Application of satellite observations for timely updates to global anthropogenic NO$_x$ emission inventories


Lok Nath Lamsal, James Dunn Building, Department of Physics and Atmospheric Science
Dalhousie University Halifax, NS, B3H 3J5, Canada (lok.lamsal@fizz.phys.dal.ca)

$^1$Department of Physics and Atmospheric
Anthropogenic emissions of nitrogen oxides ($\text{NO}_x$) can change rapidly due to economic growth or control measures. Bottom-up emissions estimated using source-specific emission factors and activity statistics require years to compile and can become quickly outdated. We present a method to use satellite observations of tropospheric NO$_2$ columns to estimate changes in NO$_x$ emissions. We use tropospheric NO$_2$ columns retrieved from the SCIAMACHY satellite instrument for 2003–2009, the response of tropospheric NO$_2$ columns to changes in NO$_x$ emissions determined from a global chemical transport model (GEOS-Chem), and the bottom-up anthropogenic NO$_x$ emissions for

Science, Dalhousie University, Halifax, NS,
B3H 3J5, Canada

$^2$Atomic and Molecular Physics Division,
Harvard-Smithsonian Center for Astrophysics, Cambridge, MA, USA

$^3$Center for Earth System Science,
Tsinghua University, Beijing, 100084, P. R. China

$^4$Environment Canada, Toronto, Ontario,
Canada

$^5$University of Alabama Huntsville,
Huntsville, AL, USA
2006 to hindcast and forecast the inventories. We evaluate our approach by comparing bottom-up and hindcast emissions for 2003. The two inventories agree within 6.0% globally and within 8.9% at the regional scale with consistent trends in western Europe, North America, and East Asia. We go on to forecast emissions for 2009. During 2006–2009, anthropogenic NO$_x$ emissions over land increase by 9.2% globally and by 18.8% from East Asia. North American emissions decrease by 5.7%.
1. Introduction

Nitrogen oxides ($\text{NO}_x = \text{NO} + \text{NO}_2$) emitted to the atmosphere mainly through fossil fuel combustion, biomass burning, soil, and lightning play a key role in atmospheric chemistry. Global anthropogenic $\text{NO}_x$ emissions are expected to change rapidly over the coming decades due to economic development and emissions controls. Monks et al. [2009] give an overview of available inventories. The bottom-up approach of estimating $\text{NO}_x$ emissions by aggregating activity data and emission factors is a major undertaking that often suffers from a time lag of years between the occurrence of emissions and completion of inventories. Timely $\text{NO}_x$ emission estimates are needed for better understanding of air pollution, acid deposition, and climate change.

Satellite observations of tropospheric $\text{NO}_2$ columns provide near-real-time and independent information on $\text{NO}_x$ emissions and their trends. Numerous studies have used space-based tropospheric $\text{NO}_2$ observations to examine temporal changes [e.g. Beirle et al., 2003; Richter et al., 2005; Kim et al., 2006; van der A et al., 2006; Zhang et al., 2007; Boersma et al., 2008a; Kaynak et al., 2009; Yoshida et al., 2010] and to provide top-down constraints on surface $\text{NO}_x$ emissions via inverse modeling [e.g. Martin et al., 2003; Jaeglé et al., 2005; Müller and Stavrakou, 2005; Napelenok et al., 2008; Chai et al., 2009; Zhao and Wang, 2009; Lin et al., 2010]. Here we present an approach to rapidly update bottom-up $\text{NO}_x$ emission inventories using top-down trend analysis of tropospheric $\text{NO}_2$ columns from the SCIAMACHY instrument [Bovensmann et al., 1999]. The SCIAMACHY tropospheric $\text{NO}_2$ column retrieval is described in section 2. In section 3, we provide a brief account of
bottom-up NO\textsubscript{x} emissions and the GEOS-Chem model. Section 4 presents our approach to construct a top-down anthropogenic NO\textsubscript{x} emission inventory.

2. SCIAMACHY Tropospheric NO\textsubscript{2} Column Retrievals

The SCIAMACHY instrument aboard the ENVISAT satellite observes solar backscatter that can be applied to retrieve tropospheric nitrogen dioxide (NO\textsubscript{2}) with a typical spatial resolution of 30 km $\times$ 60 km, achieving global coverage every 6 days [Bovensmann et al., 1999]. ENVISAT was launched in March 2002 into a sun-synchronous polar orbit, crossing the equator at 10:00 local time in the descending node.

We retrieve tropospheric NO\textsubscript{2} columns for the years 2003-2009 using the algorithms described in Martin et al. [2006] with a few updates including the use of clouds from FRESCO+ [Wang et al., 2008] in the air mass factor formulation. For this manuscript we use monthly NO\textsubscript{2} vertical profile shapes for 2006 to keep the bottom-up and top-down emissions independent. The SCIAMACHY NO\textsubscript{2} retrievals have been validated with coincident airborne in situ measurements [Martin et al., 2006] and extensively applied to understand NO\textsubscript{x} emissions [e.g. Martin et al., 2006; Sioris et al., 2007; Napelenok et al., 2008; Kaynak et al., 2009; Walker et al., 2010].

Wintertime retrievals are more error prone due to the reduced sensitivity of satellite measurements to lower tropospheric NO\textsubscript{2} at high solar zenith angle (SZA) and by uncertainties associated with snow covered scenes [O’Bryne et al., 2010]. We exclude the data for winter by excluding observations made at $>$50° SZA. To reduce retrieval errors, we use observations with cloud radiance fraction <20%.
3. Bottom-up NO\textsubscript{x} Emission Inventory and Model Description

We use the bottom-up NO\textsubscript{x} emission inventory implemented in the GEOS-Chem global three-dimensional model of atmospheric chemistry (www.geos-chem.org) to conduct sensitivity simulations and to evaluate the hindcast top-down inventory. Global anthropogenic emissions are based on the EDGAR 3.2FT2000 inventory [Olivier et al., 2001] for the year 2000. The global inventory is overwritten by regional inventories that include the CAC inventory (http://www.ec.gc.ca/pdb/cac/) for 2005 over Canada, the U.S. EPA National Emissions Inventory (NEI) for 2002 and 2005 over the United States, the BRAVO inventory [Kuhns et al., 2005] for 1999 over Mexico, the inventory from Zhang et al. [2007] for 2003 and 2006 over East Asia, and the EMEP inventory for 2003 and 2006 for Europe. Where emissions are not available for 2003 or/and 2006, emissions are scaled from the nearest year of available inventory following van Donkelaar et al. [2008]. We focus on 2006 as the most recent year with emission statistics and on 2003 as the most historical year that overlaps with SCIAMACHY observations.

Figure 1 shows bottom-up anthropogenic NO\textsubscript{x} emissions from land sources for the year 2003 (top panel) and 2006 (middle panel) and emission changes for 2003–2006 (bottom panel). Additional information on the bottom-up inventory is in the supplementary material. Global anthropogenic NO\textsubscript{x} emissions increase by 5.2% from 22.9 Tg N in 2003 to 24.1 Tg N in 2006, with global emissions growth partially counteracted by the reduction in North America and Europe. East Asian emissions increase by 25% over the three years. The changes in anthropogenic emissions in Africa, South America, and Oceania are minor (<10\textsuperscript{10} atoms N cm\textsuperscript{-2} s\textsuperscript{-1}, <0.1 Tg N).
We develop a global simulation capability for GEOS-Chem at $1^\circ \times 1.25^\circ$ (All previous global GEOS-Chem simulations were at $2^\circ \times 2.5^\circ$ or $4^\circ \times 5^\circ$). This development is applied to GEOS-Chem version 8-01-04 driven by GEOS-4 assimilated meteorology from the NASA Global Modeling and Assimilation Office. Anthropogenic NO$_x$ emissions from land sources are as described above. Other emissions have been recently described by Lamsal et al. [2010]. We conduct a simulation for the year 2006 and coincidently sample the model output for analysis of the SCIAMACHY data. The GEOS-Chem simulation of NO$_x$ has been recently compared with a variety of in situ and satellite observations [e.g. Jaeglé et al., 2005; Martin et al., 2006; Hudman et al., 2007; Wang et al., 2007; Boersma et al., 2008a, b; Zhang et al., 2008; Lamsal et al., 2008, 2010] and generally agrees to within 30% of measured NO$_x$.

4. Prediction of Emissions

Satellite observations of tropospheric NO$_2$ columns are strongly related to surface NO$_x$ emissions due to the short NO$_x$ lifetime combined with the high NO$_2$/NO$_x$ ratio in the boundary layer. We use the GEOS-Chem model to examine the relationship.

Following Walker et al. [2010], we perform two simulations, one with NO$_x$ emissions ($E$) for the year 2006 described in section 3 and another with anthropogenic NO$_x$ emissions perturbed by 15%, to establish the relationship between changes in surface NO$_x$ emissions and changes in tropospheric NO$_2$ columns ($\Omega$):

$$\frac{\Delta E}{E} = \beta \times \frac{\Delta \Omega}{\Omega}.$$ (1)
$$\Delta \Omega$$ is the change in simulated tropospheric NO$_2$ columns driven by the change in emissions $\Delta E$. The term $\beta$ represents the local sensitivity of changes in NO$_2$ columns to changes in NO$_x$, similar to the variable $\alpha$ used in top-down emission inference (E) by Martin et al. [2003]:

$$E = \alpha \times \Omega.$$ (2)

Unlike $\alpha$ (units of s$^{-1}$) that describes the direct relationship between NO$_2$ columns and NO$_x$ emissions, $\beta$ is a unitless trend factor that describes how a change in NO$_x$ emissions changes the NO$_2$ columns. $\beta$ reflects the feedback of NO$_x$ emissions on NO$_x$ chemistry. Using a perturbation of 30% (instead of 15%) changes $\beta$ by <2%. The overall error in the approach arises from the combination of errors in the NO$_2$ column trend and in $\beta$, which are the subject of ongoing work.

Figure S1 in the supplementary material shows the spatial variation of annual averaged values of $\beta$ for 2006 coincidently sampled with the SCIAMACHY data. The global mean value is 1.16. $\beta$ tends to be greater than one in remote regions where an increase in NO$_x$ emissions decreases the NO$_x$ lifetime (such as through feedback on O$_3$ and OH). In polluted regions, $\beta$ tends to be less than one since an increase in NO$_x$ emissions consumes OH and increases the NO$_x$ lifetime. Changes in the NO$_2$/NO$_x$ ratio partially compensate for changes in the NO$_x$ lifetime since increases in HO$_x$ both increase the NO$_2$/NO$_x$ ratio and decrease the NO$_x$ lifetime. A simulation at higher spatial resolution would better resolve nonlinear NO$_x$ chemistry and heterogeneous emission sources and may yield more spatial variation in $\beta$. Outside of winter, the seasonal variation of $\beta$ is <5% and interannual
variation is <3%. Increasing anthropogenic VOCs and CO by 15% increases global values
of \( \beta \) by 2.8% and 1.0%, respectively. The calculation of \( \beta \) is most accurate for regions with
homogeneous emission changes; perturbing NO\(_x\) emissions for a single grid cell in Ohio
affects \( \beta \) in neighboring grid cells by 2–6%. Zhang et al. [2007] directly compared trends
over China in bottom-up NO\(_x\) emissions with satellite NO\(_2\) columns and found a larger
trend in satellite NO\(_2\) than in NO\(_x\) emissions; it appears that accounting for \( \beta \) could help
explain the discrepancy since \( \beta \) values are less than one over regions of China with the
largest NO\(_2\) columns. A nested simulation at \( \frac{1}{2}^\circ \times \frac{1}{3}^\circ \) also yields similar \( \beta \) values for East
China [Zhang et al., in preparation].

We use monthly \( \beta \) values to translate the changes in SCIAMACHY tropospheric NO\(_2\)
columns to the changes in monthly NO\(_x\) emissions. The annual changes in NO\(_x\) emissions
are then combined with available bottom-up NO\(_x\) emissions \( E_i \) for the year \( i \) to predict
emissions \( E_j \) for the year \( j \):

\[
E_j = (1 + \beta \frac{(\Omega_j - \Omega_i)}{\Omega_i}) E_i. \tag{3}
\]

We partition the top-down NO\(_x\) emissions according to the spatial distribution of the
sources in the bottom-up inventory to derive the anthropogenic component of the pre-
dicted emissions. The error due to partitioning is minimized here by limiting our analyses
to grid boxes with large tropospheric NO\(_2\) columns (\( >1 \times 10^{15} \)) and with anthropogenic
sources dominating (>50%) total NO\(_x\) emissions. This also reduces errors in the relation
between NO\(_x\) emissions and NO\(_2\) columns. With these criteria we retain data for only
14% of land areas, but they represent 80% of anthropogenic emissions over land and 74%
of total anthropogenic emissions. No assumption is made for trends in the remaining 26% of emissions. Below we demonstrate how the 2006 inventory hindcasted to 2003 using SCIAMACHY observations compares with the bottom-up inventory for 2003, and then proceed to predict emissions for 2009.

The left column of Figure 2 shows the spatial variation of bottom-up and predicted NO\textsubscript{x} inventories of anthropogenic emissions for 2003. The spatial distribution of the two inventories is highly consistent (r = 0.90, N = 2328). The predicted NO\textsubscript{x} inventory (17.0 Tg N Y\textsuperscript{-1}) is 6.0% lower than the bottom-up (18.1 Tg N Y\textsuperscript{-1}) for regions dominated by anthropogenic NO\textsubscript{x} emissions. The two inventories exhibit regional differences of 1.7% over North America and 8.9% over OECD (Organisation for Economic Co-operation and Development) Europe, within the uncertainty in bottom-up emissions of 25% over these regions [Vestreng et al., 2009; Christian Hogrefe, personal communication, 2008]. Including changes in anthropogenic VOCs and CO in the calculation of $\beta$ decreases the hindcast inventory by <1%.

The right column of Figure 2 shows the difference between the bottom-up inventory for 2006 and the hindcast inventory for 2003 which indicates the change in NO\textsubscript{x} emissions inferred from the SCIAMACHY data. The top-down emission changes are broadly consistent with the changes in the bottom-up inventory presented in the bottom panel of Figure 1. Differences with the bottom-up inventory are shown in Figure S2 of the supplementary material. Both show significant reductions over the eastern United States and parts of Europe, and increases over eastern China. However, the hindcast inventory exhibits larger spatial heterogeneity and stronger emissions growth of 12.7% compared
with 5.5% in the bottom-up inventory. While both inventories consistently suggest a negative trend from 2003 to 2006 over Western and Central Europe, they often yield an opposite trend in the rest of Europe, implying that the current knowledge about emissions in Eastern European countries may be inadequate. Apart from some inconsistencies in southeastern China, the predicted emissions are in rather close agreement with the bottom-up inventory in East Asia, where bottom-up and predicted inventories suggest the increase of 22% and 21%, respectively. Chinese NO\textsubscript{x} emissions increase by 28% during 2003-2006 at a 9.3% annual growth rate in bottom-up inventory, in close agreement with the increase of 24% at a 8.0% annual growth rate in the predicted inventory. Varying the tropospheric NO\textsubscript{2} threshold from $1 \times 10^{15}$ molec cm\textsuperscript{-2} to $5 \times 10^{15}$ molec cm\textsuperscript{-2} increases the growth in bottom-up Chinese emissions by 28%–31% compared to 24%–39% in the predicted inventory, implying spatial variability in emissions growth. These results are in line with previous studies on Chinese NO\textsubscript{x} emissions [Richter et al., 2005; Zhang et al., 2007, 2009].

The bottom row of Figure 2 presents a forecast of anthropogenic NO\textsubscript{x} emissions for the year 2009. The predicted 2009 NO\textsubscript{x} inventory (20.9 Tg NY\textsuperscript{-1}) is 9.2% higher than the bottom-up 2006 inventory (19.1 Tg NY\textsuperscript{-1}), with most of the increase arising from East Asia. Changes in anthropogenic NO\textsubscript{x} emissions during 2006–2009 indicate a decrease of 5.7% in North America and an increase of 18.8% in East Asia, with a 6.7% annual growth rate in Chinese NO\textsubscript{x} emissions.

5. Conclusions
We developed a method to apply changes in satellite observations of the tropospheric NO$_2$ column for timely updates to bottom-up anthropogenic NO$_x$ emission inventories. We retrieved tropospheric NO$_2$ columns from SCIAMACHY for 2003-2009, and to interpret these observations we developed a global simulation capability for GEOS-Chem at a global resolution of $1^\circ \times 1.25^\circ$. The local annual scale factor was determined by examining the response of NO$_2$ columns to a small perturbation in anthropogenic NO$_x$ emissions using the GEOS-Chem model. We combined the SCIAMACHY inferred NO$_x$ emissions changes each year during 2003-2009 with the 2006 bottom-up inventory to hindcast emissions for 2003 and to forecast emissions for the year 2009. The forecast inventories for 2007–2009 serve as a temporary dataset until bottom-up inventories are developed to represent those years.

Acknowledgments.

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Figure 1. Spatial distributions of bottom-up anthropogenic NO$_x$ emissions at 1° × 1.25° for 2003 (top) and 2006 (middle). The bottom panel shows the difference between anthropogenic emissions for 2006 minus those for 2003.
Figure 2. The left column contains annual anthropogenic NO\textsubscript{x} emissions. The top panel shows the bottom-up inventory for the year 2003. Inventories predicted from SCIAMACHY observations are shown for the years 2003 (middle) and 2009 (bottom). White areas indicate where anthropogenic sources contribute <50% of total NO\textsubscript{x} emissions and tropospheric NO\textsubscript{2} columns are <1\times10^{15} molec cm\textsuperscript{-2}. Inventory totals refer to colored regions. The right column contains the changes in SCIAMACHY-derived anthropogenic NO\textsubscript{x} emissions during 2003–2006 (top) and 2006–2009 (bottom).
Supplementary Materials

1. Details on NOx emissions sources in GEOS-Chem

Table S1 shows the NOx emissions for 2003 and 2006. Natural emissions of NOx from lightning and soils are computed locally. Lightning NOx emissions are linked to deep convection following the parameterization of Price and Rind [1992] with vertical profiles taken from Pickering et al. [1998]. Following Martin et al. [2006] and Hudman et al. [2007] the midlatitude lightning NOx source is 1.6 Tg N yr\(^{-1}\). The spatial distribution of lightning flashes is scaled to OTD-LIS [Sauvage et al., 2007; Murray et al., 2009]. Soil NO\(_x\) emissions are computed as a function of vegetation type, precipitation, temperature, fertilizer usage, and a canopy reduction factor [Yienger and Levy, 1995; Wang et al., 1998]. Biomass burning emissions are from the interannual GFED2 inventory [van der Werf et al., 2006; Randerson et al., 2007]. Emissions from aircraft are based on the monthly mean emission inventory compiled by Baughcum et al. [1996]. Land surface anthropogenic emissions include emissions from fossil fuels; they exclude the contributions from all other sources. The approach to infer global annual scale factors is described in van Donkelaar et al. [2008]. In brief, scale factors have been derived using data from government statistics for North America, Europe, and Asia. Beyond these regions, NO\(_x\) emissions are scaled according to CO\(_2\) trends available from the Carbon Dioxide Information Analysis Center (CDIAC).

Table S1: Emissions for 2003 and 2006 in GEOS-Chem.

<table>
<thead>
<tr>
<th>Years/Sources</th>
<th>2003</th>
<th>2006</th>
</tr>
</thead>
<tbody>
<tr>
<td>Anthropogenic</td>
<td>24.7</td>
<td>26.1</td>
</tr>
<tr>
<td>Biomass burning</td>
<td>5.4</td>
<td>4.8</td>
</tr>
<tr>
<td>Soils</td>
<td>7.1</td>
<td>6.1</td>
</tr>
<tr>
<td>Lightning</td>
<td>5.9</td>
<td>7.3</td>
</tr>
<tr>
<td>Biofuel</td>
<td>2.2</td>
<td>2.2</td>
</tr>
<tr>
<td>Aircraft</td>
<td>0.5</td>
<td>0.5</td>
</tr>
</tbody>
</table>

2. Spatial variation of $\beta$

![Image](https://example.com/image.png)

Figure S1: Annual average value of $\beta$ (ratio of change in NO\(_x\) emissions to change in tropospheric NO\(_2\) columns) introduced in Equation 1 and calculated with GEOS-Chem. White areas indicate where
anthropogenic sources contribute <50% of total NO\textsubscript{x} emissions and tropospheric NO\textsubscript{2} columns are <1\times10^{15} molec cm\textsuperscript{-2}.

3. Difference between predicted and bottom-up emissions for 2003

Figure S2: Difference between SCIAMACHY-derived and bottom-up anthropogenic NO\textsubscript{x} emissions for 2003. White areas indicate where anthropogenic sources contribute < 50% of total NO\textsubscript{x} emissions or tropospheric NO\textsubscript{2} columns are < 1\times10^{15} molec cm\textsuperscript{-2}.

References


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