Satellite detection and model verification of NO\textsubscript{x} emissions from power plants in Northern China

Siwen Wang\textsuperscript{1,2}, David G Streets\textsuperscript{2,5}, Qiang Zhang\textsuperscript{3}, Kebin He\textsuperscript{1}, Dan Chen\textsuperscript{4}, Sicong Kang\textsuperscript{1}, Zifeng Lu\textsuperscript{2} and Yuxuan Wang\textsuperscript{1}

\textsuperscript{1} State Key Joint Laboratory of Environment Simulation and Pollution Control, Department of Environmental Science and Engineering, Tsinghua University, Beijing 100084, People’s Republic of China

\textsuperscript{2} Decision and Information Sciences Division, Argonne National Laboratory, Argonne, IL 60439, USA

\textsuperscript{3} Center for Earth System Science, Tsinghua University, Beijing 100084, People’s Republic of China

\textsuperscript{4} Department of Atmospheric and Oceanic Sciences, University of California at Los Angeles, CA 90095, USA

E-mail: dstreets@anl.gov

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Abstract

We evaluate the recently increasing tropospheric NO\textsubscript{2} columns in Northern China measured by the Ozone Monitoring Instrument (OMI) with an advanced power-plant NO\textsubscript{x} emission inventory and the NASA INTEX-B emission inventory, using a global chemical transport model (GEOS-Chem). In areas with newly built power plants the modeled and OMI-retrieved summertime average tropospheric NO\textsubscript{2} columns increased by 55% and 47%, respectively, between 2005 and 2007. A monthly average increase of 1.79 Gg NO\textsubscript{x} emissions is calculated to lead to an increase of 1.0 × 10\textsuperscript{15} molecules cm\textsuperscript{-2} in the modeled NO\textsubscript{2} columns in the study areas. Good consistency (R\textsuperscript{2} = 0.61, slope = 1.18, n = 14) between the increased modeled and OMI-retrieved summertime average NO\textsubscript{2} columns is found. These results suggest that NO\textsubscript{x} emissions from large power plants in Northern China can be identified and quantified using OMI retrievals with confidence. The NASA INTEX-B emission inventory appears to underestimate the NO\textsubscript{x} emissions from the industry and transportation sectors, making it more difficult to quantify power-plant emissions when they are co-located with large cities.

Keywords: China, NO\textsubscript{x}, emissions, GEOS-Chem, OMI

1. Introduction

Emissions of nitrogen oxides (NO\textsubscript{x} = NO\textsubscript{2} + NO) have been increasing dramatically during the past decade in China (Richter \textit{et al} 2005, Zhang \textit{et al} 2007), due to the rapidly growing economy powered by the generation of energy from fossil fuels. Since 2005 hundreds of large new electricity generating units have been constructed in China, and the total capacity of coal-fired power plants has increased by 42%, from 391 GW in 2005 to 556 GW in 2007. Under increasing pressure from acid deposition and summer photochemical smog, which are related to tropospheric NO\textsubscript{2}, NO\textsubscript{x} emission reduction has become an urgent target of legislation and government policies in China.

The magnitude and distribution of NO\textsubscript{x} emissions can change dramatically even in a couple of years in China, as a result of the booming construction activities as well as the application of emission control measures in various
sectors. The Ozone Monitoring Instrument (OMI) onboard NASA’s Aura satellite platform has proven to be an effective tool to detect NO$_2$ in the atmosphere at high temporal and spatial resolution and thus to investigate and improve NO$_x$ emissions (Boersma et al. 2008, Kim et al. 2009, Lin et al. 2010). Recently, Zhang et al. (2009a) reported increased OMI-retrieved summertime tropospheric NO$_2$ columns in response to power-plant construction that had occurred between 2005 and 2007 in Inner Mongolia, China, showing the potential for large point-source identification from the OMI retrievals. Focused on the same area, Li et al. (2010) recently analyzed OMI PBL SO$_2$ columns to identify the reduction of SO$_2$ emissions from the same power plants, due to the installation and effective operation of flue-gas desulfurization (FGD) systems. Both of these analyses compared the OMI column retrievals with emissions. The drawback to this approach is that the relationship between emissions and columns can be compromised by pollutant inflow and outflow from the region, as well as by other factors like meteorology and chemical transformation. Although Zhang et al. (2009a) and Li et al. (2010) found distinct signals of increased NO$_2$ or SO$_2$ columns associated with the newly built power plants, they were not able to give quantitative relationships between emissions and satellite retrievals, which would need a chemical transport model to account for the changes in chemistry, transport, and meteorology. In this letter we improve on the previous analytical methodologies by using the NO$_x$ point-source emission estimates to calculate NO$_2$ columns with a chemical transport model and then comparing modeled and measured columns.

To investigate the change of NO$_x$ emissions in Northern China and evaluate the impact of newly built power plants on local NO$_2$ concentrations, tropospheric NO$_2$ columns are simulated with GEOS-Chem, a global chemical transport model (CTM), for the summers (June, July, and August) of 2005 and 2007. The modeled columns are then compared with OMI-retrieved tropospheric NO$_2$ columns. This study is the first in which the modeled NO$_2$ columns from individual point sources are evaluated by comparison with satellite measurements in China.

2. Methodology

2.1. Anthropogenic NO$_x$ emissions

The 2006 NASA INTEX-B emission inventory (Zhang et al. 2009b) is adopted as a baseline emission inventory for East Asia in GEOS-Chem. This inventory has been previously validated by several model simulation studies against satellite measurements (Zhang et al. 2008) and in situ observations from the INTEX-B field campaign (van Donkelaar et al. 2008). However, due to a lack of specific information at that time about point sources in China, the INTEX-B inventory only includes individual electricity generating units ≥300 MW; smaller units are treated as area sources and distributed by socioeconomic activity data at the province level. For this work we have acquired a new, more detailed power-plant NO$_x$ emission inventory, which is based on a Chinese power sector database for 31 provinces supported by CMEP (Chinese Ministry of Environmental Protection). This database contains more than 5700 generating units of all sizes, with detailed information about boiler type, technology, geographical location, coal consumption per unit electricity supply, and the month the unit came into operation.

Annual NO$_x$ emissions are calculated by unit according to the technology and operation information, following the equation of Zhao et al. (2008):

$$E_i = \sum \sum \sum 1.4 \times U_{i,j} \times T_{i,j} \times F_{i,j} \times EF_{k,m} \times 10^{-6} \quad (1)$$

where $i, j, k, m$ stand for province, generator unit, boiler size, and emission control technology; 1.4 is the mass scaling factor from standard coal to raw coal; $E$ is the annual NO$_x$ emissions (Mg); $U$ is the unit size (MW); $T$ is the annual operation hours for unit $j$ in province $i$; $F$ is the specific coal consumption per unit electricity supply (gce kWh$^{-1}$); and EF is the emission factor. NO$_x$ emission factors for different types of units vary between 5.6 g kg$^{-1}$ coal burned and 10.5 g kg$^{-1}$ based on boiler size and the presence or absence of low-NO$_x$ burners (LNB) (Zhang et al. 2007). Using this method we calculate total NO$_x$ emissions of 8.11 Tg and 9.58 Tg for the power sector of China in 2005 and 2007, respectively. Our values are somewhat higher than the value of 6.97 Tg for 2005 obtained by Zhao et al. (2008), mostly because of differences in emission factors. NO$_x$ emissions for all other sectors for 2006 come from the INTEX-B emission inventory (v1.2, http://mic.greenresource.cn/intex-b2006). Using industrial value-added data from National Bureau of Statistics (2009) and total vehicle population data from National Bureau of Statistics (2010), scale factors of 1.32 and 1.20 for industry and 1.17 and 1.18 for transportation are established to extrapolate the NO$_x$ emissions of 2006 backward to 2005 and forward to 2007, respectively. Monthly profiles of NO$_x$ emissions in the power sector are derived from monthly electricity generation data from CMEP weighted by capacity for each province, taking into account unit operation profiles. For other sectors monthly profiles provided by Zhang et al. (2007) are adopted.

2.2. Chemical transport model

GEOS-Chem is a global, three-dimensional CTM driven by assimilated meteorological fields from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office (GMAO). In this study we use the nested-grid GEOS-Chem model (version 8-02-01, http://acmg.seas.harvard.edu/geos/) with horizontal resolution at 0.5° × 0.6667° driven by GEOS-5 meteorological fields over Southeast Asia (70°E–150°E, 11°S–55°N). This nested-grid GEOS-Chem model has already been tested in China applications and has proven to be capable of simulating individual cities with high associated emission intensities (Chen et al. 2009) and interpreting the reduction of ozone during the 2008 Beijing Olympics (Wang et al. 2009). The GEOS-5 data, which are provided every 6 h (3 h for surface fields and mixing depths), have 47 vertical hybrid eta levels, extending from the surface to 0.01 hPa with ~137 m for each of the lowest ten layers. The power-plant NO$_x$ emissions combined with NO$_x$ emissions for
Figure 1. Modeled summertime (JJA) average tropospheric NO$_2$ columns in three selected regions in Northern China for 2005 (left), 2007 (center), and ratio of NO$_2$ columns between 2007 and 2005 (right). From top down: East, Central, and West regions. Solid circles indicate the fifteen cities and towns studied. Solid triangles denote the locations of newly built power plants, with three categories from small to large: <800 MW, 800–1200 MW, >1200 MW.

other sectors from the INTEX-B emission inventory are used as the \textit{a priori} inventory to conduct tropospheric ozone–NO$_x$–hydrocarbon simulations for 2005 and 2007.

The current version of the GEOS-Chem model supports multiple-layer inputs for anthropogenic NO$_x$ emissions, which is critical for high-stack emitters such as power plants and large industrial sources. For power plants, we input the NO$_x$ emitted by generator units $\leq$100 MW into the second vertical layer (mid-altitude at $\sim$205 m) and by units $>$100 MW into the third vertical layer (mid-altitude at $\sim$335 m). For all other anthropogenic sources NO$_x$ emissions are input as surface emissions in the first level. The detailed description of default NO$_x$ emissions from soil, lightning, biomass burning, and aircraft adopted in the model can be found in Martin \textit{et al} (2002). The chemical time step in the model is 30 min. A monthly varying tropopause height is used to derive the tropospheric NO$_2$ columns. More than six months pre-simulation is conducted to obtain the initial concentration fields for each year. Finally, daily modeled tropospheric NO$_2$ columns are averaged at the local time of 13:00–15:00. To obtain average modeled NO$_2$ columns coincident with the satellite measurements, modeled columns are re-gridded to 0.25° × 0.25° horizontal resolution and sampled by following the same daily satellite scenes used in the average OMI columns.

2.3. Satellite measurements

The Dutch–Finnish OMI on board NASA's EOS Aura satellite is a nadir-viewing imaging spectrograph measuring direct and atmosphere-backscattered sunlight in the ultraviolet–visible range from 264 to 504 nm (Levelt \textit{et al} 2006). The EOS Aura spacecraft was launched on July 15, 2004, and circulates in a 98.2° inclination, sun-synchronous polar orbit at 705 km altitude, with local overpass time at $\sim$13:45. As one benefit of the two two-dimensional CCD detectors and the wide field of view (114°), OMI measures the complete spectrum with a fine ground footprint ($13 \times 24$ km$^2$ at nadir) along track and daily
global coverage. For this study, we choose the OMI standard NO2 Level-2G product (version 003) at NASA Goddard Earth Sciences Data and Information Services Center. We exclude scenes where the cloud fractions are greater than 30%, as the NASA science team did. The scenes with cloud-top pressures higher than 800 hPa in cloudy scenes are also rejected, because low clouds can give rise to inaccuracy if they intersect the NO2 air mass (Mijling et al 2009). Scenes on the edges of each swath and anomalous rows reported by NASA are also filtered out. For the summertime average, we always have one-third of the grids left with usable data after application of the filter to individual OMI daily retrievals. Finally, the NO2 columns are averaged at 0.25° × 0.25° horizontal resolution for the summers of 2005 and 2007.

3. Results and discussion

Three regions in Northern China are selected for study, referred to as East, Central, and West, which contain 42 newly built electricity generating units, with a combined capacity of 16.3 GW, installed in 18 power plants in Inner Mongolia, Ningxia, Shanxi, and Shaanxi provinces. Six of these power plants are equipped with new units ≥1000 MW. The modeled and OMI-retrieved summertime average tropospheric NO2 columns are shown in figures 1 and 2, respectively. Both figures illustrate the dense NO2 pollution in populated cities and in areas with large power plants. Ratios of summertime NO2 columns between 2005 and 2007 (right column in each figure) are well consistent and reflect significant increases in emissions in areas with new power plants that came into operation in this period. Large power plants typically have high impact on NO2 columns in a region of ~100 km around them, as can be seen in figures 1 and 2.

We assemble all of these new power-plant units into fifteen sub-regions and categorize them into three groups, as shown in table 1: city with newly built power plants (designated as Group A), city without newly built power plants (Group B), and rural area with newly built power plants (Group C), following Zhang et al (2009a) and Li et al (2010). The size of each sub-region is 0.5° × 0.5°, which is close to the original model resolution and large enough to allow for significant
Table 1. NO\textsubscript{x} emissions and NO\textsubscript{2} columns from GEOS-Chem (CTM) and OMI in summers of 2005 and 2007.

| Group\textsuperscript{a} | Region | Location\textsuperscript{b} | NO\textsubscript{x} emissions\textsuperscript{c} (Gg/month) | NO\textsubscript{2} columns (10\textsuperscript{15} molecules cm\textsuperscript{-2}) |
|-------------------------|--------|-----------------------------|------------------------|----------------------------------|
|                         |        |                             | CTM 2005 2007 OMI 2005 2007 CTM 2005 2007 OMI 2005 2007 |
| A Baotou [40.25N, 109.75E] | 4.92(0.63) 9.05(0.71) | 4.25 6.33 5.78 9.11 |
| Huhehot [40.50N, 111.50E] | 3.99(0.56) 5.49(0.54) | 2.34 3.30 4.51 6.18 |
| Wuhai [39.25N, 106.50E] | 6.03(0.91) 7.57(0.90) | 4.44 5.91 5.04 7.41 |
| Taiyuan [37.75N, 112.00E] | 0.75(0.28) 1.84(0.57) | 1.92 2.64 8.67 12.32 |
| B Datong [39.75N, 113.00E] | 2.54(0.58) 3.17(0.51) | 2.71 3.02 9.82 9.90 |
| Shuozhou [39.00N, 112.25E] | 2.34(0.84) 1.24(0.04) | 1.68 2.04 7.03 7.30 |
| Zhangjiakou [40.50N, 114.75E] | 0.87(0.06) 1.71(0.46) | 2.11 2.47 3.87 4.05 |
| Chengde [40.75N, 117.75E] | 1.70(0.62) 1.71(0.46) | 2.11 2.47 3.87 4.05 |
| C Shangdu [42.00N, 115.75E] | 0.09(0.00) 2.01(0.94) | 1.06 2.17 1.48 2.69 |
| Shenmu [38.75N, 110.00E] | 1.04(0.95) 2.57(0.97) | 1.57 2.50 4.51 7.14 |
| Tuotetuo [40.25N, 111.00E] | 3.59(0.94) 7.51(0.96) | 3.63 5.85 4.82 7.42 |
| Zhunger [39.25N, 111.00E] | 0.20(0.54) 0.87(0.85) | 1.36 1.81 4.98 6.43 |
| Zhouzi [40.50N, 112.25E] | 0.26(0.00) 3.75(0.90) | 1.50 2.60 2.28 3.18 |
| Liangcheng [40.25N, 112.25E] | 0.21(0.00) 2.10(0.86) | 1.48 2.66 2.65 3.57 |
| Lingwu [38.00N, 106.25E] | 0.30(0.36) 1.97(0.86) | 1.09 1.40 2.68 3.51 |

\textsuperscript{a} Group A denotes city with newly built power plants; Group B denotes city without newly built power plants; and Group C denotes rural area with newly built power plants.

\textsuperscript{b} The coordinates of lower left corner for each region. The size of each region is 0.5° × 0.5°.

\textsuperscript{c} The number in parentheses is the fraction of power-plant NO\textsubscript{x} emissions in total NO\textsubscript{x} emissions.

NO\textsubscript{2} transport/transformation, considering that NO\textsubscript{2} has a short lifetime of about 3.6 h in summer (Schaub \textit{et al} 2007). Table 1 also shows the NO\textsubscript{x} emissions with fractions of power-plant emissions and NO\textsubscript{2} columns from GEOS-Chem and OMI.

While the spatial distributions of NO\textsubscript{2} columns in figures 1 and 2 are broadly similar in their features, the model significantly underestimates the absolute NO\textsubscript{2} concentrations, by 46% on average, compared to the satellite measurements. This is especially obvious in the larger cities such as Taiyuan, Zhangjiakou, Shuozhou, and Datong. This kind of underestimation has been found in previous studies (Wang \textit{et al} 2007, Zhao and Wang 2009, \textit{Lin} \textit{et al} 2010). There are several possible reasons for the underestimation. One possibility is that the INTEX-B emission inventory underestimates or omits NO\textsubscript{x} emissions from smaller sources in the industry and transportation sectors. This hypothesis is supported by three aspects of our work. First, NO\textsubscript{x} emissions from the power sector scarcely changed between 2005 and 2007 in cities such as Zhangjiakou, Shuozhou, and Datong (see table 1), and in these areas GEOS-Chem underestimates the absolute NO\textsubscript{2} columns from OMI by almost 75%, significantly lower than the average. Second, the increase rates of satellite measurements are higher than those from the model in Baotou, Wuhai, and Taiyuan, implying that we might underestimate the increase of emissions. Third, the fractions of power-plant NO\textsubscript{x} emissions in total NO\textsubscript{x} emissions shown in table 1 for most Group A cities remained largely unchanged between 2005 and 2007, while those for Group B cities declined less than anticipated. Another factor could be that soil emissions adopted in GEOS-Chem could be underestimated by a factor of two in summer (Jaeglé \textit{et al} 2005), which would cause bias in the background NO\textsubscript{2} level. Finally, it has been suggested that the NASA OMI-retrieved products overestimate NO\textsubscript{2} columns by 67%–74% in summer (Lamsal \textit{et al} 2010), which might be derived in part from the annual average NO\textsubscript{2} vertical profiles used in the calculation of the air mass factor (AMF). The dense aerosol loading, especially in cities, could also lead to above-average contributions to errors in the NO\textsubscript{2} column amounts (Boersma \textit{et al} 2008).

Nevertheless, despite the differences in absolute NO\textsubscript{2} columns between model and satellite, the changes between 2005 and 2007 in both measures are consistent. Modeled summertime average NO\textsubscript{2} columns increased dramatically in areas with newly built power plants: by an average of 40% in Group A cities and 63% in Group C rural areas. The OMI-retrieved NO\textsubscript{2} columns increased by 46% and 47%,
respectively. Due to interference from the industry and transportation sectors, we do not expect all study areas to behave perfectly in the comparison. We focus on areas where NOx emissions from power plants are a large fraction of total emissions. The increase rates (see figure 3) are very consistent in areas with newly built power plants, except for Shandu, Zhuozh, and Liangcheng, because there are no NOx emissions from power plants in 2005 in these three areas. The increase rates in Group B cities are substantially lower than the modeled ones except Shuozhou. This is expected to occur, considering the biases of emissions from the industry and transportation sectors. For all fifteen areas, the R-squared value ($R^2$) of the increase rates of modeled and OMI-retrieved NO2 columns is 0.65, with a slope of 0.67. In contrast to the sharp increase of NOx emissions found by Zhang et al (2009a), the increase rates of modeled columns are more comparable with those of OMI-retrieved columns in rural areas, which provides confidence in our approach.

The correlations of absolute changes between summertime average modeled NO2 columns ($\Delta$NO2–CTM), OMI-retrieved NO2 columns ($\Delta$NO2–OMI), and monthly average NOx emissions ($\Delta$NOx) are shown in figure 4. Good correlation is found between $\Delta$NO2 and $\Delta$NO2–CTM ($R^2 = 0.77$), with a slope of 1.79, which indicates that increased NO2 emissions of 1.79 Gg/month will lead to increased local NO2 columns of $1.0 \times 10^{15}$ molecules cm$^{-2}$ in the summertime in these areas. $\Delta$NO2–OMI and $\Delta$NO2–CTM are also well correlated ($R^2 = 0.61$) if we eliminate Taiyuan, a very large capital city with many small industrial sources that are likely omitted from or underestimated in the INTEX-B inventory. The correlation between $\Delta$NO2–CTM and $\Delta$NO2–OMI ($R^2 = 0.61$, $n = 14$) in this work is of the same order of magnitude as the correlation between $\Delta$NO2 and $\Delta$NO2–OMI ($R^2 = 0.68$, $n = 9$) in Zhang et al (2009a); however, comparisons in rural areas are much improved in this work, as compared with Zhang et al (2009a), which means that we have a better representation of regional NOx emissions and the emissions from smaller sources through the use of the GEOS-Chem model. Because the estimated NOx emissions in these areas are mainly contributed by newly built power plants, the emissions from which are well known, the consistency in $\Delta$NO2–OMI, $\Delta$NO2–CTM, and $\Delta$NOx means that the satellite measurements can be used to infer NOx emissions from large point sources. Further study should be focused on the impact of newly added NOx emissions from power plants on local NO2 vertical profiles and aerosol loadings, which could play critical roles in the calculation of AMF used to retrieve satellite measurements.

We conclude that rapid changes in NOx emissions from large power plants in Northern China can be identified and quantified with confidence, as a result of the consistency between modeled and satellite-observed tropospheric NO2 columns. The NASA INTEX-B emission inventory may underestimate NOx emissions from the industry and transportation sectors, inhibiting the reliable simulation of NO2 concentrations in large cities. This suggests that NOx emissions from small sources need to be re-estimated for Northern China. Our intention now is to apply this kind of analysis for a large number of point sources throughout China to examine and accurately quantify the NOx emissions in China.

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