Atmos. Chem. Phys. Discuss., 13, 11247–11268, 2013
www.atmos-chem-phys-discuss.net/13/11247/2013/
doi:10.5194/acpd-13-11247-2013

Atmospheric Chemistry and Physics Discussions
Open Access

Trend analysis of tropospheric NO_2 column density over East Asia during 2000–2010: multi-satellite observations and model simulations with the updated REAS emission inventory

S. Itahashi[1,*], I. Uno[2], H. Irie[3], J. Kurokawa[4], and T. Ohara[5]

[1]Department of Earth System Science and Technology, Kyushu University, 6-1 Kasuga Park, Kasuga, Fukuoka, 816-8580, Japan
[2]Research Institute for Applied Mechanics, Kyushu University, 6-1 Kasuga Park, Kasuga, Fukuoka, 816-8580, Japan
[3]Center for Environmental Remote Sensing, Chiba University, 1-33 Yayoicho, Inage-ku, Chiba, 263-8522, Japan
[4]Asia Center for Air Pollution Research, 1182 Sowa, Nishi-Ku, Niigata, 950-2144, Japan
[5]National Institute for Environmental Studies, 16-2 Onogawa, Tsukuba, Ibaraki, 305-8506, Japan
[*]now at: Environmental Science Research Laboratory, Central Research Institute of Electric Power Industry, 1646 Abiko, Abiko, Chiba, 270-1194, Japan

Received: 21 March 2013 – Accepted: 21 April 2013 – Published: 25 April 2013

Correspondence to: S. Itahashi, (syuichi@riam.kyushu-u.ac.jp, isyuichi@criepi.denken.or.jp)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

Satellite observations of the tropospheric NO_2 vertical column density (VCD) are closely correlated to surface NO_x emissions and can thus be used to estimate the latter. In this study, the NO_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_2 VCD driven by expansion of anthropogenic NO_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_2 VCDs using the updated REAS emissions reasonably reproduced the annual trends observed by multi-satellites, suggesting that the NO_x emissions growth rate estimated by the updated inventory is robust. On the basis of the close linear relationship of modeled NO_2 VCD, observed NO_2 VCD, and anthropogenic NO_x emissions, the NO_x emissions in 2009 and 2010 were estimated. It was estimated that the NO_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 ($NO_x = NO + NO_2$) 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_x contributes to the formation of photochemical ozone (O_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).

ACPD
13, 11247–11268, 2013

In recent years, advances in satellite technology and the development of new instruments and algorithms have allowed for observations of the NO_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_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_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_x in the troposphere, satellite NO_2 observations are closely correlated to the surface NO_x emissions. The tropospheric NO_2 VCD retrieved from satellites has hence been successfully applied to evaluate and quantify the spatial distribution, temporal variation, and interannual trends of NO_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 trends of NO_2 VCD over eastern China in parallel with a strong increase in NO_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_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_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,

ACPD
13, 11247–11268, 2013

NO_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_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_2 VCD between 2000 and 2010 over China, Korea, and Japan were examined, with an emphasis on corresponding changes in anthropogenic NO_x emissions. By fully utilizing multi-satellite observations, variation of tropospheric NO_2 VCD in the morning and the afternoon were also investigated. Moreover, on the basis of the clear relationships between NO_2 VCD and anthropogenic NO_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

ACPD
13, 11247–11268, 2013

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 × 78 horizontal grid at a resolution of 80 km, centered at 35° N, 115° 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 ($0.5^\circ \times 0.5^\circ$), 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

ACPD
13, 11247–11268, 2013

that, the absolute values of modeled NO_2 VCD showed improved accuracy compared to those in our previous study. Modeled NO_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_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_2 VCD in both the morning and afternoon and suggested that the NO_2 VCD approximately doubled from 2001–2002 to 2008 (before the economic downturn) during both the morning and afternoon. This increased NO_2 VCD was caused by the increase in NO_x emissions from anthropogenic sources. The growth of anthropogenic NO_x emissions is discussed later.

The seasonal variation in tropospheric NO_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_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_x, the highest NO_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_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

ACPD
13, 11247–11268, 2013

NO_2 VCD covered the entire CEC region, and the model generally underestimated values in this region. There may have been underestimationf 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,

Discussion Paper | Discussion Paper | Discussion Paper | Discussion Paper |

ACPD

13, 11247–11268, 2013

Trend analysis of tropospheric NO_2 column density over East Asia

S. Itahashi et al.

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 (+0.2) $Tg\,yr^{-1}$ and 14.9 (+0.3) $Tg\,yr^{-1}$. 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 (+0.5) $Tg\,yr^{-1}$ and 32.3 (+0.7) $Tg\,yr^{-1}$, respectively. Furthermore, considering that the anthropogenic NO_x emissions from China were estimated to be 14.3 $Tg\,yr^{-1}$ 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 NO_2 VCD measured by OMI, the estimated anthropogenic NO_x 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

ACPD
13, 11247–11268, 2013

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.

Acknowledgements. This work was supported in part by the Global Environment Research Fund (S-7) of the Ministry of Environment, Japan. We are grateful for the use of the tropospheric NO_2 dataset obtained from www.temis.nl. We also thank K. Yamaji and K. Ikeda of the Research Institute for Global Change, Japan Agency for Marine-Earth Science and Technology (JAMSTEC), for their comments on the CMAQ calculations.

References

Boersma, K. F., Eskes, H. J., Brinksma, E. J.: Error analysis for tropospheric NO_2 retrieval from space, J. Geophys. Res., 109, D04311, doi:10.1029/2003JD003962, 2004.

Boersma, K. F., Eskes, H. J., Veefkind, J. P., Brinksma, E. J., van der A, R. J., Sneep, M., van den Oord, G. H. J., Levelt, P. F., Stammes, P., Gleason, J. F., and Bucsela, E. J.: Near-real time retrieval of tropospheric NO_2 from OMI, Atmos. Chem. Phys., 7, 2103–2118, doi:10.5194/acp-7-2103-2007, 2007.

Boersma, K. F., Eskes, H. J., Dirksen, R. J., van der A, R. J., Veefkind, J. P., Stammes, P., Huijnen, V., Kleipool, Q. L., Sneep, M., Claas, J., Leitão, J., Richter, A., Zhou, Y., and Brunner, D.: An improved tropospheric NO_2 column retrieval algorithm for the Ozone Monitoring Instrument, Atmos. Meas. Tech., 4, 1905–1928, doi:10.5194/amt-4-1905-2011, 2011.

Byun, D. W. and Schere, K. L.: Review of the governing equations, computational algorithms, and other components of the Model-3 Community Multiscale Air Quality (CMAQ) modeling system, Appl. Mech. Rev., 59, 51–77, 2006.

Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), Atmos. Chem. Phys., 6, 3181–3210, doi:10.5194/acp-6-3181-2006, 2006.

Han, K. M., Song, C. H., Ahn, H. J., Park, R. S., Woo, J. H., Lee, C. K., Richter, A., Burrows, J. P., Kim, J. Y., and Hong, J. H.: Investigation of NO_x emissions and NO_x-related chemistry in East Asia using CMAQ-predicted and GOME-derived NO_2 columns, Atmos. Chem. Phys., 9, 1017–1036, doi:10.5194/acp-9-1017-2009, 2009.

He, Y- J., Uno, I., Wang, Z., Ohara, T., Sugimoto. N., Shimizu, A., Richter, A., and Burrows, J. P.: Variations of the increasing trend of tropospheric NO_2 over east China during the past decade, Atmos. Environ., 41, 4865–4876, 2007.

Irie, H., Kanaya, Y., Takashima, H., Gleason, J. F., and Wang, Z.: Characterization of OMI Tropospheric NO_2 measurements in East Asia based on a robust validation comparison, SOLA, 5, 117–120, doi:10.2151/sola.2009-030, 2009.

Irie, H., Boersma, K. F., Kanaya, Y., Takashima, H., Pan, X., and Wang, Z. F.: Quantitative bias estimates for tropospheric NO_2 columns retrieved from SCIAMACHY, OMI, and GOME-2 using a common standard for East Asia, Atmos. Meas. Tech., 5, 2403–2411, doi:10.5194/amt-5-2403-2012, 2012.

ACPD
13, 11247–11268, 2013

Irie, H., Yamaji, K., Ikeda, K., Uno, I., Itahashi, S., Ohara, T., and Kurokawa, J.: An evaluation of the CMAQ reproducibility of satellite tropospheric NO_2 column observations at different local times over East Asia, submitted to acp, 2013.

Itahashi, S., Uno, I., and Kim, S- T.: Source contributions of sulfate aerosol over East Asia estimated by CMAQ-DDM, Environ. Sci. Technol., 46, 6733–6741, 2012a.

Itahashi, S., Uno, I., Yumimoto, K., Irie, H., Osada, K., Ogata, K., Fukushima, H., Wang, Z., and Ohara, T.: Interannual variation in the fine-mode MODIS aerosol optical depth and its relationship to the changes in sulfur dioxide emissions in China between 2000 and 2010, Atmos. Chem. Phys., 12, 2631–2640, doi:10.5194/acp-12-2631-2012, 2012b.

Itahashi, S., Uno, I., and Kim, S- T.: Seasonal source contributions of tropospheric ozone over East Asia based on CMAQ-HDDM, Atmos. Environ., 70, 204–217, 2013.

Kurokawa, J., Yumimoto, K., Uno, I., and Ohara, T.: Adjoint inverse modeling of NO_x emissions over eastern China using satellite observations of NO_2 vertical column densities, Atmos. Environ., 43, 1878–1887, 2009.

Kurokawa, J., Ohara, T., Morikawa, T., Hanayama, S., Greet, J.-M., Fukui, T., Kawashima, K., and Akimoto, H.: Emissions of air pollutants and greenhouse gases over Asian regions during 2000–2008: Regional Emission inventory in ASia (REAS) version 2, Atmos. Chem. Phys. Discuss., 13, 10049–10123, doi:10.5194/acpd-13-10049-2013, 2013.

Lamsal, L. N., Martin, R. V., Padmanabhan, A., van Donkelaar, A, Zhang, Q., Sioris, C. E., Chance, K., Kurosu, T. P., Newchurch, M. J.: Application of satellite observations for timely updates to global anthropogenic NO_x emission inventory, Geophys. Res. Lett., 38, L05810, doi:10.1029/2010GL046476, 2011.

Lin, J.-T.: Satellite constraint for emissions of nitrogen oxides from anthropogenic, lightning and soil sources over East China on a high-resolution grid, Atmos. Chem. Phys., 12, 2881–2898, doi:10.5194/acp-12-2881-2012, 2012.

Lin, J.-T. and McElroy, M. B.: Detection from space of a reduction in anthropogenic emissions of nitrogen oxides during the Chinese economic downturn, Atmos. Chem. Phys., 11, 8171–8188, doi:10.5194/acp-11-8171-2011, 2011.

Lin, J.-T., Liu, Z., Zhang, Q., Liu, H., Mao, J., and Zhuang, G.: Modeling uncertainties for tropospheric nitrogen dioxide columns affecting satellite-based inverse modeling of nitrogen oxides emissions, Atmos. Chem. Phys., 12, 12255–12275, doi:10.5194/acp-12-12255-2012, 2012.

ACPD
13, 11247–11268, 2013

Martin, R. V., Jacob, D. J., Chance, K., Kurosu, T. P, Palmer, P. I., and Evans, M. J.: Global inventory of nitrogen oxide emissions constrained by space-based observations of NO_2 columns, J. Geophys. Res., 108, 4537, doi:10.1029/2003JD003453, 2003.

Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., and Hayasaka, T.: An Asian emission inventory of anthropogenic emission sources for the period 1980–2020, Atmos. Chem. Phys., 7, 4419–4444, doi:10.5194/acp-7-4419-2007, 2007.

Richter, A., Burrows, J. P., Nues, H., Granier, C., and Niemeier, U.: Increase in tropospheric nitrogen dioxide over China observed from space, Nature, 437, 129–132, 2005.

Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics: from Air Pollution to Climate Change, 2nd edn., John Wiley & Sons, Inc., Hoboken, New Jersey, USA, 2006.

Sudo, K., Takahashi, M., Kurokawa, J. and Akimoto, H.: CHASER: a global chemical model of the troposphere 1. Model description, J. Geophys. Res., 107, D17, doi:10.1029/2001JD001113, 2002.

Stavrakou, T., Müller, J.-F., Boersma, K. F., van der A, R. J., Kurokawa, J., Ohara, T., and Zhang, Q.: Key chemical NO_x sink uncertainties and how they influence top-down emissions of nitrogen oxides, Atmos. Chem. Phys. Discuss., 13, 7871–7929, doi:10.5194/acpd-13-7871-2013, 2013.

Uno, I., He, Y., Ohara, T., Yamaji, K., Kurokawa, J.-I., Katayama, M., Wang, Z., Noguchi, K., Hayashida, S., Richter, A., and Burrows, J. P.: Systematic analysis of interannual and seasonal variations of model-simulated tropospheric NO_2 in Asia and comparison with GOME-satellite data, Atmos. Chem. Phys., 7, 1671–1681, doi:10.5194/acp-7-1671-2007, 2007.

van der A, R. J., Eskes, H. J., Boersma, K. F., van Noije, T. P. C., Van Roozendael, M., De Smedt, I., Peters, D. H. M. U., and Meijer, E. W.: Trends, seasonal variability and dominant NO_x source derived from a ten year record of NO_2 measured from space, J. Geophys. Res., 113, D04302, doi:10.1029/2007JD009021, 2008.

van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P. S., Morton, D. C., DeFries, R. S., Jin, Y., and van Leeuwen, T. T.: Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009), Atmos. Chem. Phys., 10, 11707–11735, doi:10.5194/acp-10-11707-2010, 2010.

Table 1. Summary of satellite-based NO_2 measurement instruments.

Instrument	GOME	SCIAMACHY	GOME-2	OMI
Satellite	ERS-2	ENVISAT	MetOp	Aura
Period	Apr. 1995–Jun. 2003	Mar. 2002–Apr. 2012	Oct. 2006	Jul. 2004
Overpass time	10:30 LT	10:00 LT	9:30 LT	13:45 LT
Nadir-view spatial resolution	40 × 320 km^2	30 × 60 km^2	13 × 24 km^2	40 × 80 km^2
Global coverage	3 days	6 days	1 day	1 day

ACPD
13, 11247–11268, 2013

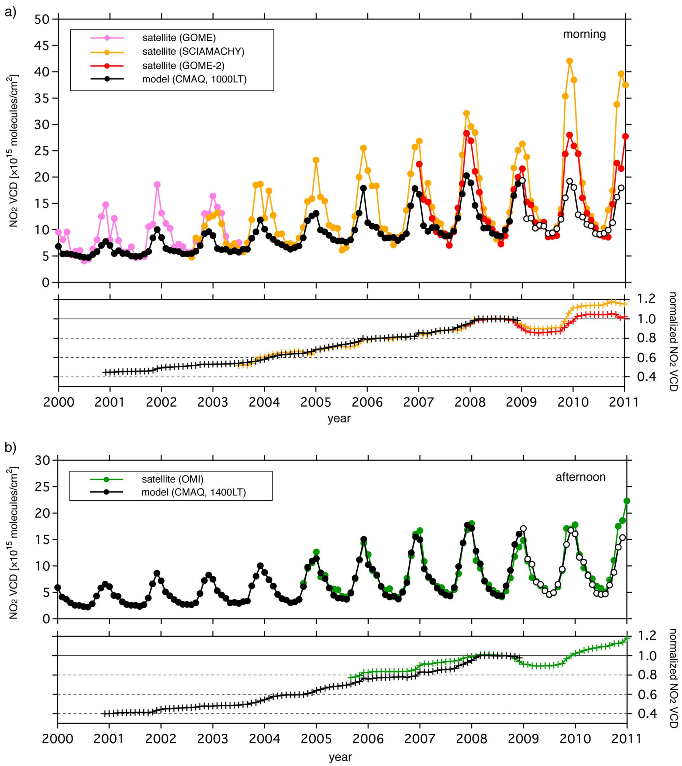

Fig. 1. Temporal variation in monthly mean tropospheric NO_2 VCD above CEC region during 2000–2010 from multi-satellite observations (GOME, SCIAMACHY, GOME-2, OMI) and model results (open circles denote sensitivity simulations with anthropogenic emissions fixed at 2008 values) in **(a)** the morning and **(b)** the afternoon. The normalized tropospheric NO_2 VCD relative to averaged values between October 2007 and September 2008 with a 12 month moving average are also shown at the bottom.

ACPD
13, 11247–11268, 2013

Trend analysis of tropospheric NO_2 column density over East Asia

S. Itahashi et al.

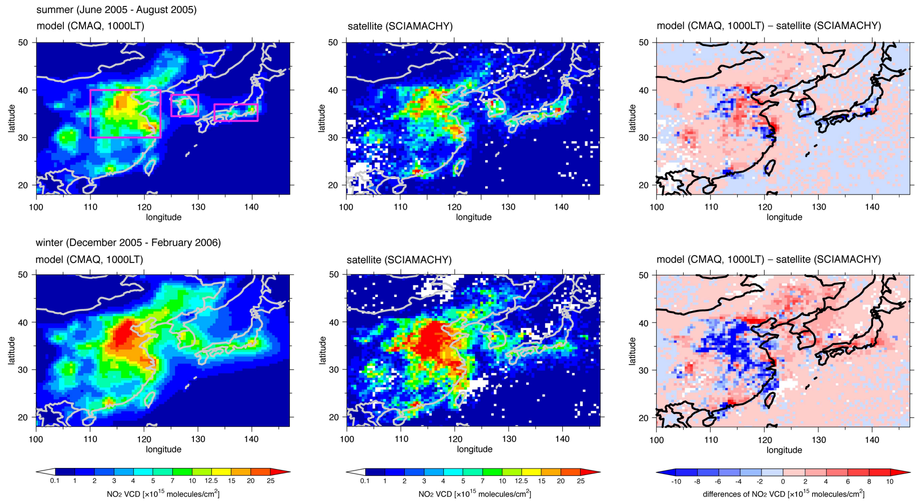

Fig. 2. Spatial distribution of tropospheric NO_2 VCD from (left) model results at 10:00 LT, (center) satellite observations by SCIAMACHY, and (right) differences between model and satellite results (top) in summer (averaged between June 2005 and August 2005) and (bottom) in winter (averaged between December 2005 and February 2006). The rectangular regions shown in the top-left figure are the investigation regions (CEC, Korea, and Japan).

ACPD
13, 11247–11268, 2013

Trend analysis of tropospheric NO_2 column density over East Asia

S. Itahashi et al.

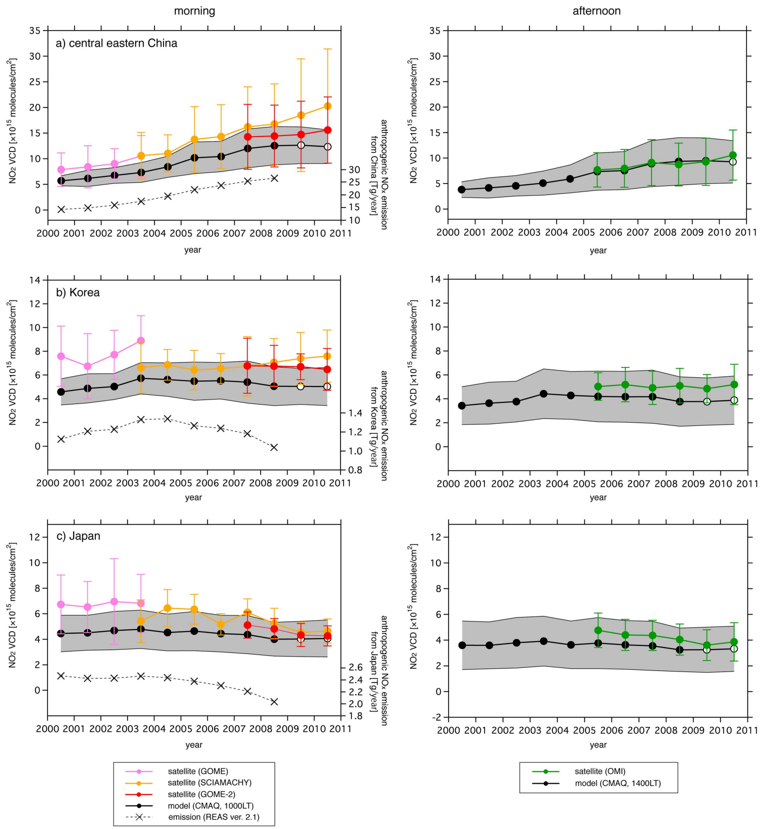

Fig. 3. Annual trend of tropospheric NO_2 VCD during 2000–2010 from the multi-satellite observations (GOME, SCIAMACHY, GOME-2, and OMI) and model results (open circles denote sensitivity simulations with anthropogenic emissions fixed at 2008 values) with 1σ standard deviation above **(a)** CEC, **(b)** Korea, and **(c)** Japan in (left) the morning and (right) the afternoon.

ACPD
13, 11247–11268, 2013

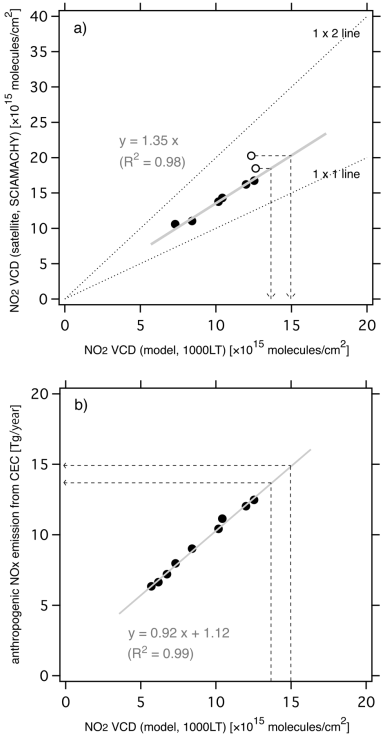

Fig. 4. Scatter diagram of **(a)** annual-mean modeled tropospheric NO_2 VCD at 10:00 LT, with annual-mean observed tropospheric NO_2 VCD by SCIAMACHY above CEC (open circles denote sensitivity simulations with anthropogenic emissions fixed at 2008 values), and **(b)** annual-mean modeled tropospheric NO_2 VCD at 10:00 LT above CEC with annual-based anthropogenic NO_x emissions from CEC.

ACPD
13, 11247–11268, 2013