Application of satellite observations for timely updates to global anthropogenic NO_x emission inventories

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[1] Anthropogenic emissions of nitrogen oxides (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using source-specific emission factors and activity statistics require years to compile and can become quickly outdated.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 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%. **Citation:** Lamsal, L. N., R. V. Martin, A. Padmanabhan, A. van Donkelaar, Q. Zhang, C. E. Sioris, K. Chance, T. P. Kurosu, and M. J. Newchurch (2011), Application of satellite observations for timely updates to global anthropogenic NO_x emission inventories, *Geophys. Res. Lett.*, *38*, L05810, doi:10.1029/2010GL046476.

1. Introduction

[2] Nitrogen oxides (NO_x = NO + 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 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 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 NO_x emission estimates are needed for better understanding of air pollution, acid deposition, and climate change.

[3] Satellite observations of tropospheric NO_2 columns provide near-real-time and independent information on NO_x emissions and their trends. Numerous studies have used space-based tropospheric 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 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 NO_x emission inventories using top-down trend analysis of tropospheric NO_2 columns from the SCIAMACHY instrument [*Bovensmann et al.*, 1999]. The SCIAMACHY tropospheric NO_2 column retrieval is described in section 2. In section 3, we provide a brief account of bottom-up NO_x emissions and the GEOS-Chem model. Section 4 presents our approach to construct a top-down anthropogenic NO_x emission inventory.

2. SCIAMACHY Tropospheric NO_2 Column Retrievals

[4] The SCIAMACHY instrument aboard the ENVISAT satellite observes solar backscatter that can be applied to retrieve tropospheric nitrogen dioxide (NO_2) with a typical spatial resolution of 30 km × 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.

[5] We retrieve tropospheric NO_2 columns for the years 2003–2009 using the algorithms described by *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_2 vertical profile shapes for 2006 to keep the bottom-up and top-down emissions independent. The SCIAMACHY NO_2 retrievals have been validated with coincident airborne in situ measurements [*Martin et al.*, 2006] and extensively applied to understand NO_x emissions [e.g., *Martin et al.*, 2006; *Sioris et al.*, 2007; *Napelenok et al.*, 2008; *Kaynak et al.*, 2009; *Walker et al.*, 2010].

[6] Wintertime retrievals are more error prone due to the reduced sensitivity of satellite measurements to lower tropospheric NO_2 at high solar zenith angle (SZA) and by uncertainties associated with snow covered scenes [*O'Byrne*

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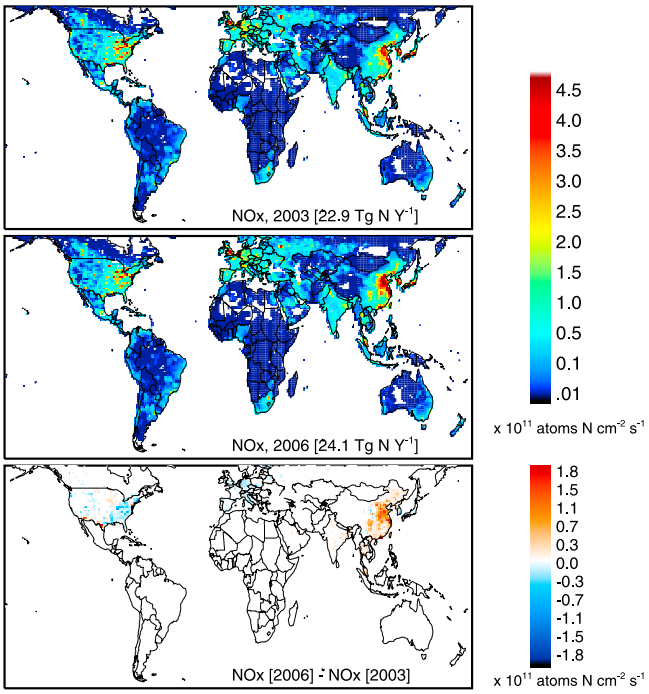

Figure 1. Spatial distributions of bottom-up anthropogenic NO_x emissions at 1° × 1.25° for (top) 2003 and (middle) 2006. (bottom) The difference between anthropogenic emissions for 2006 minus those for 2003.

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_x Emission Inventory and Model Description

[7] We use the bottom-up NO_x emission inventory implemented in the GEOS-Chem global three-dimensional model of atmospheric chemistry (http://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.

[8] Figure 1 shows bottom-up anthropogenic NO_x emissions from land sources for the year 2003 (Figure 1, top) and 2006 (Figure 1, middle) and emission changes for 2003–2006 (Figure 1, bottom). Additional information on the bottom-up inventory is in the auxiliary material.[1] Global anthropogenic NO_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 ($<10^{10}$ atoms N $cm^{-2}s^{-1}$, <0.1 Tg N).

[9] We develop a global simulation capability for GEOS-Chem at 1° × 1.25° (all previous global GEOS-Chem simulations were at 2° × 2.5° or 4° × 5°). 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 coincidentally 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, 2008b; *Zhang et al.*, 2008; *Lamsal et al.*, 2008, 2010] and generally agrees to within 30% of measured NO_x.

4. Prediction of Emissions

[10] 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.

[11] 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 (Ω):

$$\frac{\Delta E}{E} = \beta \times \frac{\Delta \Omega}{\Omega}. \quad (1)$$

$\Delta\Omega$ is the change in simulated tropospheric NO_2 columns driven by the change in emissions ΔE. The term β represents the local sensitivity of changes in NO_2 columns to changes in NO_x, similar to the variable α used in top-down emission inference (E) by *Martin et al.* [2003]:

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

Unlike α (units of s^{-1}) that describes the direct relationship between NO_2 columns and NO_x emissions, β is a unitless trend factor that describes how a change in NO_x emissions changes the NO_2 columns. β reflects the feedback of NO_x emissions on NO_x chemistry. Using a perturbation of 30% (instead of 15%) changes β by <2%. The overall error in the approach arises from the combination of errors in the NO_2 column trend and in β, which are the subject of ongoing work.

[1] Auxiliary materials are available in the HTML. doi:10.1029/2010GL046476.

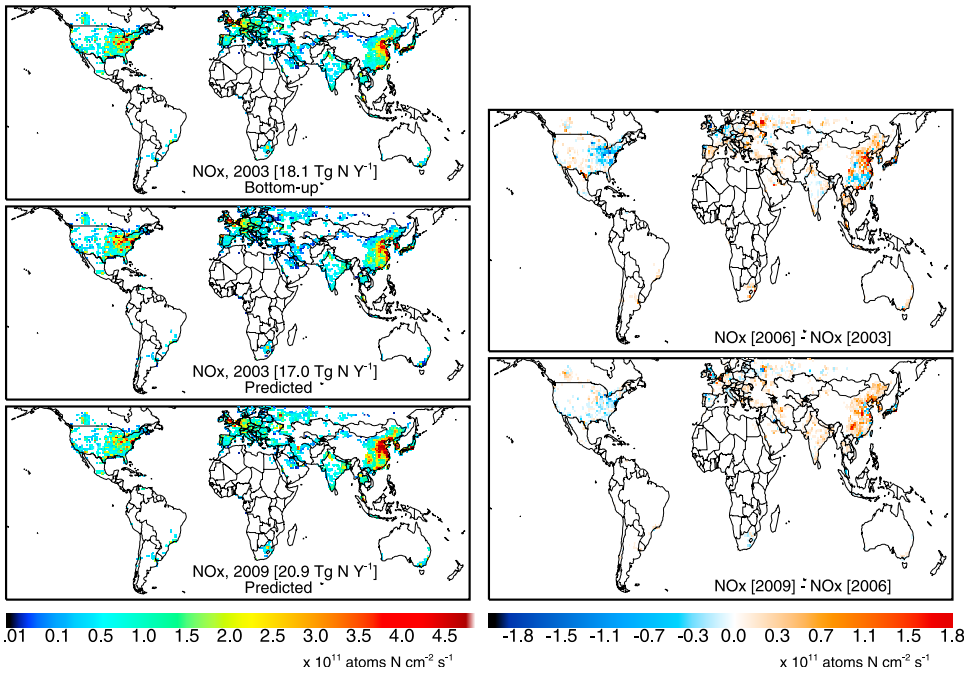

Figure 2. (left) Annual anthropogenic NO_x emissions. (top) The bottom-up inventory for the year 2003. Inventories predicted from SCIAMACHY observations are shown for the years (middle) 2003 and (bottom) 2009. White areas indicate where anthropogenic sources contribute <50% of total NO_x emissions and tropospheric NO_2 columns are $<1 \times 10^{15}$ molec cm^{-2}. Inventory totals refer to colored regions. (right) The changes in SCIAMACHY-derived anthropogenic NO_x emissions during (top) 2003–2006 and (bottom) 2006–2009.

[12] Figure S1 in the auxiliary material shows the spatial variation of annual averaged values of β for 2006 coincidentally sampled with the SCIAMACHY data. The global mean value is 1.16. β 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, β 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 β. Outside of winter, the seasonal variation of β is <5% and interannual variation is <3%. Increasing anthropogenic VOCs and CO by 15% increases global values of β by 2.8% and 1.0%, respectively. The calculation of β is most accurate for regions with homogeneous emission changes; perturbing NO_x emissions for a single grid cell in Ohio affects β 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 β could help explain the discrepancy since β values are less than one over regions of China with the largest NO_2 columns. A nested simulation at $\frac{1}{2}^\circ \times \frac{2}{3}^\circ$ also yields similar β values for East China (Q. Zhang et al., manuscript in preparation, 2011).

[13] We use monthly β 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:

$$E_j = \left(1 + \beta \frac{(\Omega_j - \Omega_i)}{\Omega_i}\right) E_i. \quad (3)$$

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 predicted 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.

[14] The left column of Figure 2 shows the spatial variation of bottom-up and predicted NO_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_x inventory (17.0 Tg N Y^{-1}) is 6.0% lower than the bottom-up (18.1 Tg N Y^{-1}) for regions dominated by anthropogenic NO_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; C. Hogrefe, personal communication, 2008]. Including changes in anthropogenic VOCs and CO in the calculation of β decreases the hindcast inventory by <1%.

[15] Figure 2 (right) shows the difference between the bottom-up inventory for 2006 and the hindcast inventory for 2003 which indicates the change in NO_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 Figure 1 (bottom). Differences with the bottom-up inventory are shown in Figure S2 of the auxiliary 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_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_2 threshold from 1×10^{15} molec cm^{-2} to 5×10^{15} molec cm^{-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_x emissions [*Richter et al.*, 2005; *Zhang et al.*, 2007, 2009].

[16] Figure 2 (bottom) presents a forecast of anthropogenic NO_x emissions for the year 2009. The predicted 2009 NO_x inventory (20.9 Tg N Y^{-1}) is 9.2% higher than the bottom-up 2006 inventory (19.1 Tg N Y^{-1}), with most of the increase arising from East Asia. Changes in anthropogenic NO_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_x emissions.

5. Conclusions

[17] 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° × 1.25°. 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.

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