Interannual and Seasonal Variations of CMAQ-simulated tropospheric NO_2 in Asia and comparison with GOME satellite data

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1. INTRODUCTION

Examination of long-term tropospheric NO_2 variation plays an important role in analysis of recent increases of NOx emissions over Asia. As the NO_2 lifetime is short and the effects of horizontal transport in the continental boundary layer are small, it is reasonable to discuss the relationship between NOx emission inventory and satellite NO_2 vertical column densities (VCDs). Richter et al. (2005) warned of the impact of rapid emission increases over China based on their Global Ozone Monitoring Experiment (GOME) satellite-derived NO_2 columns. They show that the trend of increase is approximately of the order of 7%·yr-1 from 1996 to 2002, implying an almost 40% increase within seven years. Quite similar results were also recently reported by a study including both GOME and SCanning Imaging Absorption spectrometer for Atmospheric CHartographY (SCIAMACHY) data in a statistical analysis by van der A et al. (2006). However, as discussed by Richter et al. (2005) and Noije et al. (2006), the GOME retrieval is very sensitive to several factors including cloud screening and other chemical/meteorological conditions.

Systematic comparison of satellite NO_2 VCDs and application of the chemical transport model (CTM) plays an important role for emission analysis to overcome such difficulties. Noije et al. (2006) presented a systematic comparison of NO_2 columns from 17 global CTMs and three state-of-the-art GOME retrievals for the year 2000. They report that, on average, the models underestimate the retrievals in industrial regions such as Europe, the eastern United States, and eastern China. They concluded that top-down estimations of NOx emissions from satellite retrieval are strongly dependent on the choice of model and retrieval.

The GOME retrieval (top-down approach) provides long-term data for almost 7 years (Jan. 1996 – June 2003) and CTM studies corresponding to that period with year-by-year emission estimates are absolutely necessary as a bottom-up analysis. GOME data can provide constrains for the inverse method of emission estimates (e.g., Martin et al., 2003). They also provide recent emission trends, but such a long-term CTM study has not yet been reported for Asia. As successful applications for Asian air quality studies, the community multi-scale air quality model (CMAQ; Byun and Ching, 1999) has been used intensively by Zhang et al. (2002), Uno et al. (2005), Tanimoto et al. (2005), and Yamaji et al. (2006). Here we report the results of a systematic analysis of seasonal and interannual variations of NO_2 VCDs based on GOME data and the regional scale CTM, CMAQ, and sensitivity experiments with the latest emission inventory in Asia from 1996 to 2003.

2. Outline of CMAQ simulation, Emission Inventory and GOME retrieval

In the following, we will briefly describe the regional chemical transport model, the emission inventory, the GOME NO_2 retrievals and the settings used in the numerical experiments in this paper.

(a) Chemical Transport Model, CMAQ

The three-dimensional regional-scale CTM used in this study was developed jointly by Kyushu University and the National Institute for Environmental Studies (Uno et al., 2005) based on the Models-3 CMAQ (ver. 4.4) modeling system released by the US EPA (Byun and Ching, 1999).

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Briefly, the model is driven by meteorological fields generated by the Regional Atmospheric Modeling System (RAMS) with initial and boundary conditions defined by NCEP reanalysis data (2.5° resolution and 6 h interval). The horizontal model domain for the CMAQ simulation is 6240 × 5440 km2 on a rotated polar stereographic map projection centered at 25°N, 115°E with 80 × 80 km2 grid resolution (see Fig. 1 of Tanimoto et al., 2005). For vertical resolution, 14 layers are used in the sigma-z coordinate system up to 23 km, with about seven layers within the boundary layer below 2 km. The SAPRC-99 scheme is applied for gas-phase chemistry, and the AERO3 module for aerosol calculation.

(b) REAS Emission Inventory

Reliable emission inventories of air pollutants are becoming increasingly important to assess heavy air pollution problems in Asia. An emission inventory in Asia was reported for the TRACE-P and ACE-Asia field study by Streets et al. (2003) with 1° × 1° resolution. Recently, the Regional Emission inventory in Asia (REAS; Ohara et al., 2006; Akimoto et al., 2006) was constructed based on energy data, emissions factors, and other socio-economic information between the years 1980 and 2003. It provides an Asian emission inventory for ten chemical species: NOx, SO_2, CO, CO_2, nitrous oxide (N_2O), NH_3, black carbon (BC), organic carbon (OC), methane (CH_4), and non-methane volatile organic compounds (NMVOC) from anthropogenic sources (combustion, non-combustion, agriculture, and others). The emissions estimated for district and country level were distributed into a 0.5° × 0.5° grid using index data bases of population, location of large point source (LPS), road networks, and land coverage information.

The NOx emission intensity (combustion base) of REAS version 1.1 for 2000 was estimated as 11.2 Tg-NO_2·yr-1for all of China (27.3 Tg-NO_2 for Asia). A similar number of 10.5 Tg-NO_2·yr-1 was reported from TRACE-P (Streets et al., 2003), and 13.8 Tg-NO_2·yr- from EDGAR ver. 3.2.

(c) GOME Tropospheric NO_2 Vertical Column Densities (VCDs)

GOME is a passive remote sensing instrument on board the ERS-2 satellite launched in April 1995. The GOME instrument observes the atmosphere at 10:30 local time (LT) and global coverage is achieved every 3 days with a footprint of 40 km latitude by 320 km longitude. For this study, we use the most recent version (ver. 2) of tropospheric NO_2 column data products retrieved by the University of Bremen (Richter et al., 2005). The retrieval version 2 data is based on 3D CTM, SLIMCAT data, to exclude the stratospheric NO_2 contribution, monthly AMF (air mass factor) evaluated with NO_2 profiles from a run of the global model MOZART-2 for 1997 and a surface reflectivity climatology data.

For this study, the GOME tropospheric NO_2 swath data (ver. 2) files giving the location and value for each measurement pixel are all interpolated into a 0.5° × 0.5° longitude-latitude map (as with the REAS grid resolution). The GOME tropospheric NO_2 data for the period of January 1996 – June 2003 are used in this study.

(d) Setting of numerical experiments by CMAQ/REAS

In this study, an eight-full-year simulation was conducted for 1996–2003. For this CMAQ modeling system, all emissions were obtained from 0.5° × 0.5° resolution of the REAS ver. 1.1 database. Emission intensity for the CMAQ is set as constant for each year and no seasonal variation is assumed. The initial fields and monthly averaged lateral boundary condition for most chemical tracers are provided from a global chemical transport model (CHASER; Sudo et al., 2002). This fixed lateral boundary condition is used for the eight-full-year simulation (i.e., no interannual variation of lateral conditions is assumed). The CMAQ output data are all interpolated to 0.5° × 0.5° resolution of REAS to facilitate an easy comparison.

Two sets of numerical experiments were conducted. Series E00Myy simulations used the fixed emission for 2000 with year-by-year meteorology. Series EyyMyy use both year-by-year emissions and meteorology. These two experiments were set to elucidate the sensitivity for both meteorology and changes in emission intensity. The GOME measurements in low latitudes and middle latitudes are always taken at the same LT (approximately 10:30LT). Therefore, we used the CMAQ output of 3UTC (11:00 LT for China and 12:00LT for Japan) for comparison.

3. RESULTS AND CONCLUSIONS

Systematic analyses of interannual and seasonal variations of tropospheric NO_2 vertical column densities (VCDs) based on GOME satellite data and the regional scale CTM, CMAQ, were presented over East Asia for the time period from January 1996 to June 2003. Numerical simulations with a year-by-year base of the REAS emission inventory in Asia during the same period were analyzed (details can be found in Uno et al., 2006).

The main results are:

1) The horizontal distribution of annual averaged GOME NO_2 VCDs for 2000 generally agrees with CMAQ/REAS results. However, CMAQ results underestimate GOME retrievals by factors of 2–4 over polluted industrial regions such as Central East China (CEC), the major part of Korea, Hong-Kong, and central and western areas of Japan. Examination of differences of GOME and CMAQ also suggested that the emission inventory of some regions (e.g., Taiwan, two large city region of Korea and northeastern China) demand re-examination.

2) Evolution of the tropospheric columns of NO_2 above Japan and CEC between 1996 and 2003 was examined. For the Japan region, GOME retrieval and CMAQ NO_2 show a good agreement and no clear increasing trend, which is consistent with the REAS emission inventory for Japan. For CEC, the general agreement between GOME and CMAQ is also good. Both GOME and CMAQ NO_2 show a very sharp increasing trend after 2000. The seasonal cycle of NO_2 VCDs from both CMAQ and GOME is asymmetric because of the summer monsoon exchange from the Pacific Ocean side. We also found that GOME retrievals during February–April and September–November have systematically larger dips (concave shape) than CMAQ, even considering their error bar.

3) A sensitivity experiment with a fixed emission rate for year 2000 shows that detection of emission trends over CEC from satellite data in fall or winter result in larger errors because of the variability of meteorology. Examination during summer and annual averaged NO_2 VCDs is much less sensitive to variability of meteorology and suitability of analysis of emission trends, even though it still includes 3–7% of the variability coming from meteorological variability.

4) Recent trends of annual emission increases in CEC were examined. Increasing trends of 1996–1998 and 2000–2002 for GOME and CMAQ/REAS shows a good agreement, but the increasing rate of the GOME data is approximately 10–11%·yr-1 after 2000, slightly steeper than CMAQ/REAS (8–9%·yr-1). The greatest difference was found between the years 1998 and 2000. The CMAQ/REAS shows only a few percentage points of increase, while GOME gives more than 8%·yr-1 of increase. The exact reason remains unclear, but the most likely explanation is that the REAS emission trend (based on the Chinese statistics) underestimates the rapid growth of emissions during this time period.

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