Characterization of OMI Tropospheric NO$_2$ Measurements in East Asia Based on a Robust Validation Comparison

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Abstract

In the period from June 2006 to December 2008, we measured the tropospheric nitrogen dioxide (NO$_2$) column by ground-based Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) at an urban site in China (Tai’an) and three sites in Japan, covering urban (Yokosuka), suburban (Tsukuba), and remote areas (Hedo). This robust dataset is used to characterize Ozone Monitoring Instrument (OMI) tropospheric NO$_2$ column data (the standard product, version 3). Correlations between MAX-DOAS and OMI data, both of which show very low NO$_2$ at Hedo and moderate/high levels at the other sites, have correlation coefficients ($R^2$) as high as 0.64, indicating that relative changes in OMI NO$_2$ data are reliable. However, OMI data have a negative bias of 31% on average. Assuming that these results are valid for OMI data taken over China, we find an increasing trend in tropospheric column NO$_2$ at about 5% per year on average in the industrial areas of China ($30^°-40^°$N and $110^°-123^°$E) over 2005–2008, but its spatial distribution is highly inhomogeneous.

1. Introduction

In recent decades, anthropogenic emissions of important gaseous and particulate species, including nitrogen oxides (NOx = NO + NO$_2$), have been increasing in Asian countries because of an increase in energy consumption due to rapid industrial development; hence, a future increase until 2020 has been projected (Ohara et al. 2007). In particular, the People’s Republic of China has augmented coal combustion in power plants and industrial sectors, resulting in a marked increase of anthropogenic NOx emissions. The resulting increase in tropospheric NO$_2$ vertical column density (VCD) over China has indeed been visible from satellite observations (GOME and SCIAMACHY) (e.g., Richter et al. 2005; Irie et al. 2005; van der A et al. 2006), and a linkage between changes found in the satellite-derived tropospheric column and bottom-up emission estimates has been confirmed by top-down estimation, which has been carried out by inverse modeling using a chemistry-transport model and satellite measurements by GOME and SCIAMACHY up to 2006 (Stavrakou et al. 2008).

The Ozone Monitoring Instrument (OMI) (Levelt et al. 2006) is a satellite-borne instrument measuring the tropospheric NO$_2$ VCD. Since OMI was launched onboard the Aura satellite on July 15, 2004, it has provided a unique dataset of tropospheric NO$_2$ with improved spatial ($13 \times 24$ km$^2$ at nadir) and temporal resolutions (daily global coverage) compared to those of GOME and SCIAMACHY. While OMI data are thus useful for a better understanding of NO$_2$ distributions at finer temporal and spatial resolutions, it is desirable to characterize the quality of OMI NO$_2$ data, especially in Asia, where very limited validation comparisons have been reported.

At Tai’an, located in the North China Plain, tropospheric NO$_2$ VCD data from OMI (the standard product, version 3) have been shown to agree with those derived from our ground-based Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) measurements to within the uncertainty of the OMI data (Irie et al. 2008a). However, because the comparisons have been made at only a single site and for only one month, June 2006, more robust comparisons are indispensable in establishing OMI data quality. In the present study, such robust validation comparisons are first attempted by including a new long-term data record from our MAX-DOAS measurements at three sites in Japan, covering urban (Yokosuka), suburban (Tsukuba), and remote areas (Hedo). On the basis of these comparisons, an estimate of the recent trends of tropospheric NO$_2$ VCD from 2005 to 2008 is presented.

2. MAX-DOAS

Here we briefly describe MAX-DOAS measurements performed at the aforementioned four sites. Information on these sites is summarized in Table 1, and the locations are plotted on the map of OMI tropospheric NO$_2$ data in Fig. 1. As can be seen from Fig. 1, the MAX-DOAS measurements were made at various levels of NO$_2$, covering urban, suburban, and remote areas. It is expected that temporal and spatial inhomogeneities of tropospheric NO$_2$ are higher around urban than around suburban and remote areas. Another feature of this unique set of measurements is that it is a long-term record: roughly 4.5 months at Tsukuba and 21 months both at Hedo and Yokosuka, providing a robust dataset for validating satellite NO$_2$ measurements.

At each site, we employed a miniaturized ultraviolet/visible (UV/VIS) spectrometer, a single telescope, and a movable mirror, to conduct sequential measurements of spectra of scattered sunlight at different elevation angles (ELs) every 30 minutes. In Table 1, a list of ELs used is given for each site. The spectral resolution (full width at half maximum (FWHM)) was kept at about 0.4–0.7 nm, depending on site. More information on these instruments can be found elsewhere (Irie et al. 2008a, for Tai’an; Irie et al. 2008b, 2009, for Tsukuba; Takashima et al. 2009, for Hedo). For Yokosuka, we used an instrument very similar to that of Hedo.

For all four sites, spectra measured by MAX-DOAS were analyzed by the same retrieval algorithm consisting of three steps: (1) the DOAS fit, (2) the aerosol profile/column retrieval, and (3) the NO$_2$ profile/column retrieval. Since the retrieval procedures as well as error
estimates have been described in detail by Irie et al. (2008a, 2008b, 2009), a short overview is given here.

Firstly, we applied the DOAS spectral fitting algorithm to the fitting window of 460–490 nm, retrieving the so-called differential slant column density (ΔSCD), defined as the difference between the column concentration integrated along the sunlight path measured at a low EL and that at the reference EL (70° for Hedo and 90° for the others) (Table 1). The fitting window of 460–490 nm was chosen to retrieve NO2 ΔSCD together with the oxygen collision complex (O4) ΔSCD from the same fitting window, minimizing the wavelength difference for the NO2 and O4 air mass factors. Absorption by NO2, O4, O, and H2O, and the Ring effect were taken into account. Over the whole period of measurements at Tsukuba, for example, median errors for the NO2 and O4 ΔSCD estimated from fitting residuals were ~1.8 × 1014 molecules cm−2 and ~1.0 × 1015 molecules cm−2, respectively, at solar zenith angles of 50° ± 5°.

The O4 ΔSCD values were next converted using our aerosol retrieval algorithm (e.g., Irie et al. 2008b) to aerosol optical depth (AOD) and the vertical profile of the aerosol extinction coefficient at a wavelength of 476 nm, which corresponds to the O4 cross-section-weighted mean wavelength over 460–490 nm. The aerosol retrieval method has been validated against coincident aerosol measurements by lidar and sky radiometer at Tsukuba (e.g., Irie et al. 2009) and Hedo (Takashima et al. 2009). A lookup table for the vertical profile of the box air mass factor (A box), which characterizes the ratio of the partial slant to vertical columns for a given layer, was created using a three-dimensional Monte Carlo radiative transfer model, MCARaTS (Iwabuchi 2006), and used to find the optimal aerosol and A box profiles that account for O4 ΔSCD values measured at all ELs. A box calculations by MCARaTS have been validated through comparisons with other radiative transfer models (Wagner et al. 2007).

Because the A box profile is a function of the aerosol profile, A box, is also uniquely determined by the aerosol retrieval method. Using the A box profiles and a nonlinear iterative inversion method similar to that of the aerosol retrieval (Irie et al. 2008a), the NO2 ΔSCD values were converted to tropospheric VCD values and the vertical profile of NO2.

For each retrieval, the random error was estimated from the retrieval covariance matrix constructed from the residual that arose in fitting the NO2 ΔSCD values. The systematic error was estimated as the change found when we varied the AOD by an additional ±30%, which is the dominant source of systematic error in this retrieval algorithm (Irie et al. 2008a). For data compared to OMI in the present study, the mean values of the random and systematic errors estimated in this way are summarized in Table 2.

### 3. OMI

The OMI instrument is a nadir-viewing imaging spectrometer measuring direct and atmospherically backscattered sunlight in the UV/VIS range from 270 to 500 nm (Levitt et al. 2006). It was put into a Sun-synchronous, polar orbit at an altitude of about 705 km with an equator crossing time between 13:40 and 13:50 LT. Over the OMI field of view (FOV) of 114°, 60 discrete viewing angles are distributed perpendicular to the flight direction. The FOV corresponds to a 2600-km-wide spatial swath on the Earth’s surface, achieving daily global measurements. Nadir spatial resolution ranges from 13 × 24 to 24 × 48 km2, depending on the operation mode. The present study uses the standard product (version 3) of the OMI tropospheric NO2 VCD obtained from the NASA GES-DISC. A “below cloud” NO2 column has been added for more realistic tropospheric column estimates, but only data with a cloud fraction less than 0.1 are analyzed below. For data used here, the mean values of the quoted error are given in Table 2.

### 4. Results and discussion

Recent validation studies for satellite NO2 measurements (e.g., Brinksma et al. 2008; Celarier et al. 2008; Irie et al. 2008a) have pointed out that validation...
comparisons should be made with stringent coincidence criteria for both time and space to account for the inhomogeneity of the NO$_2$ distribution. In the present study, a comparison is made only when the center of OMI measurement pixel was within 0.1° latitude and longitude of a given MAX-DOAS measurement site and the time difference between the two measurements was less than 30 minutes. Figure 2 shows an example time series of tropospheric NO$_2$ VCDs measured by MAX-DOAS and OMI at Tsukuba from November 1, 2006, to March 16, 2007. Although the MAX-DOAS measurements were made for more than 3 months at Tsukuba, only 14 coincident pairs of MAX-DOAS and OMI data fulfill the above criteria. For the selected 14 cases the mean difference (OMI minus MAX-DOAS) was $-3 \times 10^{15}$ molecules cm$^{-2}$ ($-18\%$), with a 1σ standard deviation of $4 \times 10^{15}$ molecules cm$^{-2}$ ($22\%$).

For all four sites, direct comparisons using correlation analysis are shown in Fig. 3. A regression line fit to all the data (red line) has been calculated taking into account both OMI and MAX-DOAS error ranges. The correlation coefficient for the regression analysis ($R^2$) is as high as 0.64, indicating that the relative changes seen in OMI data are very accurate. The regression line has a slope of $0.69 \pm 0.04$, indicating a negative bias of $31 \pm 4\%$ on average in the OMI data analyzed here, consistent with other validation comparisons, which suggested a negative bias of about 15–30% (e.g., Celarier et al. 2008).

To characterize the OMI NO$_2$ data also from the viewpoint of a long-term record, we investigated linear trends of the tropospheric NO$_2$ VCDs for 2005–2008, when almost complete year-round observations were made by OMI. Figures 4a and 4b show respectively the absolute and relative values of the trends derived from OMI data after the bias (components of both the slope and the intercept in Fig. 3) estimated using MAX-DOAS data is taken into account. Only estimates with trends greater than $5 \times 10^{14}$ or smaller than $-5 \times 10^{14}$ molecules cm$^{-2}$ per year, which approximately corresponds to the mean of the quoted OMI NO$_2$ errors (over the industrial region, the rectangular area in Fig. 4) divided by 4 (years), are plotted, as such changes over 4 years would be significant.

It is evident from Fig. 4 that significant increases in tropospheric NO$_2$ VCD occurred in China from 2005 to 2008. In particular, for the industrial region, which was shown by Richter et al. (2005) to have a continuous

Fig. 2. Time series of the tropospheric NO$_2$ VCD measured by MAX-DOAS at Tsukuba from November 1, 2006, to March 16, 2007 (gray). Coincident pairs of MAX-DOAS and OMI measurements are shown in black and red symbols, respectively.

Fig. 3. Correlations between the tropospheric NO$_2$ VCDs measured by MAX-DOAS and OMI for Tai’an (red), Tsukuba (blue), Hedo (green), and Yokosuka (open circles). Error bars represent the respective error ranges. The red line is a linear regression fit to all the data. Its intercept, slope, and correlation coefficient ($R^2$) are given. The 1:1 relationship is shown with the dashed line.

Fig. 4. Trends seen from the OMI tropospheric NO$_2$ VCD data for 2005–2008. (a) Absolute and (b) relative values of the trends are shown. The trends have been calculated for each grid of 0.5° × 0.5° latitude and longitude. Black rectangles represent the area of 30°–40°N and 110°–123°E investigated by Richter et al. (2005).
increase from 1996 to 2005 using GOME and SCIAMACHY data, we find a mean trend of 5 ± 3% per year (7 × 5 × 10^14 molecules cm\(^{-2}\) per year). For OMI data without corrections for biases estimated in this study, the trend was 6 ± 3% per year (5 × 3 × 10^14 molecules cm\(^{-2}\) per year). From these results, it is most likely that the tropospheric NO\(_2\) VCD over China has been increasing continuously from 1996 to 2008. In addition, a decreasing trend in tropospheric NO\(_2\) VCD was found in or near Tokyo, Japan, similar to the trend seen from GOME in 1996–2002 (Richter et al. 2005). For quantitative estimates of trends over 1996–2008, however, the local times of the satellite measurements should be taken into account.

Finally, it is interesting to note that OMI measurements, with a finer spatial resolution than those of GOME and SCIAMACHY, reveal that the spatial distribution of recent NO\(_2\) changes was inhomogeneous (Fig. 4). In particular, the rapid increases in tropospheric NO\(_2\) VCD at a rate of 10–15% per year have occurred in areas near Shenyang and Taiyuan (Fig. 4b). For a better emission evaluation in Asia, further detailed analyses of OMI data, which have been validated and characterized in this study, are highly encouraged.

5. Conclusions

We have used a robust dataset of tropospheric NO\(_2\) VCD measurements by MAX-DOAS at one site in China (Tai’an) and three sites in Japan (Tsukuba, Hedo, and Yokosuka) to validate and characterize OMI tropospheric NO\(_2\) VCD data (the standard product, version 3) over a wide range of NO\(_2\) levels in East Asia. Both MAX-DOAS and OMI data showed very low NO\(_2\) VCD values at Hedo and similar ranges at the other sites. By combining all these data, we found that OMI and MAX-DOAS NO\(_2\) values were well correlated, with R\(^2\) = 0.64. The slope of the correlations was found to be 0.69 ± 0.04, indicating a negative bias in the OMI data by 31 ± 4%. This is consistent with other validation studies (e.g., Celarier et al. 2008). After the estimated bias was taken into account, the mean trend over the industrial area of China (30°–40°N and 110°–123°E) estimated to be about 5 ± 3% per year (7 × 5 × 10^14 molecules cm\(^{-2}\) per year) for 2005–2008. OMI data additionally showed that the recent increasing trends for 2005–2008 differed between different regions in China, with the fastest increases over areas near Shenyang and Taiyuan.

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