



Inferring the anthropogenic NO_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_x emissions, NO_2 tropospheric vertical column densities (TVCDs), and NO_2 surface concentrations using model simulations for July 2011 over the contiguous United States (CONUS). The variations in NO_2 surface concentrations and TVCDs are generally consistent and reflect anthropogenic NO_x emission variations for high anthropogenic NO_x emission regions well. For low anthropogenic NO_x emission regions, however, nonlinearity in the anthropogenic-emission–TVCD relationship due to emissions from lightning and soils, chemistry, and physical processes makes it difficult to use satellite observations to infer anthropogenic NO_x emission changes. The analysis is extended to 2003–2017. Similar variations in NO_2 surface measurements and coincident satellite NO_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 in anthropogenic NO_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.

fundamental roles in the formation of ozone, acid rain, and fine particles, all of which have negative environmental impacts (Crouse et al., 2015; Kampa and Castanas, 2008; Myhre et al., 2013; Pandey et al., 2005; Singh and Agrawal, 2007). About $48.8 \text{ Tg N yr}^{-1}$ 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). In total, 3.85, 0.24, and 0.66 Tg N of anthropogenic, soil, and lightning NO_x , respectively, were emitted from the US in 2014 on the basis of the 2014 National Emission Inventory (NEI2014) and the GEOS-Chem model simulations (Silvern et al., 2019); vehicle sources and fuel combustions accounted for 93 % of the total anthropogenic NO_x emissions (EPA, 2017).

The US anthropogenic NO_x emissions during the 2010s declined dramatically compared to the mid-2000s (EPA, 2018b; 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 decreases in vehicle NO_x emission factors, NO_2 surface concentrations, nitrate wet deposition flux (Fig. S1 in the Supplement), and NO_2 tropospheric vertical column densities (TVCDs) (Bishop and Stedman, 2015; Georgoulias et al., 2019; Li et al., 2018; McDonald et al., 2018; Miyazaki et al., 2017; Russell et al., 2012; Tong et al., 2015). How-

1 Introduction

Anthropogenic emissions of nitrogen oxides ($\text{NO}_x = \text{NO}_2 + \text{NO}$) adversely affect the environment, not only because of their direct detrimental impacts on human health (Greenberg et al., 2016, 2017; Heinrich et al., 2013; Weinmayr et al., 2009) but also because of their

ever, 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_x emission trend and the anthropogenic NO_x emissions decreased steadily from 2002 to 2017 (Fig. S2), although the emission decrease rate slowed down by about 20 % after 2010 ($-5.8\% \text{ yr}^{-1}$ for 2002–2010 and $-4.7\% \text{ yr}^{-1}$ for 2010–2017; Table 1) (EPA, 2018b). Fuel-based emission estimates in Los Angeles also showed a steady decrease in anthropogenic NO_x emissions after 2000 and a small impact of the Great Recession on the anthropogenic NO_x emission decrease trend (Hassler et al., 2016). The continuous decrease in anthropogenic NO_x emissions was consistent with the ongoing reduction in vehicle emissions (McDonald et al., 2018). On the other hand, Miyazaki et al. (2017) and Jiang et al. (2018) found that the US NO_x emissions derived from satellite NO₂ 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 in NO_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_x emission trends from the observations of NO₂ surface concentrations and satellite NO₂ TVCDs is their nonlinear dependences on anthropogenic NO_x emissions (Gu et al., 2013, 2016; Lamsal et al., 2011). Although the decrease rates of both NO₂ surface concentrations and coincident OMI NO₂ 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 NO₂ surface concentrations is 12 %–79 % less than those of NO₂ TVCDs (Table 1). Secondly, the slowdown of the decrease rates of NO₂ 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 NO₂ surface concentration and OMI NO₂ TVCD trends over the CONUS.

In this study, we carefully investigate the relationships among anthropogenic NO_x emissions, NO₂ surface concentrations, and NO₂ 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) NO₂ surface observations, and NO₂ TVCD products from OMI,

GOME-2A, GOME-2B (GOME2 onboard METOP-B), and SCIAMACHY. In Sect. 3, we examine the nonlinear relationships among anthropogenic NO_x emissions, NO₂ surface concentrations, and NO₂ TVCDs using model simulations. Accounting for the effects of background sources, physical processes, and chemical nonlinearity, we then investigate the anthropogenic NO_x emission trends and changes from 2003 to 2017 over the CONUS. Finally, Sect. 4 gives a summary of the study.

2 Model and data description

2.1 REAM

REAM has been applied and evaluated in many research applications including ozone simulation and forecast, emission inversion and evaluations, and mechanistic studies of chemical and physical processes (Alkuwari et al., 2013; Cheng et al., 2017, 2018; Choi et al., 2008a, b; Gu et al., 2013, 2014; Koo et al., 2012; Liu et al., 2012, 2014; Wang et al., 2007; Yang et al., 2011; Zhang et al., 2017, 2018; Zhang and Wang, 2016; Zhao and Wang, 2009; Zhao et al., 2009a, 2010). REAM as used in this work, the model domain of which is shown in Fig. 3, has 30 vertical layers in the troposphere, and the horizontal resolution is $36 \times 36 \text{ km}^2$. 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 ($2^\circ \times 2.5^\circ$) 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 volatile organic compound (VOC) emissions are estimated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) v2.10 (Guenther et al., 2012). NO_x emissions from soils are based on the Yienger and Levy (YL) scheme (Li et al., 2019; Yienger and Levy, 1995). The cloud-to-ground (CG) lightning flashes are calculated following Choi et al. (2005) and Zhao et al. (2009a) with the parameterization of the CG flash rate as a function of convective mass fluxes and convective available potential energy (CAPE). The ratios of intra-cloud (IC) lightning flashes to CG flashes are parameterized as a function of the height between the freezing layer and the cloud top (Luo et al., 2017; Price and Rind, 1992). In this study, 250 mol of NO are emitted per CG or IC flash (Zhao et al., 2009a). As a result, on weekdays in July 2011, REAM has mean anthropogenic NO_x emissions of $7.4 \times 10^{10} \text{ molecules cm}^{-2} \text{ s}^{-1}$, mean soil NO_x emissions of $1.2 \times 10^{10} \text{ molecules cm}^{-2} \text{ s}^{-1}$, and mean

Table 1. Summary of trends of satellite NO₂ TVCD products, NO₂ surface measurements, and EPA anthropogenic NO_x emissions from different studies.

Studies	Datasets	Period 1 ¹		Period 2		Period 3		Slowdown ratio ³
		Time	Trend (yr ⁻¹) ²	Time	Trend (yr ⁻¹)	Time	Trend (yr ⁻¹)	
This study for CONUS urban sites ⁴	GOME-2B ⁵ (36 × 36 km ²)	2003–2011	−6.3 % ± 1.1 %			2013–2017	−8.2 % ± 3.0 %	
	SCIAMACHY (36 × 36 km ²)	2005–2011	−8.6 % ± 1.2 %			2011–2016	−6.1 % ± 3.6 %	−29 %
	OMI-NASA (36 × 36 km ²)	2005–2011	−8.2 % ± 1.3 %			2011–2016	−4.4 % ± 1.6 %	−46 %
	OMI-BEHR (36 × 36 km ²)	2005–2011	−7.7 % ± 1.4 %			2011–2017	−4.2 % ± 0.5 %	−46 %
	OMI-QA4ECV (36 × 36 km ²)	2005–2011	−6.5 % ± 0.8 %			2011–2017	−5.1 % ± 0.3 %	−22 %
	Updated EPA NO _x emissions ⁶	2003–2011						
This study for AQS urban sites	GOME-2B (36 × 36 km ²)	2003–2011	−7.6 % ± 1.1 %			2013–2017	−10.2 % ± 2.9 %	
	SCIAMACHY (36 × 36 km ²)	2005–2011	−9.0 % ± 0.8 %			2011–2016	−7.2 % ± 3.8 %	−20 %
	OMI-NASA (36 × 36 km ²)	2005–2011	−8.9 % ± 0.3 %			2011–2016	−6.2 % ± 2.6 %	−30 %
	OMI-BEHR (36 × 36 km ²)	2005–2011	−9.0 % ± 0.8 %			2011–2017	−5.4 % ± 0.9 %	−40 %
	OMI-QA4ECV (36 × 36 km ²)	2005–2011	−6.5 % ± 1.2 %			2011–2017	−5.9 % ± 0.8 %	−9 %
	NO ₂ surface volume mixing ratio (VMR)	2003–2011						
Russell et al. (2012) ⁸	BEHR v2.1 NO ₂ TVCD (0.05° × 0.05°) Updated EPA NO _x emissions	2005–2007	−6 % ± 5 % (−6.2 %) ⁹ −6.0 %	2007–2009	−8 % ± 5 % (−8.4 %) −10.0 %	2009–2011	−3 % ± 4 % (−3.0 %) −2.4 %	−52 % −60 %
Tong et al. (2015) ¹⁰	NASA v2.1 NO ₂ TVCD (pixels < 50 × 24 km ²)	2005–2007	−7.3 % (−7.6 %)	2008–2009	−9.2 % (−11.4 %)	2010–2012	−2.8 % (−4.4 %)	−42 %
	BEHR v2.1 NO ₂ TVCD (pixels < 50 × 24 km ²)		−8.9 % (−9.3 %)		−9.1 % (−11.8 %)		−3.6 % (−6.0 %)	−35 %
	NO ₂ surface VMR		−6.0 % (−6.2 %)		−10.8 % (−13.2 %)		−3.4 % (−5.4 %)	−13 %
	Updated EPA NO _x emissions		−6.0 %		−10.0 %		−3.4 %	−43 %
Lamsal et al. (2015) ¹¹	NASA v2.1 NO ₂ TVCD (0.1° × 0.1°) NO ₂ surface VMR	2005–2008	−4.8 % ± 1.9 % (−5.1 %) −3.7 % ± 1.5 % (−3.8 %)			2010–2013	−1.2 % ± 1.2 % (−1.2 %) −2.1 % ± 1.4 % (−2.1 %)	−76 % −45 %
	Updated EPA NO _x emissions		−6.4 %				−4.0 %	−38 %
Jiang et al. (2018) ¹¹	NASA v3 NO ₂ TVCD (0.5° × 0.667°)	2005–2009	−10.2 % ± 1.8 % (−9.8 %)			2011–2015	−3.2 % ± 1.6 % (−3.2 %)	−67 %
	DOMINO v2 ¹² NO ₂ TVCD (0.5° × 0.667°)		−9.6 % ± 1.7 % (−9.3 %)				−2.6 % ± 1.8 % (−2.6 %)	−72 %
	BEHR v2.1 NO ₂ TVCD (0.5° × 0.667°)		−8.5 % ± 1.8 % (−8.2 %)				−2.1 % ± 1.6 % (−2.1 %)	−74 %
	NO ₂ surface VMR Updated EPA NO _x emissions		−6.6 % ± 1.4 % (−6.4 %) −7.8 %				−2.6 % ± 1.5 % (−2.6 %) −5.0 %	−59 % −36 %

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: $E(y) = E_0 \times r^{y-y_0}$, where “ y ” denotes year and “ y_0 ” denotes the initial year. “ $E(y)$ ” denotes the value at year “ y ” and “ E_0 ” denotes the value at the initial year, and $r - 1$ is the relative trend. ³ Slowdown ratios = trend in “period 3”/trend in “period 1”. 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 We updated EPA anthropogenic NO_x emissions with the newest Continuous Emission Monitoring Systems (CEMS) datasets. Figure S2 shows the comparison between our updated and original EPA anthropogenic NO_x emissions (EPA, 2018b). ⁷ Denotes the averaged trends of 1300 and 1000 local time (LT) based on the values in Table 3. ⁸ The study used NO₂ TVCD from urban and power plant grid cells across the US. ⁹ 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 all the results consistent. Our results are the bold numbers in parentheses, while the numbers in normal font are from the original publications. ¹⁰ The study uses NO₂ TVCD and surface concentrations from Los Angeles, Dallas, Houston, Atlanta, Philadelphia, Washington, D.C., New York, and Boston. ¹¹ The two studies used the EPA Air Quality System (AQS) NO₂ surface measurements and coincident satellite NO₂ TVCD data over the US. ¹² DOMINO: Dutch OMI NO₂ products.

lightning NO_x emissions of 3.4×10^{10} molecules cm⁻² s⁻¹ over the CONUS.

2.2 Satellite NO₂ TVCDs

In this study, we use NO₂ TVCD products from four satellite sensors 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 instrument 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 GOME-2A) and Metop-B (GOME-2B) satellites cross the Equator at 09:30 LT and have a nadir resolution of 80×40 km². After 15 July 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 backscattered solar 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 vertical column densities (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 NO₂ SCDs; (3) dividing the NO₂ tropospheric SCDs by the tropospheric air mass factors (AMFs) to compute TVCDs.

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 NO₂ 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., 2018b). These product archives have been previously validated (Boersma et al., 2018; Drosoglou et al., 2017, 2018; Krotkov et al., 2017; Laughner et al., 2018b; 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 NO₂ TVCD datasets, we chose pixel-size NO₂ TVCD data using the criteria listed in Table S3. After the selection, we re-gridded the pixel-size data into the REAM 36×36 km² grid cells and calculated 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 August; autumn: September,

October, and November). We excluded weekend data in this study to minimize the impacts of weekend NO_x emission reduction, leading to different NO₂ TVCDs between weekdays and weekends (Fig. S3).

Satellite TVCD measurements can show large variations and apparent discontinuities due in part to the effects of cloud, lightning NO_x, the shift in satellite pixel coverage, and retrieval uncertainties (Fig. S3; 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 $RCI = (X(95\text{th percentile}) - X(5\text{th percentile})) / \text{mean}(X)$, where X is the daily NO₂ 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 NO₂ measurements

Hourly surface NO₂ measurements from 2003 to 2017 are from the EPA AQS monitoring network (archived on <https://www.epa.gov/outdoor-air-quality-data>, last access: 27 June 2018). Most AQS monitoring sites use the Federal Reference Method (FRM) gas-phase chemiluminescence to measure NO₂. 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 NO₂ is first converted to NO and then NO is measured through chemiluminescence measurement of NO₂* produced by NO + O₃. The difference is that FRM uses heated catalysts for the conversion of NO₂ to NO and FEM uses photolysis of NO₂ to NO. The conversion to NO in the FRM instruments is not specific to NO₂, and non-NO_x active nitrogen compounds (NO_z) can also be reduced by the catalysts, which would cause high biases of NO₂ measurements, while the FEM is sensitive to the photolysis conversion efficiency of NO₂ to NO (Beaver et al., 2012, 2013; Lamsal et al., 2015). The CAPS method directly determines NO₂ concentrations based on an NO₂-induced phase shift measured by a photodetector. The CAPS instrument operates at a wavelength of about 450 nm and may overestimate NO₂ concentrations due to the absorption of other molecules at the same wavelength (Beaver et al., 2012, 2013; Keababian 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 discrepancies among the above three methods. There are three sites having FRM and FEM measurements simultaneously during some periods from 2013 to 2014, two sites having both FRM and CAPS data during some periods from 2015 to 2016, and one site using all three measurement methods during some periods in 2015. Figure S4 shows the hourly averaged ratios of FEM and CAPS to FRM data

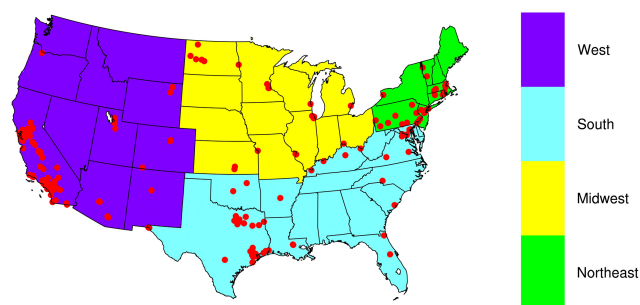


Figure 1. Region definitions and locations of NO₂ surface observation sites used in this study.

for four seasons during 2013–2016. The CAPS/FRM ratios are in the range of 0.94–1.06, and the FEM/FRM ratios are 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 NO₂ 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 NO₂ surface concentrations have significant diurnal variations (Fig. S5), we choose the data at 09:00–10:00 LT for comparison with GOME-2A/2B data, those at 10:00–11:00 LT for comparison with SCIAMACHY data, and those at 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, and thus about 1.5 % of the data is removed. We also require that the measurement site must have valid measurements in the aforementioned 3 h for at least one season from 2003 to 2017. The locations of the 179 selected sites using the site selection criteria are shown in Fig. 1. The region definitions follow the U.S. Census Bureau (https://www2.census.gov/geo/pdfs/maps-data/maps/reference/us_regdiv.pdf, last access: 6 February 2019).

3 Results and discussions

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

NO₂ surface concentrations and NO₂ TVCDs are not linearly correlated with NO_x emissions due to chemical nonlinearity, NO₂ hydrolysis on aerosols ($\text{NO}_2 \xrightarrow{\text{aerosol}, \text{H}_2\text{O}} 0.5\text{HNO}_3 + 0.5\text{HNO}_2$), dry deposition, transport effects, and background sources (Gu et al., 2013; Lamsal et al., 2011). Therefore, it is necessary to first investigate the nonlinearities among NO_x emissions, NO₂ surface concentrations, and TVCDs over the CONUS before we compare the trends between NO₂ surface

concentrations and TVCDs. The nonlinearity between NO_x emission and NO₂ TVCD is analyzed by examining the local sensitivity of NO₂ TVCD to NO_x emissions (Gu et al., 2013; Lamsal et al., 2011; Tong et al., 2015), which is defined as β in Eq. (1). We further define γ as the sensitivity of NO₂ surface concentration to NO_x emission:

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

$$\frac{\Delta E}{E} = \gamma \frac{\Delta c}{c}, \quad (2)$$

where E denotes NO_x emission and E denotes the change in NO_x emission; Ω denotes NO₂ TVCD, c denotes surface NO₂ concentration, and $\Delta \Omega$ and Δc denote the corresponding changes.

We computed β and γ values for July 2011 over the CONUS using REAM. To compute local β and γ 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 NO_x emissions were reduced by 15 %. In the 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 the group 2 tracer simulation were from the 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 the group 1 tracer simulation. Using this procedure, the effects of anthropogenic NO_x emission reduction were localized. The β and γ values were computed by the ratio of TVCD and surface concentration changes to a 15 % change in anthropogenic NO_x emissions.

Figure 2 shows the distributions of our β and γ ratios as a function of anthropogenic NO_x emissions for July 2011 over the CONUS. Results essentially the same as Fig. 2 were obtained when a perturbation of 10 % was used for anthropogenic NO_x emissions. Figure S6 shows the distributions of NO₂ TVCD fraction in the boundary layer at 13:00–14:00 LT and 10:00–11:00 LT and the fraction of soil NO_x emissions in all surface sources (soil + anthropogenic) on weekdays for July 2011. In Fig. S7, we analyzed the contributions of background sources, chemical nonlinearity, and other factors (transport, NO₂ hydrolysis on aerosols, and dry deposition) to the nonlinear relationships (β and γ) among anthropogenic NO_x emissions, NO₂ surface concentrations, and NO₂ TVCDs. While the model simulation is for 1 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.

- Both β and γ values are negatively correlated with anthropogenic NO_x emissions due to chemical non-linearity, transport, and background NO_x contributions (Figs. 2, S6, and S7) (Gu et al., 2016; Lamsal et al., 2011). It is consistent with the distribution of β as a function of NO_x emissions in China (Gu et al., 2013), although the β 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).
- The uncertainties of β and γ 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 β and γ values (Figs. 2 and S7).
- The value of γ is generally less than β , especially for low anthropogenic NO_x emission regions, which reflects the significant contribution of free tropospheric NO₂ to NO₂ TVCD but not to NO₂ surface concentrations (Figs. 2, S6, and S7).
- Generally, the standard deviations of β and γ tend to be larger at 10:00–11:00 LT than at 13:00–14:00 LT, reflecting a stronger transport effect due to weaker chemical losses in the morning (Figs. 2 and S7).
- Both β and γ values are significantly less than 1 at 13:00–14:00 LT ($\beta = 0.75$ 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 ($\beta = 0.97$ and $\gamma = 1.03$), which reflects stronger chemistry nonlinearity at noontime than in the morning (Figs. 2 and S7).
- Both background sources and non-emission factors contribute much more to β and γ values in low anthropogenic NO_x emission regions than in high anthropogenic NO_x emission regions (Fig. S7).
- Chemical nonlinearity contributes much less to β and γ values than background sources and transport effects in low anthropogenic NO_x emission regions (Fig. S7).
- Generally, non-emission factors (mainly transport) contribute more to β and γ values than background sources in low anthropogenic NO_x emission regions (Fig. S7c and d) except for the first bin where background sources contribute more to β and γ values than non-emission factors at 10:00–11:00 LT, which is partly caused by some grid cells with extremely low anthropogenic NO_x emissions, increasing the mean contributions of background sources in the first bin.

The largely varying β and γ values for anthropogenic NO_x emissions $< 10^{11}$ molecules cm⁻² s⁻¹ imply that the

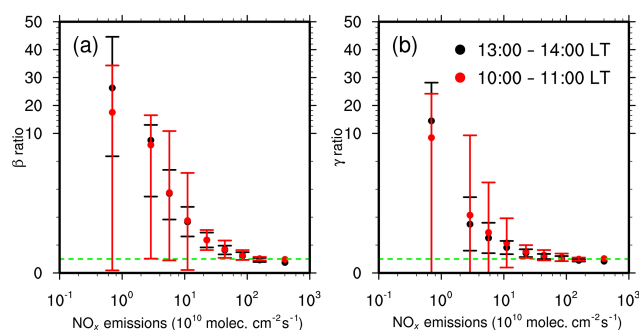


Figure 2. Distributions of β (a) and γ (b) ratios as a function of anthropogenic NO_x emissions on weekdays for July 2011 over the CONUS. “13:00–14:00 LT” is for OMI, and “10:00–11:00 LT” is for SCIAMACHY and GOME-2A/2B. The data are binned into nine groups based on anthropogenic NO_x emissions: $E \in (0, 2^1), [2^1, 2^2), [2^2, 2^3), [2^3, 2^4), [2^4, 2^5), [2^5, 2^6), [2^6, 2^7), [2^7, 2^8), [2^8, 2^9) \times 10^{10}$ molecules cm⁻² s⁻¹. Here, $(0, 2^1)$ denotes $0 < \text{emissions} < 2^1$, and $[2^1, 2^2)$ 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.

trends derived from satellite TVCD data do not directly represent anthropogenic NO_x emissions and that the variations in TVCD data may not be comparable to the corresponding surface NO₂ concentrations. We define a region a “urban” if anthropogenic NO_x emissions from NEI2011 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 (Fig. S2). 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 NO₂ AQS surface concentrations and coincident satellite NO₂ 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 remaining 22 sites are rural. Their properties are summarized in Table 2. Figure 4 shows the relative annual variations in AQS NO₂ surface measurements at 13:00–14:00 LT and coincident OMI-QA4ECV NO₂ TVCD data from 2005 to 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 NO₂ to

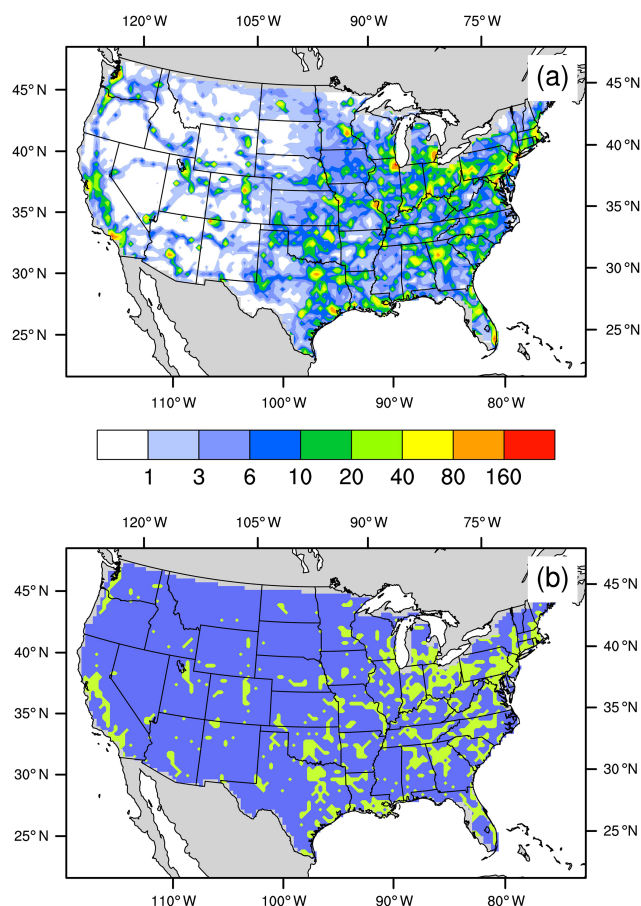


Figure 3. Spatial distributions of (a) anthropogenic NO_x emissions (unit: 10^{10} molecules $\text{cm}^{-2} \text{s}^{-1}$) and (b) urban regions satisfying our selection criteria. In (b), light green and blue denote the resulting urban and rural regions, respectively.

their corresponding 2005 values, and the resulting data are therefore unitless. Over urban regions, NO₂ surface concentrations are highly correlated with NO₂ TVCDs ($\text{TVCD} = 1.03 \times \text{AQS} + 0.11$, $R^2 = 0.98$), reflecting the comparable and stable β and γ values (Fig. 2). However, over rural regions, the scaled TVCD data significantly deviate from AQS NO₂ data ($\text{TVCD} = 1.15 \times \text{AQS} + 0.09$, $R^2 = 0.87$). 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_x emissions in winter.

We also examine the correlations of AQS NO₂ 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 (Fig. 4). SCIAMACHY and GOME-2B TVCD observations at 09:00–11:00 LT also show a large contrast between urban (SCIAMACHY: $\text{TVCD} = 0.92 \times \text{AQS} - 0.005$, $R^2 = 0.94$;

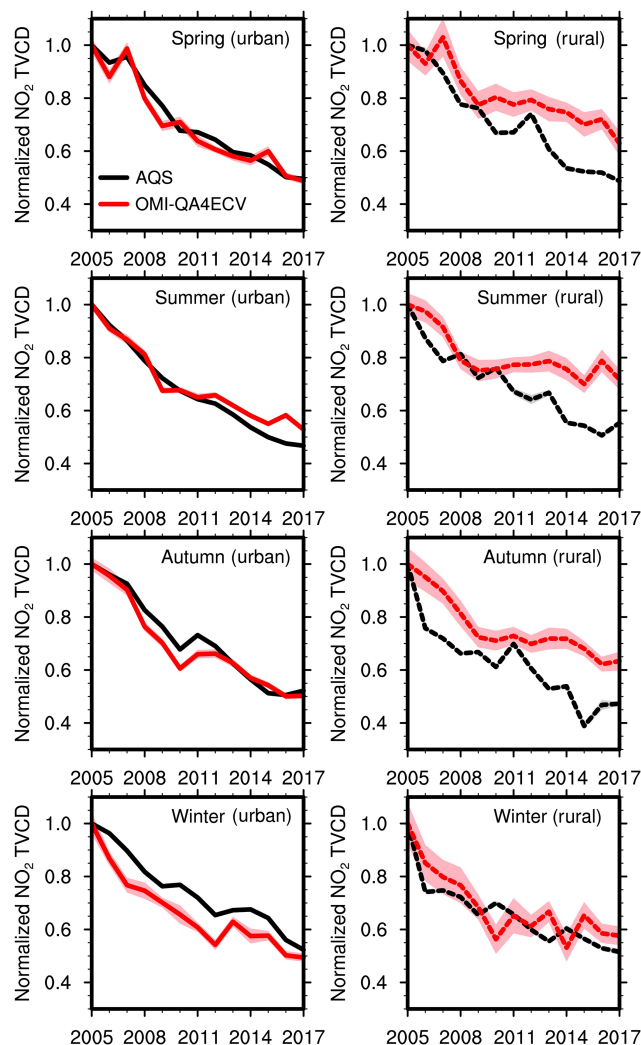


Figure 4. Relative annual variations in AQS NO₂ surface concentrations and coincident OMI-QA4ECV NO₂ TVCD in each season from 2005 to 2017 for urban (left column) and rural (right column) regions. The observation data are scaled by the corresponding 2005 values. Black and red lines denote AQS surface observations and OMI-QA4ECV NO₂ 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 may not show up.

GOME-2B: $\text{TVCD} = 0.54 \times \text{AQS} + 0.56$, $R^2 = 0.96$) and rural regions (SCIAMACHY: $\text{TVCD} = 0.77 \times \text{AQS} + 0.83$, $R^2 = 0.63$; GOME-2B: $\text{TVCD} = 0.46 \times \text{AQS} + 0.73$, $R^2 = 0.59$). The correlation of coincident GOME-2A NO₂ TVCD data with AQS surface concentrations is poor for rural ($\text{TVCD} = 0.65 \times \text{AQS} + 0.56$, $R^2 = 0.44$) and urban ($\text{TVCD} = 0.31 \times \text{AQS} + 0.56$, $R^2 = 0.21$) regions (Fig. S8), which likely reflects the degradation of the GOME-2A instrument causing a significant increase in NO₂ SCD uncertainties (Boersma et al., 2018). Therefore, we excluded GOME-2A in the analysis hereafter.

Table 2. Properties of urban and rural regions in July 2011.

Type	Surface area fraction ¹	Anthropogenic NO _x emissions ($\times 10^{10}$ molecules cm ⁻² s ⁻¹)	β at 13:00–14:00 LT	γ at 13:00–14:00 LT	β at 10:00–11:00 LT	γ at 10:00–11:00 LT
Urban/CONUS ²	17.3 %	29.9	2.5 ± 1.0	1.5 ± 0.4	2.6 ± 1.9	1.6 ± 1.2
Rural/CONUS	82.7 %	2.7	16.9 ± 16.4	8.5 ± 11.7	12.2 ± 14.0	6.4 ± 11.6
Urban/AQS	87.7 %	71.0	1.6 ± 0.8	1.2 ± 0.4	1.7 ± 1.1	1.3 ± 0.6
Rural/AQS	12.3 %	5.7	8.7 ± 9.9	5.2 ± 8.8	5.4 ± 15.1	3.8 ± 11.7

¹ “Fraction” denotes the percentages of urban or rural data points for the whole CONUS or all AQS sites. ² “Urban-CONUS” denotes CONUS urban grid cells; “Urban-AQS” denotes AQS urban site grid cells.

We further investigate OMI-QA4ECV NO₂ TVCD relative annual variations from 2005 to 2017 over the regions with different anthropogenic NO_x emissions in Fig. 5. We find clear flattening of NO₂ TVCD variations as anthropogenic NO_x emissions decrease, which is consistent with the above analysis. Similar to Fig. 4, the spread of TVCD variation is much less in winter than the other seasons. The differences between Figs. 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 Fig. 4, but all OMI-QA4ECV data are used in Fig. 5.

3.3 Trend analysis of AQS NO₂ surface concentrations, satellite TVCDs, and updated EPA NO_x emissions

We first updated the Continuous Emission Monitoring Systems (CEMS) measurement data used in the EPA NO_x emission trend datasets with the newest datasets obtained from <https://ampd.epa.gov/ampd/> (last access: 17 June 2018). As shown in Fig. S2, the updated CEMS data lead to a reduction in 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 NO₂ and SCIAMACHY TVCD products (Figs. 4, 6, and S9) and AQS NO₂ surface measurements (Figs. 4, 6, and S5) and are also noted by Russell et al. (2012) and Tong et al. (2015) (Table 1).

In Fig. 6, we show the comparisons among the relative variations in the updated EPA anthropogenic NO_x emissions, AQS NO₂ surface measurements at 10:00–11:00 LT and 13:00–14:00 LT, and coincident satellite NO₂ TVCDs for urban regions in four seasons from 2003 to 2017. Also shown are the comparisons among the updated EPA anthropogenic NO_x emissions and satellite NO₂ 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 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 Fig. 5. Including the low anthropogenic NO_x emission regions leads to underestimates of NO_x decreases. Since the area of low anthropogenic NO_x emission regions is larger than high anthropogenic NO_x emission regions (Table 2), the arithmetic averaging will lead to a large weighting of rural observations, which do not reflect anthropogenic NO_x emission changes. Miyazaki et al. (2017) and Jiang et al. (2018) included all regions in their analyses, but we exclude rural regions. Figure S9 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_x emissions was previously discussed by Zhang et al. (2018). In addition, Miyazaki et al. (2017) and Jiang et al. (2018) conducted NO_x emission inversions by using the Model for Interdisciplinary Research on Climate (MIROC)-Chem with a coarse resolution of $2.8^\circ \times 2.8^\circ$, which was insufficient to separate urban and rural regions and might distort predicted NO₂ TVCDs and inversed NO_x emissions due to nonlinear effects (Valin et al., 2011; Yu et al., 2016), which is another possible reason for their finding of flattening NO_x emission trends after 2010.

We summarize the decreasing rates of NO₂ 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 in AQS surface observations, satellite NO₂ TVCD, and updated EPA anthropogenic NO_x emissions after 2011 as in Fig. 6, but the 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 NO₂ TVCD measurements, which may reflect in part smaller γ than β values (Table 2). Our estimated slowdown magnitudes are significantly lower than Lamsal et al. (2015) and Jiang et al. (2018) (Table 1), which might be caused by their different data processing methods, such as

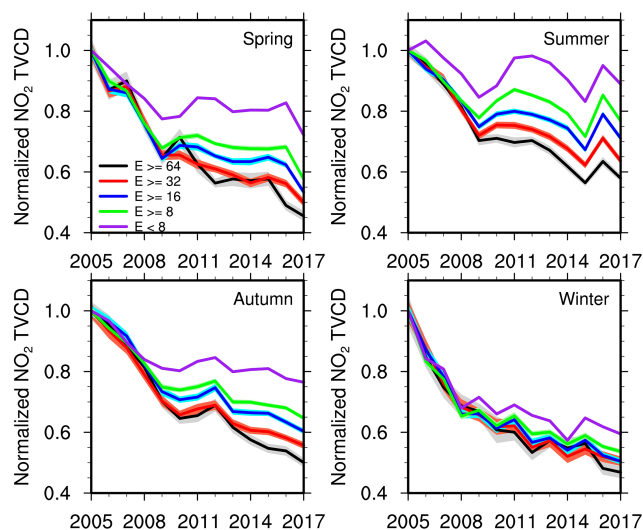


Figure 5. Relative annual variations in OMI-QA4ECV NO₂ TVCD for different anthropogenic NO_x emission groups based on NEI2011 in each season from 2005 to 2017. “ $E \geq 64$ ” denotes grid cells with anthropogenic NO_x emissions over 64×10^{10} molecules $\text{cm}^{-2} \text{s}^{-1}$. “ $E \geq 32$ ” denotes grid cells with anthropogenic NO_x emissions equal to or larger than 32×10^{10} molecules $\text{cm}^{-2} \text{s}^{-1}$ but less than 64×10^{10} molecules $\text{cm}^{-2} \text{s}^{-1}$. “ $E \geq 16$ ” and “ $E \geq 8$ ” have similar meanings as “ $E \geq 32$ ”. “ $E < 8$ ” denotes grid cells with anthropogenic NO_x emissions less than 8×10^{10} molecules $\text{cm}^{-2} \text{s}^{-1}$. 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 may not show up.

including AQS sites with incomplete measurement records (Silvern et al., 2019).

Over the CONUS urban regions, updated EPA anthropogenic NO_x emissions show a slowdown of 22 % compared to 29 %–46 % for three OMI NO₂ TVCD products. The difference is partially due to the β ratio of 2.5 ± 1.0 at 13:00–14:00 LT over the CONUS urban regions (Table 2). Satellite NO₂ TVCD measurement uncertainties also contribute to the difference. From 2013 to 2017, GOME-2B NO₂ TVCDs decrease more than OMI products, especially in spring, autumn, and winter (Tables 1 and 3). Finally, trend analyses in different regions (Fig. 7 and Table S4) indicate that generally, the Midwest has the least slowdown of the decreasing rate for urban OMI NO₂ 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

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

		Spring		Summer		Autumn		Winter	
		AQS site	CONUS	AQS site	CONUS	AQS site	CONUS	AQS site	CONUS
AQS NO ₂ VMR at 13:00–14:00 LT	2003–2011	−7.3 % ± 1.4 %		−7.4 % ± 0.9 %		−6.7 % ± 1.8 %		−5.2 % ± 0.8 %	
	2011–2017	−5.3 % ± 1.6 %		−6.4 % ± 1.2 %		−7.3 % ± 2.5 %		−6.0 % ± 2.8 %	
AQS NO ₂ VMR at 10:00–11:00 LT	2003–2011	−7.1 % ± 1.6 %		−7.6 % ± 1.5 %		−6.2 % ± 2.2 %		−4.4 % ± 1.6 %	
	2011–2017	−4.4 % ± 1.4 %		−6.1 % ± 1.8 %		−6.3 % ± 2.5 %		−5.2 % ± 2.4 %	
SCIAMACHY	2003–2011	−8.8 % ± 3.4 %	−6.9 % ± 1.1 %	−8.2 % ± 1.6 %	−5.2 % ± 1.2 %	−6.8 % ± 2.4 %	−5.6 % ± 2.1 %	−6.4 % ± 7.4 %	−7.5 % ± 5.5 %
	2011–2017								
GOME2B	2003–2011	−10.2 % ± 7.8 %	−8.3 % ± 16.9 %	−6.4 % ± 14.0 %	−5.3 % ± 4.0 %	−10.5 % ± 41.6 %	−6.9 % ± 13.2 %	−13.6 % ± 15.1 %	−12.3 % ± 78.9 %
	2013–2017								
OMI-QA4ECV	2005–2011	−9.3 % ± 5.6 %	−8.3 % ± 4.6 %	−8.3 % ± 2.4 %	−5.9 % ± 5.2 %	−10.0 % ± 4.2 %	−7.4 % ± 2.4 %	−8.3 % ± 2.1 %	−9.3 % ± 5.2 %
	2011–2017	−5.3 % ± 6.0 %	−4.3 % ± 6.5 %	−4.2 % ± 3.0 %	−4.9 % ± 9.2 %	−6.0 % ± 1.8 %	−3.8 % ± 1.8 %	−6.1 % ± 25.6 %	−3.8 % ± 3.5 %
OMI-NASA	2005–2011	−9.4 % ± 5.0 %	−9.6 % ± 5.3 %	−9.4 % ± 2.8 %	−7.1 % ± 2.9 %	−9.4 % ± 3.2 %	−8.1 % ± 2.8 %	−7.8 % ± 3.6 %	−9.5 % ± 16.6 %
	2011–2016	−4.4 % ± 18.9 %	−3.8 % ± 7.5 %	−5.7 % ± 6.7 %	−4.5 % ± 5.3 %	−6.0 % ± 3.1 %	−4.6 % ± 3.9 %	−12.8 % ± 7.8 %	−11.4 % ± 6.6 %
OMI-BEHR	2005–2011	−9.1 % ± 5.3 %	−8.9 % ± 5.8 %	−8.7 % ± 2.4 %	−6.4 % ± 3.2 %	−9.2 % ± 3.2 %	−8.0 % ± 3.1 %	−8.5 % ± 10.6 %	−9.4 % ± 23.0 %
	2011–2016	−3.8 % ± 4.4 %	−3.0 % ± 4.0 %	−5.4 % ± 7.0 %	−3.9 % ± 6.6 %	−5.6 % ± 13.2 %	−4.1 % ± 14.0 %	−9.9 % ± 5.2 %	−6.7 % ± 5.9 %
EPA	2003–2011				−6.5 % ± 0.8 %				
	2011–2017				−5.1 % ± 0.3 %				

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

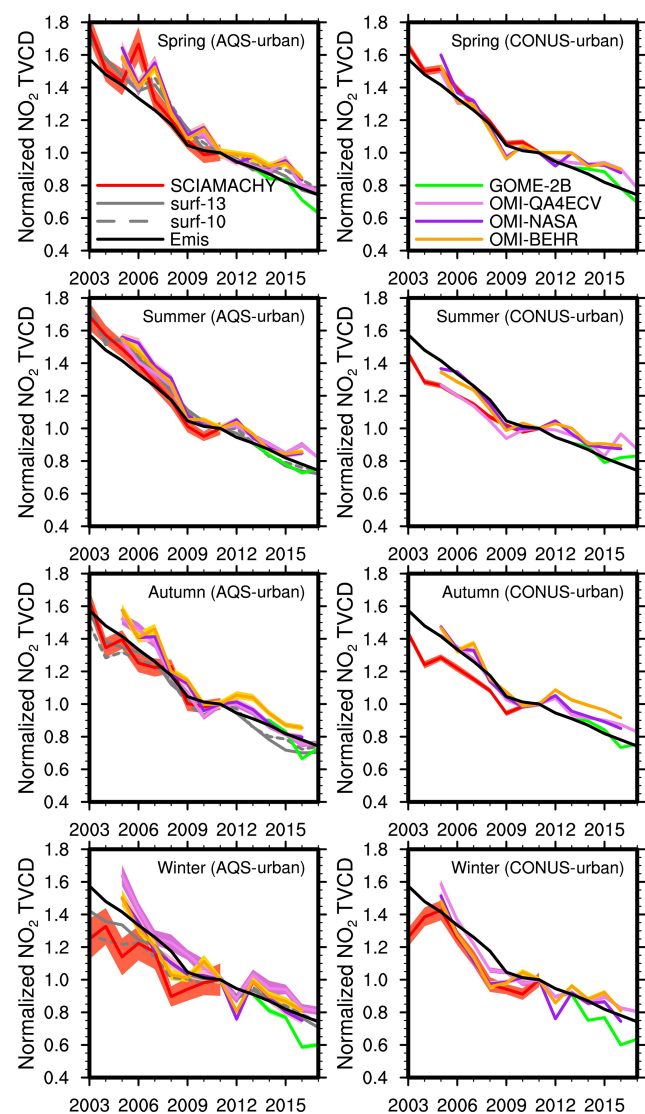


Figure 6. Relative variations in AQS NO_2 surface measurements at 13:00–14:00 LT and 10:00–11:00 LT, updated EPA anthropogenic NO_x emissions, and satellite NO_2 TVCD data over the AQS urban sites (left column) and the CONUS urban regions (right column) for four seasons. AQS NO_2 surface measurements 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 may not show up.

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_2 columns by natural emissions and make use of additional observations such as nitrate deposition fluxes. They also identified model biases

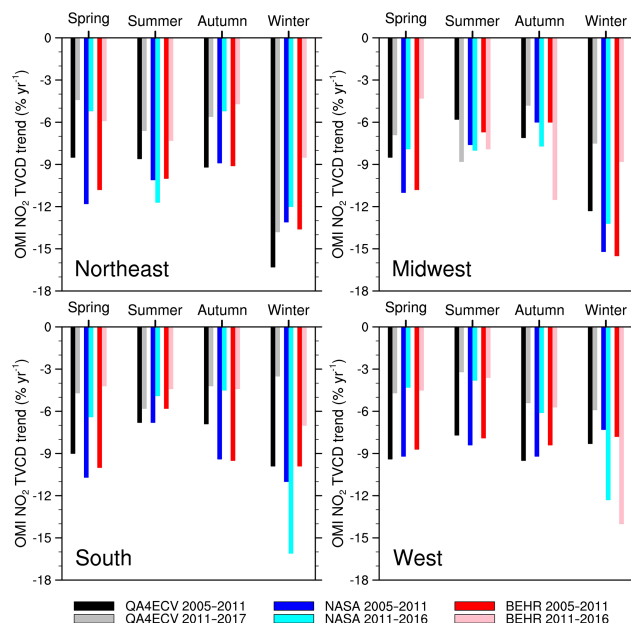


Figure 7. Pre- and post-2011 OMI NO_2 TVCD trends for four seasons in the urban regions of Northeast, Midwest, South, and West. Black bars denote OMI-QA4ECV NO_2 TVCD trends from 2005 to 2011; gray bars denote the corresponding trends during 2011–2017. Blue bars denote OMI-NASA trends from 2005 to 2011; cyan bars denote OMI-NASA trends from 2011 to 2016. Red bars denote OMI-BEHR trends from 2005 to 2011; pink bars denote OMI-BEHR trends from 2011 to 2016.

in simulating the trends of NO_2 TVCDs by missing natural emissions in the free troposphere. 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 2011, we demonstrate the nonlinear relationship of NO_2 surface concentration and TVCD with anthropogenic NO_x emissions. Over low anthropogenic NO_x emission regions, the ratios of anthropogenic NO_x emission changes to the changes in surface concentrations (γ) and TVCDs (β) have very large variations and $\beta > \gamma \gg 1$. Therefore, for the same emission changes, surface concentration and TVCD changes are much smaller and more 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 $> 10^{11} \text{ molecules cm}^{-2} \text{ s}^{-1}$ 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 to 2017 over the CONUS by using satellite

NO₂ TVCD products from GOME-2B, SCIAMACHY, OMI-QA4ECV, OMI-NASA, and OMI-BEHR, over the urban regions of CONUS. We find broad agreement among the decreases in AQS NO₂ surface observations, satellite NO₂ 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, NO₂ 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 to 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 NO₂ TVCD and surface concentration be carefully evaluated when applying satellite and surface measurements to infer the changes in anthropogenic NO_x emissions.

Data availability. The NCEP CFSv2 products are from <https://doi.org/10.5065/D61C1TXF> (last access: 10 March 2015). The EPA AQS hourly surface NO₂ measurements are downloaded from https://aqs.epa.gov/aqsweb/airdata/download_files.html#Raw (last access: 27 June 2018; EPA, 2018a). QA4ECV 1.1 NO₂ VCD products (OMI-QA4ECV, GOME-2A, and SCIAMACHY) are from <http://temis.nl/qa4ecv/no2col/data/> (last access: 20 August 2018; ESA, 2017). GOME-2B NO₂ VCD products are from <http://www.temis.nl/airpollution/no2col/no2colgome2b.php> (last access: 20 August 2018; ESA, 2013). OMI-BEHR and OMI-NASA archives are from <http://behr.cchem.berkeley.edu/DownloadBEHRData.aspx> (last access: 18 August 2018; Laughner et al., 2018a). REAM simulation results for this study are available upon request.

Supplement. The supplement related to this article is available online at: <https://doi.org/10.5194/acp-19-15339-2019-supplement>.

Author contributions. JL and YW designed the study. JL conducted model simulations and data analyses with discussions with YW. JL and YW wrote the paper.

Competing interests. The authors declare that they have no conflict of interest.

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