud radiance fraction	0.00
temperature and pressure profiles	WRF average
NO ₂ and aerosol profiles	CMAQ averages (see Fig. 2)

verted to ground values with the CMAQ profile shapes, the validation depends on the modelled NO₂ profiles which can have large uncertainties. This was studied by comparing different NO₂ profiles.

- Finally, OMI and CMAQ data sets were compared to evaluate the regional CTM with the OMI NO₂ products. We studied the spatial distribution of tropospheric NO₂ VCDs in all data sets. Furthermore, time series were analysed for the two urban areas (HKSZ and FSGZ).

4 Results

4.1 OMNO2 and HKOMI NO₂ retrieval

In this section, we look at the differences between the six OMI NO₂ data sets. After cloud filtering, 56 days with satellite observations were available covering about 50 % of all days in our study period. Figure 3 shows two examples of tropospheric NO₂ distributions (level 2). The spatial distributions are similar between the OMNO2-SP, HKOMI-1 and HKOMI-4 data sets with some differences between individual pixels. On the other hand, NO₂ magnitudes are quite

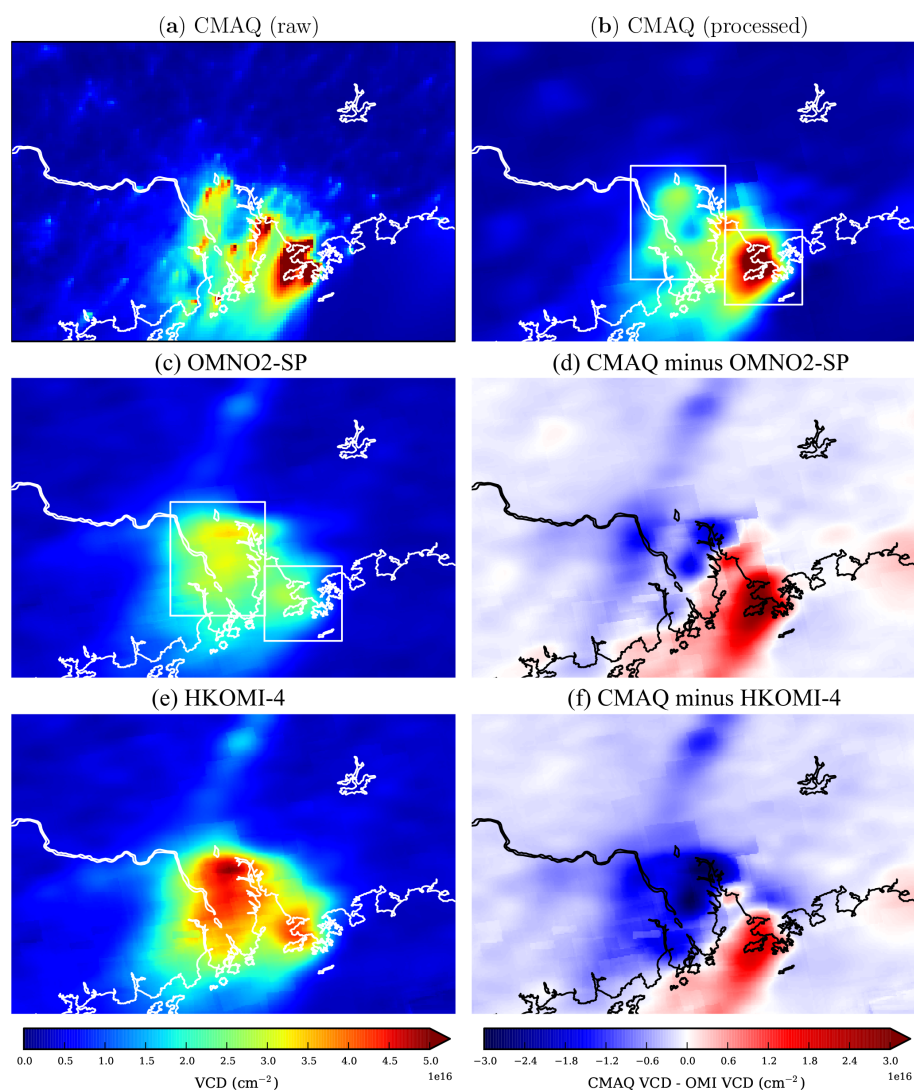


Figure 4. Four-month mean distribution of (a) raw and (b) processed CMAQ NO₂ VCDs, (c) OMNO2-SP VCDs and (d) the difference to CMAQ, (e) HKOMI-4 VCDs and (f) the difference to CMAQ.

different between the data sets. Figure 4c and e shows the averaged NO₂ distribution for whole study period. The averaged OMNO2-SP data set has a mean value of $0.45 \times 10^{16} \text{ cm}^{-2}$ and a maximum value of $3.46 \times 10^{16} \text{ cm}^{-2}$, while the HKOMI-4 data set has a mean value of $0.59 \times 10^{16} \text{ cm}^{-2}$ and a maximum value of $5.04 \times 10^{16} \text{ cm}^{-2}$. The HKOMI-2 data set has the largest mean ($0.70 \times 10^{16} \text{ cm}^{-2}$) and maximum value ($5.50 \times 10^{16} \text{ cm}^{-2}$). Table 3 shows NMB and CV between the data sets for all and the 10 % highest VCDs. Since we compared data sets which only differ by one ancillary parameter, the comparison shows the influence of each parameter. CMAQ NO₂ profiles and MODIS surface reflectance increase VCDs by about 10 %. When aerosols were included, VCDs increased between 6 and 24 % depending on the parametrization. If a priori profiles and aerosols are replaced, the increase of the VCDs is larger for the 10 % largest

VCDs. The HKOMI-4 data set is 31.0 ± 34.0 % larger than the OMNO2-SP data set.

In order to assess the differences better, we looked at the influence of the ancillary parameters using typical values. Figure 2 shows a “clean” and “polluted” NO₂ profiles from CMAQ and a profile from GEOS-Chem ($2^\circ \times 2.5^\circ$ spatial resolution) for Hong Kong (Chan et al., 2012). The polluted CMAQ NO₂ profile has an AMF of 0.82 and is about 3 % smaller than the clean profile (AMF=0.84). The GEOS-Chem profile has an AMF of 1.19 which is 41 % larger than the clean and 45 % larger than the polluted CMAQ profile. To determine the variance of the AMFs, we calculated the AMFs using all CMAQ profiles. In this sample, the largest AMF is 1.64, due to an elevated NO₂ layer in the upper troposphere, and the smallest AMF is 0.75, due to a heavily pol-

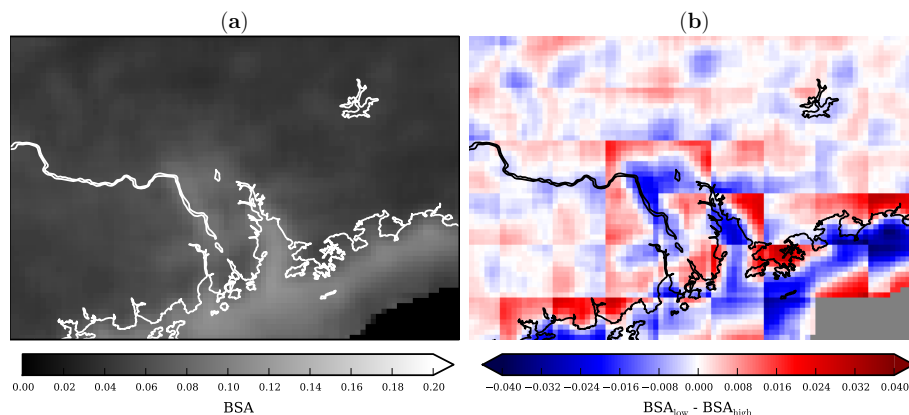


Figure 5. (a) The averaged MODIS black-sky albedo (BSA) and (b) the differences between low- and high-resolution BSAs.

Table 2. Error measures used to compare observations and model. $\langle \cdot \rangle$ is the mean value and σ_x and σ_y are standard deviations.

Index of agreement (Willmott, 1981)	$\text{IOA} = 1 - \frac{\sum_{i=1}^N (x_i - y_i)^2}{\sum_{i=1}^N (x_i - \langle y \rangle + y_i - \langle x \rangle)^2}$
Pearson's correlation coefficient	$r = \frac{1}{n-1} \sum_{i=1}^N \left(\frac{y_i - \langle y \rangle}{\sigma_y} \right) \left(\frac{x_i - \langle x \rangle}{\sigma_x} \right)$
Root mean square error	$\text{RMSE} = \sqrt{\frac{1}{N} \sum_{i=1}^N (x_i - y_i)^2}$
Coefficient of variation of the RMSE	$\text{CV} = \frac{\text{RMSE}}{\langle y \rangle}$
Mean bias	$\text{MB} = \frac{1}{N} \sum_{i=1}^N (x_i - y_i)$
Normalized mean bias	$\text{NMB} = \frac{\text{MB}}{\langle y \rangle}$

luted ground layer. The sample mean is 0.89 with a standard deviation of 0.08 (about 9 % of the mean).

Figure 5a shows the MODIS surface reflectance for a SZA of 48°. On average, the MODIS reflectance is 0.01 ± 0.02 smaller than the OMI reflectance (not shown). The BSA is smaller than the OMI climatology over land but larger over water. In order to identify the improvement due to the higher spatial resolution, we averaged the BSA to the lower spatial resolution used in the standard product ($0.5^\circ \times 0.5^\circ$) and subtracted the high-resolution BSA used in our custom product (Fig. 5b). The distribution shows areas with large differences (up to ± 0.03) along the urbanized and likely polluted coastline which would result in large biases (up to $\pm 20\%$ for a reflectance of 0.05) in the AMF calculation.

Table 4 shows the AMFs for the different aerosol cases using the clean and polluted profiles (Fig. 2). Cases 3 and 4 are equal because the CRF is zero in the test parameters. The Case 2 AMFs are 19 % smaller than Case 1 for the clean and 57 % smaller for the polluted aerosol profile. If Case 3 is compared to Case 1, the AMF is reduced between 8 and 14 % (Table 4). In Case 2, the CMAQ ground-level extinction coefficient is used as constant coefficient in the PBL (0–2 km). The β_{ext} profiles show that using the ground-level extinction coefficient is reasonable for clean but not for polluted profiles (Fig. 2), because β_{ext} is overestimated above 1 km which re-

duces the AMF because these artificial aerosols are shading the NO₂ layer below.

4.2 WRF/CMAQ validation with ground measurements

The results of the WRF validation with the automatic weather stations are tabulated in the Supplement (Table S1). The IOA between observed and simulated sea level pressure is high (IOA = 0.99) at all stations. Simulated temperature and humidity also have high agreement with the observations. The model slightly underestimates temperature and humidity at HKO and HKIA, while it slightly overestimates temperature and humidity at WGL. The agreement between modelled and observed wind speed is highest at WGL (IOA = 0.84), which is located on a remote island, and lower at HKIA (IOA = 0.68) and HKO (IOA = 0.57), which are located in complex terrain.

The results of the CMAQ validation with the RAQM network are tabulated in the Supplement (Table S2). For the 16 stations, the IOAs have an average of 0.52 and vary between 0.29 (Tap Mun) and 0.75 (Tsuen Wan). The average ground mixing ratios are shown in Fig. 6. In the measurements, two major plumes can be identified in HKSZ and in FSGZ. In the simulations, the FSGZ plume is much less pro-

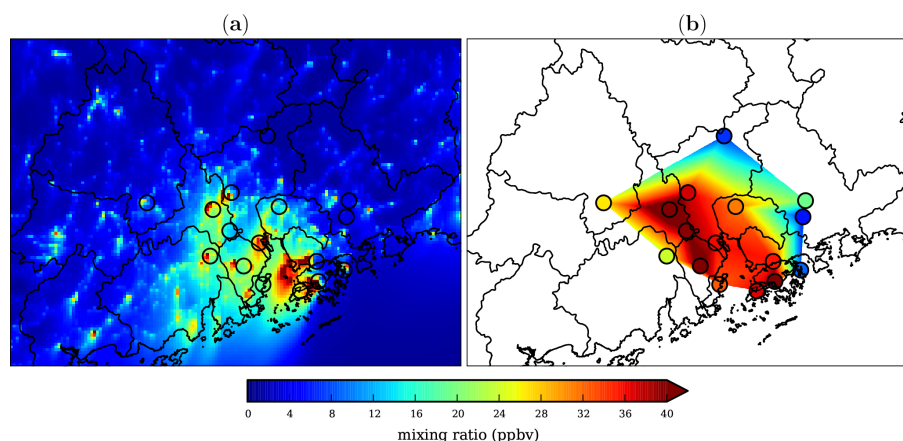


Figure 6. Ground-level NO₂ mixing ratios averaged for October 2006 to January 2007: (a) CMAQ simulation and (b) RAQM network measurements. The values between stations have been estimated by linear interpolation.

Table 3. Difference between OMI NO₂ data sets (level 2) due to different ancillary parameters.

Data set	Compared to	All VCDs		10 % highest VCDs		Different ancillary parameter
		NMB ^a	CV ^a	NMB ^a	CV ^a	
OMNO2-SW	OMNO2-SP	+11.4	13.4	+13.2	14.3	a-priori NO ₂ profile
HKOMI-1	OMNO2-SW	+11.0	20.9	+11.3	19.4	surface reflectance ^b
HKOMI-2	HKOMI-1	+24.1	24.1	+29.9	29.9	aerosols (case 2)
HKOMI-3	HKOMI-1	+8.1	9.3	+12.2	12.5	aerosols (case 3)
HKOMI-4	HKOMI-1	+6.0	8.4	+9.8	11.2	aerosols (case 4)
HKOMI-4	OMNO2-SP	+31.0	34.0	+38.3	40.0	all

^a In percent, ^b difference are also due to other difference between the OMNO2 and HKOMI retrieval, e.g. temperature profiles.

Table 4. The AMFs for different aerosol treatment cases for the standard parameters and profiles (Fig. 2).

Aerosol parametrization	Case 1	Case 2		Cases 3 & 4	
		Clean	Polluted	Clean	Polluted
NO ₂ clean	0.84	0.68	0.39	0.77	0.74
NO ₂ polluted	0.82	0.65	0.35	0.74	0.70

nounced than in the measurements, which can be seen in the averaged mean bias, which is -17.6 ppbv (-44%) for the four stations in FSGZ. The averaged mean bias is -0.0 ppbv (-5%) for the stations in HKSZ.

4.3 OMI validation with ground measurements

The comparison between OMI data sets and RAQM stations is impacted by the area averaging effect. The expected error measures were computed by comparing raw and processed CMAQ data sets (Table 5). The expected correlation coefficient is very low ($r = 0.25$) at HKSZ and good ($r = 0.67$) at FSGZ. The expected NMBs are -16 and -12% at HKSZ and FSGZ, respectively.

Table 5. The difference between raw and processed CMAQ data due to the spatial resolution of the satellite instrument (area averaging error).

	IOA	r	MB ^a	NMB ^b	RMSE ^a	CV ^b
HK & SZ	0.54	+0.25	-4.6	-16	17.6	59
FS & GZ	0.77	+0.67	-1.6	-12	7.0	52
All stations	0.69	+0.56	-2.9	-17	9.3	54

^a In ppbv; ^b in percent.

To study additive and proportional differences between satellite and ground observations, the OMI VCDs were converted to ground-level mixing ratios using the CMAQ NO₂ profiles. The mean conversion factor ($V_0/(\Delta z_0 V_{\text{trop}})$) is $(1.47 \pm 0.47) \times 10^{-3} \text{ m}^{-1}$. The conversion factor of the clean profiles is about 3% smaller than of the polluted profile. The GEOS-Chem profile has a conversion factor of $3.92 \times 10^{-3} \text{ m}^{-1}$ which is more than twice the factor computed for the CMAQ profiles. As a result, ground-level mixing ratios would be much larger if the GEOS-Chem profile were to be used for the conversion. The ground mixing ratio map is shown in Fig. 7 for 29 January 2007 using two differ-

Table 6. OMI evaluation with the RAQM network.

Data set	IOA	<i>r</i>	MB ^a	NMB ^b	RMSE ^a	CV ^b
Hong Kong and Shenzhen						
OMNO2-SP	0.31	+0.19	-17.5	-54	23.4	73
OMNO2-SW	0.34	+0.10	-13.0	-40	21.6	67
HKOMI-1	0.43	+0.24	-8.8	-27	20.6	64
HKOMI-2	0.52	+0.25	-0.7	-2	21.9	68
HKOMI-3	0.44	+0.20	-6.3	-20	20.7	64
HKOMI-4	0.45	+0.24	-7.4	-23	20.1	62
Foshan and Guangzhou						
OMNO2-SP	0.43	+0.47	-14.3	-48	20.9	71
OMNO2-SW	0.46	+0.45	-13.1	-44	20.2	68
HKOMI-1	0.48	+0.41	-10.7	-36	19.2	65
HKOMI-2	0.58	+0.42	-5.2	-18	17.8	60
HKOMI-3	0.50	+0.40	-8.5	-29	18.4	62
HKOMI-4	0.53	+0.46	-8.7	-29	17.9	60
All stations						
OMNO2-SP	0.40	+0.35	-10.7	-41	18.7	73
OMNO2-SW	0.42	+0.29	-8.2	-32	18.2	71
HKOMI-1	0.46	+0.31	-5.6	-22	18.2	71
HKOMI-2	0.53	+0.32	+0.7	+3	19.6	76
HKOMI-3	0.47	+0.29	-3.3	-13	18.6	72
HKOMI-4	0.50	+0.35	-3.9	-15	17.6	68

^a In ppbv; ^b in percent.

ent gridding algorithms. The algorithm which uses parabolic splines does not show discontinuities at the pixel boundaries and has slightly larger VCDs (Fig. 7b). The NO₂ variability below the OMI pixel size is caused by the variability of the CMAQ profile shapes showing the large impact of the model.

Table 6 shows the statistical measures for the comparison between the six OMI data sets and the ground network. Statistical measures for all 16 stations are tabulated in the Supplement (Table S3) showing that the measures vary considerably between the stations. The averaged correlation coefficients are similar with no large difference between the data sets. At FSGZ, *r* is smaller than the expected value, while at HKSZ, *r* is close to the quite small expected value (Table 5). At HKSZ, the HKOMI data sets have a slightly larger correlation coefficient than the two OMNO2 data sets. The data sets largely differ in the mean biases. The bias is largest for the OMNO2-SP and smallest for HKOMI-2 data set. The bias is closest to the expected bias for the HKOMI-3 and -4 data sets. OMNO2-SP and -SW underestimate NO₂ mixing ratios while HKOMI-2 overestimates the mixing ratios.

4.4 CMAQ evaluation with OMI NO₂ data sets

The modelled NO₂ VCDs were evaluated with the OMI NO₂ data sets. Figure 4a and b show the difference between the raw and the processed CMAQ data set. In the processed data set, the spatial NO₂ distribution is smoothed below the size of the OMI ground pixel. Therefore, CMAQ (processed) and

OMI data sets can be directly compared without artefacts due to different spatial resolutions.

Table 7 shows the mean values at RAQM stations for CMAQ and OMI data sets. If the model performance were good, CMAQ (raw) should agree well with RAQM and CMAQ (processed) should agree well with the OMI data sets. The former were already compared in Sect. 4.2 showing a small bias at HKSZ and a large bias at FSGZ. The results are similar between CMAQ (processed) and HKOMI-4 having similar mean values at HKSZ but a large bias at FSGZ.

The NO₂ distribution of the HKOMI-4 data set has one plume at FSGZ and a second plume at HKSZ (Fig. 4e). In addition, the OMI data set has increased NO₂ near the northern edge of the model domain and at the Pearl River in the western part of the domain. The differences between CMAQ and OMI are shown in Fig. 4d and f. The modelled VCDs are underestimated at FSGZ and overestimated at HKSZ. The form of the plume at HKSZ differs between OMI and CMAQ. In CMAQ, the plume has an elliptic shape with a strong, south-western outflow. In the OMI data sets, this feature is less distinct.

The results of the time series analysis are shown in Table 8. The correlation coefficient is smaller at FSGZ than at HKSZ. At FSGZ, CMAQ VCDs are much smaller than the HKOMI-4 VCDs (NMB = -40 %). This bias is close to the bias between CMAQ and RAQM (-44 %). On the other hand, CMAQ VCDs are larger than the OMI VCDs (+15 %) at HKSZ.

5 Discussions

5.1 Influence of ancillary parameters

Ancillary parameters have a large influence on the retrieval of tropospheric NO₂ VCDs. In our study, their influence on the AMFs is similar to findings for other retrieval algorithms (e.g. Zhou et al., 2010; Russell et al., 2011; Lin et al., 2014). A direct comparison is difficult because the influence depends on regional factors. For example, we found a very strong gradient in surface reflectance along the coastline in the PRD region which has a large influence on tropospheric NO₂ VCDs. In other regions, the variability of surface reflectance can be smaller, influencing VCDs less.

In our retrieval, CMAQ NO₂ profiles were used which have a quite different shape compared to GEOS-Chem profiles (Fig. 2). Since NO₂ profile measurements were not available for the PRD region, the modelled profiles were not validated in this study. As a consequence, AMF uncertainties due to NO₂ profiles are difficult to quantify. However, it can be argued that the regional CMAQ CTM provides more accurate vertical distributions than the global model, because of higher spatial resolution and more detailed PBL, vertical advection and diffusion schemes. On the other hand, different schemes exist for regional CTM and these might result in dif-

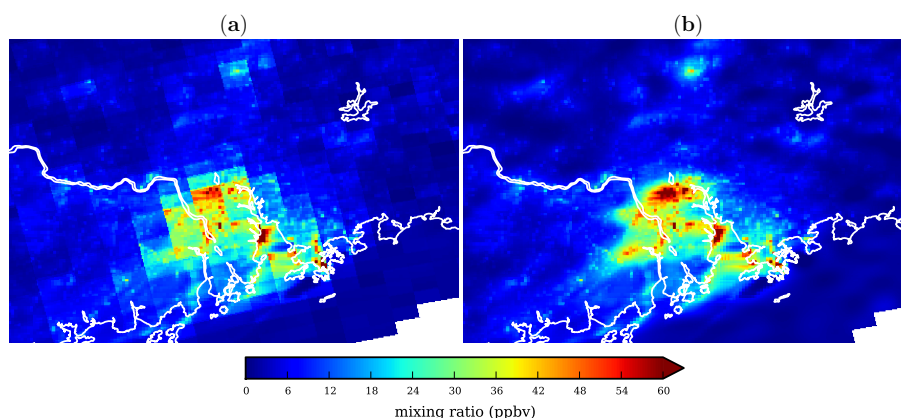


Figure 7. OMI ground mixing ratios from orbit number 13513 on 29 January 2007 comparing a (a) “standard” and (b) newly developed gridding algorithm (Kuhlmann et al., 2014). The discontinuous map created by the standard algorithm is difficult to interpret while the new algorithm makes an analysis of the spatial distribution easier.

Table 7. NO₂ mean values (in ppbv) at the RAQM network stations for CMAQ, network and OMI data sets.

Stations	CMAQ (raw)	CMAQ (processed)	RAQM	OMNO2-SP	OMNO2-SW	HKOMI-1	HKOMI-2	HKOMI-3	HKOMI-4
HKSZ	29.3	25.1	32.9	14.9	19.3	23.4	31.6	25.9	24.8
FSGZ	13.7	12.0	29.5	15.2	16.5	18.9	24.4	21.0	20.8
All stations	18.6	15.7	25.3	14.5	16.9	19.4	25.6	21.6	21.0

Table 8. Evaluation of the time series of CMAQ with OMI NO₂ VCDs in the two areas marked in Fig. 4.

Data set	OMI Mean ^a	IOA	<i>r</i>	MB ^a	NMB ^b
Foshan and Guangzhou (area)					
OMNO2-SP	2.5	0.57	0.37	−0.5	−18
OMNO2-SW	2.8	0.57	0.39	−0.7	−26
HKOMI-1	3.1	0.55	0.35	−1.1	−35
HKOMI-2	4.0	0.51	0.35	−2.0	−49
HKOMI-3	3.5	0.56	0.43	−1.5	−42
HKOMI-4	3.4	0.54	0.37	−1.3	−40
Hong Kong and Shenzhen (area)					
OMNO2-SP	1.7	0.51	0.57	+1.2	+73
OMNO2-SW	2.2	0.64	0.59	+0.8	+37
HKOMI-1	2.4	0.65	0.51	+0.6	+24
HKOMI-2	3.2	0.66	0.45	−0.2	−8
HKOMI-3	2.7	0.75	0.58	+0.2	+9
HKOMI-4	2.6	0.71	0.56	+0.4	+15

^a In 10¹⁶ molecules cm^{−2}; ^b in percent.

ferent NO₂ profiles (e.g. Tang et al., 2011; Xie et al., 2012). Therefore, the validation of model profiles is important to better estimate the uncertainties for model evaluation with satellite observations. In this study, the CMAQ NO₂ profiles have a strong vertical gradient in the PBL which is expected

for strong emissions near the surface. The qualitative shape of the profile agrees reasonably with measured and simulated urban NO₂ profiles used in other studies (e.g. Schaub et al., 2006; Leitão et al., 2010).

Aerosols have a large influence on the retrieved VCDs. In the HKOMI retrieval, good agreement with ground measurements was found when calculating β_{ext} profiles and reducing CRF based on AOT in the model (Case 4). β_{ext} calculation and CRF reduction are based on empirical formulas which have the advantage that they are easy to use. However, the IMPROVE formula was not developed to calculate β_{ext} profiles in an urban environment which can result in uncertainties. An improved IMPROVE formula is available which we consider to use in an updated version of our retrieval (Zhang et al., 2013). Alternatively, optical properties can be calculated by Mie theory. The aerosol information was not validated with ground- or satellite observations which is also planned to be implemented in an updated version. Nevertheless, the shape of the aerosol profiles is in reasonable agreement with LIDAR measurements in Hong Kong (He et al., 2008). The empirical cloud correction formula is also very simple and, as mentioned before, should be improved in future. In conclusion, the impact of aerosols and clouds is an important factor for unbiased retrieval of tropospheric NO₂ VCDs. Unfortunately, a complete treatment of aerosols and clouds is complex and often not feasible for air quality model evaluations.

The OMI surface reflectance climatology provides scene reflectance, which includes both the surface and the presence of boundary layer haze or aerosols, and minimum reflectance, which is the lowest retrieved reflectance value (Kleipool et al., 2008). Since scattering by weakly absorbing aerosols at or below an NO₂ layer increases AMFs, it has been argued that scene reflectance should be used for satellite-based trace gas retrieval, if no information about aerosols is available (Herman et al., 2001). Furthermore, the minimum reflectance can be underestimated due to ground- and cloud shading or darkening by rainfall. For these reasons, scene reflectance is used in the standard product (Boersma et al., 2011). However, if an NO₂ layer is mixed with highly absorbing aerosols, the AMF is not increased but decreased (Leitão et al., 2010). In the HKOMI retrieval, we have highly absorbing ($\omega_0 = 0.82$) urban aerosols and thus AMFs are decreased in the presence of aerosols. As a result, if scene reflectance is used for an urban area, NO₂ VCDs can be strongly underestimated.

Since ancillary parameters have a large influence on the VCDs, it is important to quantify their uncertainties. This is often difficult because vertical profiles are required which are rarely available. In the view of model evaluation, it is also helpful if surface reflectance, aerosols and clouds are provided independently, because it makes it possible to use modelled aerosol profiles in the retrieval.

5.2 WRF/CMAQ validation with ground measurements

The evaluation of the meteorological fields shows good agreement between model and observations within the expected limitations due to the model resolution. The agreement is lower for the wind fields due to the impact of local topography. Unfortunately, no meteorological data for the whole PRD region were available. However, due to the high agreement in Hong Kong, similar model performance is expected in the complete model domain. The meteorological fields are sufficient to provide input for the chemistry transport simulations.

The agreement between CMAQ and RAQM network is low for various reasons. First, the point measurement may not be representative for the grid cell because of the influence of local sources and sinks as well as local topography and station height. Second, the differences can arise from the model due to inaccurate wind fields, limited parametrization of the chemistry and, in particular, insufficient knowledge about strength and distribution of emissions. The model performance is similar to the result by Wu et al. (2012) who also evaluated CMAQ in the PRD region using a similar emission inventory.

The low model performance can largely be explained by problems with the emission inventory. For example, the low IOA at Tap Mun is the result of several peaks in the simulated time series which were not found in the measure-

ments. The peaks were traced back to NO_x emissions at the Dapend Peninsula about 15 km east of Tap Mun. As a second example, the large bias between model and observations at FSGZ is mainly due to underestimated NO_x emissions in this region. The smaller bias in HKSZ is thought to be the result of the updated emission inventory with more accurate information for Hong Kong. It should be noted that updated emission inventories exist for the PRD region which would improve the model performance. However, our objective was not an accurate CMAQ simulation but a model evaluation with satellite observations. For this application, the used emissions inventory was found to be very useful.

5.3 OMI validation with ground measurements

The OMI data sets were validated with the RAQM network. The HKOMI retrieval does not change the correlation coefficient considerably compared to the standard products, while they are generally increased in other custom products (e.g. Lin et al., 2014; Russell et al., 2011; Zhou et al., 2009). The non-existent improvement has two main reasons. First, the low CTM performance can result in random errors impacting ancillary parameters and conversion factors. Second, ground-based point values were compared with satellite-based area-averaged values in an urban area with high NO₂ variability. The expected discrepancies between ground- and satellite-based observations were estimated using CMAQ NO₂ distributions showing that the expected correlation coefficients were quite small reducing possible improvements.

On the other hand, the HKOMI retrieval significantly reduces the bias between ground- and satellite-based observations. However, systematic errors in the parameters can still cause biases in the data sets. For example, if the model underestimates emissions, NO₂ and β_{ext} profiles might be described better by the polluted than the clean profiles (Fig. 2). As a consequence, the AMFs are reduced by about 9 % and the conversion factor by about 3 %. Therefore, NO₂ mixing ratios would be underestimated by 10 to 20 % which can partly explain the large NMB at FSGZ.

Besides these limitations, the HKOMI retrieval shows the possibility for unbiased NO₂ observations using satellite instruments. Of the four aerosol cases, HKOMI-4 performed best with the highest correlation coefficient and the smallest NMB. Furthermore, HKOMI-4 makes the most reasonable assumptions, although very simple, about aerosols and clouds. Correlation coefficients and mean biases are expected to improve further with better CTM performance.

5.4 Model evaluation and further applications

The model evaluation with OMI NO₂ data sets demonstrated that it is possible to study the spatial distribution and the magnitude of NO₂ VCDs with satellite observations. The biases between CMAQ and HKOMI-4 are consistent with the bias found between CMAQ and RAQM giving us further confi-

dence that the HKOMI-4 retrieval results in smaller systematic errors. Small systematic errors are important for various applications.

In air quality studies, satellite observations can be used to obtain ground-level NO₂ concentrations (e.g. Lamsal et al., 2008). To estimate the impact on human health, these concentrations need to be unbiased because otherwise the air quality is misinterpreted. This is particular important in polluted areas, where NO₂ VCDs can be biased due to the presence of absorbing aerosols. Our retrieval reduces this bias in particular for high NO₂ values making it more suitable for satellite-based air quality studies in urban areas.

Satellite observations can also be used for estimating NO_x emissions by inverse modelling (e.g. Mijling et al., 2013; Gu et al., 2014). Systematic errors in the satellite observations can cause biases in the derived emission inventory (e.g. Connor et al., 2008). To reduce this problem, Gu et al. (2014) derived emissions by iteratively updating the a priori NO₂ profiles used in satellite retrieval. Their method reduces differences between emissions derived from GOME-2 and OMI. However, we and other showed that NO₂ VCDs can still be biased due to surface reflectance, aerosols and clouds. Therefore, we think it is necessary to also update AKs in the inverse method and to characterize the spatiotemporal distribution of systematic errors in the satellite retrieval. The latter was done, for example, for the retrieval of carbon dioxide (Buchwitz et al., 2013).

6 Conclusions

In this paper, we evaluated biases in a regional CTM (3 km × 3 km spatial resolution) with ground- and satellite-based NO₂ observations. Atmospheric chemistry was simulated with the CMAQ modelling system and ground measurements were taken from the RAQM network. Six OMI NO₂ data sets were compiled with NASA's standard retrieval (OMNO2-SP and -SW) and our custom retrieval (HKOMI-1 to -4). In the OMNO2-SW data set, a priori NO₂ profiles were replaced by CMAQ profiles using the scattering weights provided with the product. In the HKOMI retrieval, we recalculated tropospheric AMFs using updated ancillary parameters of a priori NO₂ profiles, surface reflectance and aerosol profiles. The HKOMI data sets differ in how aerosols were implemented in the retrieval.

The updated ancillary parameters increased tropospheric NO₂ VCDs by (+11.4 ± 13.4) % (a priori NO₂ profiles), (+11.0 ± 20.9) % (surface reflectance) and (+6.0 ± 8.4) % (HKOMI-4 aerosol parametrization). As a result, the normalized mean bias (NMB) between satellite and ground observations was significantly reduced from -41 % (OMNO2-SP) to -15 % (HKOMI-4). The remaining biases can be explained by CMAQ model biases, resulting from the smoothed model terrain, the finite layer structure, emission errors etc., and the area averaging effect, i.e. OMI's inability to resolve complex

structures due to its ground pixel size. If only the a priori profiles are replaced in the standard product, which is recommended for model evaluation studies, the NMB is only reduced to -32 %.

Since ancillary parameters have such a strong influence on the VCDs, the parameters need to be well known to obtain accurate NO₂ VCDs and to estimate their uncertainties. In our study, NO₂ and aerosol profile observations were not available making it difficult to estimate uncertainties and their impact on the AMF calculations. Future studies are necessary to address these limitations.

The CTM performance was low mainly due to the underestimated emissions causing a large bias in Foshan and Guangzhou (NMB = -40 %). However, the results the model evaluation with the RAQM network and the OMI NO₂ data sets were consistent. We also estimated that our custom retrieval could still underestimate VCDs by 10 to 20 % in some areas which is important to remember when validating a model with satellite products. In general, we expect an improved HKOMI product with better CTM simulation.

To conclude, our study demonstrates that the data sets created by the HKOMI retrieval are suitable for the evaluation of the spatial distributions and the magnitudes of NO₂ concentrations in the model. We showed that a retrieval, which updates not only a priori NO₂ profiles, reduces the biases in urban areas. Since the ancillary parameters have a large impact on the retrieval, tools need to be developed to keep track of their influence on the final product. Such tools could improve CTM evaluations with satellite observations.

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