



# Unexpected slowdown of US pollutant emission reduction in the past decade

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Edited by John H. Seinfeld, California Institute of Technology, Pasadena, CA, and approved March 30, 2018 (received for review January 22, 2018)

Ground and satellite observations show that air pollution regulations in the United States (US) have resulted in substantial reductions in emissions and corresponding improvements in air quality over the last several decades. However, large uncertainties remain in evaluating how recent regulations affect different emission sectors and pollutant trends. Here we show a significant slowdown in decreasing US emissions of nitrogen oxides (NO<sub>x</sub>) and carbon monoxide (CO) for 2011–2015 using satellite and surface measurements. This observed slowdown in emission reductions is significantly different from the trend expected using US Environmental Protection Agency (EPA) bottom-up inventories and impedes compliance with local and federal agency air-quality goals. We find that the difference between observations and EPA's NO<sub>x</sub> emission estimates could be explained by: (i) growing relative contributions of industrial, area, and off-road sources, (ii) decreasing relative contributions of on-road gasoline, and (iii) slower than expected decreases in on-road diesel emissions.

nitrogen oxides | emission regulations | decadal scale variation

To achieve and maintain air-quality standards, US regulations have required significant reductions in the key ozone (O<sub>3</sub>) precursor emissions of NO<sub>x</sub> and CO since the 1960s (1). These emission reductions, confirmed by both ground (2–4) and satellite measurements (5–7), have resulted in substantial improvement in air quality in the last few decades through reduction in surface O<sub>3</sub> in many populated areas (8, 9). In addition to emission regulations, technology innovations and changes in patterns of human activity also alter energy demand, industrial practices, goods movement, and vehicular travel, and thus have important and complicated effects on pollutant emissions. For example, a recent study (10) has demonstrated larger vehicular primary NO<sub>2</sub> emission reduction in Europe than assumed in policy projections.

In October 2015, the US Environmental Protection Agency (EPA) revised the O<sub>3</sub> standard (11) from 75 ppb (2008 standard) to 70 ppb. The new O<sub>3</sub> standard requires stricter controls on O<sub>3</sub> precursor emissions in the subsequent years; for example, the South Coast Air Quality Management District recently released the Air Quality Management Plan (12), and requires 45% reduction of NO<sub>x</sub> emissions in Southern California in the period of 2016–2023. To better understand the variation of O<sub>3</sub> precursor emissions, we evaluate trends in EPA's NO<sub>x</sub> and CO emission inventory data (*Methods*) between 2005 and 2015 by combining datasets including top-down anthropogenic NO<sub>x</sub> and CO emission estimates from inverse analysis studies (6, 7), remotely sensed NO<sub>2</sub> measurements from the Ozone Monitoring Instrument (OMI), CO measurements from Measurement of Pollution in the Troposphere (MOPITT), surface in situ NO<sub>2</sub>, CO, and O<sub>3</sub> measurements from the US Air Quality System (AQS), and emission estimation using fuel-based bottom-up methods.

## Results

**Comparison of Top-Down and Bottom-Up Estimates of NO<sub>x</sub> Emission Changes.** In a recent study, Miyazaki et al. (6) estimated global NO<sub>x</sub> emissions in the period of 2005–2015 using multiple satellite measurements (*SI Appendix*). The top-down NO<sub>x</sub> emissions were obtained using an ensemble Kalman filter, while improving the representation of the chemical system (e.g., NO<sub>x</sub> lifetime) affecting tropospheric NO<sub>2</sub> by assimilating multiple chemical species including CO and O<sub>3</sub> concentrations. Fig. 1A (green line) shows percent changes of the top-down anthropogenic NO<sub>x</sub> emissions (normalized at 2009), indicating a dramatic slowdown (76%) in US NO<sub>x</sub> emissions reduction from  $-7.0 \pm 1.4\%/y$  (2005–2009) to  $-1.7 \pm 1.4\%/y$  (2011–2015), as shown in Table 1. Uncertainties represent 1  $\sigma$  and include the error budget described in *SI Appendix*. Average top-down anthropogenic NO<sub>x</sub> emissions for the 11-y period are shown in Fig. 2A, demonstrating the strongest emissions in the northeast United States. Fig. 2B and C shows the differences of top-down anthropogenic NO<sub>x</sub> emissions from 2005–2006 to 2008–2009, and from 2011–2012 to

## Significance

Emissions of nitrogen oxides (NO<sub>x</sub>) have a large impact on air quality and climate change as precursors in the formation of ozone and secondary aerosols. We find that NO<sub>x</sub> emissions have not been decreasing as expected in recent years (2011–2015) when comparing top-down estimates from satellites and surface NO<sub>2</sub> measurements to the trends predicted from the US Environmental Protection Agency's emission inventory data. The discrepancy can be explained by the growing relative contribution of industrial, area, and off-road mobile sources of emissions, decreasing relative contribution of on-road gasoline vehicles, and slower than expected decreases in on-road diesel NO<sub>x</sub> emissions, with implications for air-quality management.

Author contributions: Z.J., H.W., J.R.W., D.K.H., and D.B.A.J. designed research; Z.J. and B.C.M. performed research; Z.J., B.C.M., K.M., and Z.Q. analyzed data; and Z.J., B.C.M., H.W., J.R.W., K.M., Z.Q., D.K.H., D.B.A.J., A.F.A., E.V.F., L.Z., and K.F.B. wrote the paper.

The authors declare no conflict of interest.

This article is a PNAS Direct Submission.

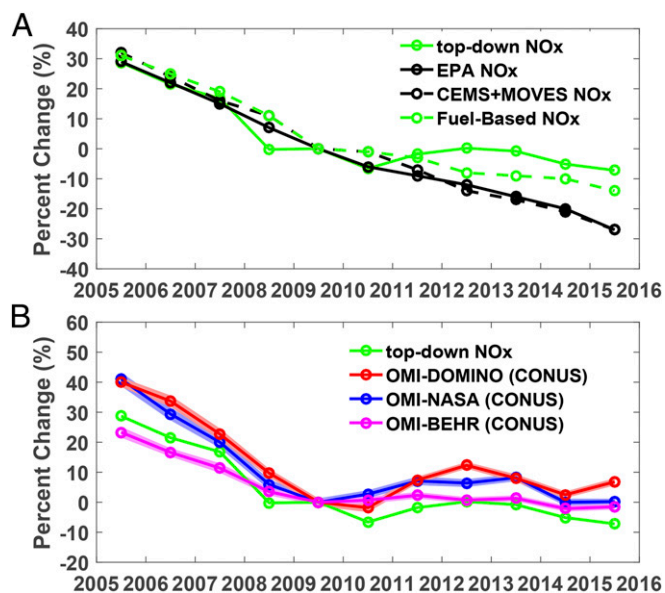
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This article contains supporting information online at [www.pnas.org/lookup/suppl/doi:10.1073/pnas.1801191115/-DCSupplemental](http://www.pnas.org/lookup/suppl/doi:10.1073/pnas.1801191115/-DCSupplemental).

Published online April 30, 2018.



**Fig. 1.** (A) Percent changes (normalized at 2009) of top-down US anthropogenic  $\text{NO}_x$  emission estimates from inverse analysis (green line), EPA's emissions trends report data of  $\text{NO}_x$  (black solid line), revised EPA emission estimates including CEMS and MOVES national-scale data (black dashed line, *SI Appendix, Table S1*), and revised industrial, on-road, off-road emission estimates using fuel-based methodologies (green dashed line, *SI Appendix, Table S1*). (B) Percent changes of top-down US anthropogenic  $\text{NO}_x$  emission estimates and tropospheric OMI  $\text{NO}_2$  columns over CONUS. The shaded areas represent  $1\text{-}\sigma$  uncertainties for random and sampling errors.

2014–2015, respectively. We find pronounced changes in the reduction of anthropogenic  $\text{NO}_x$  emissions for these two periods, throughout the continental contiguous United States (CONUS).

For comparison, we evaluate EPA's bottom-up emission trends over the same time periods. Fig. 1A (black solid line) shows percent changes of EPA's bottom-up emission estimates (*Methods*). As shown in Table 1, trends of top-down anthropogenic  $\text{NO}_x$  emission estimates ( $-7.0 \pm 1.4\%/y$ ) and EPA's emission estimates ( $-6.4\%/y$ ) are consistent within the top-down uncertainty estimates in the period of 2005–2009. However, for 2011–2015, top-down ( $-1.7 \pm 1.4\%/y$ ) and bottom-up ( $-5.3\%/y$ )  $\text{NO}_x$  emissions trends are inconsistent. Between the periods of 2005–2009 and 2011–2015, the slowdown predicted by the EPA's emissions is only 16%, from  $-6.4\%/y$  to  $-5.3\%/y$ , which is much smaller than the slowdown observed by the top-down estimates (76%).

#### Changes in Tropospheric Column (Satellite) and Surface $\text{NO}_2$ Abundance.

Fig. 1B shows percent changes of the top-down anthropogenic  $\text{NO}_x$  emissions and tropospheric OMI  $\text{NO}_2$  columns from National Aeronautics and Space Administration (NASA), Dutch OMI  $\text{NO}_2$  (DOMINO), and Berkeley High-Resolution (BEHR) products (*SI Appendix*) over CONUS. The interannual variation of top-down  $\text{NO}_x$  emissions generally follows the variation in OMI  $\text{NO}_2$  measurements

as expected, since the OMI DOMINO product is included in the assimilated data (6). Since each point in Fig. 1B represents an average over the CONUS for each year, the precision errors are relatively small; however, differences in the NASA, DOMINO, and BEHR products provide an estimate of the accuracy in tropospheric  $\text{NO}_2$  interannual variations. Fig. 3A–F displays maps of the differences of mean tropospheric OMI  $\text{NO}_2$  columns from 2005–2006 to 2008–2009, and from 2011–2012 to 2014–2015, respectively, for the different OMI data products, demonstrating a consistent slowdown of the reduction in tropospheric  $\text{NO}_2$  columns.

To corroborate the satellite observations of tropospheric  $\text{NO}_2$  columns, we perform a similar analysis using surface in situ AQS measurements (*SI Appendix*). Fig. 4A and B shows the differences of mean surface  $\text{NO}_2$  concentrations, as measured by the AQS network, from 2005–2006 to 2008–2009, and from 2011–2012 to 2014–2015. Fig. 5A shows percent changes of the surface in situ AQS  $\text{NO}_2$  measurements and tropospheric OMI  $\text{NO}_2$  columns sampled at the times and locations of AQS measurements (based on monthly averages) over all CONUS AQS sites. Consistent with previous studies (3, 13), the sampled OMI  $\text{NO}_2$  data demonstrate good agreement with AQS  $\text{NO}_2$  measurements. Fig. 5B–D demonstrates agreement between AQS and OMI  $\text{NO}_2$  measurements within their uncertainties over three distinct US regions. Similar to our analysis, the EPA Air Trend data (14) show a 42% slowdown of  $\text{NO}_2$  concentration reduction from  $-3.3\%/y$  to  $-1.9\%/y$ .

The similar slowdown of the reductions of observed  $\text{NO}_2$  abundances demonstrates the slowdown of estimated  $\text{NO}_x$  emission reduction (6) is reasonable. In addition, the relation between changes in  $\text{NO}_x$  emissions and  $\text{NO}_2$  abundances may be affected by the nonlinear chemistry (15, 16). In a recent study, Jin et al. (17) indicated that some US megacities have changed from volatile organic compounds (VOCs) to  $\text{NO}_x$  limited in recent years, and thus, the same  $\text{NO}_x$  emission reduction may result in slower reduction in  $\text{NO}_2$  abundance through an increase in  $\text{NO}_x$  lifetime. However, we do not expect a significant influence due to changes in urban  $\text{NO}_x$  chemistry because the slowdown (Fig. 3A–F) is observable throughout much of CONUS. Furthermore, we tested the role of  $\text{NO}_x$  emissions in controlling  $\text{NO}_2$  abundance with a sensitivity study where global surface  $\text{NO}_x$  emissions were reduced by 20% compared with the standard simulation in the chemical atmospheric general circulation model (AGCM) for study of atmospheric environment and radiative forcing (CHASER) for 2015. This resulted in a 16–20% decrease in annual mean surface  $\text{NO}_2$  concentrations (*SI Appendix, Fig. S1*), demonstrating that variations in  $\text{NO}_2$  abundances are dominated by changes in emissions.

**Changes in CO Emissions.** Recent studies (1, 18, 19) have demonstrated that a synthesis of  $\text{NO}_x$  and CO measurements can provide an effective constraint on trends in anthropogenic emission inventories because both are coemitted byproducts of combustion. Warneke et al. (20) also showed that trends of VOCs found in gasoline are also highly correlated with trends in CO. Consequently, we also investigate the decadal variation of CO to evaluate the changes in anthropogenic  $\text{NO}_x$  emissions. In a recent study, Jiang et al. (7) constrained global CO emissions in the

**Table 1.** Trends and uncertainties for all  $\text{NO}_x$  datasets

Period	EPA $\text{NO}_x$	Top-down $\text{NO}_x$	OMI (NASA)	OMI (DOMINO)	OMI (BEHR)	AQS $\text{NO}_2$
2005–2009 (CONUS)	-6.4%	$-7.0 \pm 1.4\%$	$-8.8 \pm 1.0\%$	$-8.6 \pm 0.9\%$	$-5.4 \pm 1.0\%$	
2011–2015 (CONUS)	-5.3%	$-1.7 \pm 1.4\%$	$-1.9 \pm 0.8\%$	$-1.0 \pm 0.9\%$	$-1.0 \pm 0.8\%$	
2005–2009 (sampled)			$-10.2 \pm 1.8\%$	$-9.6 \pm 1.7\%$	$-8.5 \pm 1.8\%$	$-6.6 \pm 1.4\%$
2011–2015 (sampled)			$-3.2 \pm 1.6\%$	$-2.6 \pm 1.8\%$	$-2.1 \pm 1.6\%$	$-2.6 \pm 1.5\%$

All trends are relative to the average of each data period (2005–2009 and 2011–2015) cover the whole US and based on a linear trend model. Uncertainties represent  $1\text{-}\sigma$  and include the error budget discussed in *SI Appendix*. OMI (sampled) represents OMI  $\text{NO}_2$  measurements sampled at AQS  $\text{NO}_2$  measurement locations and times based on monthly averages.

**Table 2. Trends and uncertainties for CO datasets and eastern US AQ5 O<sub>3</sub>**

Period	EPA CO	Top-down CO	MOPITT CO	AQS CO	AQS O <sub>3</sub>
2005–2009	−7.0%	−4.5 ± 1.1%	−2.7 ± 0.6%	−7.9 ± 1.3%	−1.6 ± 1.0%
2011–2015	−4.6%	−1.4 ± 1.1%	−1.4 ± 0.6%	−2.7 ± 1.3%	−0.4 ± 0.9%

All trends are relative to the average of each data period (2005–2009 and 2011–2015) and based on a linear trend model. Uncertainties represent 1  $\sigma$  and include the error budget discussed in *SI Appendix*. AQS O<sub>3</sub> includes measurements over eastern US only (eastward of 100°W), whereas other datasets cover the whole US.

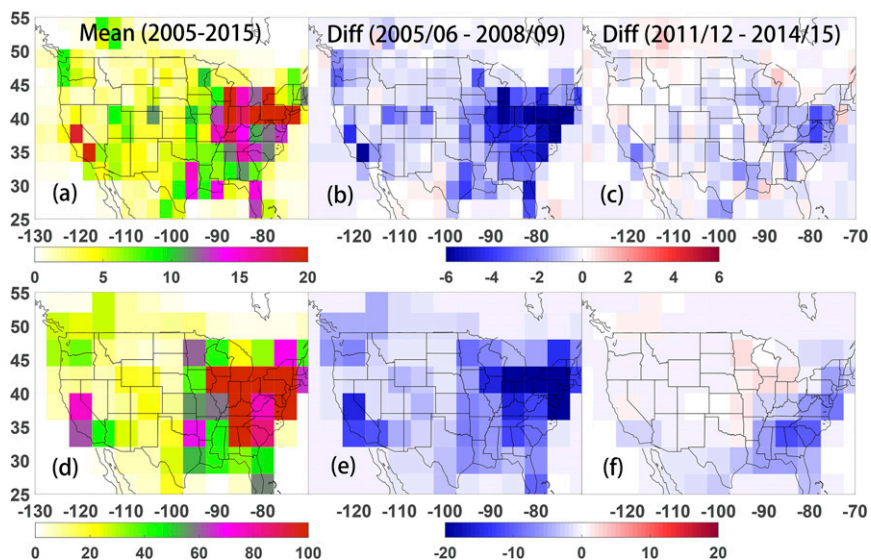
period of 2001–2015 using MOPITT CO measurements (*SI Appendix*). The top-down CO emissions were obtained using a four-dimensional variational approach, and the role of long-range transport was accounted for by optimizing boundary conditions around the North American continent. Fig. 2D shows the 11-y averages of top-down anthropogenic CO emissions (7), excluding biomass burning and oxidation sources. Fig. 2E and F shows the differences of top-down anthropogenic CO emissions from 2005–2006 to 2008–2009, and from 2011–2012 to 2014–2015, respectively. In the first period, 2005–2009, we observe a large decrease in both NO<sub>x</sub> and CO emissions.

Fig. 3G and H shows the differences of mean MOPITT surface layer CO mixing ratio, from 2005–2006 to 2008–2009, and from 2011–2012 to 2014–2015, respectively. These show a similar slowdown of the decrease of CO mixing ratios in the most recent years, particularly over the northeast United States. However, unlike OMI NO<sub>2</sub> retrievals, MOPITT CO retrievals (even surface layer CO mixing ratio) are not an ideal proxy for local emissions, because of the longer CO lifetime (compared with NO<sub>x</sub> lifetime) and the coarse vertical resolution of MOPITT profile retrievals (21). For example, *SI Appendix, Fig. S2C* shows a significant reduction in top-down biomass burning CO emissions (7) in Mexico in the most recent years. These emissions influence CO concentrations in the southeast United States through regional transport, and explain the continued decrease of CO emissions in 2011–2015 for the southeast United States (Fig. 2F).

Fig. 4C and D shows the differences of mean surface CO concentrations, as measured by the AQS network, from 2005–2006 to 2008–2009, and from 2011–2012 to 2014–2015, respectively. As shown in Table 2, the trends in the MOPITT surface layer CO mixing ratio, AQS in situ CO measurements, and

top-down US anthropogenic CO emission estimates from Jiang et al. (7) all exhibit similar slowdowns in reduction in recent years. Besides NO<sub>2</sub> and CO, AQS in situ O<sub>3</sub> measurements over the eastern United States (Table 2) show a similar 75% slowdown of surface O<sub>3</sub> concentration reduction from −1.6%/y to −0.4%/y, suggesting the importance of diminishing rates of decrease for anthropogenic CO, VOCs, and NO<sub>x</sub>.

**Revisions to Bottom-Up Emission Estimates.** What are the potential explanations for this dramatic slowdown of reductions of US anthropogenic NO<sub>x</sub> and CO emissions in the recent years? For CO emissions, a slowdown in reductions is expected due to diminishing returns to improved three-way catalytic converters on gasoline engines (22). Past studies have shown that transportation emissions of CO are highly correlated with VOCs found in gasoline fuel and tailpipe exhaust (20, 22), implying that decreases in gasoline-related VOC emissions are also slowing down as well. However, the slowdown in anthropogenic NO<sub>x</sub> emissions is surprising. Since the late 1990s, large decreases in NO<sub>x</sub> emissions were driven by efforts to regulate power plant emissions (23), fuel switching of electric power generation from coal to natural gas (24), and controls on transportation emissions (25). Since 2005, stack monitors suggest that NO<sub>x</sub> emissions from power plants are still declining (*SI Appendix, Table S1*), tailpipe emission standards on light-duty gasoline vehicles have gotten stricter, and selective catalytic reduction (SCR) systems have begun to be installed on 2010 model year and later heavy-duty diesel trucks. Therefore, US NO<sub>x</sub> emissions are expected to decline at a similar rate in the 2011–2015 time period as during 2005–2009.



**Fig. 2.** (A) Mean top-down anthropogenic NO<sub>x</sub> emissions from inverse analysis in the period 2005–2015. (B and C) Difference of top-down anthropogenic NO<sub>x</sub> emissions from 2005–2006 to 2008–2009, and from 2011–2012 to 2014–2015. (D–F) same as A–C, but for top-down anthropogenic CO emissions. The unit is 10<sup>10</sup> mole/cm<sup>2</sup>/s.

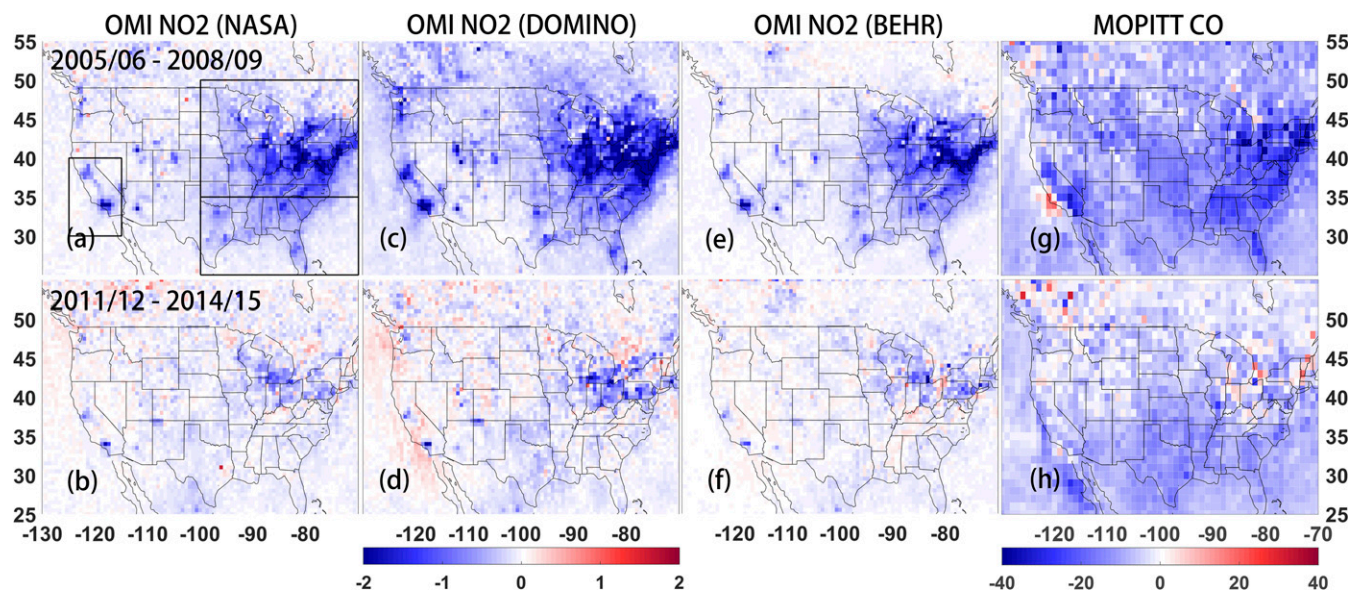


Fig. 3. (A–F) Difference of mean tropospheric OMI NO<sub>2</sub> columns from 2005–2006 to 2008–2009, and from 2011–2012 to 2014–2015. The unit is  $10^{15}$  mole/cm<sup>2</sup>. (G and H) same as A–F, but for MOPITT surface layer CO measurements with unit ppb (parts per billion). A also indicates the southwest, southeast, and northeast US regions for sampling satellite observations at the AQS sites used in Fig. 5 comparison.

Fig. 1A shows EPA's emissions trend report data across all anthropogenic sources (black solid line). To attain higher sectoral-level information, we substitute on-road emissions from the trends report with national-scale outputs from the EPA Motor Vehicle Emission Simulator (MOVES) model, as well as utilize Continuous Emission Monitoring Systems (CEMS) data directly for electric power generation (black dashed line). We also propose three further modifications to help explain the observed NO<sub>x</sub> trend:

- i) We estimate industrial, residential, and area source NO<sub>x</sub> emissions in a consistent manner using a fuel-based methodology outlined by Xing et al. (26), and off-road mobile source emissions following a fuel-based approach described previously (27, 28). Based on these results (*SI Appendix, Table S1*), industrial, area, and off-road mobile source NO<sub>x</sub> emissions are shown to be decreasing at a slower rate in the 2011–2015 relative to the 2005–2009 time period.
- ii) We estimate on-road gasoline emissions using a fuel-based approach (25). While NO<sub>x</sub> emissions are consistently declining by  $\sim 8\%/y$  from 2005 to 2015 in this analysis (*SI Appendix, Table S1*), the main effect of this revision is to reduce on-road gasoline emissions by  $\sim 40\%$  relative to output from the EPA MOVES model, and consistent with recent atmospheric modeling studies (29–32). This increases the relative contribution of other anthropogenic sectors whose emissions may not be declining as quickly as for on-road gasoline vehicles. We note that a recent report suggests that gasoline vehicles are now reaching the point of diminishing returns in reducing NO<sub>x</sub> emissions (33), which would also contribute to a slowdown.
- iii) We estimate on-road diesel emissions using a fuel-based approach (25). While NO<sub>x</sub> emissions are declining throughout the 2005–2015 time period, the decreases in 2011–2015 are approximately half the rate of those in the EPA inventory (*SI Appendix, Table S1*). Recent chassis dynamometer and portable testing of heavy-duty trucks show that under local/urban driving conditions, NO<sub>x</sub> emissions are significantly elevated relative to in-use certification limits (34, 35). Recent roadside measurements of NO<sub>x</sub> emission factors (36) also indicate that the emission reductions from SCR systems may not be as large as anticipated by emission certification tests (*SI Appendix, Fig. S3*).

Combining these three modifications (green dashed line in Fig. 1A) gives a slowdown with the reduction rate of NO<sub>x</sub> emissions from  $-6.7\%/y$  for 2005–2009 to  $-2.9\%/y$  for 2011–2015 (*SI Appendix, Table S1*), consistent with the observed slowdown.

The above revisions to bottom-up emission estimates provide reasonable explanations for the observed slowdown of emission reduction at a national scale. However, as shown in Fig. 5, we might expect regional variability in trends due to regional differences in air-quality management practices. The reduction rates of AQS surface in situ NO<sub>2</sub> measurements are  $-4.1 \pm 2.2\%/y$  (2005–2009) and  $-3.9 \pm 2.5\%/y$  (2011–2015) for the southwest United States (particularly from California), suggesting relatively stable reductions in this region. By contrast, the

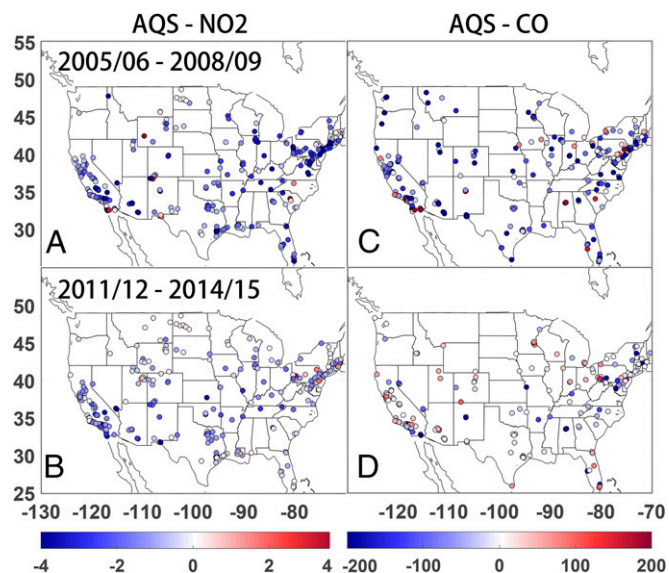


Fig. 4. (A and B) Difference of mean NO<sub>2</sub> concentrations of surface in situ NO<sub>2</sub> measurements (AQS stations) from 2005–2006 to 2008–2009, and from 2011–2012 to 2014–2015. (C and D) Same as A and B, but for surface in situ CO measurements. The unit is ppb.

reduction rates of AQS surface in situ  $\text{NO}_2$  measurements are  $-7.8 \pm 2.0\%/y$  (2005–2009) and  $-2.6 \pm 2.1\%/y$  (2011–2015) for the northeast United States, and  $-6.7 \pm 2.3\%/y$  (2005–2009) and  $-0.1\% \pm 2.6\%/y$  (2011–2015) for the southeast United States, indicating a dramatic slowdown. Similar to AQS measurements, the slowdown of emission reductions over the southwest US suggested by OMI tropospheric  $\text{NO}_2$  columns (e.g., NASA product sampled at AQS  $\text{NO}_2$  measurement locations and times in Fig. 5) is also much weaker: the reduction rates are  $-8.6 \pm 4.0\%/y$  (2005–2009) and  $-5.6 \pm 3.6\%/y$  (2011–2015) over the southwest United States, compared with  $-10.2 \pm 1.8\%/y$  (2005–2009) and  $-3.2\% \pm 1.6\%/y$  (2011–2015) over CONUS.

California is expected to have more stringent emission regulations than other states of the United States. For example, California is accelerating the turnover of the heavy-duty vehicle fleet, such that by 2023, almost all truck and buses operating in the state will require a 2010 engine or later model year. In other regions of the United States, there has been increasing scrutiny of glider-kit trucks, which are heavy-duty trucks with refurbished engines installed on a new chassis. However, EPA suggests that  $\text{NO}_x$  emissions from such glider-kit trucks significantly exceed the emission standards promulgated in 2010 (37), which could contribute to a slowdown in  $\text{NO}_x$  emission reductions in regions where glider-kit trucks are operating in significant numbers.

There is also regional variability in trends of  $\text{NO}_x$  emissions from energy generation. Stack monitors on power plants indicate that  $\text{NO}_x$  emissions have consistently declined by 7–10% over the 2005–2009 and 2011–2015 time periods in the Northeast and Southeast regions, consistent with reporting under the Acid Rain Program and the Cross State Air Pollution Rule (38). However, in the Southwest region, the decrease in power plant emissions of  $\text{NO}_x$  has slowed from  $-20\%$  in 2005–2009 to  $-8\%$  in 2011–2015. In some oil and natural gas basins, including in Texas and North Dakota, satellite  $\text{NO}_2$  columns have been shown to be increasing (5).

## Conclusions

Using a synthesis of recently estimated top-down anthropogenic  $\text{NO}_x$  and CO emissions from inverse analysis studies (6, 7), remotely sensed  $\text{NO}_2$  measurements from OMI, CO measurements from MOPITT, surface in situ  $\text{NO}_2$  and CO measurements from AQS, and emission estimation using fuel-based bottom-up methods, we evaluate trends in EPA’s emission inventory data between 2005 and 2015. In contrast to the larger European emission reduction as suggested by Grange et al. (10), we find an unexpected, significant slowdown in the reductions of US  $\text{NO}_x$  and CO emissions in the most recent years. The similar slowdown of surface  $\text{O}_3$  concentration reduction suggests a potential important influence from variations in pollutant emissions on the formation of secondary pollutants, and consequent socioeconomic costs resulting from degraded air quality.

Our analysis suggests the slowdown in decreasing  $\text{NO}_x$  emissions observed in 2011–2015 is mainly driven by the growing relative contribution of industrial, area, and off-road mobile sources of emissions, decreasing relative contribution of on-road gasoline vehicles, and slower than expected decreases in on-road diesel  $\text{NO}_x$  emissions. Meanwhile, the slowdown in decreasing CO emissions is likely due to diminishing returns from the large fraction of gasoline vehicles that have already significantly reduced CO emissions. While this study demonstrates the large-scale effects of changing emission trends and identifies the likely causes of the observed slowdown in declining pollution trends, a more quantitative attribution of emission changes for  $\text{NO}_x$  and CO and their subsequent effects on  $\text{O}_3$  and other air pollutants will require models and data with finer (e.g., urban and roadway environments) spatial scales. This work highlights the importance of satellite and model inversion technologies to monitor changes in pollutant emissions and interpret the effects of regulations and economic activities.

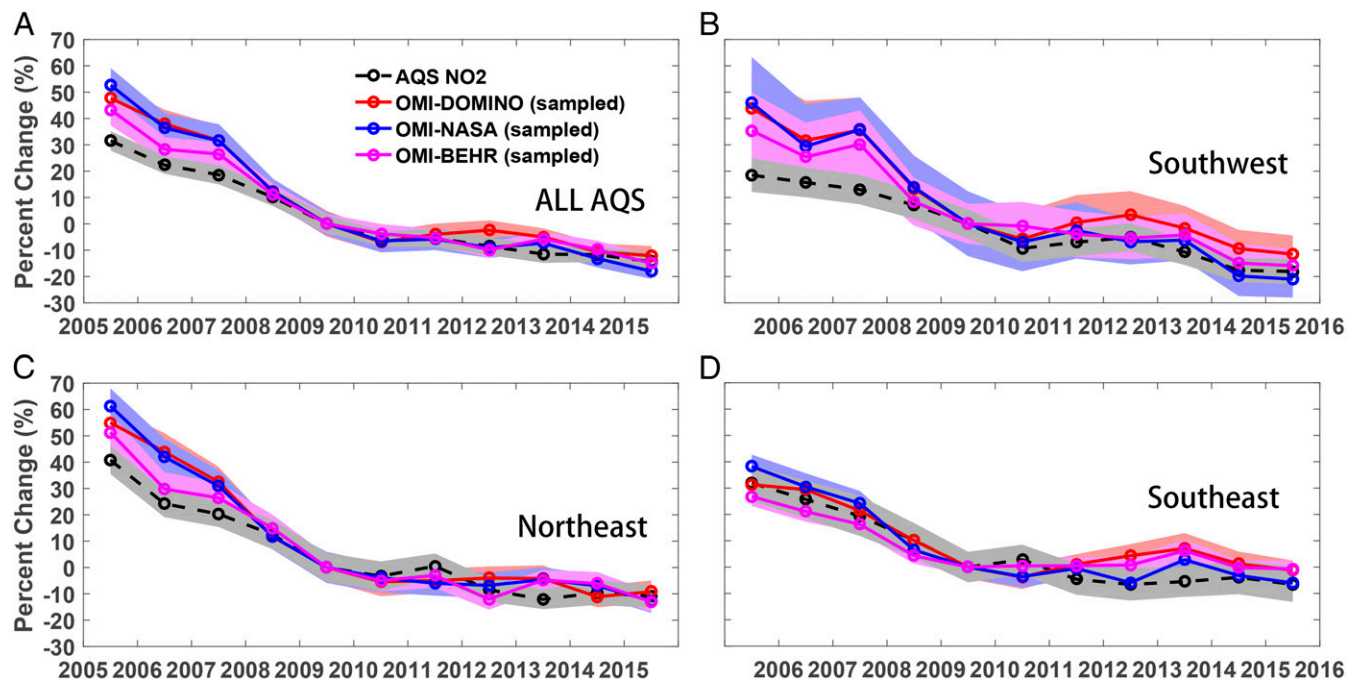


Fig. 5. (A–D) Percent changes (annual means normalized at 2009) of AQS surface in situ  $\text{NO}_2$  measurements and tropospheric OMI  $\text{NO}_2$  columns for various regions. Both AQS and OMI measurements are averaged with monthly resolution; the averaged OMI (monthly) data are sampled at AQS  $\text{NO}_2$  (monthly) measurement locations and times; annual means are calculated based on monthly means. The region definition is shown in Fig. 3A. The shaded areas represent  $1\text{-}\sigma$  uncertainties for random and sampling errors.

## Methods

**Bottom-Up NO<sub>x</sub> Emission Data.** The EPA inventory used in this study is from the Air Pollutant Emissions Trends Data downloaded at: <https://www.epa.gov/air-emissions-inventories/air-pollutant-emissions-trends-data>. The emissions are updated through the NEI 2014v1. To better reconcile bottom-up emission inventories with top-down observations for NO<sub>x</sub>, we modify anthropogenic emissions only. First, we update electric power generation emissions with the latest CEMS data downloaded at: <https://ampd.epa.gov/ampd/>. Xing et al. (26) outlined a fuel-based methodology to consistently estimate industrial, residential, and commercial fuel combustion emissions for long-term atmospheric modeling simulations (1990–2010). We employ their approach here, and update energy use statistics through 2015 (39). The largest decreases in industrial NO<sub>x</sub> emission factors occur before 2005 and are relatively constant thereafter (26). We maintain this trend and hold NO<sub>x</sub> emission factors constant after 2010. Other emissions associated with industrial processes are left unmodified from the EPA inventory.

We revise mobile source emissions using a fuel-based approach for estimating both on-road (1, 25) and off-road engines (27, 28). Briefly, fuel-use statistics for on-road and off-road engines are available annually from the Federal Highway Administration and Energy Information Administration (40–42). Emission factors (in g/kg fuel) are based on a metaanalysis of roadway studies (1, 25), laboratory measurements of off-road gasoline engines (43–45), and the EPA NONROAD model for off-road diesel engines.

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More details about emission factors for on-road vehicles are provided in *SI Appendix*.

**Other Datasets and Statistical Analysis.** The descriptions for the top-down NO<sub>x</sub> and CO emission data, tropospheric OMI NO<sub>2</sub> column data, MOPITT CO data, AQS surface in situ measurements, and statistical analysis associated with trends and uncertainties are provided in *SI Appendix*.

**ACKNOWLEDGMENTS.** We acknowledge useful discussions with Vivienne H. Payne, Benjamin Gaubert, and Forrest Lacey. We thank the EPA for providing their national NO<sub>x</sub> and CO emission data and surface in situ NO<sub>2</sub>, CO, and O<sub>3</sub> measurements (AQS). We acknowledge the OMI tropospheric NO<sub>2</sub> column data from <https://disc.sci.gsfc.nasa.gov>, [www.temis.nl](http://www.temis.nl), and [behr.cchem.berkeley.edu/DownloadBEHRData.aspx](http://behr.cchem.berkeley.edu/DownloadBEHRData.aspx). The MOPITT team also acknowledges the Canadian Space Agency for the instrument finance, the Natural Sciences and Engineering Research Council and Environment Canada (formerly the Meteorological Service of Canada) for help with the data processing, COMDEV (the prime contractor), and ABB BOMEM. The National Center for Atmospheric Research (NCAR) MOPITT project is supported by the NASA Earth Observing System Program. NCAR is sponsored by the National Science Foundation. Part of this work was carried out at the Jet Propulsion Laboratory, California Institute of Technology, under a contract with NASA. Support for Z.Q. and D.K.H. was provided by NASA Health and Air Quality Applied Science Team NNX16AQ26G.

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