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Evaluating NOx emission inventories for regulatory air quality modeling using satellite and air quality model data

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HIGHLIGHTS

- We used OMI and CAMx NO_2 columns to estimate NOx emissions over the southeast U.S.
- NOx emissions estimates were developed using DOMINO v2.0 and NASA SP2 retrievals.
- The two top–down NOx estimates were quite different over the southeast U.S.
- These disparities were due to differences in the two NO_2 retrievals.
- It was not possible to constrain the TCEQ's NOx inventory with these estimates.

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ABSTRACT

The purpose of this study was to assess the accuracy of NOx emissions in the Texas Commission on Environmental Quality's (TCEQ) State Implementation Plan (SIP) modeling inventories of the southeastern U.S. We used retrieved satellite tropospheric NO_2 columns from the Ozone Monitoring Instrument (OMI) together with NO_2 columns from the Comprehensive Air Quality Model with Extensions (CAMx) to make top–down NOx emissions estimates using the mass balance method. Two different top–down NOx emissions estimates were developed using the KNMI DOMINO v2.0 and NASA SP2 retrievals of OMI NO_2 columns. Differences in the top–down NOx emissions estimates made with these two operational products derived from the same OMI radiance data were sufficiently large that they could not be used to constrain the TCEQ NOx emissions in the southeast. The fact that the two available operational NO_2 column retrievals give such different top–down NOx emissions results is important because these retrievals are increasingly being used to diagnose air quality problems and to inform efforts to solve them. These results reflect the fact that NO_2 column retrievals are a blend of measurements and modeled data and should be used with caution in analyses that will inform policy development. This study illustrates both benefits and challenges of using satellite NO_2 data for air quality management applications. Comparison with OMI NO_2 columns pointed the way toward improvements in the CAMx simulation of the upper troposphere, but further refinement of both regional air quality models and the NO_2 column retrievals is needed before the mass balance and other emission inversion methods can be used to successfully constrain NOx emission inventories used in U.S. regulatory modeling.

1. Introduction

Texas has two metropolitan areas, Houston-Galveston-Brazoria and Dallas-Fort Worth, which do not attain the National Ambient Air Quality Standard (NAAQS) for ozone. The Texas Commission on Environmental Quality (TCEQ) carries out ozone modeling as part of the Texas State Implementation Plan (SIP) that prescribes emissions controls that will allow these areas to attain the NAAQS in the

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future. Ozone transport has been shown to play an important role in determining ozone levels in Texas (Berlin et al., 2013). The southeastern U.S. sometimes is a source of ozone transport into Texas. A systematic high bias is present in the TCEQ's modeling of southeast ozone that can confound efforts to quantify the effects of ozone transport on Texas (ENVIRON, 2011). A high bias in modeled ozone in the southeast has been noted across multiple models (e.g. Herwehe et al., 2011; Chai et al., 2013). Model overestimates of ozone in the southeast may result, at least in part, from biased NOx emissions. The purpose of this study was to assess the accuracy of the NOx emissions data in the TCEQ's SIP modeling inventories, which are based on the 2005 National Emission Inventory (NEI) in the southeast.

Satellite NO_2 column retrievals have been used together with chemistry-transport models to provide constraints on global and regional NOx emission inventories by many investigators (e.g. Leue et al., 2001; Martin et al., 2003, 2006; Konovalov et al., 2006; Zhang et al., 2007; Boersma et al., 2008; Napelenok et al., 2008; Kim et al., 2009; Lin et al., 2010; Miyazaki et al., 2012; Tang et al., 2013, 2014; Vinken et al., 2014). A review of methods is provided in Streets et al. (2013).

We evaluated the feasibility of using satellite NO_2 column data from the Ozone Monitoring Instrument (OMI) together with NO_2 columns from a regional air quality model to constrain the TCEQ's NOx emission inventory for the southeast. Following the mass balance method (Martin et al., 2003, 2006; Boersma et al., 2008; Lamsal et al., 2010; Tang et al., 2013), the ratio of the observed NO_2 columns to modeled NO_2 columns from the Comprehensive Air quality Model with extensions (CAMx; ENVIRON, 2014) was used together with the existing TCEQ bottom–up NOx emission inventory to estimate top–down NOx emissions. The top–down emission estimates are given by:

$$NOx\ Emiss_{top\ down} = \frac{\Omega_{satellite}}{\Omega_{model}} \times \left(NOx\ Emiss_{bottom\ up}\right) \times (Smoothing\ Factor) \quad (1)$$

where Ω are integrated tropospheric NO_2 vertical column densities (VCD; molecules cm^{-2}) from the satellite retrieval and the air quality model. The smoothing factor accounts for the potential for NOx emissions to be transported out of a given grid cell and influence NO_2 columns beyond the grid cell in which emissions occurred.

In many previous studies, a single satellite retrieval was used to determine top–down NOx emissions constraints. In order to understand the uncertainty introduced into the top–down NOx emission estimates by the choice of retrieval, we used two independent operational retrievals to develop two different sets of top–down NOx emissions estimates. This is the first study we are aware of that compares top–down gridded regional emissions estimates developed using multiple satellite retrievals in order to assess method uncertainties.

The two sets of calculated top–down NOx emissions were compared with the TCEQ NOx emission inventory across the southeast. The comparison was carried out for May 31–July 2, 2006. This June 2006 episode is used by the TCEQ for regulatory ozone modeling.

2. Methods

2.1. Tropospheric NO_2 vertical column density data

2.1.1. OMI tropospheric NO_2 column retrieval

NO_2 column data are derived from measurements of backscattered solar radiation made by OMI, a nadir-viewing UV–visible spectrometer (Levelt et al., 2006). OMI flies aboard NASA's Aura satellite, a polar orbiter that provides daily global coverage (Schoeberl et al., 2006; Boersma et al., 2007, 2011). Aura is in a sun-synchronous orbit with overpass at ~1:40 pm local time.

OMI does not measure NO_2 directly, but measures both direct sunlight and sunlight that is backscattered from the earth's atmosphere. Conversion of measured radiation from the OMI instrument to vertical tropospheric NO_2 columns is a multi-step process known as a retrieval. There is no unique solution for the vertical NO_2 column density given a set of measured OMI reflectances. The goal of the retrieval is to calculate the integrated vertical column of NO_2 that best reproduces the reflectances measured by OMI, given our knowledge of the characteristics of the instrument, the satellite viewing geometry and the relevant properties of the earth and the atmosphere.

2.1.2. DOMINO and SP2 retrievals

The first retrieval used in this study is the Derivation of OMI tropospheric NO_2 product (DOMINO; available from the Tropospheric Emission Monitoring Internet Service [TEMIS] at http://www.temis.nl/) product developed by KNMI (Koninklijk Nederlands Meteorologisch Instituut). The DOMINO v2.0 OMI NO_2 column data product was the most recent version of the DOMINO product available at the time of this study (Boersma et al., 2011, Boersma and van der A, 2011). The second retrieval used here is version 2 of the NASA Standard Product (SP2; Bucsela et al., 2013). Although the two retrievals begin with the same slant NO_2 columns, they differ in some of their input data, their atmospheric mass factor calculations, and their stratospheric NO_2 column separation methods (Boersma et al., 2011; Bucsela et al., 2013) (See Supporting information for additional description of the retrieval process). There can be significant differences between the NO_2 columns produced by two different retrievals given the same OMI radiance data (Lamsal et al., 2010; Bucsela et al., 2008; Herron-Thorpe et al., 2010). It is not possible to say, based on available evaluations of the retrievals (e.g. Lamsal et al., 2014), whether one is more accurate than the other across the southeast during the June 2006 episode.

2.2. CAMx air quality model

CAMx was used to model the continental United States using nested 36/12/4 km resolution grids (See map in Supporting information). CAMx is a three-dimensional chemical-transport grid model and is used for ozone air-quality planning in Texas (TCEQ, 2010). The model was applied from the earth's surface upward to a height of approximately 15 km (vertical grid structure is shown in the Supporting information).

The June 2006 CAMx modeling databases were developed by the TCEQ for regulatory modeling of ozone. Meteorological input data for CAMx were developed using the Weather Research and Forecasting Model version 3.2 (WRF; Skamarock and Klemp, 2007). Boundary conditions for the outermost (36 km) grid were derived from a GEOS-Chem (Bey et al., 2001) global chemistry-transport model simulation of 2006. Emissions of VOCs, NOx, and CO from the TCEQ's 2006 emission inventory (TCEQ, 2010) were used. Throughout the modeling domain, emissions for U.S. power plants that report continuous emissions monitoring data to the U.S. EPA's Clean Air Markets Database were modeled at actual hourly June 2006 levels. For sources other than power plants, the TCEQ based the 2006 inventory on the 2005 NEI outside Texas and developed a detailed 2006 emission inventory based on local data for Texas. The Carbon Bond 6 chemical mechanism (CB6; Yarwood et al., 2010, 2012) was used.