Unexpected slowdown of US pollutant emission reduction in the past decade

Zhe Jiang[a,b,1], Brian C. McDonald[c,d], Helen Worden[a], John R. Worden[e], Kazuyuki Miyazaki[f], Zhen Qu[g], Daven K. Henze[g], Dylan B. A. Jones[h], Avelino F. Arellano[i], Emily V. Fischer[j], Liye Zhu[j,2], and K. Folkert Boersma[k,l]

[a]Atmospheric Chemistry Observations and Modeling Laboratory, National Center for Atmospheric Research, Boulder, CO 80301; [b]School of Earth and Space Sciences, University of Science and Technology of China, Hefei, Anhui, 230026, China; [c]Chemical Sciences Division, National Oceanic and Atmospheric Administration Earth System Research Laboratory, Boulder, CO 80305; [d]Cooperative Institute for Research in Environmental Sciences, University of Colorado Boulder, Boulder, CO 80309; [e]Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA 91009; [f]Research and Development Center for Global Change, Japan Agency for Marine-Earth Science and Technology, Yokohama, 236-0001, Japan; [g]Department of Mechanical Engineering, University of Colorado Boulder, Boulder, CO 80309; [h]Department of Physics, University of Toronto, Toronto, ON M5S 1A7, Canada; [i]Department of Hydrology and Atmospheric Sciences, University of Arizona, Tucson, AZ 85721; [j]Department of Atmospheric Science, Colorado State University, Fort Collins, CO 80523; [k]Department of Meteorological and Air Quality, Wageningen University, Wageningen, 6700, The Netherlands; and [l]Satellite Observations Department, Royal Netherlands Meteorological Institute, De Bilt, 3731, The Netherlands

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_x) 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_x 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_3) precursor emissions of NO_x 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_3 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_2 emission reduction in Europe than assumed in policy projections.

In October 2015, the US Environmental Protection Agency (EPA) revised the O_3 standard (11) from 75 ppb (2008 standard) to 70 ppb. The new O_3 standard requires stricter controls on O_3 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_x emissions in Southern California in the period of 2016–2023. To better understand the variation of O_3 precursor emissions, we evaluate trends in EPA's NO_x and CO emission inventory data (*Methods*) between 2005 and 2015 by combining datasets including top-down anthropogenic NO_x and CO emission estimates from inverse analysis studies (6, 7), remotely sensed NO_2 measurements from the Ozone Monitoring Instrument (OMI), CO measurements from Measurement of Pollution in the Troposphere (MOPITT), surface in situ NO_2, CO, and O_3 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_x Emission Changes. In a recent study, Miyazaki et al. (6) estimated global NO_x emissions in the period of 2005–2015 using multiple satellite measurements (*SI Appendix*). The top-down NO_x emissions were obtained using an ensemble Kalman filter, while improving the representation of the chemical system (e.g., NO_x lifetime) affecting tropospheric NO_2 by assimilating multiple chemical species including CO and O_3 concentrations. Fig. 1*A* (green line) shows percent changes of the top-down anthropogenic NO_x emissions (normalized at 2009), indicating a dramatic slowdown (76%) in US NO_x emissions reduction from -7.0 ± 1.4%/y (2005–2009) to -1.7 ± 1.4%/y (2011–2015), as shown in Table 1. Uncertainties represent 1 σ and include the error budget described in *SI Appendix*. Average top-down anthropogenic NO_x emissions for the 11-y period are shown in Fig. 2*A*, demonstrating the strongest emissions in the northeast United States. Fig. 2 *B* and *C* shows the differences of top-down anthropogenic NO_x emissions from 2005–2006 to 2008–2009, and from 2011–2012 to

Significance

Emissions of nitrogen oxides (NO_x) have a large impact on air quality and climate change as precursors in the formation of ozone and secondary aerosols. We find that NO_x emissions have not been decreasing as expected in recent years (2011–2015) when comparing top-down estimates from satellites and surface NO_2 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_x 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.

[1]To whom correspondence should be addressed. Email: zhejiang@ustc.edu.cn.

[2]Present address: Department of Atmospheric and Oceanic Sciences, University of California, Los Angeles, CA 90095.

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

Published online April 30, 2018.

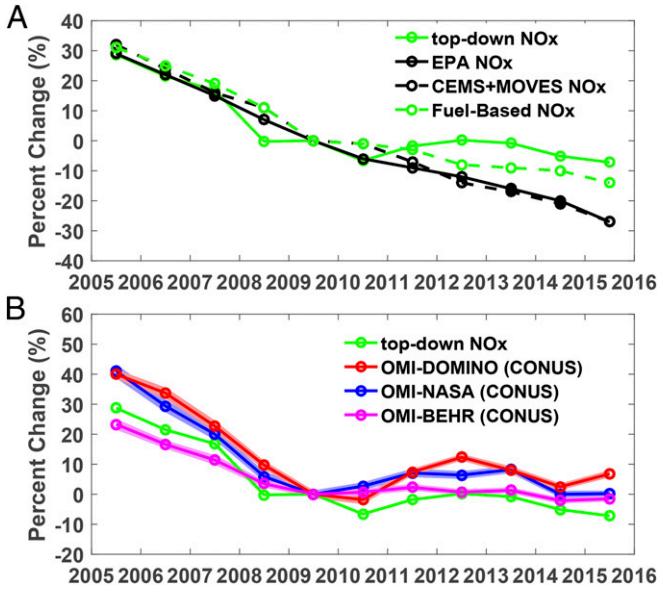

Fig. 1. (*A*) Percent changes (normalized at 2009) of top-down US anthropogenic NO_x emission estimates from inverse analysis (green line), EPA's emissions trends report data of NO_x (black solid line), revised EPA emission estimates including CEMS and MOVES national-scale data (black dashed line, *SI 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 NO_x emission estimates and tropospheric OMI NO_2 columns over CONUS. The shaded areas represent 1-σ uncertainties for random and sampling errors.

2014–2015, respectively. We find pronounced changes in the reduction of anthropogenic 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. 1*A* (black solid line) shows percent changes of EPA's bottom-up emission estimates (*Methods*). As shown in Table 1, trends of top-down anthropogenic NO_x emission estimates (−7.0 ± 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 ± 1.4%/y) and bottom-up (−5.3%/y) 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 NO_2 Abundance. Fig. 1*B* shows percent changes of the top-down anthropogenic NO_x emissions and tropospheric OMI NO_2 columns from National Aeronautics and Space Administration (NASA), Dutch OMI NO2 (DOMINO), and Berkeley High-Resolution (BEHR) products (*SI Appendix*) over CONUS. The interannual variation of top-down NO_x emissions generally follows the variation in OMI NO_2 measurements as expected, since the OMI DOMINO product is included in the assimilated data (6). Since each point in Fig. 1*B* 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 NO_2 interannual variations. Fig. 3 *A*–*F* displays maps of the differences of mean tropospheric OMI 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 NO_2 columns.

To corroborate the satellite observations of tropospheric NO_2 columns, we perform a similar analysis using surface in situ AQS measurements (*SI Appendix*). Fig. 4 *A* and *B* shows the differences of mean surface NO_2 concentrations, as measured by the AQS network, from 2005–2006 to 2008–2009, and from 2011–2012 to 2014–2015. Fig. 5*A* shows percent changes of the surface in situ AQS NO_2 measurements and tropospheric OMI 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 NO_2 data demonstrate good agreement with AQS NO_2 measurements. Fig. 5 *B*–*D* demonstrates agreement between AQS and OMI 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 NO_2 concentration reduction from −3.3%/y to −1.9%/y.

The similar slowdown of the reductions of observed NO_2 abundances demonstrates the slowdown of estimated NO_x emission reduction (6) is reasonable. In addition, the relation between changes in NO_x emissions and 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 NO_x limited in recent years, and thus, the same NO_x emission reduction may result in slower reduction in NO_2 abundance through an increase in NO_x lifetime. However, we do not expect a significant influence due to changes in urban NO_x chemistry because the slowdown (Fig. 3 *A*–*F*) is observable throughout much of CONUS. Furthermore, we tested the role of NO_x emissions in controlling NO_2 abundance with a sensitivity study where global surface 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 NO_2 concentrations (*SI Appendix*, Fig. S1), demonstrating that variations in NO_2 abundances are dominated by changes in emissions.

Changes in CO Emissions. Recent studies (1, 18, 19) have demonstrated that a synthesis of 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 NO_x emissions. In a recent study, Jiang et al. (7) constrained global CO emissions in the

Table 1. Trends and uncertainties for all NO_x datasets

Period	EPA NO_x	Top-down NO_x	OMI (NASA)	OMI (DOMINO)	OMI (BEHR)	AQS NO_2
2005–2009 (CONUS)	−6.4%	−7.0 ± 1.4%	−8.8 ± 1.0%	−8.6 ± 0.9%	−5.4 ± 1.0%	
2011–2015 (CONUS)	−5.3%	−1.7 ± 1.4%	−1.9 ± 0.8%	−1.0 ± 0.9%	−1.0 ± 0.8%	
2005–2009 (sampled)			−10.2 ± 1.8%	−9.6 ± 1.7%	−8.5 ± 1.8%	−6.6 ± 1.4%
2011–2015 (sampled)			−3.2 ± 1.6%	−2.6 ± 1.8%	−2.1 ± 1.6%	−2.6 ± 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 σ and include the error budget discussed in *SI Appendix*. OMI (sampled) represents OMI NO_2 measurements sampled at AQS NO_2 measurement locations and times based on monthly averages.

Table 2. Trends and uncertainties for CO datasets and eastern US AQS O_3

Period	EPA CO	Top-down CO	MOPITT CO	AQS CO	AQS O_3
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 σ and include the error budget discussed in *SI Appendix*. AQS O_3 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. 2*D* shows the 11-y averages of top-down anthropogenic CO emissions (7), excluding biomass burning and oxidation sources. Fig. 2 *E* 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_x and CO emissions.

Fig. 3 *G* 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_2 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_x lifetime) and the coarse vertical resolution of MOPITT profile retrievals (21). For example, *SI Appendix*, Fig. S2*C* 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. 2*F*).

Fig. 4 *C* 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_2 and CO, AQS in situ O_3 measurements over the eastern United States (Table 2) show a similar 75% slowdown of surface O_3 concentration reduction from −1.6%/y to −0.4%/y, suggesting the importance of diminishing rates of decrease for anthropogenic CO, VOCs, and NO_x.

Revisions to Bottom-Up Emission Estimates. What are the potential explanations for this dramatic slowdown of reductions of US anthropogenic NO_x 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_x emissions is surprising. Since the late 1990s, large decreases in NO_x 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_x 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_x emissions are expected to decline at a similar rate in the 2011–2015 time period as during 2005–2009.

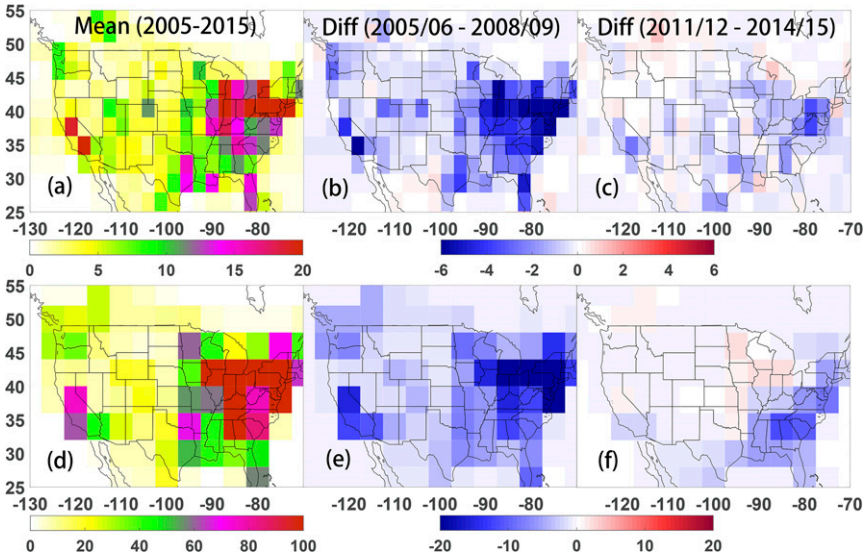

Fig. 2. (*A*) Mean top-down anthropogenic NO_x emissions from inverse analysis in the period 2005–2015. (*B* and *C*) Difference of top-down anthropogenic NO_x emissions from 2005–2006 to 2008–2009, and from 2011–2012 to 2014–2015. (*D*–*F*) same as *A*–*C*, but for top-down anthropogenic CO emissions. The unit is 10^{10} mole/cm^2/s.

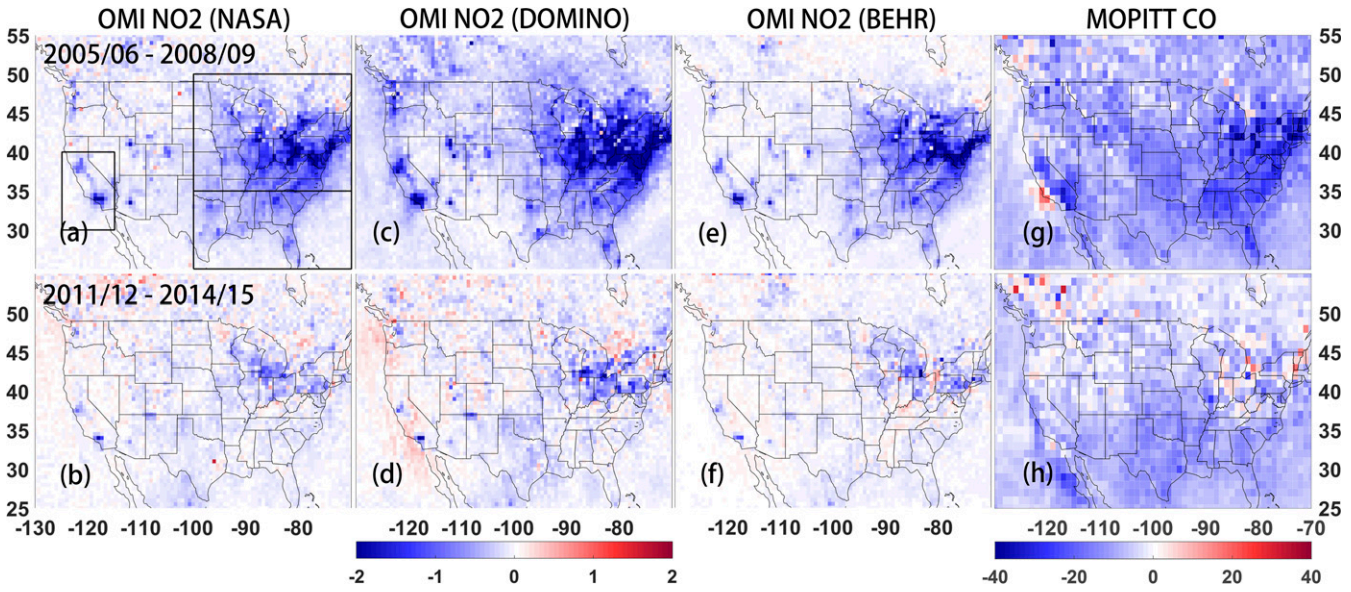

Fig. 3. (*A*–*F*) Difference of mean tropospheric OMI NO_2 columns from 2005–2006 to 2008–2009, and from 2011–2012 to 2014–2015. The unit is 10^{15} mole/cm^2. (*G* and *H*) same as *A*–*F*, but for MOPITT surface layer CO measurements with unit ppb (parts per billion). *A* also indicates the southwest, southeast, and northeast US regions for sampling satellite observations at the AQS sites used in Fig. 5 comparison.

Fig. 1*A* 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_x trend:

i) We estimate industrial, residential, and area source NO_x 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_x 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_x emissions are consistently declining by ∼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 ∼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_x emissions (33), which would also contribute to a slowdown.

iii) We estimate on-road diesel emissions using a fuel-based approach (25). While NO_x 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_x emissions are significantly elevated relative to in-use certification limits (34, 35). Recent roadside measurements of NO_x 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. 1*A*) gives a slowdown with the reduction rate of NO_x 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_2 measurements are −4.1 ± 2.2%/y (2005–2009) and −3.9 ± 2.5%/y (2011–2015) for the southwest United States (particularly from California), suggesting relatively stable reductions in this region. By contrast, the

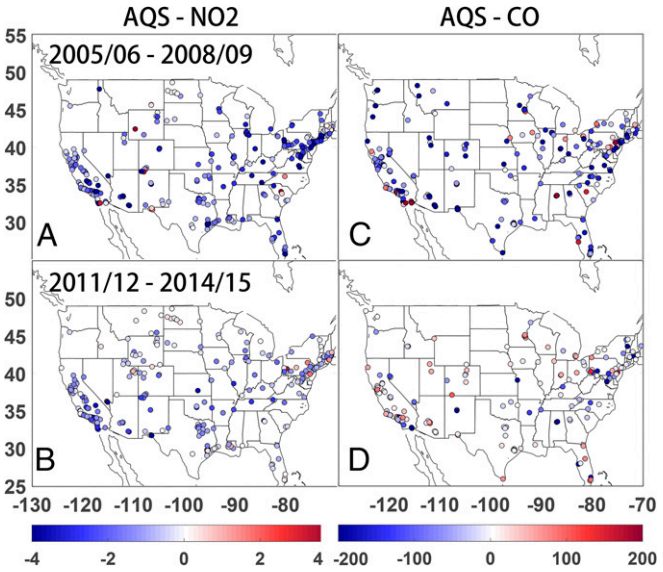

Fig. 4. (*A* and *B*) Difference of mean NO_2 concentrations of surface in situ NO_2 measurements (AQS stations) from 2005–2006 to 2008–2009, and from 2011–2012 to 2014–2015. (*C* and *D*) Same as *A* and *B*, but for surface in situ CO measurements. The unit is ppb.

reduction rates of AQS surface in situ NO_2 measurements are −7.8 ± 2.0%/y (2005–2009) and −2.6 ± 2.1%/y (2011–2015) for the northeast United States, and −6.7 ± 2.3%/y (2005–2009) and −0.1% ± 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 NO_2 columns (e.g., NASA product sampled at AQS NO_2 measurement locations and times in Fig. 5) is also much weaker: the reduction rates are −8.6 ± 4.0%/y (2005–2009) and –5.6 ± 3.6%/y (2011–2015) over the southwest United States, compared with −10.2 ± 1.8%/y (2005–2009) and −3.2% ± 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 NO_x emissions from such glider-kit trucks significantly exceed the emission standards promulgated in 2010 (37), which could contribute to a slowdown in NO_x emission reductions in regions where glider-kit trucks are operating in significant numbers.

There is also regional variability in trends of NO_x emissions from energy generation. Stack monitors on power plants indicate that 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 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 NO_2 columns have been shown to be increasing (5).

Conclusions

Using a synthesis of recently estimated top-down anthropogenic NO_x and CO emissions from inverse analysis studies (6, 7), remotely sensed NO_2 measurements from OMI, CO measurements from MOPITT, surface in situ 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 NO_x and CO emissions in the most recent years. The similar slowdown of surface 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 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 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 NO_x and CO and their subsequent effects on 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.

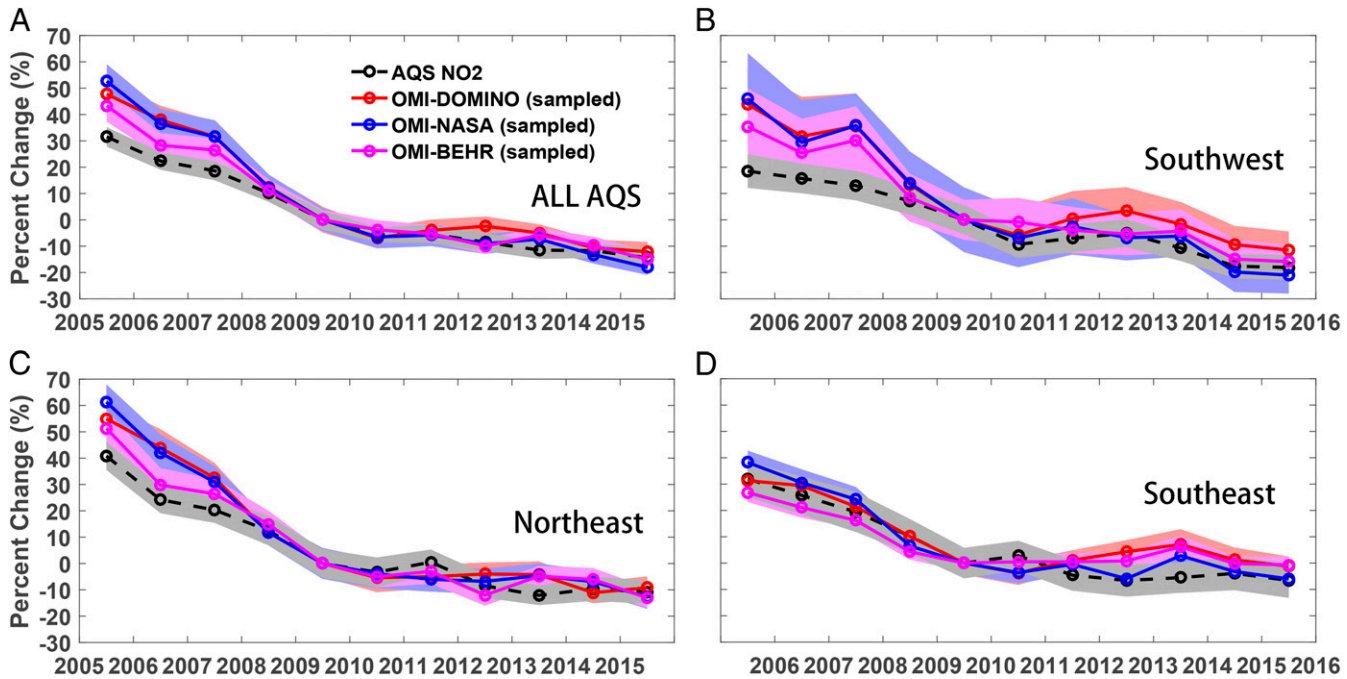

Fig. 5. (*A–D*) Percent changes (annual means normalized at 2009) of AQS surface in situ NO_2 measurements and tropospheric OMI NO_2 columns for various regions. Both AQS and OMI measurements are averaged with monthly resolution; the averaged OMI (monthly) data are sampled at AQS NO_2 (monthly) measurement locations and times; annual means are calculated based on monthly means. The region definition is shown in Fig. 3*A*. The shaded areas represent 1-σ uncertainties for random and sampling errors.

Methods

Bottom-Up NO_x 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_x, 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_x emission factors occur before 2005 and are relatively constant thereafter (26). We maintain this trend and hold NO_x 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. 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_x and CO emission data, tropospheric OMI NO_2 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_x and CO emission data and surface in situ NO_2, CO, and O_3 measurements (AQS). We acknowledge the OMI tropospheric NO_2 column data from https://disc.sci.gsfc.nasa.gov, www.temis.nl, and 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.

1. Hassler B, et al. (2016) Analysis of long-term observations of NO_x and CO in megacities and application to constraining emissions inventories. *Geophys Res Lett* 43:9920–9930.
2. Gratz LE, Jaffe DA, Hee JR (2015) Causes of increasing ozone and decreasing carbon monoxide in springtime at the Mt. Bachelor Observatory from 2004 to 2013. *Atmos Environ* 109:323–330.
3. Kharol SK, et al. (2015) Assessment of the magnitude and recent trends in satellite-derived ground-level nitrogen dioxide over North America. *Atmos Environ* 118:236–245.
4. Tong D, et al. (2015) Long-term NO_x trends over large cities in the United States during the great recession: Comparison of satellite retrievals, ground observations, and emission inventories. *Atmos Environ* 107:70–84.
5. Duncan B, et al. (2016) A space-based, high-resolution view of notable changes in urban NO_x pollution around the world (2005–2014). *J Geophys Res Atmos* 121:976–996.
6. Miyazaki K, et al. (2017) Decadal changes in global surface NO_x emissions from multi-constituent satellite data assimilation. *Atmos Chem Phys* 17:807–837.
7. Jiang Z, et al. (2017) A 15-year record of CO emissions constrained by MOPITT CO observations. *Atmos Chem Phys* 17:4565–4583.
8. Cooper O, Gao R, Tarasick D, Leblanc T, Sweeney C (2012) Long-term ozone trends at rural ozone monitoring sites across the United States, 1990–2010. *J Geophys Res Atmos* 117:D22307.
9. Strode S, et al. (2015) Trends and variability in surface ozone over the United States. *J Geophys Res Atmos* 120:9020–9042.
10. Grange SK, Lewis AC, Moller SJ, Carslaw DC (2017) Lower vehicular primary emissions of NO_2 in Europe than assumed in policy projections. *Nat Geosci* 10:914–918.
11. United States Environmental Protection Agency (2017) NAAQS Table Available at https://www.epa.gov/criteria-air-pollutants/naaqs-table. Accessed April 10, 2018.
12. South Coast Air Quality Management District (2016) Air Quality Management Plan. Available at www.aqmd.gov/docs/default-source/clean-air-plans/air-quality-management-plans/2016-air-quality-management-plan/final-2016-aqmp/final2016aqmp.pdf?sfvrsn=15. Accessed April 10, 2018.
13. Lamsal L, et al. (2015) U.S. NO_2 trends (2005–2013): EPA Air Quality System (AQS) data versus improved observations from the Ozone Monitoring Instrument (OMI). *Atmos Environ* 110:130–143.
14. United States Environmental Protection Agency (2017) Air Trend. Available at https://www3.epa.gov/cgi-bin/broker?_service=data&_program=dataprog.aqplot_data_2016.sas&parm=42602&stat=P98V&styear=2000&endyear=2016&pre=val&region=99. Accessed April 10, 2018.
15. Valin L, Russell A, Cohen R (2013) Variations of OH radical in an urban plume inferred from NO_2 column measurements. *Geophys Res Lett* 40:1856–1860.
16. Valin L, Russell A, Cohen R (2014) Chemical feedback effects on the spatial patterns of the NO_x weekend effect: A sensitivity analysis. *Atmos Chem Phys* 14:1–9.
17. Jin X, et al. (2017) Evaluating a space-based indicator of surface ozone-NO_x-VOC sensitivity over midlatitude source regions and application to decadal trends. *J Geophys Res Atmos* 122:10439–10461.
18. Konovalov I, et al. (2016) Estimation of fossil-fuel CO_2 emissions using satellite measurements of 'proxy' species. *Atmos Chem Phys* 16:13509–13540.
19. Silva S, Arellano A (2017) Characterizing regional-scale combustion using satellite retrievals of CO, NO_2 and CO_2. *Remote Sens* 9:744.
20. Warneke C, et al. (2012) Multiyear trends in volatile organic compounds in Los Angeles, California: Five decades of decreasing emissions. *J Geophys Res Atmos* 117:D00V17.
21. Deeter M, Edwards D, Gille J, Worden H (2015) Information content of MOPITT CO profile retrievals: Temporal and geographical variability. *J Geophys Res Atmos* 120:12723–12738.
22. McDonald BC, Gentner DR, Goldstein AH, Harley RA (2013) Long-term trends in motor vehicle emissions in U.S. urban areas. *Environ Sci Technol* 47:10022–10031.
23. Frost G, et al. (2006) Effects of changing power plant NOx emissions on ozone in the eastern United States: Proof of concept. *J Geophys Res Atmos* 111:D12306.
24. Gouw J, Parrish D, Frost G, Trainer M (2014) Reduced emissions of CO_2, NO_x, and SO_2 from U.S. power plants owing to switch from coal to natural gas with combined cycle technology. *Earths Futur* 2:75–82.
25. McDonald B, Dallmann T, Martin E, Harley R (2012) Long-term trends in nitrogen oxide emissions from motor vehicles at national, state, and air basin scales. *J Geophys Res Atmos* 117:D00V18.
26. Xing J, et al. (2013) Historical gaseous and primary aerosol emissions in the United States from 1990 to 2010. *Atmos Chem Phys* 13:7531–7549.
27. Kean AJ, Sawyer RF, Harley RA (2000) A fuel-based assessment of off-road diesel engine emissions. *J Air Waste Manag Assoc* 50:1929–1939.
28. Dallmann TR, Harley RA (2010) Evaluation of mobile source emission trends in the United States. *J Geophys Res Atmos* 115:D14305.
29. Anderson D, et al. (2014) Measured and modeled CO and NO_y in DISCOVER-AQ: An evaluation of emissions and chemistry over the eastern US. *Atmos Environ* 96:78–87.
30. Canty T, et al. (2015) Ozone and NO_x chemistry in the eastern US: Evaluation of CMAQ/CB05 with satellite (OMI) data. *Atmos Chem Phys* 15:10965–10982.
31. Souri A, et al. (2016) Constraining NO_x emissions using satellite NO_2 measurements during 2013 DISCOVER-AQ Texas campaign. *Atmos Environ* 131:371–381.
32. Travis K, et al. (2016) Why do models overestimate surface ozone in the Southeast United States? *Atmos Chem Phys* 16:13561–13577.
33. Coordinating Research Council (2017) On-road remote sensing of automobile emissions in the Chicago area: Fall 2016. Available at https://crcao.org/reports/recentstudies2017/E-106/E-106%20Chicago%202016%20Final%20Report%20v4_Sept%202017.pdf. Accessed April 10, 2018.
34. Thiruvengadam A, et al. (2015) Emission rates of regulated pollutants from current technology heavy-duty diesel and natural gas goods movement vehicles. *Environ Sci Technol* 49:5236–5244.
35. Dixit P, et al. (2017) Differences between emissions measured in urban driving and certification testing of heavy-duty diesel engines. *Atmos Environ* 166:276–285.
36. Haugen MJ, Bishop GA (2017) Repeat fuel specific emission measurements on two California heavy-duty truck fleets. *Environ Sci Technol* 51:4100–4107.
37. United States Environmental Protection Agency (2017) Chassis dynamometer testing of two recent model year heavy-duty on-highway diesel glider vehicles. Available at https://www.dieselnet.com/misc/201711_epa_glider_report.pdf. Accessed April 10, 2018.
38. United States Environmental Protection Agency (2017) Annual NOx emissions from CSAPR and ARP sources. Available at https://www3.epa.gov/airmarkets/progress/reports/emissions_reductions_nox.html#figure1. Accessed April 10, 2018.
39. United States Energy Information Administration (2016) State Energy Data System (SEDS) 1960-2014. Available at https://www.eia.gov/state/seds/. Accessed April 10, 2018.
40. Federal Highway Administration (2016) Highway Statistics 2015, Table MF-2: Motor-Fuel Volume Taxed by States. Available at https://www.fhwa.dot.gov/policyinformation/statistics/2015/mf2.cfm. Accessed April 10, 2018.
41. Federal Highway Administration (2016) Highway Statistics 2015, Table MF-24: Non-highway use of gasoline. Available at https://www.fhwa.dot.gov/policyinformation/statistics/2015/mf24.cfm. Accessed April 10, 2018.
42. United States Energy Information Administration (2016) Fuel Oil and Kerosene Sales 2015. Available at https://www.eia.gov/petroleum/fueloilkerosene/archive/2015/pdf/foks.pdf. Accessed April 10, 2018.
43. Volckens J, et al. (2007) Emissions profile from new and in-use handheld, 2-stroke engines. *Atmos Environ* 41:640–649.
44. Burgard DA, Bria CR, Berenbeim JA (2011) Remote sensing of emissions from in-use small engine marine vessels. *Environ Sci Technol* 45:2894–2901.
45. Gordon TD, et al. (2013) Primary gas- and particle-phase emissions and secondary organic aerosol production from gasoline and diesel off-road engines. *Environ Sci Technol* 47:14137–14146.