Trend analysis of tropospheric NO\textsubscript{2} column density over East Asia during 2000–2010: multi-satellite observations and model simulations with the updated REAS emission inventory

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Trend analysis of tropospheric NO$_2$ column density over East Asia

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Abstract

Satellite observations of the tropospheric NO\textsubscript{2} vertical column density (VCD) are closely correlated to surface NO\textsubscript{x} emissions and can thus be used to estimate the latter. In this study, the NO\textsubscript{2} VCDs simulated by a regional chemical transport model with data from the updated Regional Emission inventory in ASia (REAS) version 2.1 were validated by comparison with multi-satellite observations (GOME, SCIAMACHY, GOME-2, and OMI) between 2000 and 2010. Rapid growth in NO\textsubscript{2} VCD driven by expansion of anthropogenic NO\textsubscript{x} emissions was revealed above the central eastern China region, except during the economic downturn. In contrast, slightly decreasing trends were captured above Japan. The modeled NO\textsubscript{2} VCDs using the updated REAS emissions reasonably reproduced the annual trends observed by multi-satellites, suggesting that the NO\textsubscript{x} emissions growth rate estimated by the updated inventory is robust. On the basis of the close linear relationship of modeled NO\textsubscript{2} VCD, observed NO\textsubscript{2} VCD, and anthropogenic NO\textsubscript{x} emissions, the NO\textsubscript{x} emissions in 2009 and 2010 were estimated. It was estimated that the NO\textsubscript{x} emissions from anthropogenic sources in China beyond doubled between 2000 and 2010, reflecting the strong growth of anthropogenic emissions in China with the rapid recovery from the economic downturn during late 2008 and mid-2009.

1 Introduction

Nitrogen oxides (NO\textsubscript{x} = NO + NO\textsubscript{2}) emitted from anthropogenic sources (e.g., fossil fuel combustion, transportation, power plants) and natural sources (e.g., microbiological processes in soil, lightning) play key roles in tropospheric chemistry, with important implications for air quality and climate change. In particular, NO\textsubscript{x} contributes to the formation of photochemical ozone (O\textsubscript{3}) and secondary aerosols. It is also involved in the chemical formation of other atmospheric species through feedback on hydroxyl radicals (OH) (Seinfeld and Pandis, 2006).
In recent years, advances in satellite technology and the development of new instruments and algorithms have allowed for observations of the NO\textsubscript{2} vertical column density (VCD) from space, providing useful information for air quality research (e.g., Richter et al., 2005; van der A et al., 2008; Irie et al., 2009). A global picture of the spatial distribution of the NO\textsubscript{2} VCD is now available because satellite measurements provide global coverage in a very short time (between 1 and 6 days, depending on the instrument and cloud cover). Operational observations of tropospheric NO\textsubscript{2} VCD have been performed for more than ten years continuously by the Global Ozone Monitoring Experiment (GOME), the SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY), the Ozone Monitoring Instrument (OMI), and the GOME-2. Because of the short lifetime of NO\textsubscript{x} in the troposphere, satellite NO\textsubscript{2} observations are closely correlated to the surface NO\textsubscript{x} emissions. The tropospheric NO\textsubscript{2} VCD retrieved from satellites has hence been successfully applied to evaluate and quantify the spatial distribution, temporal variation, and interannual trends of NO\textsubscript{x} emissions (He et al., 2007; Uno et al., 2007; van der A et al., 2008; Han et al., 2009; Lamsal et al., 2011).

A large positive trend of NO\textsubscript{2} VCD over eastern China in parallel with a strong increase in NO\textsubscript{x} emissions from 1996 to 2005 due to increases in industry and traffic has been found using a combination of GOME and SCIAMACHY observations (Richter et al., 2005). Based on a systematic analysis of NO\textsubscript{2} VCD measured by GOME and simulated by a regional chemical transport model with anthropogenic emission data from the Regional Emission inventory in ASia (REAS) version 1.1 (Ohara et al., 2007), it was revealed that the anthropogenic emissions would have underestimated the rapid growth in NO\textsubscript{x} emissions from China, especially during 1998–2000 (Uno et al., 2007). The emissions estimated by bottom-up methods have large uncertainties because of the uncertainties in activity data such as energy consumption, removal efficiencies, emission factors, and others. A combination of top-down methods is one potential alternative approach (e.g., Martin et al., 2003). By developing a data assimilation system,
NO\textsubscript{x} emissions in an a priori REAS emission inventory was optimized to catch up the rapid growth of emissions from China (Kurokawa et al., 2009).

Recently, the REAS emission inventories were updated to version 2.1 based on the same methodology as previous versions, but with updated activity data and parameters. The new dataset covers the years from 2000 to 2008 with expanded target areas, including Russia and Central Asia. The dataset contains monthly variations in the individual sources and source categories with a 0.25° × 0.25° grid resolution (Kurokawa et al., 2013). Evaluations of emission inventories estimated from the bottom-up method by comparing ground-based and/or space-borne observations are needed to improve our knowledge of air quality. Emission data are necessary inputs for chemical transport models, although large uncertainties remain in such data. In this study, the tropospheric NO\textsubscript{2} VCD over East Asia observed by four satellite observations (GOME, SCIAMACHY, GOME-2, and OMI) and simulated by a regional chemical transport model with the updated REAS inventory version 2.1 were combined and analyzed. Trends in tropospheric NO\textsubscript{2} VCD between 2000 and 2010 over China, Korea, and Japan were examined, with an emphasis on corresponding changes in anthropogenic NO\textsubscript{x} emissions. By fully utilizing multi-satellite observations, variation of tropospheric NO\textsubscript{2} VCD in the morning and the afternoon were also investigated. Moreover, on the basis of the clear relationships between NO\textsubscript{2} VCD and anthropogenic NO\textsubscript{x} emissions, simplified inverse estimations were also proposed.

2 Modeling setup and satellite observations

2.1 Model description

Three-dimensional numerical simulations over East Asia were conducted using the regional chemical transport model of the Community Multi-scale Air Quality (CMAQ) modeling system version 4.7.1 (Byun and Schere, 2006) released by the US Environmental Protection Agency (EPA). The CMAQ modeling systems have been used...
extensively for atmospheric environmental research over East Asia (e.g., He et al., 2007; Uno et al., 2007; Han et al., 2009; Itahashi et al., 2012a, b, 2013). In this study, the meteorological fields were generated by the Weather Research and Forecasting (WRF) model version 3.3 with the National Centers for Environmental Prediction (NCEP)/National Center for Atmospheric Research (NCAR) reanalysis data (FNL; http://rda.ucar.edu/datasets/ds083.2/). The model was constructed with a $98 \times 78$ horizontal grid at a resolution of 80 km, centered at $35^\circ$ N, $115^\circ$ E on a Lambert conformal projection, with 37 vertical grids extending from the surface to 50 hPa. Anthropogenic emissions data were obtained from the newly updated REAS version 2.1 (Kurokawa et al., 2013). Natural sources of NO$_x$ from soil were also prepared from the updated REAS version 2.1, but from lightning were not considered in this study due to the difficulty of determining its magnitude and variations. Emissions from biogenic and biomass burning, with monthly variations, were obtained from the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006) and the Global Fire Emissions Database (GFED) version 3.1 (van der Werf et al., 2010), respectively. CMAQ was configured to use mass-conserving scheme for advection, multiscale horizontal diffusion and eddy vertical diffusion. The Statewide Air Pollution Research Center version 99 (SAPRC-99) chemical mechanism and AERO5 were respectively adopted for production of gas-phase chemistry and aerosol chemistry. The initial conditions were provided in the CMAQ default dataset, and lateral boundary conditions were generated using the results of the CHemical Atmospheric general circulation model for Study of atmospheric Environment and Radiative forcing (CHASER) global chemistry model (Sudo et al., 2002), with a monthly-mean basis. Numerical simulations with the updated REAS emission were performed from 2000 to 2008, and the emissions from anthropogenic sources were fixed for 2009 and 2010 using the same inventory as 2008 in a sensitivity simulation.
2.2 Observation data

GOME, SCIAMACHY, GOME-2, and OMI data were analyzed in this study. The specifications of these instruments are summarized in Table 1. The GOME, SCIAMACHY, and GOME-2 data were used for morning observations, and the OMI data were used for afternoon observations. The level 2 products of TM4NO2A version 2.0 were used. These were developed at the Royal Netherlands Meteorological Institute (KNMI) and are available from the Tropospheric Emission Monitoring Internet Service (TEMIS) at www.temis.nl. The retrieval process for tropospheric NO$_2$ VCD was as follows. (1) The NO$_2$ slant column density (SCD) was obtained from the reflectance spectra using the differential optical absorption spectroscopy (DOAS) technique. (2) The stratospheric and tropospheric contributions to the NO$_2$ SCD were separated by assimilating the NO$_2$ SCD into the TM4 chemistry-transport model. (3) The tropospheric air mass factor (AMF) was incorporated to convert the tropospheric NO$_2$ SCD into the tropospheric NO$_2$ VCD (e.g., Boersma et al., 2007). The improvements in the version 2.0 products compared to version 1.0 included an updated albedo database, improved AMF and surface height calculations, and a new version of the TM4 chemistry-transport model (e.g., Boersma et al., 2011).

The observed NO$_2$ VCDs were analyzed from January 2000 to July 2003 for GOME, from August 2002 to December 2010 for SCIAMACHY, from January 2007 to December 2010 for GOME-2, and from October 2004 to December 2010 for OMI. The SCIAMACHY, GOME-2, and OMI measurement results had no significant biases compared to ground-based Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) observations during daytime (Irie et al., 2012). For the multi-satellite observation comparisons, model results at 10:00 LT and 14:00 LT were used to represent morning and afternoon results, respectively. The satellite observation dataset giving the location and value for each measurement pixel were interpolated into the same model resolution (0.5° × 0.5°), with only used during cloud-free conditions (cloud fraction less than 20 %). The swath path and obtained pixels of the satellite observations were not taken into
account in the analysis of the model results, the satellite record therefore has a clear-sky bias. However, a largest difference of NO$_2$ VCD with regard to the cloud fraction is found to be up to 30–35%, which is much smaller than the quoted uncertainty in satellite retrieval (Uno et al., 2007; Irie et al., 2013). Averaging kernel information were not applied in the model analysis, but profile uncertainties which lead to changes in the retrieval are approximately 10% (Boersma et al., 2004).

3 Results and discussion

3.1 Tropospheric NO$_2$ column trends

Tropospheric NO$_2$ VCDs above central eastern China (CEC; 110–123° E, 30–40° N), Korea (125–130° E, 34.5–39° N), and Japan (133–141° E, 33.5–37° N) were examined (see top-left panel in Fig. 2). In Fig. 1, the temporal variations in monthly mean tropospheric NO$_2$ VCD above CEC are shown for (a) morning and (b) afternoon. Both the model results and satellite observations clearly illustrate the rapid increase in NO$_2$ VCD above the CEC between 2000 and 2010, except during the economic downturn between late 2008 and mid-2009. The peak levels of monthly mean NO$_2$ VCD in the winter of 2008–2009 were significantly lower than those in the winter of 2007–2008. However, satellite observations indicated a rapid recovery during the winter of 2009–2010. CMAQ modeling results reproduced the observed temporal variation but greatly underestimated the high NO$_2$ VCD in the mornings during winter. However, they remarkably captured the absolute values throughout the year between 2000 and 2008 in the afternoon. In the model simulations, a sensitivity study during 2009–2010 with anthropogenic emission fixed at 2008 levels was conducted. The large differences between model and satellite observations also implied the recovery of the Chinese economy and hence increasing emissions during 2009–2010 compared to 2008 values. These differences were larger and clearer in the morning than in the afternoon because the morning was considered to have a more direct relation to emissions. Here we note
that, the absolute values of modeled NO\textsubscript{2} VCD showed improved accuracy compared to those in our previous study. Modeled NO\textsubscript{2} VCDs were reported to be lower than observed values by factors of 2–4 over polluted CEC (Uno et al., 2007). Possible reasons for the improved accuracy in the present study are changes in vertical resolution, the updating of CMAQ modeling system (including the chemical reaction scheme and improvements to vertical diffusion), and the use of the updated REAS inventory.

To further investigate the effect of the economic downturn, the normalized tropospheric NO\textsubscript{2} VCD to averaged values between October 2007 and September 2008 with a 12 month moving window were analyzed using the same manner as Lin et al. (2012). Results are shown in the lower part of Fig. 1. The model results obtained using the updated REAS emission inventory remarkably reproduced the growth rate of NO\textsubscript{2} VCD in both the morning and afternoon and suggested that the NO\textsubscript{2} VCD approximately doubled from 2001–2002 to 2008 (before the economic downturn) during both the morning and afternoon. This increased NO\textsubscript{2} VCD was caused by the increase in NO\textsubscript{x} emissions from anthropogenic sources. The growth of anthropogenic NO\textsubscript{x} emissions is discussed later.

The seasonal variation in tropospheric NO\textsubscript{2} VCD above CEC is shown as summer minima and winter maxima (Fig. 1). Modeled results greatly underestimated in the morning during winter above the CEC region. Comparisons between the spatial distribution of tropospheric NO\textsubscript{2} VCD in the morning (model results at 10:00 LT and retrieved data from SCIAMACHY) are presented in Fig. 2, with focus on the summer (June 2005 to August 2005) and winter (December 2005 to February 2006) seasons. In summer, due to the short lifetime of NO\textsubscript{x}, the highest NO\textsubscript{2} VCDs were found around megacities such as Beijing, Shanghai, Hong Kong, Seoul, Tokyo, and the North China Plain region. Above the North China Plain, the model reproduced the temporal variation in the NO\textsubscript{2} VCD over the CEC region, but overestimated it in Hebei Province and underestimated it in Shanxi Province. Revision of emissions at the province scale may be required, and considerations of lightning emissions would partly improve the underestimation of model results especially above northern part of CEC (e.g., Lin, 2012). In winter, high
NO$_2$ VCD covered the entire CEC region, and the model generally underestimated values in this region. There may have been underestimation in the NO$_x$ emission estimations and uncertainty in the NO$_x$-related chemistry of model simulations, along with potential problems in the NO$_2$ retrievals from satellite observations (e.g., Han et al., 2009). In the model simulations, more detailed studies based on sensitivity tests (e.g., spatial resolution, emission intensity) with CMAQ were conducted (Irie et al., 2013). However, the reasons for the difference remain unclear. The averaged NO$_2$ VCD values above the CEC were improved in a 20% emission increment sensitivity run, but the diurnal variation (ratio of morning to afternoon values) still showed discrepancies between the model and satellite data. Modeling uncertainties in meteorology and chemistry are also important factors. Modeled cloud optical depth, the uptake coefficient of HO$_2$ on aerosols, and some rate constant have found to be large impacts on NO$_2$ VCD in global chemistry model of GEOS-Chem, and these issues would be also relevant to CMAQ model (Lin et al., 2012). From the viewpoint of satellite observations, there may be difficulties in retrieving wintertime NO$_2$ VCDs with the same quality as summertime NO$_2$ VCDs, for example, due to more stable, shallow boundary layer conditions in winter. The quoted uncertainty in satellite NO$_2$ VCD appears to be usually larger in winter than in summer (Irie et al., 2013).

The annual mean tropospheric NO$_2$ VCDs over the CEC, Korea, and Japan regions (see rectangular region in Fig. 2) between 2000 and 2010 are illustrated in Fig. 3, along with the temporal variation of anthropogenic NO$_x$ emissions of the total in China, Korea, and Japan estimated by the updated REAS inventory. Over China, as has been discussed, the model tended to underestimate the tropospheric NO$_2$ VCD in morning during winter. The annual mean NO$_2$ VCD was therefore also underestimated by the model. However, the increases were closely reproduced by the model, and the growth rates of tropospheric NO$_2$ VCD during 2002–2008 were $+10.6\%$ yr$^{-1}$ (model, 10:00 LT) and $+9.7\%$ yr$^{-1}$ (satellite, SCIAMACHY), respectively. During the same period, the updated REAS estimated a $+8.7\%$ yr$^{-1}$ growth in NO$_x$ emissions from anthropogenic sources. The tropospheric NO$_2$ VCD also exhibited growth of $+8.9\%$ yr$^{-1}$ (CMAQ,
14:00 LT) and +5.1 % yr\(^{-1}\) (satellite, OMI), respectively, between 2005 and 2008 in the afternoon. The differences between satellite observations and models with sensitivity simulations in 2009 and 2010 (open circles in Fig. 3) again showed emission growth in 2009–2010 after the recovery from the economic downturn. Above Korea, both satellite observations and model results showed complex variability. The tropospheric NO\(_2\) VCD showed an initial peak during 2003–2004, and then a decrease toward 2006. Following this, the satellite observations exhibited an increasing trend from 2007 by SCIAMACHY and a flat trend by GOME-2. The model produced a slightly decreasing trend during 2007–2008 according to the reduction in anthropogenic NO\(_x\) emissions. In the afternoon, the tropospheric NO\(_2\) VCDs were continuously constant above Korea. In Japan, slightly decreasing trends of tropospheric NO\(_2\) VCD between 2000 and 2010 were revealed, along with a decline in emissions both in the morning and afternoon. To summarize, for the annual trend in NO\(_2\) VCD, the model simulations utilizing the updated REAS emission inventory reasonably reproduced the tropospheric NO\(_2\) VCD observed by multi-satellites. To clarify the discrepancies between satellite observations and model results found over Korea after 2007, and to validate the model results with updated REAS emissions, a comparison study with other emission inventories would be helpful (e.g., Han et al., 2009).

3.2 Inverse estimation of anthropogenic NO\(_x\) emissions

The NO\(_2\) VCD modeled with the updated REAS emission inventory version 2.1 captured the temporal variation in the observed NO\(_2\) VCD from multi-satellite observations, and closely corresponded with the fluctuations in NO\(_x\) emissions from anthropogenic sources above CEC, Korea, and Japan. A simple inverse estimation of NO\(_x\) emissions using tropospheric NO\(_2\) VCD observation is proposed, focusing on the CEC region. The relationships among modeled NO\(_2\) VCD, observed NO\(_2\) VCD, and anthropogenic NO\(_x\) emissions above CEC estimated by the updated REAS inventory are presented in Fig. 4. In this comparison, only the SCIAMACHY data were used because that dataset
for the morning was considered to have a more direct relationship with emissions, and SCIAMACHY has longer archives than the other satellites. Because of the underestimation of modeled NO$_2$ VCD in winter, the slope of the linear regression results on an annual basis was larger than unity. However, a close linear relationship can be seen between the model and satellite observations. Furthermore, the relationships between the modeled NO$_2$ VCD and anthropogenic NO$_x$ emissions from CEC were also found to be clearly linear (see Fig. 4b). The modeled NO$_2$ VCD was calculated using various emission sources (e.g., anthropogenic, biomass burning, and biogenic emissions), but was mainly attributable to anthropogenic sources (Itahashi et al., 2013).

Hereafter, a simple estimate of NO$_x$ emissions from the CEC region based on the observed NO$_2$ VCD is introduced. Because of the close linear relationship between observed and modeled NO$_2$ VCD, the modeled NO$_2$ VCD in 2009 and 2010 could be estimated from the linear regression results in Fig. 4a. After the modeled NO$_2$ VCD in 2009 and 2010 were estimated, the anthropogenic emissions from CEC were assessed based on the close linear relationship in Fig. 4b. The results showed that the estimated anthropogenic NO$_x$ emissions from CEC in 2009 and 2010 were respectively 13.6 ($\pm 0.2$) Tgyr$^{-1}$ and 14.9 ($\pm 0.3$) Tgyr$^{-1}$. The results in parentheses indicate that the differences in the y-intercept are not forced to zero in Fig. 4a. Considering that the ratio of the CEC emissions to the total in China is around 46%, anthropogenic NO$_x$ emissions from China in 2009 and 2010 were estimated to be 29.7 ($\pm 0.5$) Tgyr$^{-1}$ and 32.3 ($\pm 0.7$) Tgyr$^{-1}$, respectively. Furthermore, considering that the anthropogenic NO$_x$ emissions from China were estimated to be 14.3 Tgyr$^{-1}$ in 2000, anthropogenic NO$_x$ emissions from China beyond doubled between 2000 and 2010. The observed tropospheric NO$_2$ VCD declined during the economic recession between late 2008 and mid-2009, but the rapid recovery in winter of 2009–2010 led to a clear increase in anthropogenic NO$_x$ emissions.

Our results remarked that 7.2 % yr$^{-1}$ growth of anthropogenic NO$_x$ emissions from 2006 to 2009 in China. This is consistent to a research by Lamsal et al. (2011) which estimated a annual growth rate of 6.7% in China during the same period based on
their β method using a model based sensitivity and satellite observation data. Our methodology based on a close linear relation provides a simple estimate of emissions, but can be valuable for examining the rapid changes in emissions resulting from economic shifts and/or control measures. Here we note that, when we used the observed NO2 VCD measured by OMI, the estimated anthropogenic NOx emissions from CEC in 2009 and 2010 were respectively 12.6 (+0.5) Tg yr\(^{-1}\) and 14.3 (+1.2) Tg yr\(^{-1}\), well consistent with the result using the SCIAMACHY data.

In our previous study, anthropogenic SO\(_2\) emissions from China were inversely estimated based on the observed SO\(_2\) column density above China and aerosol optical thickness in the downwind region (Itahashi et al., 2012a, b). It was assessed that anthropogenic SO\(_2\) emissions from China in 2009 was equivalent to that in 2004 owing to widespread installation of fuel-gas desulfurization. As we have examined through this study, the anthropogenic NO\(_x\) emissions had strong positive increase during 2000–2010. In contrast, the regulations imposed by the 12th Five-Year Plan (2011–2015) of the Chinese government aimed to reduce NO\(_x\) emissions, therefore, it is considered that air quality over East Asia would be faced to complex variability. Combining numerical simulations with satellite observations would offer insight into changes in air quality over East Asia.

4 Conclusions

In this study, tropospheric NO\(_2\) VCD between 2000 and 2010 over East Asia was investigated based on a combined analysis of multi-satellite observations (GOME, SCIAMACHY, GOME-2, and OMI) and CMAQ model simulations with an updated REAS emission inventory version 2.1. A rapid increase in tropospheric NO\(_2\) VCD along with expanding anthropogenic emissions was revealed above the CEC region in both model results and satellite observations, except during the economic downturn between late 2008 and mid-2009. The model simulations closely reproduced the growth of NO\(_2\) VCD both in the morning and afternoon and showed an approximate doubling of
tropospheric NO$_2$ VCD from 2001–2002 to 2008 (before the economic downturn). In contrast to the situation above the CEC, the NO$_2$ VCD showed a slight decreasing trend over Japan with continuous reduction in anthropogenic emissions. Above Korea, the NO$_2$ VCD increased toward 2003–2004, before decreasing to 2006. However, the following situation was different between the model results and satellite observations in the morning. Both the model results and satellite observations revealed a constant trend of NO$_2$ VCD above Korea in the afternoon. The tropospheric NO$_2$ VCD simulated with the updated REAS emission inventory reasonably reproduced the observed NO$_2$ VCD from multi-satellite observations. However, more research, such as comparison studies with other emission inventories, will be necessary to establish a more reliable emission inventory over East Asia. Modeling uncertainties should also be considered in light of recent studies in forthcoming study (Lin et al., 2012; Stvrakou et al., 2013).

The differences between multi-satellite observations and model sensitivity simulations in 2009 and 2010 with fixed emissions in 2008 implied a growth of NO$_x$ emissions from 2008 levels. The model results and satellite observations of tropospheric NO$_2$ VCD were closely correlated, with a linear relationship to anthropogenic NO$_x$ emissions. Hence a simple inverse estimate of NO$_x$ emissions was proposed. On the basis of the NO$_2$ VCD observed by satellite, the anthropogenic NO$_x$ emissions in 2009 and 2010 were estimated using linear regression results. By this approach, we estimated that the anthropogenic NO$_x$ emissions from China beyond doubled between 2000 and 2010. This means that there was a strong increase in anthropogenic emissions in China and rapid recovery of the Chinese economy from economic recession.

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Table 1. Summary of satellite-based NO$_2$ measurement instruments.

<table>
<thead>
<tr>
<th>Instrument</th>
<th>GOME</th>
<th>SCIAMACHY</th>
<th>GOME-2</th>
<th>OMI</th>
</tr>
</thead>
<tbody>
<tr>
<td>Satellite</td>
<td>ERS-2</td>
<td>ENVISAT</td>
<td>MetOp</td>
<td>Aura</td>
</tr>
<tr>
<td>Overpass time</td>
<td>10:30 LT</td>
<td>10:00 LT</td>
<td>9:30 LT</td>
<td>13:45 LT</td>
</tr>
<tr>
<td>Nadir-view spatial resolution</td>
<td>40 × 320km$^2$</td>
<td>30 × 60km$^2$</td>
<td>13 × 24km$^2$</td>
<td>40 × 80km$^2$</td>
</tr>
<tr>
<td>Global coverage</td>
<td>3 days</td>
<td>6 days</td>
<td>1 day</td>
<td>1 day</td>
</tr>
</tbody>
</table>
Fig. 1. Temporal variation in monthly mean tropospheric NO\textsubscript{2} VCD above CEC region during 2000–2010 from multi-satellite observations (GOME, SCIAMÁCHY, GOME-2, OMI) and model results (open circles denote sensitivity simulations with anthropogenic emissions fixed at 2008 values) in (a) the morning and (b) the afternoon. The normalized tropospheric NO\textsubscript{2} VCD relative to averaged values between October 2007 and September 2008 with a 12 month moving average are also shown at the bottom.
Fig. 2. Spatial distribution of tropospheric NO$_2$ VCD from (left) model results at 10:00 LT, (center) satellite observations by SCIAMACHY, and (right) differences between model and satellite results (top) in summer (averaged between June 2005 and August 2005) and (bottom) in winter (averaged between December 2005 and February 2006). The rectangular regions shown in the top-left figure are the investigation regions (CEC, Korea, and Japan).
Fig. 3. Annual trend of tropospheric NO$_2$ VCD during 2000–2010 from the multi-satellite observations (GOME, SCIAMACHY, GOME-2, and OMI) and model results (open circles denote sensitivity simulations with anthropogenic emissions fixed at 2008 values) with 1σ standard deviation above (a) CEC, (b) Korea, and (c) Japan in (left) the morning and (right) the afternoon.
Fig. 4. Scatter diagram of (a) annual-mean modeled tropospheric NO$_2$ VCD at 10:00 LT, with annual-mean observed tropospheric NO$_2$ VCD by SCIAMACHY above CEC (open circles denote sensitivity simulations with anthropogenic emissions fixed at 2008 values), and (b) annual-mean modeled tropospheric NO$_2$ VCD at 10:00 LT above CEC with annual-based anthropogenic NO$_x$ emissions from CEC.