Inferring the anthropogenic NO\textsubscript{x} emission trend over the United States during 2003 - 2017 from satellite observations: Was there a flattening of the emission trend after the Great Recession?

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Abstract

We illustrate the nonlinear relationships among anthropogenic NO\textsubscript{x} emissions, NO\textsubscript{2} tropospheric vertical column densities (TVCDs), and NO\textsubscript{2} surface concentrations using model simulations for July 2011 over the contiguous United States (CONUS). The variations of NO\textsubscript{2} surface concentrations and TVCDs are generally consistent and reflect well anthropogenic NO\textsubscript{x} emission variations for high-anthropogenic-NO\textsubscript{x} emission regions. For low-anthropogenic-NO\textsubscript{x} emission regions, however, nonlinearity in the emission-TVCD relationship makes it difficult to use satellite observations to infer anthropogenic NO\textsubscript{x} emission changes. The analysis is extended to 2003 – 2017. Similar variations of NO\textsubscript{2} surface measurements and coincident satellite NO\textsubscript{2} TVCDs over urban regions are in sharp contrast to the large variation differences between surface and satellite observations over rural regions. We find a continuous decrease of anthropogenic NO\textsubscript{x} emissions after 2011 by examining surface and satellite measurements in CONUS urban regions, but the decreasing rate is lower by 9\% - 46\% than the pre-2011 period.
1. Introduction

Anthropogenic emissions of nitrogen oxides (NO$_x$ = NO$_2$ + NO) adversely affect the environment, not only because of their direct detrimental impacts on human health (Greenberg et al., 2016; Greenberg et al., 2017; Heinrich et al., 2013; Weinmayr et al., 2009), but also their fundamental roles in the formation of ozone, acid rain, and fine particles which are unfavorable to human health, ecosystem stabilities, and climate change (Crouse et al., 2015; Kampa and Castanas, 2008; Myhre et al., 2013; Pandey et al., 2005; Singh and Agrawal, 2007). About 48.8 Tg N yr$^{-1}$ of NO$_x$ are emitted globally from both anthropogenic (77%) and natural (23%) sources, such as fossil fuel combustion, biomass and biofuel burning, soil bacteria, and lightning (Seinfeld and Pandis, 2016). 3.85 Tg N and 0.24 Tg N of anthropogenic and natural NO$_x$, respectively, were emitted from the U.S. in 2014 on the basis of the 2014 National Emission Inventory (NEI2014); vehicle sources and fuel combustions accounted for 93% of the total anthropogenic NO$_x$ emissions (EPA, 2017).

The U.S. anthropogenic NO$_x$ emissions during the 2010s declined dramatically compared to the mid-2000s (EPA, 2018; Xing et al., 2013) due to stricter air quality regulations and emission control technology improvements, such as the phase-in of Tier II vehicles during 2004 – 2009 and the switch of power plants from coal to natural gas (De Gouw et al., 2014; McDonald et al., 2018). The overall reduction (about 30% - 50%) of anthropogenic NO$_x$ emissions from the mid-2000s to the 2010s was corroborated by observed decreasing of vehicle NO$_x$ emission factors, NO$_2$ surface concentrations, nitrate wet deposition flux, and NO$_2$ tropospheric vertical column densities (TVCDs) (Bishop and Stedman, 2015; Li et al., 2018; McDonald et al., 2018; Miyazaki et al., 2017; Russell et al., 2012; Tong et al., 2015). However, the detailed NO$_x$ emission changes after the Great Recession (from December 2007 to June 2009) are highly uncertain. On the one hand, the U.S. Environmental Protection Agency (EPA) estimated that the Great Recession had a
slight impact on the anthropogenic NO\textsubscript{x} emission trend, and the anthropogenic NO\textsubscript{x} emissions decreased steadily from 2002 to 2017 (Figure S1), although the emission decrease rate slowed down by about 20\% after 2010 (-5.8\% yr\textsuperscript{-1} for 2002 – 2010, and -4.7\% yr\textsuperscript{-1} for 2010 – 2017, Table 1) (EPA, 2018). Fuel-based emission estimates in Los Angeles also showed a steady decrease of anthropogenic NO\textsubscript{x} emissions after 2000 and a small impact of the Great Recession on anthropogenic NO\textsubscript{x} emission decrease trend (Hassler et al., 2016). The continuous decrease of anthropogenic NO\textsubscript{x} emissions was consistent with the ongoing reduction of vehicle emissions (McDonald et al., 2018). On the other hand, Miyazaki et al. (2017) and Jiang et al. (2018) found that the U.S. NO\textsubscript{x} emissions derived from satellite NO\textsubscript{2} TVCDs, including OMI (the Ozone Monitoring Instrument), SCIAMACHY (SCanning Imaging Absorption SpectroMeter for Atmospheric CHartography), and GOME-2A (Global Ozone Monitoring Experiment – 2 onboard METOP-A), were almost flat from 2010 - 2015 and suggested that the decrease of NO\textsubscript{x} emissions was only significant before 2010, which was completely different from the bottom-up and fuel-based emission estimates.

A complicating factor in inferring anthropogenic NO\textsubscript{x} emission trends from the observations of NO\textsubscript{2} surface concentrations and satellite NO\textsubscript{2} TVCDs is the nonlinearity in NO\textsubscript{x} chemistry (Gu et al., 2013; Gu et al., 2016). Although the decrease rates of both NO\textsubscript{2} surface concentrations and coincident OMI NO\textsubscript{2} TVCDs slowed down after the Great Recession over the United States, Tong et al. (2015), Lamsal et al. (2015) and Jiang et al. (2018) found that the slowdown of the decrease rates derived from NO\textsubscript{2} surface concentrations is 12\% - 79\% less than those of NO\textsubscript{2} TVCDs (Table 1). Secondly, the slowdown of the decrease rates of NO\textsubscript{2} surface concentrations and OMI TVCDs over cities and power plants (Russell et al., 2012; Tong et al., 2015) is significantly less than those over the whole contiguous United States (CONUS) (Jiang et al., 2018; Lamsal et al., 2015). Moreover, Zhang et al. (2018) found that filtering out lightning-
affected measurements could significantly improve the comparison of NO$_2$ surface concentration and OMI NO$_2$ TVCD trends over the CONUS.

In this study, we carefully investigate the relationships among anthropogenic NO$_x$ emissions, NO$_2$ surface concentrations, and NO$_2$ TVCDs over the CONUS and evaluate the impact of the relationships on inferring anthropogenic NO$_x$ emission changes and trends from surface and satellite observations. Section 2 describes the model and datasets used in this study, including the Regional chEmistry and trAnsport Model (REAM), the EPA Air Quality System (AQS) NO$_2$ surface observations, and NO$_2$ TVCD products from OMI, GOME-2A, GOME-2B (GOME2 onboard METOP-B), and SCIAMACHY. In Section 3, we examine the nonlinear relationships among anthropogenic NO$_x$ emissions, NO$_2$ surface concentrations, and NO$_2$ TVCDs using model simulations. Accounting for the effects of chemical nonlinearity, we then investigate the anthropogenic NO$_x$ emission trends and changes from 2003 – 2017 over the CONUS. Finally, section 4 gives a summary of the study.

2. Model and Data Description

2.1 REAM

The REAM model has been applied and evaluated in many research applications including ozone simulation and forecast, emission inversion and evaluations, and mechanical studies of chemical and physical processes (Alkuwari et al., 2013; Cheng et al., 2017; Cheng et al., 2018; Choi et al., 2008a; Choi et al., 2008b; Gu et al., 2013; Gu et al., 2014; Koo et al., 2012; Liu et al., 2012; Liu et al., 2014; Wang et al., 2007; Yang et al., 2011; Zhang et al., 2017; Zhang et al., 2018; Zhang and Wang, 2016; Zhao and Wang, 2009; Zhao et al., 2009a; Zhao et al., 2010). REAM used in this work has 30 vertical layers in the troposphere, and the horizontal resolution is $36 \times 36$ km$^2$. The model is driven by meteorology fields from a Weather and Research
Forecasting (WRF, version 3.6) model simulation initialized and constrained by the NCEP coupled forecast system model version 2 (CFSv2) products (Saha et al., 2011). The chemistry mechanism is based on GEOS-Chem v11.01 with updated reaction rates and aerosol uptake of isoprene nitrates (Fisher et al., 2016). Chemistry boundary conditions and initializations are from a GEOS-Chem (2° × 2.5°) simulation. Hourly anthropogenic emissions on weekdays are based on the 2011 National Emission Inventory (NEI2011), while weekend anthropogenic emissions are set to be two-thirds of the weekday emissions (Beirle et al., 2003; Choi et al., 2012). Biogenic VOC emissions are estimated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) v2.10 (Guenther et al., 2012). NOx emissions from soils are based on the Yienger and Levy (YL) scheme (Li et al., 2019; Yienger and Levy, 1995).

2.2 Satellite NO2 TVCDs

In this study, we use NO2 TVCD products from four satellite measurements in the past decade, including SCIAMACHY, GOME-2A, GOME-2B, and OMI, the spectrometers onboard sun-synchronous satellites to monitor atmospheric trace gases. The SCIAMACHY onboard the Environmental Satellite (ENVISAT) has an equator overpass time of 10:00 Local time (LT) and a nadir pixel resolution of 60 × 30 km². The GOME-2 instruments on Metop-A (named as GOME-2A) and Metop-B (GOME-2B) satellites cross the equator at 9:30 LT and have a nadir resolution of 80 × 40 km². After July 15, 2013, the nadir resolution of GOME-2A became 40 × 40 km² with a smaller scanning swath. The OMI onboard the EOS-Aura satellite has a nadir resolution of 24 × 13 km² and overpasses the equator around 13:45 LT. More detailed information about these instruments is summarized in Table S1. These instruments measure transmitted, backscattered, and reflected radiation from the atmosphere in the ultraviolet and visible wavelength. The radiation measurements in the wavelength of 402 - 465 nm are then used to retrieve NO2 VCDs.

The retrieval process consists of three steps: 1) converting radiation observations to NO2 slant column densities (SCDs) by using the Differential Optical Absorption Spectroscopy (DOAS)
spectral fitting method; 2) separating tropospheric SCDs and stratospheric SCDs from the total NO\textsubscript{2} SCDs; 3) dividing the NO\textsubscript{2} tropospheric SCDs by the tropospheric air mass factors (AMF) to compute VCDs.

The product archives we use in this study include GOME-2B (TM4NO2A v2.3), SCIAMACHY (QA4ECV v1.1), GOME-2A (QA4ECV v1.1), OMI (QA4ECV v1.1, hereafter referred to as OMI-QA4ECV), OMNO2 (SPv3, hereafter referred to as OMI-NASA), and the Berkeley High-Resolution NO\textsubscript{2} products (v3.0B, hereafter referred to as OMI-BEHR). OMI-BEHR uses the tropospheric SCDs from OMI-NASA products but updates some inputs for the tropospheric AMF calculation (Laughner et al., 2018). These product archives have been previously validated (Boersma et al., 2018; Drosoglou et al., 2017; Drosoglou et al., 2018; Krotkov et al., 2017; Laughner et al., 2018; Wang et al., 2017; Zara et al., 2018). Generally, the pixel-size uncertainties of these products are > 30\% over polluted regions under clear-sky conditions. We summarize the basic information about these products in Table S2. To keep the high quality and sampling consistency of NO\textsubscript{2} TVCD datasets, we chose pixel-size NO\textsubscript{2} TVCD data using the criteria listed in Table S3. After the selection, we re-grid the pixel-size data into the REAM 36 × 36 km\textsuperscript{2} grid cells and calculate the seasonal means of each grid cell with corresponding daily values on weekdays (winter: January, February, and December; spring: March, April, and May; summer: June, July, and Autumn; autumn: September, October, and November). We excluded weekend data in this study to minimize the impacts of weekend NO\textsubscript{x} emission reduction, leading to different NO\textsubscript{2} TVCDs between weekdays and weekends (Figure S2).

Satellite TVCD measurements can show large variations and apparent discontinuities due in part to the effects of cloud, lightning NO\textsubscript{x}, the shift of satellite pixel coverage, and retrieval uncertainties (Figure S2; e.g., (Boersma et al., 2018; Zhang et al., 2018)). However, continuous and consistent measurements are required for reliable trend analyses. In addition to the criteria of...
data selection in Table S3, we compute the seasonal relative 90th percentile confidence interval, defined as $\text{RCI} = (X(95\text{th percentile}) - X(5\text{th percentile})) / \text{mean}(X)$, where $X$ is the daily NO$_2$ TVCD for a given season. To compute the seasonal trend, we require that RCI is < 50% for the selected season every year in the analysis period (Table S3). About 45% of data are removed as a result.

2.3 Surface NO$_2$ measurements

Hourly surface NO$_2$ measurements from 2003 - 2017 are from the EPA AQS monitoring network (archived on https://www.epa.gov/outdoor-air-quality-data). Most AQS monitoring sites use the Federal Reference Method (FRM) — gas-phase chemiluminescence to measure NO$_2$. Few sites use the Federal Equivalent Method (FEM) — photolytic-chemiluminescence or the Cavity Attenuated Phase Shift Spectroscopy (CAPS) method. FRM and FEM are indirect methods, in which NO$_2$ is first converted to NO and then NO is measured through chemiluminescence measurement of NO$_2^*$ produced by NO + O$_3$. The difference is that FRM uses heated reducers/catalysts for the conversion of NO$_2$ to NO and FEM uses photolysis of NO$_2$ to NO. The conversion to NO in the FRM instruments is not specific to NO$_2$, and non-NO, active nitrogen compounds (NO$_x$) can also be reduced by the catalysts, which would cause high biases of NO$_2$ measurements, while the FEM method is sensitive to the photolysis conversion efficiency of NO$_2$ to NO (Beaver et al., 2012; Beaver et al., 2013; Lamsal et al., 2015). The CAPS method directly determines NO$_2$ concentrations based on a NO$_2$-induced phase shift measured by a photodetector. The CAPS instrument operates at a wavelength of about 450 nm and may overestimate NO$_2$ concentrations due to absorption of other molecules at the same wavelength (Beaver et al., 2012; Beaver et al., 2013; Kebabian et al., 2005).

Due to the different characteristics of the above three methods and demonstrated biases between the FRM and the FEM by Lamsal et al. (2015), we firstly investigate the measurement
There are discrepancies among the above three methods. There are three sites having FRM and FEM measurements simultaneously during some periods from 2013 - 2014, two sites having both FRM and CAPS data during some periods from 2015 – 2016, and one site using all three measurement methods during some periods in 2015. Figure S3 shows the hourly averaged ratios of FEM and CAPS to FRM data, respectively, for 4 seasons during 2013 – 2016. The CAPS/FRM ratios are in the range of 0.94 – 1.06 and the FEM/FRM ratios of 0.86 – 1.11. Furthermore, Zhang et al. (2018) discussed that the relative trends are not affected by scaling the observation data. As in the work by Zhang et al. (2018), we analyze the relative trends in the surface NO$_2$ data. We, therefore, did not scale the FRM data. At sites with FEM or CAPS measurements, we use these measurements in place of FRM data. If both FEM and CAPS data are available, we use the averages of the two datasets.

Since NO$_2$ surface concentrations have significant diurnal variations (Figure S4), we choose the data at 9:00-10:00 LT for comparison with GOME-2A/2B data, 10:00-11:00 LT for comparison with SCIAMACHY data, and 13:00-14:00 LT for OMI data. The seasonal $RCI < 50\%$ requirement is also used here to be consistent with the analysis of satellite TVCD data. We also require that the measurement site must have valid measurements in the aforementioned 3 hours for at least one season from 2003 – 2017. The locations of the 179 selected sites using the site selection criteria are shown in Figure 1. The region definitions follow the U.S. Census Bureau (https://www2.census.gov/geo/pdfs/maps-data/maps/reference/us_regdiv.pdf).
3. Results and Discussions

3.1 Nonlinear relationships among anthropogenic NOx emissions, NO2 surface concentrations, and NO2 TVCDs

NO2 surface concentrations and NO2 TVCD are not linearly correlated with NOx emissions due in part to chemical nonlinearity (Gu et al., 2013; Lamsal et al., 2011). Therefore, it is necessary to first investigate the nonlinearities among NOx emissions, NO2 surface concentrations, and TVCDs over the CONUS before we compare the trends between NO2 surface concentrations and TVCDs. The nonlinearity between NOx emission and NO2 TVCD is analyzed by examining the local sensitivity of NO2 TVCD to NOx emissions (Gu et al., 2013; Lamsal et al., 2011; Tong et al., 2015), which is defined as $\beta$ in Equation (1). We further define $\gamma$ as the sensitivity of NO2 surface concentration to NOx emission:

\[
\frac{\Delta E}{E} = \beta \frac{\Delta \Omega}{\Omega}
\]

\[
\frac{\Delta E}{E} = \gamma \frac{\Delta c}{c}
\]

where $E$ denotes NOx emission and $\Delta E$ denotes the change of NOx emission; $\Omega$ denotes NO2 TVCD, $c$ denotes surface NO2 concentration, and $\Delta \Omega$ and $\Delta c$ denote the corresponding changes.

We computed $\beta$ and $\gamma$ values for July 2011 over the CONUS using REAM. To compute local $\beta$ and $\gamma$ values, we added another independent group of chemistry species ("group 2") in REAM in order to compute the standard and sensitivity simulations concurrently. The original chemical species in the model ("group 1") were used in the standard simulation. For group 2 chemical species, anthropogenic NOx emissions were reduced by 15%. In model simulation, we first computed the advection of group 1 tracers. The horizontal tracer fluxes were therefore
available. All influxes into a grid cell for group 2 tracer simulation were from group 1 tracer simulation; only outfluxes were computed using group 2 tracers. The outflux was one way in that nitrogen species were transported out but the transport did not affect adjacent grid cells because the influxes were from group 1 tracer simulation. Using this procedure, the effects of anthropogenic NO$_x$ emission reduction were localized. The $\beta$ and $\gamma$ values were computed by the ratio of TVCD and surface concentration changes to 15% change of anthropogenic NO$_x$ emissions, respectively.

Figure 2 shows the distributions of our $\beta$ and $\gamma$ ratios as a function of anthropogenic NO$_x$ emissions for July 2011 over the CONUS. Results essentially the same as Figure 2 were obtained when a perturbation of 10% was used for anthropogenic NO$_x$ emissions. While the model simulation is for one summer month, several key points on the surface and column concentration sensitivities to anthropogenic NO$_x$ emissions have implications for comparing the trends of AQS and satellite TVCD data. (1) Both $\beta$ and $\gamma$ values are negatively correlated with anthropogenic NO$_x$ emissions due to chemical nonlinearity and background NO$_x$ contributions (Gu et al., 2016; Lamsal et al., 2011). It is consistent with the distribution of $\beta$ as a function of NO$_x$ emissions in China (Gu et al., 2013), although the $\beta$ ratios for the US are generally larger than for China due primarily to different emission distributions of NO$_x$ and VOCs and regional circulation patterns (Zhao et al., 2009b). (2) The uncertainties of $\beta$ and $\gamma$ values increase significantly as anthropogenic NO$_x$ emissions decrease, which means regions with low anthropogenic NO$_x$ emissions are more sensitive to environmental conditions, such as NO$_x$ transport from nearby regions which may even produce negative $\beta$ and $\gamma$ values. (3) The value of $\gamma$ is generally less than $\beta$, especially for low-anthropogenic-NO$_x$ emission regions, which reflects the significant contribution of free tropospheric NO$_2$ to NO$_x$ TVCD but not to NO$_2$ surface concentrations. (4) The variations of $\beta$ and $\gamma$ values in anthropogenic NO$_x$ emission bins tend to be larger at 10:00 – 11:00 than at 13:00 – 14:00 LT, reflecting a stronger transport effect due to weaker chemical
losses at 10:00 – 11:00. (5) Both $\beta$ and $\gamma$ values are significantly less than 1 at 13:00 – 14:00 LT ($\beta = 0.74$ and $\gamma = 0.84$) when anthropogenic NO$_x$ emissions are $> 4 \times 10^{12}$ molecules cm$^{-2}$ s$^{-1}$, but they are close to 1 at 10:00 – 11:00 LT ($\beta = 0.96$ and $\gamma = 1.02$), which reflect stronger chemistry nonlinearity at 13:00 – 14:00 than in the morning.

The largely varying $\beta$ and $\gamma$ values for anthropogenic NO$_x$ emissions $< 10^{11}$ molecules cm$^{-2}$ s$^{-1}$ imply that the trends derived from satellite TVCD data do not directly represent anthropogenic NO$_x$ emissions and that the variations of TVCD data may not be comparable to the corresponding surface NO$_2$ concentrations. We define a region “urban” if anthropogenic NO$_x$ emissions are $> 10^{11}$ molecules cm$^{-2}$ s$^{-1}$. All the other regions are defined as “rural”. Figure 3 shows the distributions of anthropogenic NO$_x$ emissions and urban and rural regions defined in this study. Such defined urban regions account for 69.8% of the total anthropogenic NO$_x$ emissions over the CONUS, the trend of which is, therefore, representative of anthropogenic emission changes. A caveat is that some “urban” regions would become “rural” if anthropogenic NO$_x$ emissions decreased after 2011 as the EPA anthropogenic NO$_x$ emission trend suggested (Figure S1). In a sensitivity study, we define an urban region using a stricter criterion of anthropogenic NO$_x$ emissions $> 2 \times 10^{11}$ molecules cm$^{-2}$ s$^{-1}$ and the analysis results are similar to those shown in the next section.

3.2 Trend comparisons between NO$_2$ AQS surface concentrations and coincident satellite NO$_2$ tropospheric VCD over urban and rural regions

By using anthropogenic NO$_x$ emissions of $10^{11}$ molecules cm$^{-2}$ s$^{-1}$ as the threshold value, 157 AQS sites are urban, and the rest 22 sites are rural. Their properties are summarized in Table 2. Figure 4 shows the relative annual variations of AQS NO$_2$ surface measurements at 13:00 – 14:00 and coincident OMI-QA4ECV NO$_2$ TVCD data from 2005 – 2017 in each season for urban and rural regions. The contrast between the two regions is apparent in all seasons. For comparison
purposes, we scale the time series of TVCD and AQS surface NO\textsubscript{2} to their corresponding 2005 values, and the resulting data are therefore unitless. Over urban regions, NO\textsubscript{2} surface concentrations are highly correlated with NO\textsubscript{2} TVCDs (TVCD = 1.03 × AQS + 0.11, R\textsuperscript{2} = 0.98), reflecting the comparable and stable β and γ values (Figure 2). However, over rural regions, the scaled TVCD data significantly deviate from AQS NO\textsubscript{2} data (TVCD = 1.15 × AQS + 0.09, R\textsuperscript{2} = 0.87). It is noteworthy that the discrepancies between urban and rural data are smaller in winter than in spring, summer, and autumn due to a more dominant role of transport than chemistry and lower natural NO\textsubscript{x} emissions in winter.

We also examine the correlations of AQS NO\textsubscript{2} surface concentrations with coincident OMI-NASA, OMI-Behr, SCIAMACHY, GOME-2A, and GOME-2B TVCD measurements. The results of OMI-NASA and OMI-Behr are similar to those of OMI-QA4ECV (Figure 4).

SCIAMACHY and GOME-2B TVCD observations at 9:00-11:00 LT also show large contrast between urban (SCIAMACHY: TVCD = 0.92 × AQS - 0.005, R\textsuperscript{2} = 0.94; GOME-2B: TVCD = 0.54 × AQS + 0.56, R\textsuperscript{2} = 0.96) and rural regions (SCIAMACHY: TVCD = 0.77 × AQS +0.83, R\textsuperscript{2} = 0.63; GOME-2B: TVCD = 0.46 × AQS + 0.73, R\textsuperscript{2} = 0.59). The correlation of coincident GOME-2A NO\textsubscript{2} TVCD data with AQS surface concentrations is poor for rural (TVCD = 0.65 × AQS + 0.56, R\textsuperscript{2} = 0.44) and urban (TVCD = 0.31 × AQS + 0.56, R\textsuperscript{2} = 0.21) regions (Figure S5), which likely reflects the degradation of the GOME-2A instrument causing significant increase of NO\textsubscript{2} SCD uncertainties (Boersma et al., 2018). Therefore, we excluded GOME-2A in the analysis hereafter.

We further investigate the sensitivities of OMI-QA4ECV NO\textsubscript{2} TVCD relative annual variations from 2005 - 2017 to different anthropogenic NO\textsubscript{x} emissions over the CONUS in Figure 5. We find clear flattening of NO\textsubscript{2} TVCD variations as anthropogenic NO\textsubscript{x} emissions decrease, which is consistent with the above analysis. Similar to Figure 4, the spread of TVCD variation is much less in winter than the other seasons. The differences between Figures 5 and 4 are due to a
much larger dataset used in the former than the latter. Only coincident AQS and OMI-QA4ECV data are used in Figure 4, but all OMI-KMNI data are used in Figure 5.

3.3 Trend analysis of AQS NO$_2$ surface concentrations, satellite TVCDs, and updated EPA NOx emissions

We first updated the CEMS measurement data used in the EPA NO$_x$ emission trend datasets with the newest datasets obtained from https://ampd.epa.gov/ampd/. As shown in Figure S1, the updated CEMS data lead to a reduction of anthropogenic NO$_x$ emissions during the Great Recession (2008 – 2009) and a recovery period in 2010 – 2011. The sharp drop during the Great Recession and the flattening trend right after the Great Recession are captured by OMI NO$_2$ and SCIAMACHY TVCD products (Figures 4, 6, and S6) and AQS NO$_2$ surface measurements (Figures 4, 6, and S4) and are also noted by Russell et al. (2012) and Tong et al. (2015) (Table 1).

In Figure 6, we show the comparisons among the relative variations of the updated EPA anthropogenic NO$_x$ emissions, AQS NO$_2$ surface measurements at 10:00-11:00 and 13:00-14:00, and coincident satellite NO$_2$ TVCDs for urban regions in 4 seasons from 2003 to 2017. Also shown are the comparisons among the updated EPA anthropogenic NO$_x$ emissions and satellite NO$_2$ TVCDs. There are many more data points for the latter comparison because the data selection is no longer limited to those coincident with the AQS surface data, and therefore, the uncertainty spread is much lower. The comparisons, in general, show consistent results that the updated EPA anthropogenic NO$_x$ emissions, AQS surface measurements, and satellite TVCD data are in agreement. The agreement of decreasing trends among the datasets is just as good for the post-2011 period as the pre-2011 period. This result differs from Miyazaki et al. (2017) and Jiang et al. (2018), who suggested no significant decreasing trend for OMI TVCD data and inversed NO$_x$ emissions after 2010. The disagreement can be explained by the results of Figure 5. Including the low anthropogenic NO$_x$ emission regions leads to underestimates of NO$_x$ decreases.
Since the area of low anthropogenic NO\textsubscript{x} emission regions is larger than high anthropogenic NO\textsubscript{x} emission regions (Table 2), the arithmetic averaging will lead to a large weighting of rural observations, which do not reflect anthropogenic NO\textsubscript{x} emission changes. Miyazaki et al. (2017) and Jiang et al. (2018) included all regions in their analyses, but we exclude rural regions. Figure S6 shows the seasonal variations if the TVCDs over rural regions are included; the result shows a much lower decreasing rate of TVCDs over the CONUS. The much slower satellite TVCD trends for regions with low NO\textsubscript{x} emissions was previously discussed by Zhang et al. (2018). In addition, Miyazaki et al. (2017) and Jiang et al. (2018) conducted NO\textsubscript{x} emission inversions by using the Model for Interdisciplinary Research on Climate (MIROC)-Chem with a coarse resolution of 2.8° \times 2.8°, which was insufficient to separate urban and rural regions and might distort predicted NO\textsubscript{2} TVCDs and inversed NO\textsubscript{x} emissions due to nonlinear effects (Valin et al., 2011; Yu et al., 2016), which is another possible reason for their find of flattening NO\textsubscript{x} emission trends after 2010.

We summarize the decreasing rates of NO\textsubscript{2} after the Great Recession in Table 3. To minimize the effect of the sharp decrease and the subsequent recovery, we chose to analyze the post-2011 period. Table 3 summarizes the results for each season, while Table 1 gives the averaged annual decreasing trends. Generally, Tables 1 and 3 confirm the continuous decreases of AQS surface observations, satellite NO\textsubscript{2} TVCD, and updated EPA anthropogenic NO\textsubscript{x} emissions after 2011 as in Figure 6, but decreasing rates are lower than the pre-2011 period. Over the AQS urban sites, the slowdown magnitudes are 9% for AQS surface observations and 20% - 40% for satellite NO\textsubscript{2} TVCD measurements, which may reflect in part smaller $\gamma$ than $\beta$ values (Table 2). Our estimated slowdown magnitudes are significantly lower than Lamsal et al. (2015) and Jiang et al. (2018) but comparable to the results by Tong et al. (2015) (Table 1). The agreement with Tong et al. (2015) is because we select urban AQS sites based on anthropogenic NO\textsubscript{x} emissions and they chose eight large cities, while Lamsal et al. (2015) and Jiang et al. (2018) used all AQS sites.
Over the CONUS urban regions, updated EPA anthropogenic NO\textsubscript{x} emissions show a slowdown of 22% compared to 29% - 46% for three OMI NO\textsubscript{2} TVCD products. The difference is partially due to the $\beta$ ratio of $2.3 \pm 0.9$ at 13:00 – 14:00 over the CONUS urban regions (Table 2). Satellite NO\textsubscript{2} TVCD measurement uncertainties also contribute to the difference. From 2013 – 2017, GOME-2B NO\textsubscript{2} TVCDs decrease more than OMI products, especially in spring, autumn and winter (Tables 1 and 3). Finally, trend analyses in different regions (Figure 7 and Table S4) indicate that generally, the Midwest has the least slowdown of the decreasing rate for urban OMI NO\textsubscript{2} TVCD (-14% on average) after 2011 compared to the Northeast (-30%), South (-34%), and West (-28%).

The results presented in this study are qualitatively in agreement with the work by Silvern et al. (2019). The two studies were independent. Therefore, the foci of the studies are different despite reaching similar conclusions. While we focused on understanding the detailed data analysis of Jiang et al. (2018) and limited the use of model simulation results so that our results can be compared to the previous study directly, Silvern et al. (2019) relied more on multi-year model simulations. As a result, Silvern et al. (2019) can clearly identify the contributions of the NO\textsubscript{2} columns by natural emissions and make use of additional observations such as nitrate deposition fluxes. They also identified model biases in simulating the trends of NO\textsubscript{2} TVCDs by natural emissions. Our study, on the other hand, explored the data analysis procedure through which the trend of anthropogenic emissions can be derived from satellite observations and its limitations.

### 4. Conclusions

Using model simulations for July 2017, we demonstrate the nonlinear relationship of NO\textsubscript{2} surface concentration and TVCD with anthropogenic NO\textsubscript{x} emissions. Over low anthropogenic NO\textsubscript{x} emission regions, the ratios of anthropogenic NO\textsubscript{x} emission changes to the changes of
surface concentrations ($\gamma$) and TVCDs ($\beta$) have very large variations and $\beta > \gamma \gg 1$.

Therefore, for the same emission changes, surface concentration and TVCD changes are much smaller and variable than urban regions, making it difficult to use the observations to directly infer anthropogenic NO$_x$ emission trends. We find that defining urban regions where anthropogenic NO$_x$ emissions are $> 10^{11}$ molecules cm$^{-2}$ s$^{-1}$ and using surface and TVCD observations over these regions can infer the trends that can be compared with the EPA emission trend estimates.

We evaluate the anthropogenic NO$_x$ emission variations from 2003 – 2017 over the CONUS by using satellite NO$_2$ TVCD products from GOME-2B, SCIAMACHY, OMI-QA4ECV, OMI-NASA, and OMI-BEHR, over the urban regions of CONUS. We find broad agreements among the decreases of AQS NO$_2$ surface observations, satellite NO$_2$ TVCD products, and the EPA anthropogenic NO$_x$ emissions with the CEMS dataset updated. After 2011, they all show a slowdown of the decreasing rates. Over the AQS urban sites, NO$_2$ surface concentrations have a slowdown of 9% and OMI products show a slowdown of 20% - 40%. Over the CONUS urban regions, OMI TVCD products indicate a slowdown of 29% - 46%, and the updated EPA anthropogenic NO$_x$ emissions have a slowdown of 22%. The different slowdown magnitudes between OMI TVCD products and the other two datasets may be caused by the nonlinear response of TVCD to anthropogenic emissions and the uncertainties of satellite measurements (e.g., GOME-2B TVCD data show a larger decreasing trend than OMI products from 2013 – 2017).

We did not find observation evidence supporting the notion that anthropogenic NO$_x$ emissions have not been decreasing after the Great Recession. In future studies, we recommend that the nonlinear relationships of NO$_x$ emissions with NO$_2$ TVCD and surface concentration be
carefully evaluated when applying satellite and surface measurements to infer the changes of anthropogenic NO$_x$ emissions.

**Data availability**

The EPA AQS hourly surface NO$_2$ measurements are downloaded from https://aqs.epa.gov/aqswah/airdata/download_files.html#Raw. QA4ECV 1.1 NO$_2$ VCD products (OMI-QA4ECV, GOME-2A, and SCIAMACHY) are from http://temis.nl/qa4ecv/no2col/data/. GOME-2B NO$_2$ VCD products are from http://www.temis.nl/airpollution/no2col/no2colgome2b.php. OMI-BEHR and OMI-NASA archives are from http://behr.chem.berkeley.edu/DownloadBEHRData.aspx. REAM simulation results for this study are available upon request.

**Author contribution**

JL and YW designed the study. JL conducted model simulations and data analyses with discussions with YW. JL and YW wrote the manuscript.

**Competing interests**

The authors declare that they have no conflict of interest.

**Acknowledgments**

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We updated EPA emissions. Since different studies used different time division methods, we list the period of each study in the table.

<table>
<thead>
<tr>
<th>Studies</th>
<th>Datasets</th>
<th>Time</th>
<th>Period 1</th>
<th>Period 2</th>
<th>Period 3</th>
<th>Trend (yr)^{-1}</th>
<th>Slowdown ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>This study for CONUS “urban” sites</td>
<td>GOME-2B (36 × 36 km^2)</td>
<td>2003 – 2011</td>
<td>-6.3 ± 1.1</td>
<td>2013 - 2017</td>
<td>-8.2 ± 3.0%</td>
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<tr>
<td>SCIAMACHY (36 × 36 km^2)</td>
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<td>-8.6 ± 1.2</td>
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<tr>
<td>OMI-NASA (36 × 36 km^2)</td>
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<td>-8.2 ± 1.3</td>
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<tr>
<td>OMI-BEHRSch (36 × 36 km^2)</td>
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<tr>
<td>Updated EPA NOx emissions</td>
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<td>-6.5 ± 0.8</td>
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<tr>
<td>This study for AQS “urban” sites</td>
<td>GOME-2B (36 × 36 km^2)</td>
<td>2003 - 2011</td>
<td>-7.6 ± 1.1</td>
<td>2013 - 2017</td>
<td>-10.2 ± 2.9%</td>
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<tr>
<td>SCIAMACHY (36 × 36 km^2)</td>
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<td>-9.0 ± 0.8</td>
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<td>OMI-NASA (36 × 36 km^2)</td>
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<td>OMI-BEHRSch (36 × 36 km^2)</td>
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<td>-9.0 ± 0.8</td>
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<td>OMI-SCIAMACHY (36 × 36 km^2)</td>
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<td>-6.5 ± 1.2</td>
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<tr>
<td>(Russel et al., 2012)^8</td>
<td>BEHR v2.1 NOx TVCD (0.05°×0.05°)</td>
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<td>-6.0%</td>
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<tr>
<td>Updated EPA NOx emissions</td>
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<tr>
<td>NASA v2.1 NOx TVCD (pixels &lt; 50 × 24 km^2)</td>
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<tr>
<td>BEHR v2.1 NOx TVCD (pixels &lt; 50 × 24 km^2)</td>
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<td>-7.3% (-7.6%)</td>
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<tr>
<td>NOx surface VMR</td>
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<td>-8.9% (-9.3%)</td>
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<td>Updated EPA NOx emissions</td>
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<td>-7.3% (-7.6%)</td>
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<td>(Lamsal et al., 2015)^10</td>
<td>NASA v2.1 NOx TVCD (0.1°×0.1°)</td>
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<tr>
<td>NOx surface VMR</td>
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<td>Updated EPA NOx emissions</td>
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<tr>
<td>(Jiang et al., 2018)^11</td>
<td>BEHR v2.1 NOx TVCD (0.5°×0.667°)</td>
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<td>NOx surface VMR</td>
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</tbody>
</table>

1. Since different studies used different time division methods, we list the period of each study in the table.
2. Trends are based on an exponential model: (E) \( \left(E_0 \times r^{-1}\right)^n \). "E" denotes the value at year "y" and "E_i" denotes the value at the initial year; "r-1" is the relative trend.
4. Trends in our study are calculated based on the national seasonal trends shown in Table 3.
5. The information on satellite products used in this study is summarized in Table S2.
6. Denote the averaged trends of 13:00 and 10:00 LT based on the values in Table 3.
The study used NO\textsubscript{2} TVCD from urban and power plant grid cells across the U.S. Since previous studies used linear models to calculate trends and the results are sensitive to their calculation methods and the selection of initial years, we recalculate the trends based on the above exponential model, which makes the results consistent. Our results are those bold numbers inside the parentheses, while the numbers in normal fonts are from the original publications.

The study uses NO\textsubscript{2} TVCD and surface concentrations from Los Angeles, Dallas, Houston, Atlanta, Philadelphia, Washington, D.C., New York, and Boston.

The two studies used the EPA Air Quality System (AQS) NO\textsubscript{2} surface measurements and coincident satellite NO\textsubscript{2} TVCD data over the U.S.
<table>
<thead>
<tr>
<th>Type</th>
<th>Surface Area Fraction</th>
<th>Anthropogenic NO\textsubscript{x} Emissions (×10\textsuperscript{10} molecules cm\textsuperscript{-2} s\textsuperscript{-1})</th>
<th>(\gamma) at 13:00 – 14:00 LT</th>
<th>(\beta) at 10:00 – 11:00 LT</th>
<th>(\gamma) at 10:00 – 11:00 LT</th>
</tr>
</thead>
<tbody>
<tr>
<td>Urban/CONUS</td>
<td>17.3%</td>
<td>29.9</td>
<td>2.3 ± 0.9</td>
<td>1.4 ± 0.3</td>
<td>2.4 ± 1.8</td>
</tr>
<tr>
<td>Rural/CONUS</td>
<td>82.7%</td>
<td>2.7</td>
<td>8.1 ± 8.7</td>
<td>3.1 ± 3.9</td>
<td>5.9 ± 8.0</td>
</tr>
<tr>
<td>Urban/AQS</td>
<td>87.7%</td>
<td>71.0</td>
<td>1.5 ± 0.7</td>
<td>1.2 ± 0.4</td>
<td>1.7 ± 1.0</td>
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<tr>
<td>Rural/AQS</td>
<td>12.3%</td>
<td>5.7</td>
<td>5.0 ± 2.0</td>
<td>2.5 ± 1.3</td>
<td>4.3 ± 3.2</td>
</tr>
</tbody>
</table>

1. "Fraction" denotes the percentages of "urban" or "rural" data points for the whole CONUS or all AQS sites.
2. "Urban-CONUS" denote CONUS "urban" grid cells; "Urban-AQS" denote AQS "urban" site grid cells.
<table>
<thead>
<tr>
<th>AQS site</th>
<th>Spring</th>
<th>CONUS</th>
<th>Summer</th>
<th>CONUS</th>
<th>Autumn</th>
<th>CONUS</th>
<th>Winter</th>
<th>CONUS</th>
</tr>
</thead>
<tbody>
<tr>
<td>AQS NOx VMR at 13:00 - 14:00</td>
<td>2003 - 2011</td>
<td>-7.3 ± 1.4%</td>
<td>13:00 - 14:00</td>
<td>-7.4 ± 0.9%</td>
<td>2011 - 2017</td>
<td>-5.2 ± 0.8%</td>
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<tr>
<td>AQS NOx VMR at 10:00 - 11:00</td>
<td>2003 - 2011</td>
<td>-7.1 ± 1.6%</td>
<td>10:00 - 11:00</td>
<td>-7.6 ± 1.5%</td>
<td>2011 - 2017</td>
<td>-4.4 ± 1.4%</td>
<td></td>
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</tr>
<tr>
<td>SCIAMACHY</td>
<td>2003 - 2011</td>
<td>-8.8 ± 3.4%</td>
<td>-6.9 ± 1.1%</td>
<td>-6.2 ± 1.2%</td>
<td>2011 - 2017</td>
<td>-5.2 ± 2.4%</td>
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<tr>
<td>GOME2B</td>
<td>2003 - 2011</td>
<td>-10.2 ± 7.8%</td>
<td>-8.3 ± 16.9%</td>
<td>-8.3 ± 14.0%</td>
<td>2011 - 2017</td>
<td>-10.5 ± 41.6%</td>
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<tr>
<td>OMI-QA4ECV</td>
<td>2005 - 2011</td>
<td>-9.3 ± 5.6%</td>
<td>-8.3 ± 4.6%</td>
<td>-5.9 ± 5.2%</td>
<td>2011 - 2017</td>
<td>-10.0 ± 4.2%</td>
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<tr>
<td>OMI-NASA</td>
<td>2005 - 2011</td>
<td>-9.4 ± 5.0%</td>
<td>-9.6 ± 5.3%</td>
<td>-9.4 ± 2.8%</td>
<td>2011 - 2016</td>
<td>-9.4 ± 3.2%</td>
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<tr>
<td>OMI-BEHR</td>
<td>2005 - 2011</td>
<td>-9.1 ± 5.3%</td>
<td>-8.9 ± 5.8%</td>
<td>-8.7 ± 2.4%</td>
<td>2011 - 2016</td>
<td>-6.9 ± 3.2%</td>
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<tr>
<td>EPA</td>
<td>2003 - 2011</td>
<td>-3.8 ± 4.4%</td>
<td>-3.0 ± 4.0%</td>
<td>-5.4 ± 7.0%</td>
<td>2011 - 2017</td>
<td>-6.7 ± 5.9%</td>
<td></td>
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</tr>
</tbody>
</table>

We calculate trends by using the exponential model described in Table 1.

Table 3. Summary of national trends of updated EPA anthropogenic NOx emissions, AQS NO2 surface concentrations at 13:00 – 14:00 and 10:00 – 11:00 LT, and satellite NO2 TVCD products for 4 seasons during different periods.

1 We calculate trends by using the exponential model described in Table 1.
Figure 1. Region definitions and locations of NO$_2$ surface observation sites used in this study.
Figure 2. Distributions of \( \beta \) (panel a) and \( \gamma \) (panel b) ratios as a function of anthropogenic NO\(_x\) emissions on weekdays for July 2011 over the CONUS. “13:00 – 14:00 LT” is for OMI, and “10:00 – 11:00” LT is for SCIAMACHY and GOME-2A/2B. The data are binned into nine groups based on anthropogenic NO\(_x\) emissions: \( E \in (0, 2^1), [2^1, 2^2), [2^2, 2^3), [2^3, 2^4), [2^4, 2^5), [2^5, 2^6), [2^6, 2^7), [2^7, 2^8), [2^8, 2^9) \times 10^{10} \) molecules cm\(^{-2}\) s\(^{-1}\). Here, \( (0, 2^1) \) denotes \( 0 < \text{emissions} < 2^1 \), \( [2^1, 2^2) \) denotes \( 2^1 \leq \text{emissions} < 2^2 \), similar to other intervals. The green dashed line denotes a value of 1. Error bars denote standard deviations.
Figure 3. Spatial distributions of (a) anthropogenic NO\textsubscript{x} emissions (unit: 10\textsuperscript{10} molecules cm\textsuperscript{-2} s\textsuperscript{-1}) and (b) “urban” regions satisfying our selection criteria. In (b), light green and blue denote the resulting urban and rural regions, respectively.
Figure 4. Relative annual variations of AQS NO\textsubscript{2} surface concentrations and coincident OMI-QA4ECV NO\textsubscript{2} TVCD in each season from 2005 – 2017 for urban (left panel) and rural (right panel) regions. The observation data are scaled by the corresponding 2005 values. Black and red lines denote AQS surface observations and OMI-QA4ECV NO\textsubscript{2} TVCDs, respectively. Shading in a lighter color is added to show the standard deviation of the results; when uncertainty is small due in part to a large number of data points, shading area may not show up.
Figure 5. Relative annual variations of OMI-QA4ECV NO2 TVCD for different anthropogenic NOx-emission groups in each season from 2005 – 2017. “E >= 64” denotes grid cells with anthropogenic NOx emissions over $64 \times 10^{10}$ molecules cm$^{-2}$ s$^{-1}$. “E >= 32” denotes grid cells with anthropogenic NOx emissions equal to or larger than $32 \times 10^{10}$ molecules cm$^{-2}$ s$^{-1}$ but less than $64 \times 10^{10}$ molecules cm$^{-2}$ s$^{-1}$. “E >= 16” and “E >= 8” have similar meanings as “E >= 32”. “E < 8” denotes grid cells with anthropogenic NOx emissions less than $8 \times 10^{10}$ molecules cm$^{-2}$ s$^{-1}$. Shading in a lighter color is added to show the standard deviation of the results; when uncertainty is small due in part to a large number of data points, shading area may not show up.
Figure 6. Relative variations of AQS NO$_2$ surface measurements at 13:00-14:00 and 10:00-11:00 LT, updated EPA anthropogenic NO$_x$ emissions, and satellite NO$_2$ TVCD data over the AQS urban sites (left column) and the CONUS urban regions (right column) for 4 seasons. AQS NO$_2$ surface measurements are not included in the right column. All datasets are scaled by their corresponding values in 2011 except for GOME-2B. For GOME-2B, we firstly normalized the values in each season to the corresponding 2013 values and plotted the relative changes from the 2013 EPA point of each season to make the GOME-2B relative variations comparable to the other datasets. Shading in a lighter color is added to show the standard deviation of the results; when uncertainty is small due in part to a large number of data points, shading area may not show up.