Satellite-observed U.S. power plant NO_x emission reductions and their impact on air quality

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[1] Nitrogen oxide (NO_x) emissions resulting from fossil fuel combustion lead to unhealthy levels of near-surface ozone (O_3). One of the largest U.S. sources, electric power generation, represented about 25% of the U.S. anthropogenic NO_x emissions in 1999. Here we show that space-based instruments observed declining regional NO_x levels between 1999 and 2005 in response to the recent implementation of pollution controls by utility companies in the eastern U.S. Satellite-retrieved summertime nitrogen dioxide (NO_2) columns and bottom-up emission estimates show larger decreases in the Ohio River Valley, where power plants dominate NO_x emissions, than in the northeast U.S. urban corridor. Model simulations predict lower O_3 across much of the eastern U.S. in response to these emission reductions. **Citation:** Kim, S.-W., A. Heckel, S. A. McKeen, G. J. Frost, E.-Y. Hsie, M. K. Trainer, A. Richter, J. P. Burrows, S. E. Peckham, and G. A. Grell (2006), Satellite-observed U.S. power plant NO_x emission reductions and their impact on air quality, *Geophys. Res. Lett.*, *33*, L22812, doi:10.1029/2006GL027749.

1. Introduction

[2] In the lower troposphere, high ozone (O_3) mixing ratios are a key component of photochemical smog that frequently occurs during summertime under high pressure systems, near or downwind of industrial and urban areas. Elevated levels of O_3 are hazardous to human health and vegetation and are a primary focus of pollution control strategies. Nitric oxide (NO) and nitrogen dioxide (NO_2), which together are known as NO_x, play a crucial role in producing O_3 *via* photochemistry that is further controlled by the amount of volatile organic compounds (VOC), carbon monoxide, sunlight, and temperature [e.g., *Crutzen*, 1979; *Haagen-Smit*, 1952; *McKeen et al.*, 1991; *Ryerson et al.*, 2001].

[3] In the eastern United States (U.S.), summertime O_3 often exceeds the U.S. Environment Protection Agency (EPA) 8-hour standard [*Environmental Protection Agency* (*EPA*), 2005a, 2005b]. O_3 exceedances in the northeast U.S. have been associated with NO_x emissions from point sources, especially coal-burning power plants, in the Ohio River Valley (EPA, Ozone transport assessment group, final report, vol. I: Executive summary, 1997, available at http://capita.wustl.edu/otag). As a result, numerous programs to control point source NO_x emissions in the eastern U.S. have been implemented [*EPA*, 2005a]. In particular, the recent control program, the 1998 NO_x State Implementation Plans Call, focused on the summertime reduction of power industry NO_x emissions to limit ozone formation [*EPA*, 2005b]. With the aid of these programs, 2004 point source NO_x emissions across the eastern U.S. dropped by about 45% from 1990 levels during a period when utility companies actually increased the amount of electric power generated [*EPA*, 2005a]. These assessments of reductions in power plant NO_x emissions are based primarily on Continuous Emission Monitoring System (CEMS) observations collected at each plant. Power plants in 1999 accounted for a quarter of the U.S. total NO_x emissions, which are dominated by mobile sources [*Frost et al.*, 2006]. Regional changes in both NO_x and VOC emissions will determine the trend in O_3 across the eastern U.S. However, U.S. NO_x emission trends have not yet been investigated with observations on a regional scale.

[4] The main goal of this study is to assess the change of regional NO_x emissions in the eastern U.S. by comparing NO_2 columns derived from the SCIAMACHY (Scanning Imaging Absorption spectroMeter for Atmospheric CHartographY) satellite instrument [*Bovensman et al.*, 1999] with three-dimensional regional scale chemical transport model results that include the estimated NO_x emission changes. Satellite observations have been widely used to estimate global scale NO_x emissions and their trends [e.g., *Martin et al.*, 2003; *Richter et al.*, 2005]. This study is the first in which satellite measurements are utilized to investigate the regional NO_x emission changes in the U.S. The air quality benefits from these NO_x emission changes are explored with a chemical-transport model.

2. Method

2.1. Satellite Measurements

[5] Measurements of tropospheric NO_2 from space have been available since GOME (Global Ozone Monitoring Experiment) [*Burrows et al.*, 1999] and SCIAMACHY became operational. The ultraviolet/visible nadir measurements of SCIAMACHY are similar to those from GOME, with the main difference being the improved spatial resolution (320×40 km^2 vs. 60×30 km^2) [*Richter et al.*, 2005]. The finer resolution of SCIAMACHY relative to GOME is necessary to resolve regional scale NO_2 distributions. Because of their availability and similar horizontal resolu-

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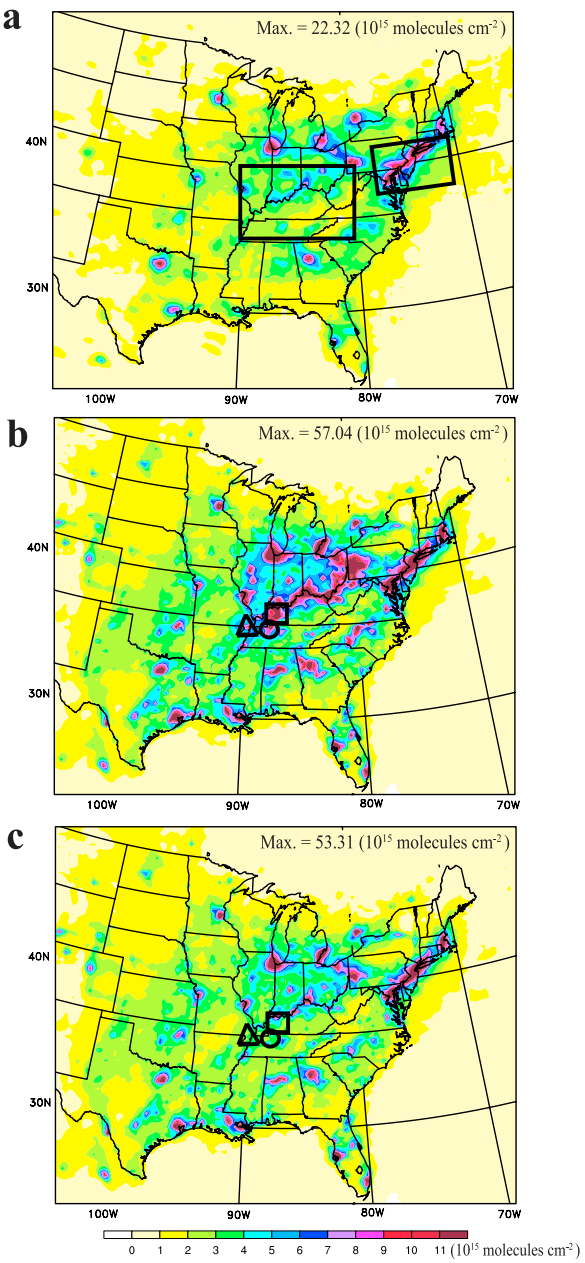

Figure 1. Spatial distribution of NO_2 columns time-averaged over June–August 2004 from (a) SCIAMACHY satellite observations, (b) WRF-Chem reference emission case runs, and (c) WRF-Chem updated emission case runs. Two rectangular boxes in Figure 1a represent the Ohio River Valley and the northeast U.S. urban corridor. Box, circle, and triangle symbols in Figures 1b and 1c highlight Paradise, Cumberland, and New Madrid power plants, respectively.

tion to the model simulations, we use SCIAMACHY measurements in comparisons with WRF-Chem simulations for 2004. However, to get the long term evolution of NO_2 columns between 1997 and 2005, both GOME and SCIAMACHY data are utilized. In previous studies, output from a global chemical transport model, MOZART-2 [*Horowitz et al.*, 2003], has been used as *a priori* information for both GOME and SCIAMACHY measurements [*Richter et al.*, 2005]. In this study, we utilize our chemical transport model to retrieve the SCIAMACHY observations, since our simulations are limited to the eastern U.S. and have much finer resolution (27 × 27 km^2) than the approximately 2° × 2° resolution of the global model.

2.2. Chemistry-Transport Model

[6] The Weather Research and Forecasting Chemistry (WRF-Chem) model adopted in this study is an extension of the earlier MM5-Chem regional scale chemical transport model to the WRF architecture [*Grell et al.*, 2005]. WRF-Chem retrospective simulations consist of 24-hour forecasts starting at 0000 UTC each day between 1 April and 31 October 2004. Two WRF-Chem retrospective cases are performed. The reference emission case uses the EPA 1999 National Emission Inventory version 3 (NEI99), which was released in December 2003 and was, until April 2006, the latest fully vetted and complete EPA inventory for air quality modeling. In the updated emission case NEI99 NO_x and SO_2 emissions for about 1000 power plants were updated using 2004 month-specific CEMS data [*Frost et al.*, 2006].

3. Results

[7] The satellite and the model spatial distributions of NO_2 columns time-averaged over summer 2004 (June–August) are shown in Figure 1. For this comparison, the model data are sampled by following daily satellite tracks under clear sky conditions. 2004 satellite-retrieved NO_2 columns show maxima in areas with mixed urban, industrial, and power plant sources, such as the northeast U.S. urban corridor, Chicago, Pittsburgh, Detroit, Toronto, Atlanta, Dallas, and Houston (Figure 1a). WRF-Chem simulated NO_2 columns using the NEI99 emissions show a similar distribution pattern to the satellite data in many urban areas (Figure 1b). However, reference case model NO_2 columns in the Ohio River Valley (larger box in Figure 1a) are much larger than those observed by the satellite. WRF-Chem model simulations with power plant NO_x emissions updated to 2004 levels (Figure 1c) agree more closely with satellite data in the Ohio River Valley. The slopes (spatial correlation coefficients) from the least squares linear fit between all model and satellite data shown in Figure 1 are 1.48 (0.84) and 1.15 (0.85) for the reference and the updated emission cases, respectively. The 6-state Ohio River region (IL, IN, OH, PA, KY and WV) produces about one-third of national power plant NO_x emissions. CEMS data show that emissions from this region in 2004 were reduced to 50% of 1999 levels by pollution control programs [*Frost et al.*, 2006]. Particularly striking is the change in NO_2 plumes from the largest power plants, such as Paradise, Cumberland, and New Madrid (symbols in Figures 1b and 1c), between the WRF-Chem reference and updated emission cases. Because the NEI99 does not capture the impact of recent power plant NO_x controls, there are large discrepancies in the Ohio River Valley between SCIAMACHY and the WRF-Chem reference case.

[8] Two boxes in Figure 1a delineate power plant (Ohio River Valley) and motor vehicle (northeast U.S. urban corridor) dominated NO_x source areas. Figure 2 shows the 1997–2005 trends in summertime (June–August) mean

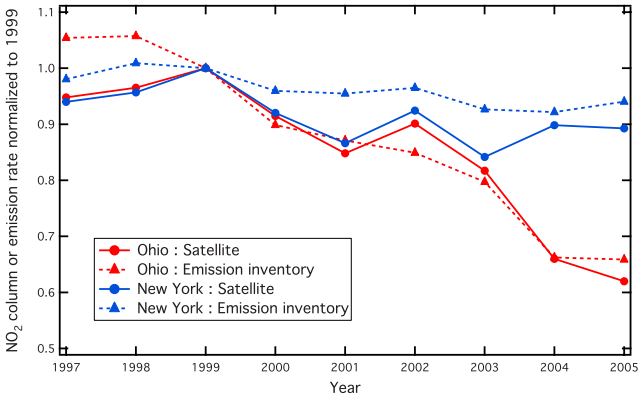

Figure 2. The trends in summertime (June–August) mean NO_2 columns from the GOME and SCIAMACHY satellites and the bottom-up NO_x emission rates in the Ohio River Valley and the northeast U.S. urban corridor during 1997–2005. SCIAMACHY data are used for 2003–2005, while GOME data are utilized for the earlier period. Data are normalized to 1999 values.

NO_2 columns normalized to the 1999 value retrieved from GOME and SCIAMACHY satellite observations in these two representative regions. Also included in Figure 2 are the trends in the bottom-up NO_x emissions normalized to the 1999 value, derived from CEMS reports time-averaged for these same months in each year and from an assumption that all other NO_x sources remained constant at NEI99 summer levels throughout this period. While there is debate about trends in mobile source emissions over the past decade, recent studies [*Harley et al.*, 2005; *Parrish*, 2006] indicate that mobile NO_x emissions have not changed as drastically as emissions from power plants. These findings are supported by the northeast corridor satellite NO_2 columns which, like the NO_x emission inventory, show no clear trend. In contrast, by 2005, both Ohio River Valley NO_2 columns and NO_x emissions decreased by 38% and 34% of 1999 levels, respectively. NO_x emission reductions in a region dominated by power plants that have implemented controls over the past decade are thus clearly evident in the satellite data.

[9] The impact of power plant emission controls is also seen in the seasonal trends in NO_2 columns from the WRF-Chem simulations and the satellite observations, which are compared quantitatively (14 day running means) during April–October 2004 in the two regions (Figure 3). The satellite observations show generally lower NO_2 columns everywhere in summer compared with those in spring and fall, which the model attributes to decreased NO_2 lifetimes in response to seasonal increases in solar actinic flux. In addition to this natural cycle, we also expect seasonal emission fluctuations in some regions since recent point source NO_x control programs [*EPA*, 2005b] specifically target summer NO_x emissions. In the Ohio River Valley, discrepancies in NO_2 columns between the WRF-Chem reference case and the satellite observations are large during summer and fall after the onset of power plant emission controls. The model simulation with the updated emission inventory shows much better agreement with satellite measurements throughout the entire period April–October. The average correlation coefficient between the model and the satellite NO_2 column time series in this region improves noticeably from 0.59 to 0.70 when using the updated power plant emissions. The satellite measurements capture the model-predicted seasonal changes of NO_2 columns due both to the natural photochemical cycle and to the summertime use of power plant pollution controls. As expected for a region dominated by mobile source emissions, there is little difference between model NO_2 columns in the northeast corridor using either 1999 or 2004 power plant emissions. Using either emission inventory, WRF-Chem simulated NO_2 columns in this region agree well with satellite observations in the summertime, with average correlation coefficients of 0.83 for both emission cases.

[10] Finally, the impact of power plant NO_x emission controls in the eastern U.S. on the modeled distribution of boundary layer NO_2 and O_3 is examined. Figure 4 demonstrates the differences in WRF-Chem NO_2 and O_3 between the updated and the reference emission cases at 20 UTC (1500 EST) averaged vertically from the surface to 1 km level and time-averaged for all days June–August 2004. The well-mixed planetary boundary layer generally extends to at least 1 km altitude during mid-afternoon over continental areas in the model domain. The most significant boundary layer NO_2 reductions occur in the vicinity of

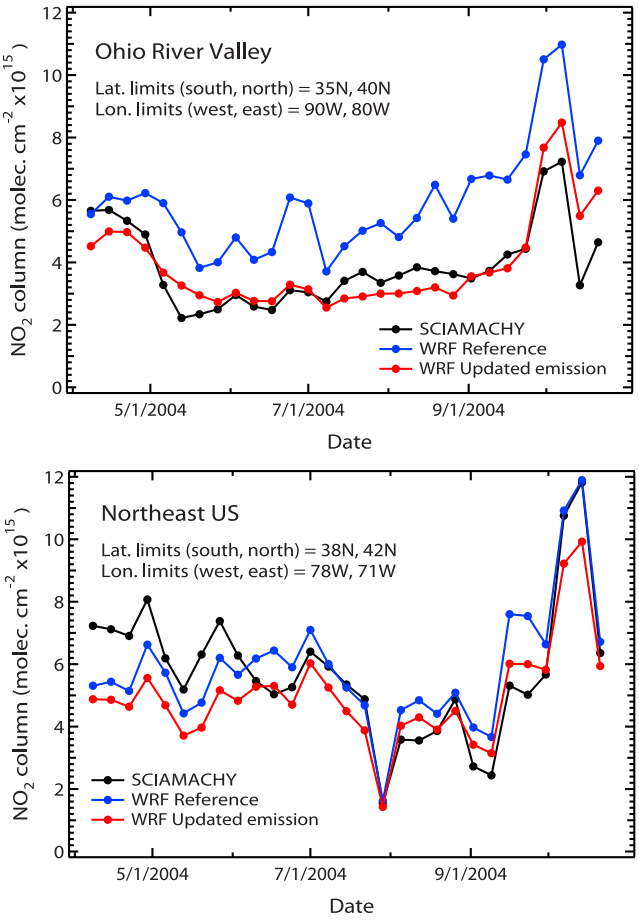

Figure 3. The time evolution of 14 day running means of WRF-Chem modelled and SCIAMACHY observed NO_2 columns in the Ohio River Valley and the northeast U.S. urban corridor during April–October 2004. The model data are sampled by following daily satellite tracks under clear sky conditions.

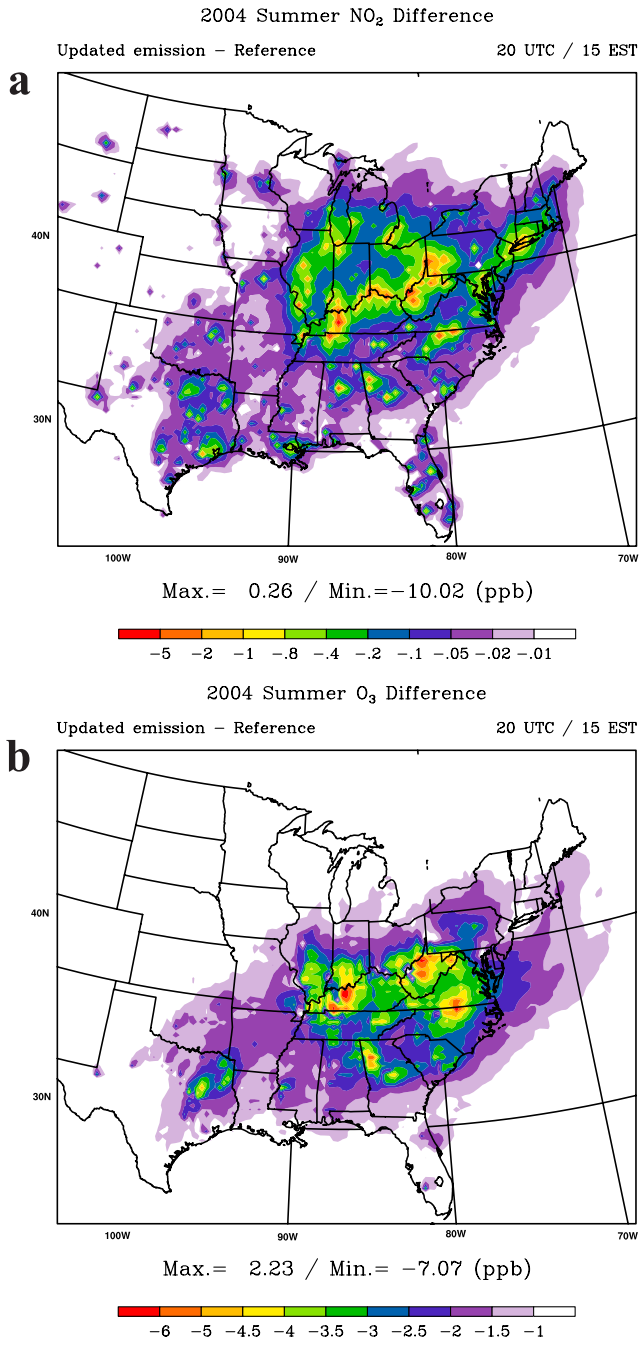

Figure 4. The differences between the updated and the reference emission cases of modelled (a) NO_2 and (b) O_3 averaged vertically from the surface to 1 km and time-averaged using all simulation data at 20 UTC (1500 EST) during June–August 2004. A nonlinear color scale has been used because of the large horizontal variability of NO_2 and O_3 differences.

power plants in the Ohio River Valley, North Carolina, Virginia, Georgia, Alabama, the New York City area, Texas and Florida. The maximum NO_2 decrease in the mid-afternoon summertime average is large, about 10 ppbv (parts per billion by volume) from a 12 ppbv NO_2 mixing ratio in the reference case. O_3 changes have a different spatial pattern than those in NO_2, because both transport and photochemistry involving VOC as well as NO_2 play important roles in the formation of O_3. The modeled O_3 generally decreases in response to the NO_x emission reductions. The largest O_3 reductions occur in the Ohio River Valley, North Carolina, Virginia, Georgia, and Texas. The maximum O_3 decrease seen in Figure 4 is about 7 ppbv out of 72 ppbv O_3 in the reference case. The variability in the O_3 response to comparable NO_2 decreases (for example, in eastern Texas) illustrates the strong dependence of O_3 on VOC emissions as well as those of NO_x. The lack of a strong O_3 response to NO_x changes in the northern U.S. states will require additional study.

4. Conclusions

[11] Control of NO_x pollution from coal-burning power plants has had a measurable impact on air quality in the eastern U.S. The satellite observations clearly detect both year-to-year and summertime NO_2 column decreases in regions impacted by power plants that have implemented NO_x controls over the past decade. In response to the power plant NO_x emission reductions, the model predicts about 4–10% decreases of O_3 across the Ohio River Valley, Virginia and North Carolina, and smaller O_3 changes in the northeast U.S. during the summer of 2004. O_3 decreases of 9–16% over the period 1990–2004 have been detected by EPA surface monitors throughout the eastern U.S. (EPA, Air trends Web site, 2006, http://www.epa.gov/airtrends/ozone.html). The O_3 monitor trends are complicated by annual differences in meteorological conditions and changes in NO_x and VOC emissions from a variety of sources besides power plants. Air quality model simulations over a wider time period than the current study will ultimately be needed to understand the cause of the trends in the surface O_3 observations. While air quality models have played a crucial role in determining environmental policy regarding O_3, model errors such as positive O_3 biases [*McKeen et al.*, 2005] may complicate the interpretation of the model results.

[12] While the satellite observations presented here demonstrate the recent decline in NO_x emissions from power generation, they also indicate that comparable decreases in NO_x emitted by mobile sources have either not occurred or are not being detected. Assuming the former inference is true, it is therefore likely that power generation now accounts for less than the 25% contribution it made to total U.S. anthropogenic NO_x emissions in 1999. These findings suggest that further substantive reductions in eastern U.S. tropospheric NO_x levels will require decreases in mobile source NO_x emissions.

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