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Research Report 195

**Impacts of Regulations on Air Quality and Emergency
Department Visits in the Atlanta Metropolitan Area, 1999–2013**

Russell et al.

**APPENDIX D. EVIDENCE FOR A POTENTIAL OVERESTIMATION
IN MOBILE SOURCE NO_x EMISSIONS**

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Correspondence may be addressed to Dr. Armistead (Ted) Russell, Department of Civil and Environmental Engineering, Georgia Institute of Technology, 311 Ferst Drive, Atlanta, GA 30322; e-mail: trussell@ce.gatech.edu.

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HEI Research Report 195 Russell Appendix D (Available on the HEI Website)

APPENDIX D. EVIDENCE FOR A POTENTIAL OVERESTIMATION IN MOBILE SOURCE NO_x EMISSIONS

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INTRODUCTION

Accurate estimation of oxides of nitrogen (NO_x*) emissions is critical to understanding the impact of mobile sources on atmospheric chemistry and human health (Gego et al. 2008; Kim et al. 2009; Stavrou et al. 2008). On-road mobile sources, one of the largest emission sources of health-related air pollutants, are typically estimated using bottom-up methods. These methods can have large uncertainties due to the complexities of the models used and the uncertainties in both the model inputs and parameters, (e.g., uncertainties in determining emission factors and human activity intensity (Kim et al. 2009)). However, identifying and quantifying potential biases that may be present in estimated emissions rates and activities for large fleets (such as Atlanta's) that are made up of multiple vehicle types is difficult. Snyder et al. (2014) found that differences in vehicle miles traveled (VMT) estimates from two models translated into +45% to -35% uncertainty in emissions. They noted that uncertainty in the ratios of heavy duty to light duty automobiles would affect certain pollutants, such as particulate matter $\leq 2.5 \mu\text{m}$ in aerodynamic diameter (PM_{2.5}) and NO_x, more than others, such as carbon monoxide (CO). Adjusting the percent of diesel vehicles from 5.2% to 9.2% on one specific

* A list of abbreviations and other terms appears at the end of this appendix.

type of roadway in their simulations changed total estimated PM_{2.5} and NO_x by 53% and 29%, respectively. Boroujeni and Frey (2014) investigated road grade parameterizations and Sandhu et al. (2014) addressed refuse truck emissions rates. No studies to date have combined uncertainty estimates of each of the models within MOVES (U.S. EPA Motor Vehicle Emissions Simulator) to estimate uncertainty across multiple inputs. Hanna et al. (2001) estimated uncertainty in various air quality inputs using an expert elicitation, and applied $\pm 100\%$ for mobile sources. Using a variety of methods and comparisons — including fuel and mileage-based emissions factors, ground-based observations, satellite inverse modeling, and regional chemical transport modeling — multiple studies have found large variability between modeled and a priori estimates (Dallmann and Harley 2010; Deguillaume et al. 2007; Konovalov et al. 2006; Napelenok et al. 2008; Parrish 2006).

There is evidence from other areas of the country that the mobile source NO_x emissions estimates may be biased high by as much as a factor of two (Anderson et al. 2014; Fujita et al. 2012; Kim et al. 2009; SW Kim et al. 2016). Evidence for the magnitude of the overestimate is derived from other modeling studies and from satellite-based observations. (Anderson et al. 2014) suggests the overestimate may be due to MOVES's treatment of degrading catalysts in aging automobiles. At the recent Community Modeling and Analysis System (CMAS) conference, multiple presentations discussed the potential bias issue (e.g., Frost 2016: See www.cmascenter.org/conference/2016/agenda.cfm).

While there is evidence of a potential bias in mobile source NO_x (and CO) emissions, there is also evidence that modeled emissions capture air pollutant trends accurately enough as to allow air quality models to provide adequate estimates of air quality and changes over time (Foley et al. 2015a,b; Gego et al. 2008; Gilliland et al. 2008; Simon et al. 2012). For some modeling applications (such as statistical models), capturing trends in emissions changes is more important than the absolute emissions amount (e.g., the empirical modeling approach taken here). Previous studies have found that modeled mobile emissions trends match expected trends based on ambient air quality observations (Blanchard et al. 2012; Pachon et al. 2012).

For other applications, however, a major bias (under or overestimate) of emissions in the Southeast or elsewhere has major implications on this work and air quality management generally. First, if it is true that the actual mobile source emissions are now lower than estimated, that would indicate that the regulations are more effective than believed. Second, and possibly more important, is that the amount of NO_x emissions to be further reduced is lower, which has implications on how to assess potential future interventions and regulatory effectiveness. There are other ramifications (e.g., air quality model evaluation). It is difficult to overstate the importance of better understanding and quantifying mobile source NO_x emissions, particularly in the light of the major reductions in utility NO_x emissions and the tightening ozone standard. Understanding how NO_x-limited the region is will be critical to identifying the most effective strategies to further reduce ozone (and particulate matter [PM]), and quantify the degree of further controls required and from what sources such reductions may come.

Here we examined this issue in multiple ways, including using air quality modeling evaluations of multiple models, empirical trend analysis of NO_x concentrations using both ground- and satellite-based observations and using a ratio-of-ratios method. Our results are suggestive of a

potential bias, but the evidence here is not strong and consistent between the different approaches and can be obscured by the more complex chemistry of the large biogenic fluxes in the Southeast, e.g., leading to organonitrates, which are not typically measured in ground level monitoring networks, and are not observed from space. The satellite data and ground level observations don't follow the same trends as the NO_x emissions. The ratio-of-ratios method is suggestive of a potential low bias as ratios of the ambient concentrations of NO_x to those of another directly emitted species (elemental carbon [EC]) have been reduced proportionally less in relationship to their emissions. In addition, we find a high bias in our simulated NO_x (and CO) concentrations in our air quality modeling as compared to observations.

METHODS

We use three general approaches in this subanalysis. These include comparing air quality modeling results to observations, comparing emissions changes to concentration changes, as well as a ratio-of-ratios approach, and direct comparison of observed and modeled trends. Data sources include satellite observations, in-situ air quality observations, and emissions inventories.

Ground-Based Air Quality Data

Ground-based air quality data used in our analyses are taken from U.S. Environmental Protection Committee's (U.S. EPA) Air Quality System (<https://www.epa.gov/ttn/airs/airsaqs/detaildata/downloadaqsddata.htm>), and supplemented with data from the Southeastern Aerosol Research and Characterization (SEARCH) study. More details can be found in Henneman et al. 2017b and in the main body of the report.

Satellite Data Source

As part of this analysis, we also include observations from space. Three satellite products are used to compare with surface monitoring and emissions in Atlanta, GA (Boersma et al. 2004, 2010). Each satellite product recorded tropospheric nitrogen dioxide (NO₂) columns during their operating period, and the overall time series of the three satellites covered the whole period between 1997 and 2013 (GOME 06/1996–06/2003, SCIAMACHY 08/2002–03/2012, OMI 10/2014–12/2013). Monthly mean tropospheric NO₂ columns were downloaded directly from the Web site (European Space Agency 2016). The satellite crossing times over Atlanta, Georgia, were approximately 3:00 PM (local time). Considering the NO₂ diurnal variation, we chose daily surface concentrations monitored at 3:00 PM to conduct the comparison. On-road mobile emissions (hereafter referred to as *mobile*) included in the analysis were modeled using MOVES 2010b (U.S. EPA 2012) for the 20-county Atlanta PM_{2.5} nonattainment area. Electricity generating unit (EGU) emissions were collected from plants located in the same region and downloaded from the U.S. EPA's Air Markets Program Database (U.S. EPA 2016). The emissions data sets are described in Henneman et al. 2017b). Both interannual temporal trends and monthly variations were compared across satellite platforms, ambient concentrations, and emissions.

Data Analysis Approaches

Ratio of Ratios Analysis The ratio-of-ratios (RR) method is used to assess mobile emissions trends over time and provides information on potential biases, both by considering how trends deviate over time and also how close the RR value is to one. The RR analysis calculates the ratio between two pollutant species in emissions to observations. Using NO_x and EC as an example, the RR value is calculated as follows,

$$RR = \frac{\left[\frac{NO_x}{EC}\right]_{emiss}}{\left[\frac{NO_x}{EC}\right]_{obs}}$$

where the emission ratio (*emiss*) and the observation ratio (*obs*) are the ratios of NO_x to EC, and RR is the ratio of the two ratios. The units used should be consistent between the species, and it is important to consider how emissions are reported (e.g., NO_x emissions are reported as NO₂). We calculated temporal trends of three RR values (CO:NO_x, NO_x:EC, and EC:CO) between different compounds over the period from 1999 to 2013 at the Jefferson Street monitoring station (JST) in Atlanta, Georgia. Gaseous species were converted from molar ratios (ppb) to mass using atomic mass ratios between the species of interest and air (48 g/mol). NO_x mass was taken as the mass of NO₂ to align with the method taken in MOVES emissions modeling (U.S. EPA 2010), as well as how emissions are reported in the National Emission Inventory. Assuming the emission estimate being accurate and the air degradation rates of the two compounds being the same, the RR value of the two compounds would be close to 1. A value higher or lower than 1 suggests either a disparity in air degradation rates between the two compounds or a bias in emission estimate, though evidence from this test alone cannot verify a bias.

R-LINE Simulation The Research LINE-source (R-LINE) dispersion model for near-surface releases is a dispersion model developed to line sources impacts on ambient air quality (Snyder et al. 2013). R-LINE is a steady-state dispersion model that simulates physical dispersion processes using wind speed, wind direction, Monin-Obukhov length for turbulence, surface friction velocity, and other meteorological parameters. In an application in Atlanta, 10 years of annual average concentration fields for PM_{2.5}, CO, and NO_x in a 20-county area in Atlanta are developed using an annual average approach that accounts for the frequencies of different meteorological conditions and emission diurnal changes (Zhai et al. 2016). The emissions used are link emissions of 2010 at 43,712 links based on the Atlanta Regional Commission's 20-county activity-based travel demand model in the Atlanta Roadside Emissions Exposure Study (AREES) (Kim et al. 2009). The emissions are adjusted using annual variability in the MOVES-modeled emission factors.

R-LINE estimates are based on mobile sources but are compared to total observations of CO and NO_x since mobile sources are the major sources of the two species (i.e., mobile sources are estimated to consist of 88% and 73% of ground level CO and NO_x emissions). For PM_{2.5}, the R-LINE estimates are calibrated to mobile-source impacts estimated using chemical mass balance with gas constraints (CMB-GC) based on observational data. Regression approaches are explored here in linear and log-transformed forms using the jackknife resampling method (Sahinler and Topuz 2007), which estimates regression parameters with each available observation data point withheld one-at-a-time. In

total, available observations in 10 years were used at three sites for PM_{2.5}, five sites for CO, and seven sites for NO_x in Atlanta.

Community Multiscale Air Quality Model Simulations We conducted Community Multiscale Air Quality (CMAQ) version 5.0.2 simulation for 2011 at 36 and 12 km resolution, respectively. Detailed information on the 12-km simulation and evaluation data can be found in Henneman and colleagues (2017b). The 36-km simulation covered the overall contiguous United States, and the emission inventory was compiled based on emission data from the National Emission Inventory using the Sparse Matrix Operator Kernel Emissions (SMOKE) model version 3.5.1. The emission data source is the same as for the 12-km simulation to exclude the influence of emissions on differences between the two simulation results. It should be noted, the high bias in NO_x emissions estimates is likely greater in later years, thus the choice of using 2011 results, but more recent model applications would be preferable and have been used in informing this discussion. However, those simulations were not conducted as part of the HEI Accountability study. For example, we conducted detailed modeling of the 2013 Southern Oxidants and Aerosol Study intensive period. This period, however, was only one month long, so is less complete than the 2011 modeling period used here. Further, we considered information from recent regulatory-oriented modeling conducted for the Southeast.

Ozone sensitivities to NO_x are estimated using CMAQ in July 2011 for two cases: a base case with actual emissions, and an adjusted case with mobile emissions reduced by 50%. This exercise tests both the effect of the magnitude of NO_x emissions on NO₂ and ozone levels and the model's ability to capture nonlinearities in the concentrations vs. emissions curve.

RESULTS

1. Comparison of Observational Trends to Mobile Source and Utility NO_x Emissions

The Long-Term Temporal Trends of Satellite Records are Consistent with Surface NO₂ Monitoring

Given that the satellite product algorithms do not rely on surface NO₂ observations (Boersma et al. 2004, 2011), we first compared the trends of satellite-based vertical column density (VCD) and surface NO₂ concentrations during the period from 1999 to 2013 (Figure D.1). It was found that long-term temporal trends from these two data sources agreed well with each other at both the JST and the Yorkville, Georgia (YRK) sites. For instance, both satellite records and surface monitoring showed a valley point appearing in 2001, a peak point in 2005, and a continuous decreasing trend afterwards at the JST site. Similarly, both data sources indicated a gradually decreasing trend at the YRK site during the last decade. We also noticed a higher interannual VCD variability reported by SCIAMACHY satellite data compared with OMI satellite data at the JST site during the overlapped period, which might be caused by the instrumental and retrieval differences between the two datasets (Boersma et al. 2008).

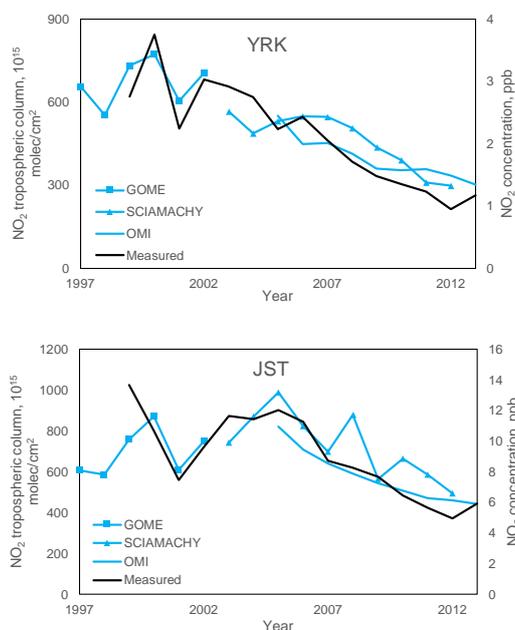


Figure D.1. Comparisons of long-term temporal trends between satellite-monitored tropospheric NO₂ columns and (1) surface-monitoring NO₂ concentrations at YRK site; (2) surface-monitored NO₂ concentrations at JST site.

Monthly variations were also derived from each data source for intercomparison (Figure D.2). At the JST (urban) site, satellite-based monthly variation agrees well with surface monitoring, with both showing higher concentrations in winter, and lower concentrations in summer. At YRK (rural site), however, surface concentrations show a slightly higher increase ratio in winter than the NO₂ VCD, likely caused by seasonal changes of planetary boundary layer height and enhanced local emissions for heating demand. Generally, the satellite-based NO₂ VCD are consistent with the surface-measured NO₂ in the long-term temporal trends and monthly variations in both urban and rural regions within the study area.

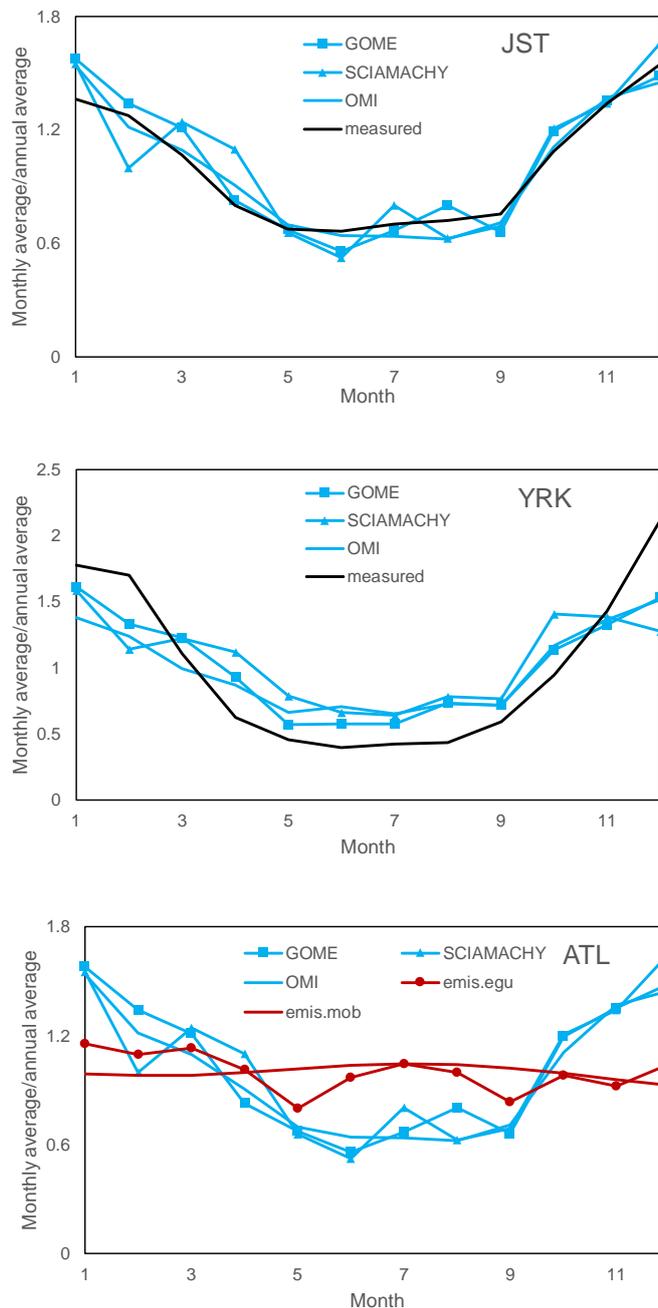


Figure D.2. Comparisons of monthly variations between satellite-monitored tropospheric NO₂ columns and (1) surface-monitoring NO₂ concentrations at YRK site; (2) surface-monitoring NO₂ concentrations at JST site; (3) NO_x emissions in Atlanta.

It should be noted that, due to the relatively coarse resolution (0.25 degree), satellite NO₂ shows a much lower spatial variation. The average NO₂ VCD in the urban site was only 45% higher than that at the rural site, while the surface-monitoring concentration was 220% higher. Similar results have been found in a previous study showing that the satellite records with coarse footprint-pixel sizes tended to smooth the NO₂ variations between the urban core and outside (HC Kim et al. 2016).

Comparison Between NO_x Observation and Emission Estimates Shows Inconsistent Temporal Trends

Inconsistencies in monthly and interannual variations and the longer-term trends are found in the comparison between NO₂ observations with NO_x emissions in Atlanta. Observations have a higher monthly-to-annual-average ratio in winter and a slower interannual decreasing trend compared with emissions (Figure D.2). Additionally, no peak point was found in emissions around

2005 (Figure D.3). Inconsistencies in monthly variation can be caused by seasonal difference in meteorological conditions, since NO_2 tends to be constrained in a relatively low level in summer by rapid photochemical reactions. However, for interannual variation, the influence of meteorological factors is limited (Andersson et al. 2007). The inconsistency in interannual variation is highly likely to be raised by potential biases inherent in emission estimation. Mobile emissions and EGU are the two major sources in Atlanta. Compared with mobile emissions, EGU emission estimates are expected to have much less uncertainty, because the emission amounts are determined through a real-time monitoring system. Mobile emissions, however, are based on a bottom-up method and could be quite susceptible to bias. Bottom-up emission estimates are usually highly uncertain. Large uncertainty could be caused by uncertainty in emission factors as well as the activity data that feed into estimation (van Aardenne and Pulles 2002).

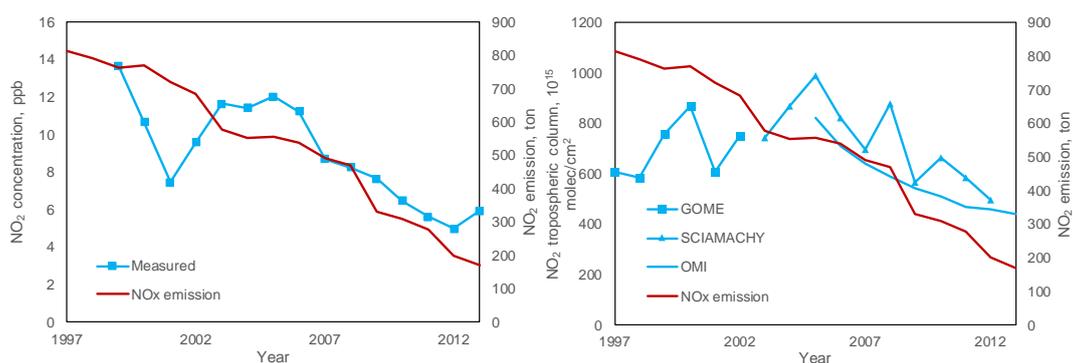


Figure D.3. Comparisons of NO_x/NO_2 trends (1) between total NO_x emissions (EGU + mobile) in Atlanta and surface NO_2 concentrations at JST; **(2)** between total NO_x emissions in Atlanta and tropospheric NO_2 columns.

2. Ratio of Ratios Analysis

We calculated a time series of mean RRs of $\text{CO}:\text{NO}_x$, $\text{NO}_x:\text{EC}$, and $\text{EC}:\text{CO}$, respectively, from 1999 to 2013 along with the annual 95% confidence interval (Figure D.4). The RR's are calculated first without correction for the CO background and are then calculated when accounting for the CO background (CO is naturally produced from the oxidation of biogenic volatile organic compounds [VOCs]). If the uncorrected CO concentrations are used, the RRs of $\text{EC}:\text{CO}$ are all close to 1 (indeed, 95% confidence intervals of each of the ratios for all years except for 2012 in the YRK-adjusted and 2009 in the unadjusted values contain 1), suggesting that there is no major bias in the emissions, or that the CO and NO_x emissions have similar biases.

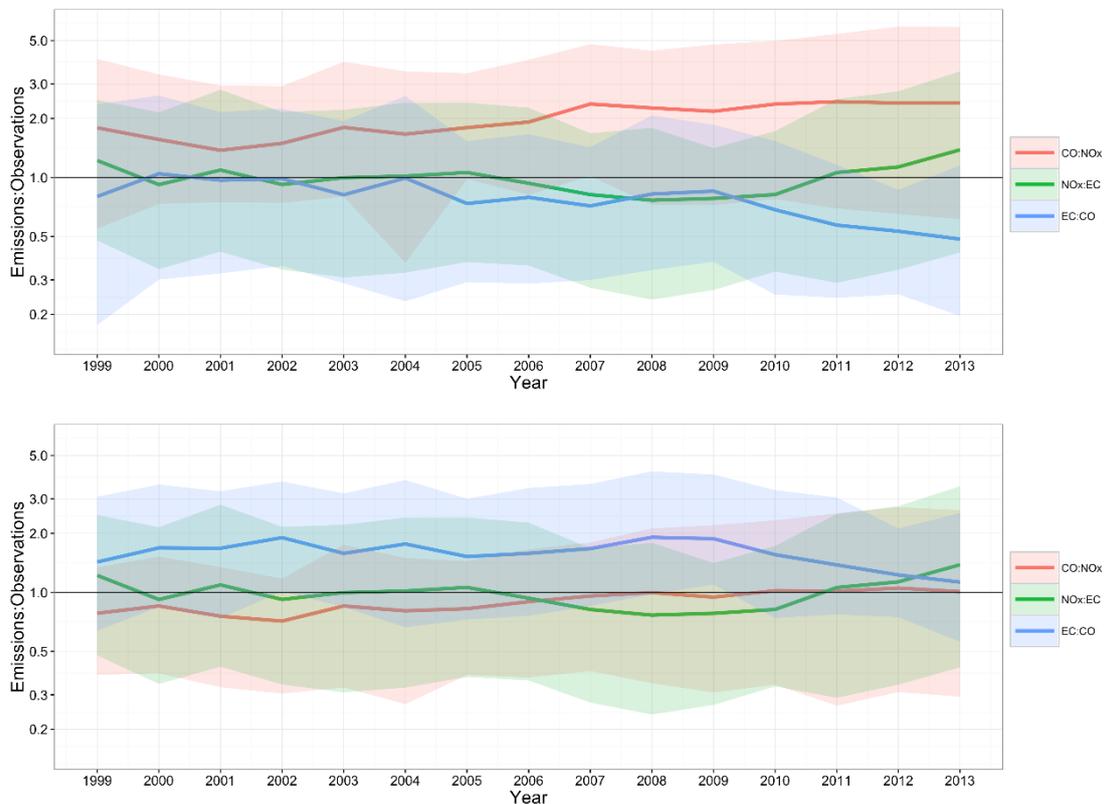


Figure D.4. Temporal trends of the ratios of ratios of CO:NO_x, NO_x:EC, and EC:CO from 1999 to 2013. Top: CO is corrected for background using YRK measurements, **Bottom:** CO is taken as raw observations at JST. Shaded areas correspond to a 95% confidence interval around the mean, taken from annual distributions of daily values. Vertical axis is on a log scale. (Zhai et al. 2016. Creative Commons license:

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However, if the CO (and NO_x) values are corrected, there is a major bias in emissions CO:NO_x ratios as compared to ambient concentrations. The correction is conducted using the YRK values as the background. In this case, the difference between the JST and YRK observations are used to account for the natural (biogenic) background. This is an imperfect comparison because air quality at YRK is impacted somewhat by anthropogenic Atlanta emissions (Blanchard et al. 2012), though the background values obtained are in alignment with estimates of background CO (e.g., Seinfeld and Pandis 2005). After adjustment for the background, the elevated CO:NO_x RR in this plot provides evidence that NO_x emissions are underestimated compared to CO. It should also be noted that there is a recent upwards trend in the NO_x-to-EC ratio, also suggesting a trend towards an increasing bias in the NO_x emissions compared to EC emissions estimates.

The analysis is based on the assumption that air degradation rates between compounds are similar. Air quality models can be further applied to provide the modeled RR values for reference. Stronger evidences are expected based on the ratio-of-ratios analysis.

3. R-LINE

As part of a variety of studies (Sarnat et al. In press; Zhai et al. 2016), we have been applying R-LINE to estimate mobile-source emissions and the related air quality impacts in Atlanta. We have found that R-LINE simulated values of NO_x , CO, and mobile-source derived $\text{PM}_{2.5}$ (we use CMB-GC modeling to derive the PM from mobile sources: see Zhai et al. 2016, for details) are biased high in these applications, so they have been calibrated to observations. If NO_x requires a different adjustment than the other pollutants, that would indicate there is a different bias in the emissions of NO_x than of other pollutants.

Regression relationships (Figure D.5) indicate an overestimation of R-LINE estimates compared to CMB-GC for $\text{PM}_{2.5}$, and observations in Atlanta for daily 1-hour maximum CO and NO_x , with slopes of 0.54, 0.69, and 0.30 for $\text{PM}_{2.5}$, CO, and NO_x , respectively. R-LINE estimates were higher by factors of 1.8, 1.3, and 4.2 on average, respectively, for $\text{PM}_{2.5}$, CO, and NO_x . The normalized root mean square error (NRMSE) and normalized mean bias (NMB) are especially high for NO_x (Table D.1), indicating the highest bias for NO_x .

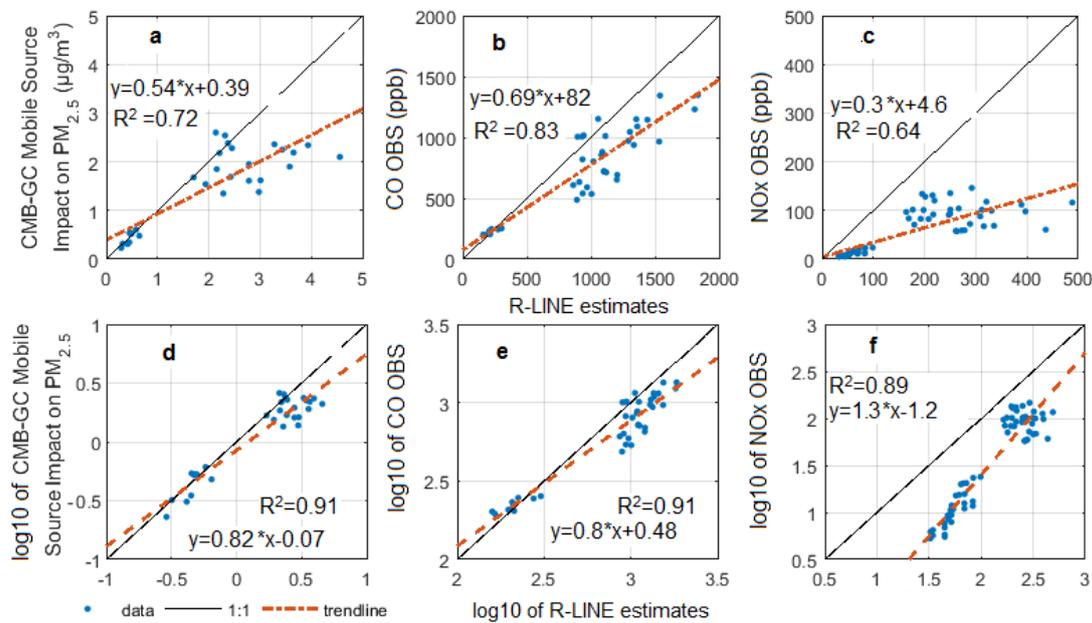


Figure D.5. Regressions between R-LINE estimates and CMB-GC estimates of mobile source $\text{PM}_{2.5}$ and observations for CO and NO_x in Atlanta, performed on a linear basis in the top row and log basis in bottom row. (Zhai et al. 2016. Creative Commons license:

<https://s100.copyright.com/AppDispatchServlet?publisherName=ELS&contentID=S1352231016308111&orderBeanReset=true>).



Figure D.6. Near-road measurement location. The concentration field shown is raw R-LINE PM_{2.5} mobile concentrations of 2015 (A 3 km by 3.5 km area in central Atlanta in 250-m resolution, µg/m³). The black square denotes the JST site location. The diamond denotes the near-road site by Georgia Environmental Protection Division, located next to the maximum concentration.

The near roadway estimates from R-LINE model are very high compared to measurements from 2015 at a near-road site located next to a major highway (I-85/I-75 connector) in Midtown Atlanta and next to the peak location, as shown in Figure D.6. The 2015 annual average observations are lower than R-LINE estimates by a factor of 3.1 and 7.4, respectively, suggesting large near-road biases in R-LINE model results. More importantly, here, is that the bias in NO_x estimates are higher than for CO and PM_{2.5}, suggesting a higher bias in those emissions estimates. However, R-LINE estimates are rather biased across the board, so other factors are likely playing a role.

Table D.1. Error and bias of R-LINE compared to CMB-GC mobile source impacts for PM_{2.5}, and observations for CO and NO_x.

	NRMSE	NMB
PM _{2.5}	39%	29%
CO	33%	22%
NO _x	326%	303%

Zhai et al. 2016. Creative Commons license:

<https://s100.copyright.com/AppDispatchServlet?publisherName=ELS&contentID=S1352231016308111&orderBeanReset=true>.

4. CMAQ Model Assessment: Comparison of Simulated and Observed NO_x and Resolution and Sensitivity Analysis

As part of this study, we conducted an evaluation of CMAQ using observations from the eastern United States, with a focus on the Southeast. The simulations were conducted at 12 km, though we also consider a 36 km simulation here to explore potential biases. We found a tendency for the modeled NO_x to be biased high (43%), suggesting that NO_x emission estimates are biased high. Given that EGU NO_x is unlikely to have large biases, this would suggest an even larger bias in mobile source emissions estimates. Henneman and colleagues (2017b) found an even larger bias in the comparison of simulated to observed NO₂ at JST over the entire four years modeled in the project (over 300%) and also a high bias in nitrate (over 40%), further suggesting a high bias in the emissions estimates.

Table D.2. Model performance for NO₂ for the Southeastern U.S. Normalized mean bias (NMB), mean normalized bias (MNB), the slope (B) and the semi-95% confidence interval range for the slope (B_{int}) of CMAQ NO_x simulation at 36 and 12 km resolutions compared to observations.

	NMB	MNB	B	B _{int}
36 km	13%	30%	1.18	0.17
12 km	24%	43%	1.30	0.13

In addition to emissions biases, a mismatch in spatial resolution could introduce uncertainty and bias in the model evaluation. Previous studies have shown that model simulation at coarse resolutions can underestimate concentrations of compounds with a relatively short atmospheric life time, such as polycyclic aromatic hydrocarbon compounds and black carbon (Shen et al. 2014; Wang et al. 2014). To evaluate the CMAQ model and determine how the emissions and model resolutions contribute to model uncertainty respectively, we conducted CMAQ simulation at two different resolutions of 12 km and 36 km and compared the modeled annual average NO₂ concentrations with near-surface observations (Figure D.7). In both cases, the predictions trend higher than the observations. Given the relatively higher resolution, it is reasonable to believe that spatial variations in NO₂ distribution was better captured by the 12-km simulation, which resulted in decreased model uncertainty relative to the 36-km simulation. However, assuming the emissions were accurate, mean bias should also be reduced by the 12-km simulation, because most of the observation sites are located in cities or surrounding areas where emissions are intensified and varied, and they are poorly captured by coarse resolutions such as 36 km. The increased mean bias indicates a potential overestimation of NO₂ emissions.

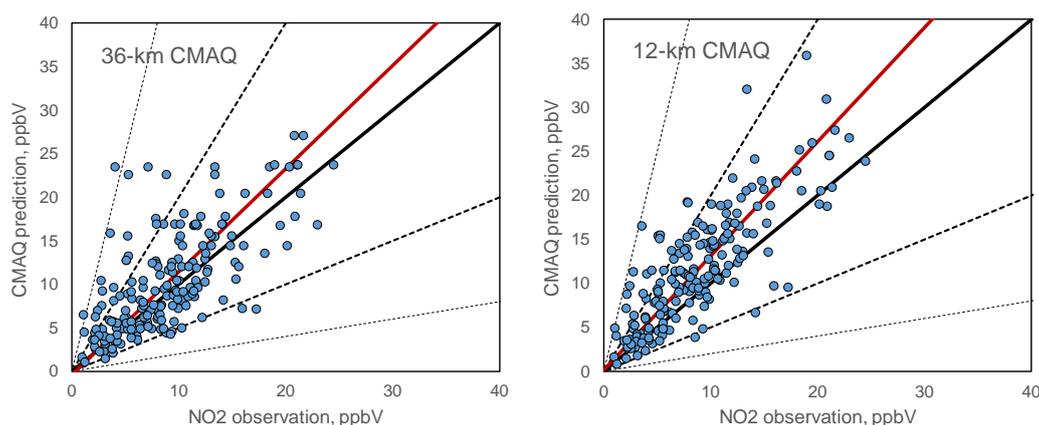


Figure D.7. Comparisons of annual average CMAQ-modeled NO₂ concentrations with observations in the Southeastern United States. 1: model simulation at 36 km resolution, **2:** model simulation at 12 km resolution. Each plot represents a monitoring site. Black solid lines are the 1:1 lines. Black dashed lines show 0.5~2 times range and grey dashed lines show 0.2~5 times range. Reduced-major-axis regression lines (red solid lines) are also shown.

12-km CMAQ simulations with the Decoupled Direct Method can provide modeled time series with source contributions. The time series of NO₂ concentrations is at least two times higher than the

observed level at the JST site in both the 2001–2002 and the 2011–2012 periods (see figures in Appendix A). Additionally, NO₂ is one of the worst fits for EGU and mobile sensitivities compared with the empirical model derived in this study. The time series shows the seasonality being essentially opposite for CMAQ vs. empirical mobile sensitivities. Generally, these results suggest an overestimate of NO₂ emissions that is mostly induced by mobile emissions.

As part of a recent regulatory-oriented modeling study in the Southeast, CMAQ was applied to 2007 (see Odman and Adelman 2014). They found a 43% high bias in their simulated NO₂ and they also found a high bias in the wet deposition of oxidized nitrogen, further suggesting a high bias in the emissions. It should be noted that this was for a 2007 modeling period, so any further deviation of the emissions estimates from actual emissions would likely accentuate the bias.

DISCUSSION

In this study, we first confirmed the long-term temporal trends of NO_x observations at JST and YRK by comparing with satellite-based tropospheric NO₂ columns compared with emissions yield disparity in temporal trends between observations and emissions. This disparity indicates a potential bias in NO_x emission estimates. Ratio-of-ratios analysis also suggests a bias, after correction for background concentrations. Mobile emissions contribute a large amount of NO_x emission in Atlanta, as do EGUs, though emission estimates in mobile sources are associated with higher uncertainty, hence, the bias of emission temporal trends is very likely caused by bias in the mobile emission estimate.

Using the R-LINE model with mobile emissions only, we simulated annual average concentrations of various pollutants in Atlanta. Even considering only mobile emissions, NO_x concentrations were overestimated by the model. It should be noted that the overestimation of the R-LINE model compared to observations can be caused by several factors, such as the formulation of the model, the properties of the pollutants, the impact from other sources, and the uncertainties in the models and data. However, the results clearly indicate a difference in NO_x and the other two species (i.e., PM_{2.5} and CO), with approximately 10 times higher NRMSE and NMB. Besides the difference of species properties, the over-estimation of modeled emissions for NO_x can be a leading factor, as has been found in several studies (Anderson et al. 2014; Fujita et al. 2012; HC Kim et al. 2016; SW Kim et al. 2016; Liu and Frey 2015). The overestimation in emissions can lead to large bias in the modeling estimates at finer resolutions when used in ambient air quality management and epidemiology studies.

We further assessed the influence of model resolution on NO_x concentrations at monitoring sites in the southeastern United States. It was found that the finer resolution enhanced modeled concentrations at these sites, because most sites are located within or around cities where the concentrations are higher than average. Therefore, model performance at the finer resolution (12 km) was expected to be more accurate. However, compared with observations, the 12-km simulation actually overestimated NO₂ concentrations, indicating a potential overestimate of emissions. Modeled time series at 12-km resolution showed a similar seasonal trend with observations, but the level was at least double the observed. Further research should examine the change in modeled results at resolutions finer than 12-km (e.g., 4 km or 250 m) to see to what extent the modeled concentrations

could be further enhanced. This may help to determine the amount of overestimation of mobile emissions.

IMPLICATIONS

Emission inventories play a key role in atmospheric chemical transport models and air quality assessment. However, bottom-up emission estimation could introduce large uncertainties, because the emission factors — the amount of pollutant emitted per unit activity — vary widely and are highly uncertain for some sources such as mobile sources. Based on observation and air quality models, our work provides further evidence of a potential overestimate of mobile NO_x emissions in the southeastern United States, including Atlanta. Further work is needed to better address whether NO_x emissions from mobile sources are overestimated and, if so, by how much.

The implications of a bias, high or low, in the mobile source inventory of a factor of about 2 or more is large, and is supported by current evidence:

1. Mobile source emissions controls are being more effective than estimated.
2. A reduced response is found in photochemical model applications (this is seen in this study, and is also found in other dynamic model analyses, (e.g., Dunker et al. 2016).
3. Potentially most important, the mass of NO_x emissions left to reduce is smaller than has been estimated, thus further mobile-source controls would not be as effective as potentially simulated. This would be particularly important in mobile source-dominated environments and is increasingly important as EGUs are controlling NO_x emissions.
4. Estimated responses of ozone and other pollutants to nonmobile sources will also be impacted.

One avenue to better estimate on-road emissions is to reduce the uncertainties being introduced into emission factor estimates and conduct further comparisons of emissions of specific cars to observations. The emission factor of NO_x is a function of engine type, load and age, fuel type and quality, driving mode, and emission control, among other things. Influence of each factor on NO_x emissions should be addressed accurately. On-road monitoring can be carried out to capture emissions from a large number of various motor vehicles in different driving conditions. A second approach would be long-term monitoring of not only NO_x, but also CO₂ and other traffic-related air pollutants (e.g., CO, specific metals, mobile source VOCs, semivolatile VOCs and/or intermediate-volatility VOCs and EC), taking advantage of U.S. EPA's new near-road network and the advances in inexpensive air quality monitors (e.g., for CO₂ and CO). This latter approach will not be able to provide a direct link between a specific vehicle and its emissions, but will provide fleet-integrated emissions. Further application of both fine and coarser scale models to identify model biases in not only ambient species, but deposition of related species, is warranted. Finally, satellite data is proving to be a very powerful approach to assessing emissions trends, so continued use of such data is suggested.

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ABBREVIATIONS AND OTHER TERMS

CMB-GC	chemical mass balance with gas constraints
CMAQ	Community Multiscale Air Quality
CMAS	Community Modeling and Analysis System
CO	carbon monoxide
EC	elemental carbon
EGU	electricity generating unit
JST	Jefferson Street monitoring station, Atlanta
MOVES	U.S. EPA MOtor Vehicle Emissions Simulator
NMB	normalized mean bias
NRMSE	normalized root mean square error
NO ₂	nitrogen dioxide
NO _x	oxides of nitrogen
PM	particulate matter
PM _{2.5}	particulate matter ≤ 2.5 μm in aerodynamic diameter
R-LINE	research LINE-source dispersion model for near-surface releases
RR	ratio-of-ratios
SEARCH	SouthEastern Aerosol Research and Characterization
U.S. EPA	U.S. Environmental Protection Agency
VCD	vertical column density
VMT	vehicle miles traveled

VOC volatile organic compound

YRK Yorkville, Georgia site