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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.

The existing TCEQ SIP modeling June 2006 emission inventory was augmented with Fire Emission Inventory from NCAR (FINN) day-specific fire emissions derived from satellite observations (Wiedinmyer et al., 2011), Emissions Database for Global Atmospheric Research (EDGAR) aircraft emissions and lightning NOx (LNOx) emissions. LNOx emissions were calculated using the parameterization of Koo et al. (2010). LNOx emissions were allocated to grid cells where modeled convection occurred using convective precipitation as a proxy for lightning activity and were distributed in the vertical using the profiles of Ott et al. (2010). A description of both inventories is provided in the Supporting information.

CAMx was run for the June 2006 episode, and a model performance evaluation for ozone, NO_2 and NOx was carried out for monitoring sites in Texas and the southeast. The model performance evaluation showed that CAMx generally performed well in simulating ozone in urban areas of East Texas, but had an overall high bias (see Supporting information; we refer to this simulation as the initial CAMx run).

2.3. Processing of retrieval data

We began with the NASA SP2 and DOMINO Level II datasets. Because the presence of clouds can cause significant bias in NO_2 columns (~60%; Boersma et al., 2004), we used only pixels with a cloud radiance fraction less than a threshold of 0.3 (Boersma et al., 2011). Pixels flagged as potentially containing artifacts due to ice, sun glint, eclipses, etc. were also discarded.

In order to compare the modeled CAMx NO_2 columns with the retrieved columns and use the mass balance method to prepare top–down emissions estimates, it is necessary to apply the OMI averaging kernel (AK) to the CAMx NO_2 VCD profiles before integrating the CAMx VCD through the depth of the troposphere to produce the final modeled VCD estimate (Rodgers, 2000; Eskes and Boersma, 2003; Boersma et al., 2011). The DOMINO v2.0 product provides the AK for each pixel. The NASA SP2 product provides scattering weights from which the AK can be calculated. We calculated AKs for the NASA SP2 retrieval for all pixels within the OMI swaths used for the June 2006 episode (see Supporting information for description of method). Next, we applied the AKs for each retrieval to the CAMx output to produce two sets of modeled VCD. The first was a set of CAMx modeled VCD to which the DOMINO AK had been applied; this set of columns was compared to the DOMINO retrieved columns. The second was a set of CAMx modeled VCD to which the SP2 AK had been applied; this set of columns was compared to the SP2 retrieved columns.

We compared the retrieved OMI tropospheric VCD with CAMx modeled VCD. Across the eastern U.S. and throughout the modeling episode, CAMx NO_2 columns were consistently lower than the OMI NO_2 columns from both retrievals (not shown). A low bias relative to OMI NO_2 columns has been noted for other regional air quality models (Huijnen et al., 2010; Allen et al., 2012). CAMx surface performance for NO_2 was evaluated against research-grade surface NO_2 measurements at SouthEastern Aerosol Research and Characterization (SEARCH; Hansen et al., 2003) monitoring sites in the southeast as well as against INTEX-A NO_2 measurements from the DC-8 aircraft (Singh et al., 2007) (see Supporting information). The evaluation showed that CAMx did not have a persistent low bias relative to SEARCH NO_2 measurements; this suggested that the surface NOx emission inventory was not the cause for the CAMx NO_2 column low bias relative to OMI NO_2 columns. The model had a low bias for upper tropospheric NO_2, which we addressed through addition of downward transport of NOy and ozone from the stratosphere, an updated aircraft emission inventory, a new version of the CB6 chemical mechanism (Yarwood et al., 2012), and parameterized vertical transport of chemical species within clouds. The effects of each of these changes were evaluated separately and are discussed in the Supporting information. The model updates that had the largest effect on the vertical profiles of NOy in the upper troposphere and tropospheric NO_2 VCD were the addition of a stratospheric NOy source and a more detailed aircraft emission inventory.

Following these changes, NO_2 performance in the upper troposphere improved, with the modeled NO_2 profile taking on the "C" shape seen in the observations, and a reduction in normalized bias near the tropopause from −66% to −23%. This improved agreement allowed the study to move forward, but does not demonstrate that the CAMx NO_2 columns are correct or that they show improved agreement with the retrieved VCD for the right reasons.

3. Results

3.1. Comparison of the DOMINO and SP2 episode average retrieved NO_2 VCD

Once the final model run had been completed, we recalculated the CAMx VCD and compared the modeled and retrieved columns (Fig. 1). The overall patterns of VCD are similar for the DOMINO and SP2 retrievals, with generally higher VCD over urban areas such as Chicago, Houston and the northeast corridor, and lower VCD over water and rural areas. The left panel of Fig. 2 compares the DOMINO and SP2 episode average retrieved VCD for all grid cells. The retrievals are well-correlated ($r^2 = 0.81$), with the DOMINO retrieval showing larger VCD for grid cells with the highest VCD located in the New York and Chicago urban areas.

There are significant differences between the two retrievals, most notably over the southeast. Fig. 1 shows that the SP2 retrieval has higher VCD over much of the southeast, including the Atlanta area, whose maximum is higher in SP2 than in DOMINO. The right hand panel of Fig. 2 indicates that the two retrievals are reasonably well-correlated over the southeast ($r^2 = 0.55$) but the SP2 VCD are generally larger than the DOMINO VCD. The reason for this difference is not clear.

Comparison of the CAMx VCD following application of the SP2 and DOMINO AKs (Fig. 1) shows that the modeled VCD has similar patterns of high and low VCD to the retrieved VCD, with higher VCD over urban areas than in rural regions and higher VCD over the continent than offshore. Fig. 3 compares the retrieved VCD with CAMx modeled VCD using DOMINO and SP2 AKs. The CAMx VCD is well-correlated with both retrievals ($r^2 = 0.77$ and 0.78 for DOMINO and SP2 retrievals, respectively) with better agreement for lower values of the retrieved VCD. Using both the DOMINO and SP2 AKs, CAMx has urban maxima that are more intense than in the retrievals (e.g. Houston, Atlanta, Chicago, Northeast corridor). This is likely because CAMx has higher spatial resolution than the models used to develop the retrievals and better resolves regions of high urban NO_2.

When the ratios (Retrieved VCD/CAMx VCD) are taken using the two retrievals (right panels of Fig. 1), striking differences emerge. In the southeast, the area of interest for the NOx emission inventory assessment, the SP2/CAMx VCD ratio is generally >1 outside urban areas. Outside urban areas, the DOMINO/CAMx VCD ratio is generally close to 1 or <1 in the southeast. This difference in the retrieval/CAMx ratios occurs because the DOMINO VCD is lower than the SP2 retrieval throughout much the southeast (Fig. 2, right panel), while the CAMx VCD is fairly similar in the southeast following application of the SP2 and DOMINO AKs. The differences noted above in the retrieval/CAMx VCD ratios directly affect top–down emissions estimates developed via the mass balance method.

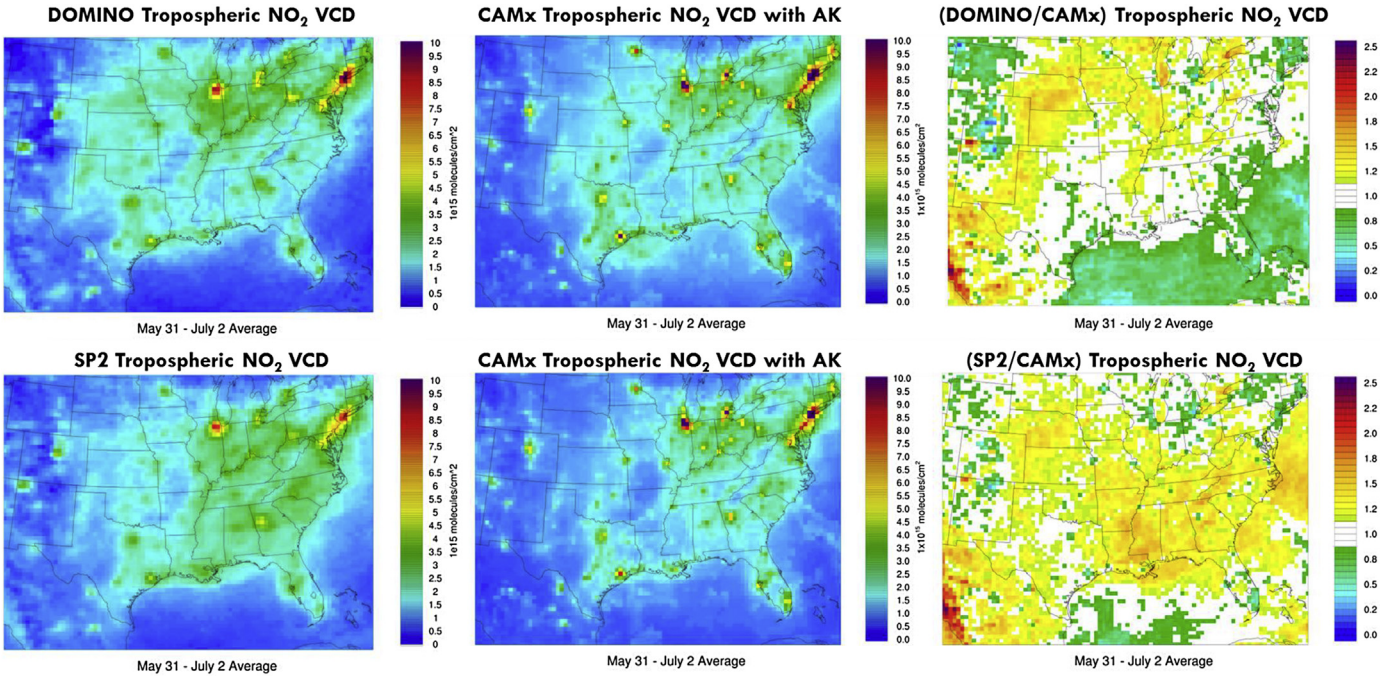

Fig. 1. Upper panels: (Left) DOMINO retrieved episode average tropospheric NO_2 VCD. (Middle) CAMx episode average tropospheric NO_2 VCD using DOMINO averaging kernel (AK). (Right) Ratio of DOMINO to CAMx episode average tropospheric NO_2 VCD. Lower panels: (Left) SP2 retrieved episode average tropospheric NO_2 VCD. (Middle) CAMx episode average tropospheric NO_2 VCD using SP2 AK. (Right) Ratio of SP2 to CAMx episode average tropospheric NO_2 VCD.

3.2. Top–down NOx emission inventory estimates

We developed two separate top–down NOx emission inventories using CAMx together with the DOMINO and SP2 retrievals. For the DOMINO case, the top–down emissions estimates were calculated for each grid cell using:

$$(Top\ Down\ NOx\ Emissions)_{DOMINO} = \left(\frac{\Omega_{DOMINO}}{\Omega_{CAMx}^{DOMINO}}\right) \times TCEQ\ NOx\ Emissions \times Smoothing \quad (2)$$

Ω_{CAMx}^{DOMINO} is the CAMx column with the DOMINO AK applied to it. A second top–down emissions estimate was prepared using the NASA SP2 retrieval:

$$(Top\ Down\ NOx\ Emissions)_{SP2} = \left(\frac{\Omega_{SP2}}{\Omega_{CAMx}^{SP2}}\right) \times TCEQ\ NOx\ Emissions \times Smoothing \quad (3)$$

Ω_{CAMx}^{SP2} is the CAMx column with the NASA SP2 AK applied to it. The two top–down emissions estimates derived using the SP2 and DOMINO retrievals were then compared to the augmented version

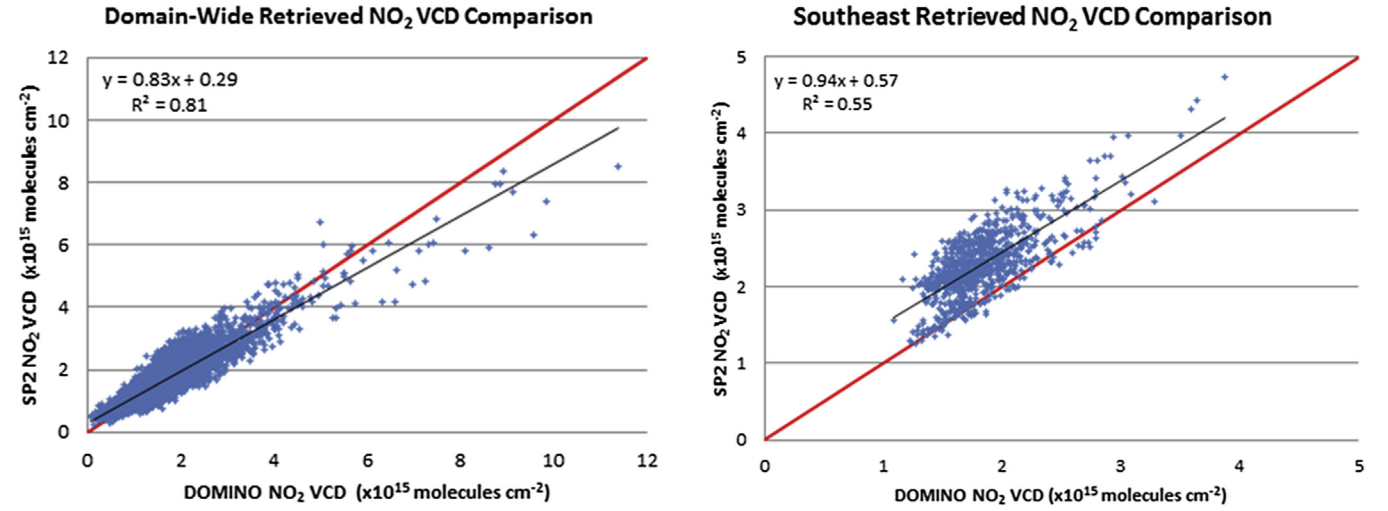

Fig. 2. Comparison of episode average retrieved tropospheric NO_2 VCD for entire domain shown in Fig. 1 (left) and southeast region consisting of Louisiana, Arkansas, Mississippi, Alabama, Georgia, Florida, South Carolina, and Tennessee (right). Red line is 1:1 line. Black line is linear best fit. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

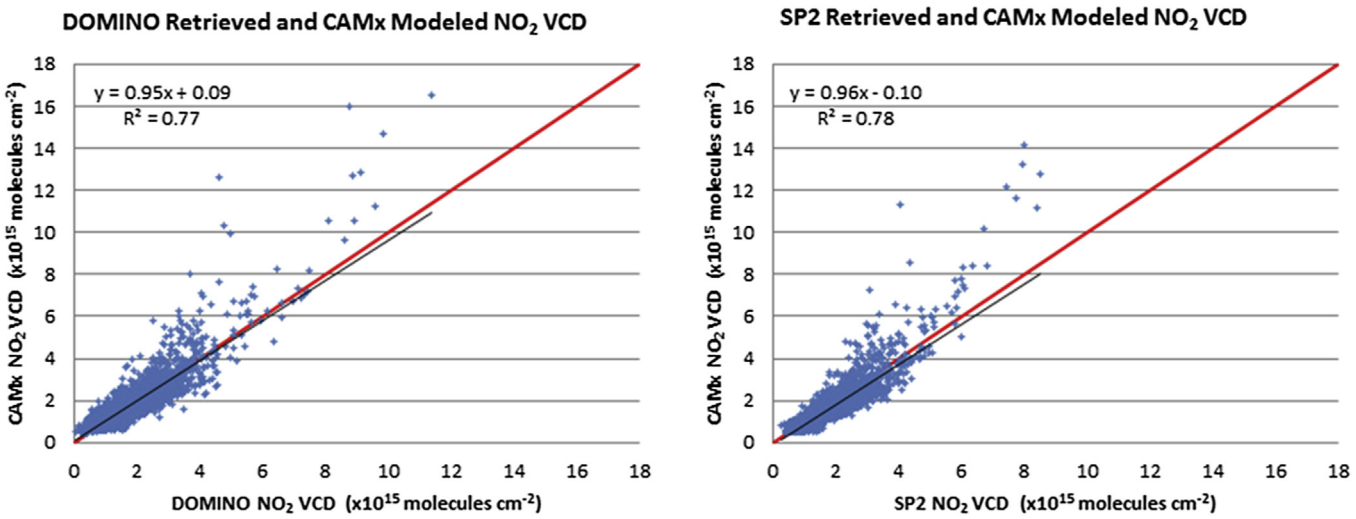

Fig. 3. Comparison of episode average retrieved and modeled tropospheric NO_2 VCD shown in Fig. 1. Left: DOMINO and CAMx VCD. Right: SP2 and CAMx VCD. Red line is 1:1 line. Black line is linear best fit. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

of the TCEQ NOx emission inventory that contains emissions from lightning, wildfires and aircraft.

The lifetime of NOx over the U.S. in summer during OMI overpass hours is on the order of 7 h (Lamsal et al., 2010). For the 36 km grid cells in the TCEQ modeling domain, it is possible that the NO_2 column for a given grid cell may be influenced by NOx emissions from neighboring grid cells. If a grid cell with large NOx emissions lies adjacent to a cell with small NOx emissions, the retrieved NO_2 column in the grid cell with small emissions may be affected by emissions in the cell with large emissions via transport. The top–down emissions estimate will therefore spuriously attribute emissions to the grid cell with small NOx emissions. Others have identified this problem and compensated by spatially smoothing the emission inventories (Martin et al., 2003; Boersma et al., 2008; Tang et al., 2013).

As part of the emission inventory development, we investigated several different methods for spatial smoothing of the bottom–up emission inventory. Ideally, the smoothing would approximate the effect of advecting NOx emissions from the grid cell in which they are emitted into surrounding cells. We determined that the spatial smoothing method that appears in the literature (e.g. Boersma et al., 2008; Tang et al., 2013) does not reproduce the expected characteristics of a NOx emissions field smoothed by advection and gives top–down emission inventory results that contain numerical artifacts and are not useful in constraining the TCEQ's SIP modeling emission inventory. Therefore, the smoothing factor in equation (1) was set to 1 in the results that are presented here, although the lack of smoothing introduces error into the top–down emissions estimates through attribution of NO_2 to grid cells adjacent to sources of NOx emissions. The magnitude of this error is dependent on the intensity of the NOx emissions and the local wind field, and we discuss its effect on the results of this study below.

The TCEQ column-integrated episode average bottom–up NOx emissions are shown in Fig. 4(a). The NOx emissions show maxima coincident with large cities and major point sources such as the power plants in the Four Corners Area. Emissions from aircraft and ships are evident over the Gulf of Mexico and the Atlantic Ocean. Fig. 4(b) presents the difference between the top–down NOx emission inventories produced using equations (2) and (3) and the bottom–up NOx emission inventory shown in Fig. 4(a). The two lower panels of Fig. 4 show regions where the OMI data may indicate biases in the TCEQ bottom–up inventory. There are regions where the top–down NOx emission inventories are consistent in their difference from the bottom–up NOx inventory, such as Dallas, Houston and Miami. Over the southeast, however, the two top–-down inventories are not consistent in their differences from the bottom–up inventory. The top–down inventory developed using CAMx and the SP2 retrieval (EI_{SP2}; lower right panel) has higher NOx emissions than the TCEQ bottom–up inventory across broad areas of the southeast, while the top–down inventory developed using CAMx and the DOMINO retrieval (EI_{DOMINO}) is either generally comparable to (Mississippi, Louisiana, Alabama, Georgia) or is lower than (North and South Carolina) the bottom–up inventory.

Fig. 5 compares the two top–down emission inventories over the southeast. The two inventories are very well-correlated ($r^2 = 0.99$), as expected given that they are both based on the same CAMx simulation and bottom–up inventory, but EI_{SP2} is consistently higher than EI_{DOMINO}.

It is not known which top–down inventory is more accurate, so it is not possible to determine whether there is a systematic bias in the bottom–up NOx emission inventory based on these results.

As noted above, the top–down emission inventories were calculated without spatial smoothing. The inventories are influenced only by the ratio of modeled columns to satellite columns, and may contain error due to attribution of emissions into grid cells near NOx sources due to advection of NOx out of the grid cells into which they were emitted. Previous studies have employed spatial smoothing to approximate the advection of NOx emissions from the grid cell in which they are emitted into surrounding cells. Our analysis was for a month-long period over which time local wind fields varied substantially in response to many factors. Rather than attempting to use smoothing factors to describe effects of varying wind fields, we took a different approach of integrating comparisons over geographic areas (states) that are sufficiently large as to encompass urban areas that could require smoothing. Integrating over states has the advantage of comparability with emission inventories. We performed a state-level comparison of the VCD and top–down and bottom–up inventories to minimize the effect of the attribution error described above. The method and results are described in the Supporting information, and the results are consistent with those shown in Figs. 1–5. We conclude that this attribution error does not significantly affect our emission inventory results over the southeast.

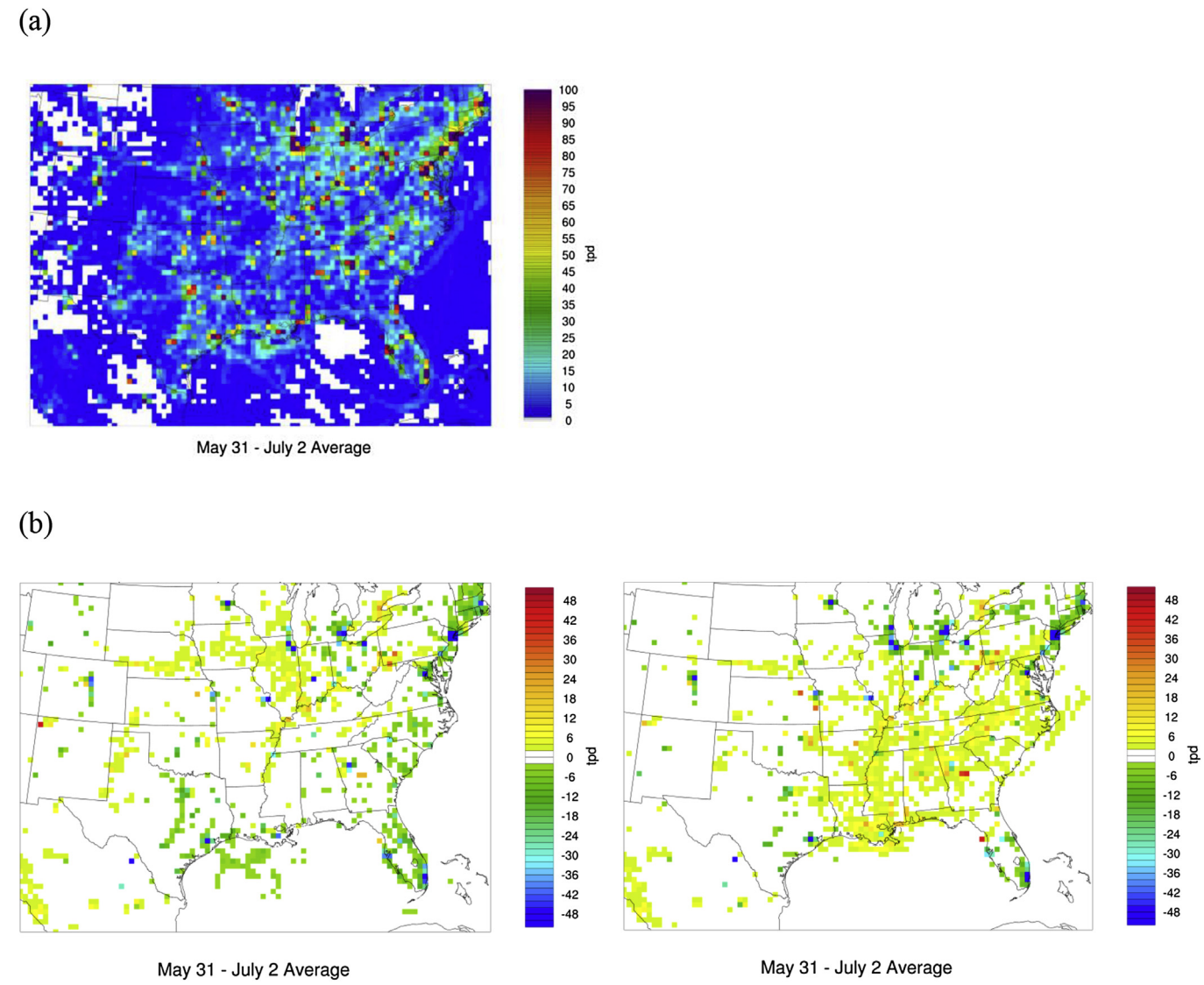

Fig. 4. (a): Episode average emissions for the TCEQ June 2006 episode NOx emission inventory (bottom–up NOx emission inventory). (b): Top–down NOx emission inventory – bottom–up NOx emission inventory using CAMx with DOMINO (lower left panel) and SP2 (lower right panel) retrievals. No smoothing was performed in the calculation of these emissions estimates. Emissions estimates are in tons per day (tpd).

4. Discussion

We evaluated the feasibility of using DOMINO and SP2 tropospheric NO_2 column retrievals together with CAMx NO_2 columns to make two top–down NOx emissions estimates for the southeast to constrain the TCEQ's SIP modeling NOx emission inventories. The two sets of NOx emissions estimates were found to have significant differences over the southeast. These differences were large enough to confound efforts to gain information from the top–down estimates that could constrain the TCEQ inventory and stemmed from differences in the SP2 and DOMINO retrieved VCD over 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. Determining the factor(s) that cause differences between retrievals is an area for future research and refinement of the retrievals.

This study points out the dependence of top–down emission inventory results on the choice of retrieval. A technique that is becoming more prevalent in the application of the mass balance method is to use the air quality model's NO_2 profile to calculate a new atmospheric mass factor, essentially developing a new retrieval which has as its *a priori* NO_2 profile the modeled NO_2 profile shape (e.g. Lamsal et al., 2010; Tang et al., 2014; Vinken et al., 2014). Lamsal et al. (2010) note that this method allows a more consistent comparison with the air quality model. The utility of developing such a retrieval depends on confidence in the NOx emission inventory and the model's simulation of NO_2 and its reservoir species. This study points out the dependence of the retrievals on the assumptions that go into them and reflects the fact that they are a blend of measurements and modeled data and should be used with caution in analyses that will inform policy development.

This study also shows how satellite data used together with aircraft measurements can be a powerful tool for evaluating and improving air quality models. Underestimates of retrieved VCD in CAMx noted in the comparison of the initial CAMx modeled NO_2

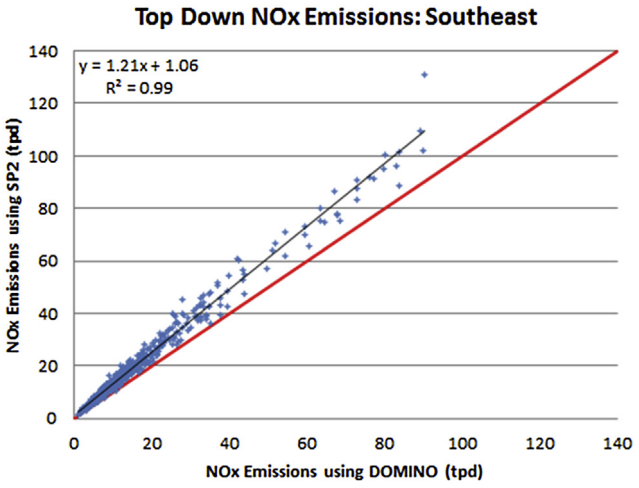

Fig. 5. Comparison of top–down NOx emissions calculated using CAMx together with DOMINO and SP2 retrievals for southeast region consisting of Louisiana, Arkansas, Mississippi, Alabama, Georgia, Florida, South Carolina, and Tennessee. Red line is 1:1 line. Black line is linear best fit. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

columns with retrieved columns led to the first evaluation of CAMx against aircraft observations of the upper troposphere. This comparison showed that, like other regional and global models, CAMx underestimated NO_2, NOx, and NOy in the upper troposphere. Once this bias had been identified, we were able to take steps to reduce it, allowing the model to be used in further comparisons with satellite NO_2 VCD retrievals.

The addition of transport of NOy from the stratosphere to the troposphere was the model update that had the largest effect on the upper tropospheric NO_2 and VCD, and spurred development of CAMx capability to read in a spatially/temporally varying top concentration obtained from a global model simulation. This allows for an improved simulation of cross-tropopause transport and upper tropospheric NOy. Another need identified was for CAMx to include a simulation of vertical transport of chemical species by deep convection. Given the importance of convection in venting polluted air from the surface layer and transporting chemical species to the free troposphere, it is critical that a physically-based parameterization of convective transport of chemical species be integrated into CAMx. This work is underway. With these important components of the simulation of the upper troposphere in place, the CAMx model will be better able to simulate NO_2 columns required for top–down emissions estimation. Models that do not contain an accurate simulation of processes affecting the upper troposphere, such as cross-tropopause transport and a detailed characterization of aircraft cruise emissions, should not be used for emissions evaluation using column-integrated satellite products.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2015.07.002.

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