

Summertime State-Level Source-Receptor Relationships between Nitrogen Oxides Emissions and Surface Ozone Concentrations over the Continental United States

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Interstate transport of ozone (O_3) and its precursors can contribute substantially to state-level surface O_3 concentrations, making it difficult for some states to meet the National Ambient Air Quality Standards (NAAQS) for O_3 by limiting only their own emissions. We analyze the effect of interstate transport on surface O_3 in each continental U.S. state in July 1996 using the community multiscale air quality (CMAQ) model. By examining the difference between a baseline simulation and perturbation simulations in which each state's nitrogen oxides (NO_x) emissions are removed, we establish for the first time a summertime source-receptor matrix for all 48 continental states. We find that for 16 (20) states at least one neighboring state's NO_x emissions are responsible for a larger increase in monthly mean peak 8 h (all-hour) O_3 concentrations than the state's own emissions. For over 80% of the contiguous states, interstate transport is more important than local emissions for summertime peak O_3 concentrations. Our source-receptor matrices indicate that the geographic range of the clean air interstate rule (CAIR) was sufficient to address interstate transport of O_3 in most of the states included in the program. However, the exclusion of Texas, which has particularly large NO_x emissions, from the CAIR O_3 program left emission sources uncontrolled that contribute more than 1 ppbv to the July mean of peak 8 h O_3 concentrations in over a dozen states.

1. Introduction

Tropospheric ozone (O_3) is produced by the catalytic reactions of nitrogen oxides ($NO_x = NO + NO_2$) and volatile organic compounds (VOCs). It has direct adverse effects on human health, natural ecosystems, and agriculture (1). O_3 exposure is associated with premature mortality (2) and a variety of morbidity health end points (3). In addition, elevated

concentrations of surface O_3 account for up to 90% of air pollution-related crop loss in the United States (4, 5).

To protect public health and welfare the U.S. Clean Air Act (CAA) requires the U.S. Environmental Protection Agency (EPA) to set National Ambient Air Quality Standards (NAAQS) for O_3 (6, 7). Modest downward trends in the frequency with which NAAQS for surface O_3 are exceeded have occurred across the United States (8). However, approximately 140 million people in the United States currently live in areas which do not attain the 0.08 ppmv 8 h O_3 NAAQS (9).

Until recently, no federal rule required a state in compliance with the NAAQS to reduce its emission of O_3 precursors regardless of whether they affected a downwind state. Interstate transport of O_3 and its precursors, however, can contribute significantly to persistent nonattainment at many locations. Such transport can make it difficult for states that fail to meet the NAAQS to reach compliance by simply reducing their own emissions.

Because emissions from upwind states affect the ability of downwind states to comply with the O_3 NAAQS, a coalition of 13 northeastern and mid-Atlantic states established the Ozone Transport Commission (OTC) under the CAA and created a multistate NO_x cap and trade program called the OTC NO_x Budget Program. This trading program applied a cap on the total NO_x emissions allowed from large combustion sources within the region during summer while permitting emission trading among emitters (6). The program ran from 1999 to 2003 and reduced NO_x emissions from large stationary sources in the OTC region by approximately 70% from 1990 baseline levels. In 2004, the NO_x SIP Call program extended the OTC domain to southeastern and midwestern states (1). In March 2005, the Clean Air Interstate Rule (CAIR) was finalized by the U.S. EPA though, in July 2008, the U.S. Court of Appeals for the D.C. Circuit found "fatal flaws" in CAIR and dismantled it. Again using the cap-and-trade approach, CAIR provided a federal framework to reduce state-level sulfur dioxide (SO_2) and nitrogen oxide (NO_x) emissions from large stationary sources across 28 eastern states requiring reductions in NO_x emissions from large stationary sources across the regulated region by 53% in 2009 and by 61% in 2015 from 2003 levels (10). However, the Court determined that "CAIR must include some assurance that it achieves something measurable towards the goal of prohibiting sources "within the State" from contributing to non-attainment in "any other state" (24)". Our work is directly relevant to the Court's concern and demonstrates the efficacy of source state-level reductions of NO_x emissions on reducing receptor-state O_3 concentrations.

To simplify trading among emitters, the OTC, SIP Call and CAIR NO_x trading programs focus on reducing total NO_x emissions without considering geographic or episodic factors such as meteorology, local biogenic VOC emissions or population distributions. However, because O_3 precursors (NO_x and VOCs) are not well-mixed, O_3 photochemistry is nonlinear, and meteorology is variable, the impact of a fixed quantity of NO_x emissions on O_3 production is spatially and temporally variable (11–13). Recent studies by Mauzerall et al. (12), and Tong et al. (13) have demonstrated that the damages caused by NO_x emissions vary with location and time, and have suggested that NO_x emitters be charged for the actual damages their emissions cause rather than for the emissions themselves. Recent work by Bergin et al. (14), examined the impacts of statewide emissions of NO_x , SO_2 , and VOCs on O_3 and fine particulate matter in the eastern United States. They find that on average 77% of each state's

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surface O₃ that is sensitive to precursor emissions is produced by emissions from other states.

This study goes beyond the earlier work and establishes source-receptor matrices (SRMs) among all pairs of the continental U.S. states for monthly mean all-hour and peak 8 h surface O₃ concentrations. The SRMs describe, for July 1996, both how much the NO_x emitted from each state contributes to the surface O₃ concentrations of all states, and where the O₃ produced by the NO_x emitted from each state goes. These mean state-level metrics are a more conservative criteria for evaluating interstate influence on surface O₃ concentrations than is a peak influence metric at a particular location. They are not directly indicative of whether nonattainment at a particular monitor is due to interstate transport. However, these SRMs can be used to characterize the ability of a state to influence mean surface O₃ concentrations within its borders by controlling only its own emissions. The SRMs can also help identify the geographical range beneficial for control strategies.

2. Model Description and Simulation Design

2.1. Model Description. The Community Multiscale Air Quality (CMAQ) model (version 4.2) (15) is used to simulate the transport, chemical transformation, and deposition of tropospheric O₃ and its precursors for July 1996 over the continental United States. We use the same model configuration here as was evaluated in Tong and Mauzerall (16) and summarize key features in the Supporting Information (SI).

The CMAQ horizontal resolution is 36 × 36 km² with one grid cell comparable in size to a typical northeastern county, but smaller than a western county (e.g., Los Angeles County, CA covers 21 cells). Twelve vertical layers extend from the surface to approximately 15 km with half the layers in the lowest kilometer and a surface layer of approximately 38 m. Our CMAQ simulations are run from July 1 to 31, 1996 using meteorology generated by the MM5 model. Lateral boundary conditions for O₃ and other species are obtained for July from the average of a multiyear simulation of the global chemical transport model MOZART-2 (17). The CMAQ model assumes zero-flux at the top boundary while using a relatively low model top (200 mb); therefore, the contribution of stratosphere–troposphere exchange (STE) to tropospheric O₃ concentrations is not included (16).

2.2. Model Evaluation. O₃ concentrations simulated by this version of CMAQ have been compared with surface measurements from 987 Air Quality System (AQS) (the former AIRS) and 123 Clean Air Status and Trends Network (CAST-Net) sites as well as with vertical O₃ profiles from ozonesonde data over the continental United States (16). We summarize key findings in the Supporting Information (SI), section S3.

2.3. Simulation Design. To quantify the source-receptor relationships between NO_x emissions from one state and surface O₃ concentrations in downwind states, we conduct one base and 48 perturbation simulations. The baseline simulation uses standard emissions for July 1996. For each perturbation simulation we remove all anthropogenic and biogenic NO_x emissions from one of the 48 continental states. Although biogenic NO_x emissions are often categorized as natural, the majority of the NO_x emissions from this sector originate from fertilized agricultural lands and we hence consider them anthropogenic (see SI section S2 for details). We use the difference between the baseline and a perturbation simulation to quantify the monthly mean change in surface O₃ concentrations across all states resulting from the NO_x emissions from a single state. Each perturbation simulation uses July 1 of the base simulation for initial conditions and runs from July 2 to 31, 1996. The first two days of each perturbation simulation are used for model spin-up and are discarded to minimize the effect of initial

concentrations. We report the change in monthly mean maximum 8 h and all-hour surface O₃ concentrations over each state resulting from the NO_x emissions for July 4–31, 1996 (adjusted to represent all 31 days in July) from each of the 48 continental states. We developed a GIS-based tool to manipulate model input and output to analyze source and receptor areas of arbitrary shape (i.e., individual states). Changes (base minus perturbation) in mean O₃ concentrations over a state are obtained by first averaging the change in O₃ concentration in all grid cells over in each state each day and then by averaging the daily change to obtain monthly means. Our state-level source-receptor matrices then take a spatially weighted average of the data over each state.

3. Results

3.1. State-Level NO_x Emissions and Resulting Changes in Surface O₃ Concentrations. Figure 1 shows annual state-level NO_x emissions and the resulting simulated monthly mean increase in maximum 8 h and all-hour surface O₃ concentrations over the continental United States in July 1996. We include both O₃ metrics because 8 h peak surface concentrations are currently used for regulatory purposes, whereas all-hour average concentrations indicate the cumulative impact of state-level NO_x emissions. State level NO_x emissions only loosely correlate with net O₃ contributions as spatial variation in biogenic hydrocarbon emissions, NO_x/VOC ratios, nonlinearity in O₃ production efficiency (OPE) (11), and meteorological conditions (18) are also important.

NO_x emitted from Texas and California, the two largest emitters, cause the largest increases over the continental U.S. in both peak 8 h and all-hour surface O₃ concentrations. Although most other western states have low NO_x emissions, many make relatively large contributions to surface O₃ because their NO_x emissions are relatively dispersed and the region has a low NO_x to VOC ratio (see Figure S2d in SI) resulting in higher OPE (11). In addition, O₃ that is produced from a western state's NO_x is likely to have a two to three times longer lifetime (19), and to be transported further over land than O₃ produced from NO_x emitted from eastern states.

The contribution of New England states to surface O₃ concentrations over the continental United States is small because NO_x emissions are low, OPE is relatively low, and prevailing southwesterly winds in July advect much of the O₃ that is produced over the Atlantic Ocean. Similarly, states with large NO_x emissions which are located close to the national borders also result in lower source-receptor ratios than more centrally located states with similar emissions. For example, although NO_x emissions from Florida are the fifth largest in the nation while those from Oregon are one-fifth the size, NO_x emissions from both states result in similar changes in surface O₃ concentrations over the continental U.S. because much O₃ resulting from NO_x emitted from Florida is transported over ocean. Compared to states in the northeast, southeastern states with similar NO_x emissions generally produce more surface O₃ due to abundant biogenic VOC emissions (see Figure S2 in SI), lower NO_x concentrations, and frequent stagnant high pressure systems (20).

3.2. Spatial Distribution of Surface O₃ in Source and Receptor States. Figure 2 and Figure S4 (in SI) show the spatial changes in mean July peak 8 h and all-hour O₃ concentrations resulting from NO_x emissions from 10 states distributed across the U.S. These states include the five largest NO_x emitters (TX, CA, OH, IL, and FL), two ranked in the top 25% (PA and TN), and three with moderate emissions (NY, NJ, and CO). Except in locations of large local NO_x emissions, where O₃ decreases due to reaction with NO, NO_x emissions increase O₃ concentrations in both source and downwind states. NO_x emissions from Texas and California make positive contributions to surface O₃ concentrations over a larger continental region (See Figure 2a and c and Figure S4a and

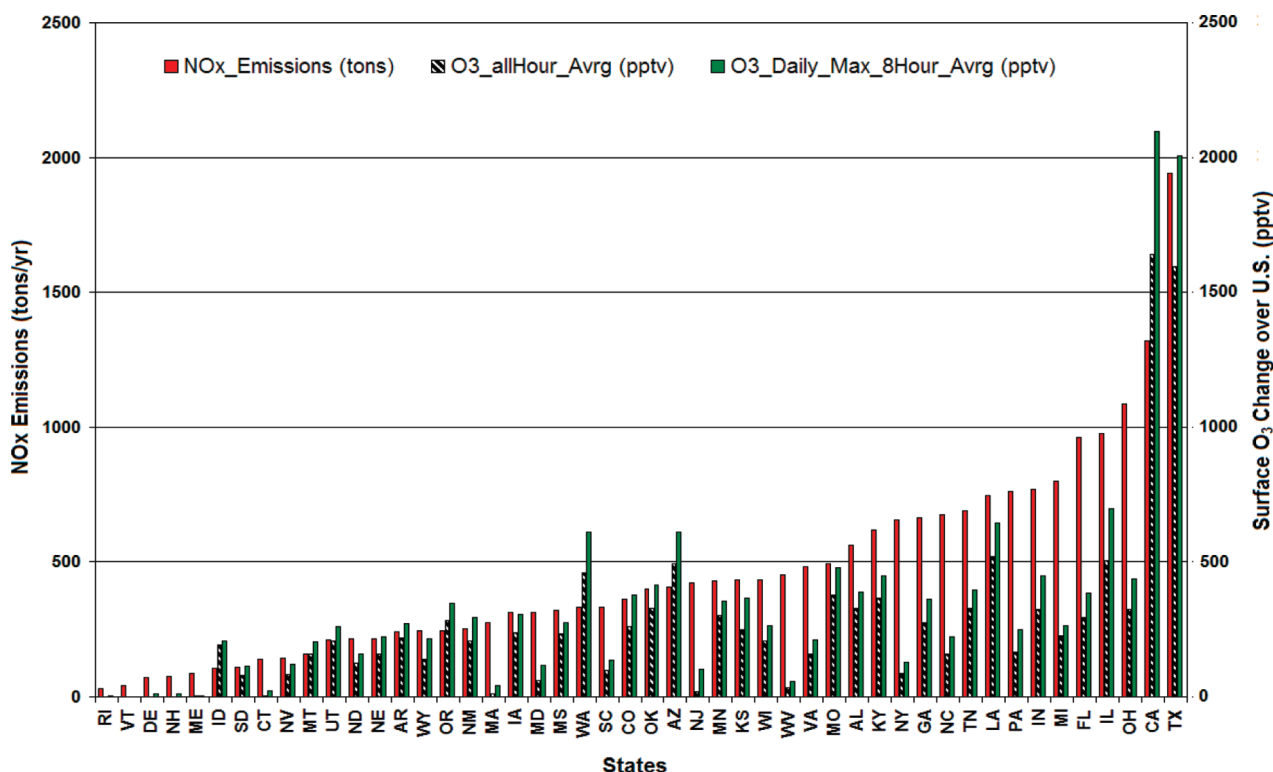


FIGURE 1. Annual state-level NO_x emissions and resulting changes in monthly mean all-hour and peak 8 h surface O_3 concentrations over the continental United States (land-only) due to the NO_x emitted from each state in July 1996.

c) than other states due both to their large emissions, western location and strong surface winds. High-pressure circulation over Florida carries most surface O_3 its NO_x emissions generate off-shore (Figure 2f and and SI figure S4f).

O_3 can be lost by reaction with freshly emitted NO . Decreases in O_3 concentrations caused by local emissions are seen in urban centers and around large point sources inside source states. Our simulations show that if California, Texas or New York NO_x emissions are removed, monthly mean O_3 concentrations would increase by 14 ppbv in Los Angeles, 20 ppbv in Houston, and 22 ppbv in New York City, respectively (SI Figure S4).

3.3. State-Level Source-Receptor Relationships. To summarize the effect NO_x emissions from one state (source state) have on surface O_3 concentrations over itself and other states (receptor states), we create two source receptor matrices (SRMs) from our model results. Table 1 and Table 2 show the SRMs for July 1996 mean daily peak 8 h and all-hour O_3 concentration changes, respectively. At the bottom of Table 1 (Table 2) we sum the total O_3 derived from NO_x emitted from the 48 continental states ("sum"), and provide the monthly mean 8 h peak (all-hour) surface O_3 concentrations over each receptor state ("base"). The difference between the "sum" and "base" concentrations is due to background O_3 from out-of-U.S. emission sources and the stratosphere (19) which is partially included in the simulations through our boundary conditions from the global MOZART-2 model.

Table 1 is a SRM for monthly mean maximum 8 h average O_3 concentrations for July 1996. It shows intrastate emissions contribute to O_3 increases ranging from 0.4 to 22.0 ppbv. Several northeastern states (NJ, MA, RI, CT, DE) NO_x emissions increase intrastate monthly mean 8 h peak O_3 concentrations by 10.8, 1.8, 1.4, 2.2, and 4.0 ppbv, respectively, whereas decreasing intrastate monthly mean all-hour surface O_3 concentrations by -1.4 , -1.4 , -1.1 , -0.9 , and -0.1 ppbv, respectively (Tables 1 and 2). Spatial variability exists for these changes within each state, however. For example, NO_x emissions from NJ decrease mean all-hour O_3 concentrations over western NJ by 5–20 ppbv (SI Figure S4j) but increase

peak 8 h O_3 concentrations over eastern NJ by 10–20 ppbv (Figure 2j). Surface O_3 resulting from NO_x emissions from NJ increases state-average monthly mean peak 8 h O_3 concentrations over most of the mid-Atlantic and New England states by approximately 1–4 ppbv (see Figure 3 and Table 1). NJ NO_x also contributes up to 17 ppbv to the monthly mean peak 8 h surface O_3 concentration at a single grid cell in a neighboring state (see SI Table S1). This indicates that reductions in NJ NO_x emissions would also assist other states meet the O_3 NAAQS.

For peak 8 h O_3 concentrations we find 16 states for which at least one neighboring state's NO_x emissions are responsible for a larger increase in surface O_3 concentrations than the state's own emissions. These states include eight northeastern states (CT, DE, ME, MA, NH, NJ, RI, and VT), three midwestern states (AR, MS, OK), three western states (OR, UT, and NV), and two southeastern states (SC and WV) (see Table 1). For a receptor state, the number of source states that can contribute 1 ppbv or more to monthly mean peak 8 h O_3 concentrations vary from 1 to 11. For Texas, Washington, and North Dakota, no U.S. state except itself contributes more than 1 ppbv to peak O_3 . In five other states (CA, MT, NV, OK, and OR), only one state, besides the source state, contributes more than 1 ppbv. These eight states are either located upwind of main transport pathways, along the national boundaries, or downwind of a single large NO_x emitting state. However, the majority of receptor states (32) receive at least 1 ppbv peak O_3 from interstate transport from three to ten states. The most interconnected states are located in the eastern United States where a source state several states away can contribute considerably to another state's peak O_3 .

Table 2 is a SRM for monthly mean all-hour surface O_3 for July 1996. It shows that intrastate NO_x emissions increase all-hour average O_3 concentrations in 40 states by 1 to 15 ppbv and decrease all-hour average O_3 concentrations in five northeastern states (CT, DE, MA, NJ, and RI) by 0.1–1.4 ppbv. These five northeastern states are either surrounded by or downwind of large NO_x emitting states, causing

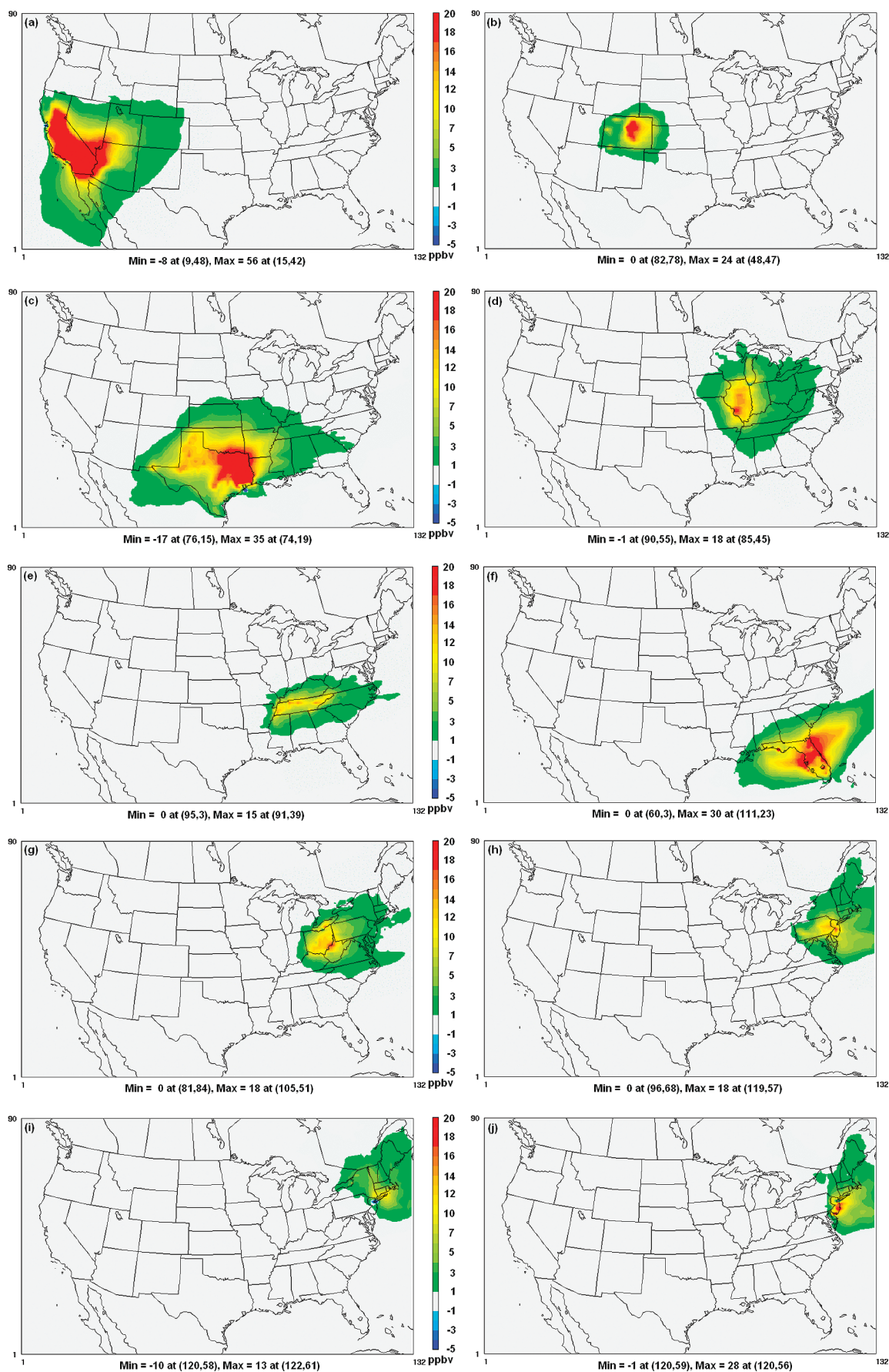


FIGURE 2. Changes in monthly mean maximum 8 h surface O_3 concentrations for July 1996 resulting from NO_x emissions from: (a) California; (b) Colorado; (c) Texas; (d) Illinois; (e) Tennessee; (f) Florida; (g) Ohio; (h) Pennsylvania; (i) New York; and (j) New Jersey.

TABLE 1. State-Level Source-Receptor Relationships between NO_x Emissions and Changes in Monthly Mean Maximum 8 h Surface O_3 Concentrations among 48 U.S. states in July 1996

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^a Each row represents O_3 changes in each state resulting from NO_x emissions from the source state; each column represents O_3 changes in a receptor state due to NO_x emissions from all source states. The diagonal shows O_3 changes due to a state's own NO_x emissions. The bottom two rows, "Sum" and "Base", provide the sum of NO_x -derived O_3 concentrations from all 48 states, and the base case surface O_3 concentration over each state. SRC: source; RCP: receptor.

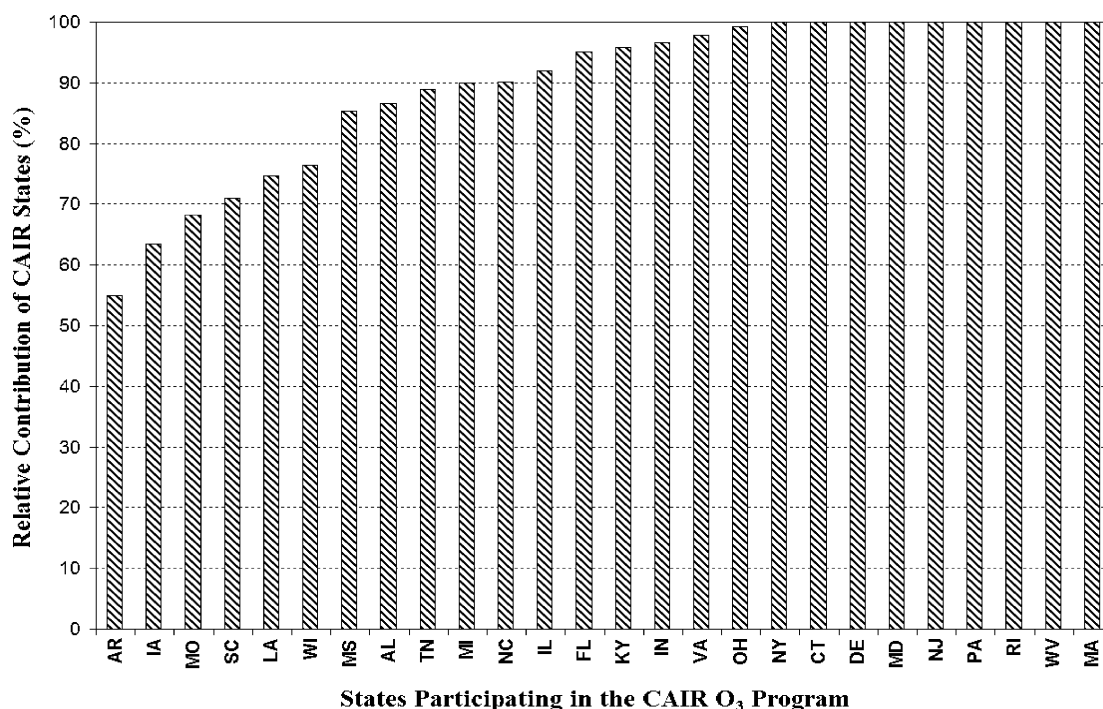


FIGURE 3. Relative contributions of NO_x emissions from CAIR-regulated states to monthly mean daily maximum 8 h surface O_3 concentrations in July 1996 over each CAIR state.

additional NO_x emissions from intrastate sources to decrease O_3 concentrations locally. While intrastate emissions in most states contribute to a larger increase in local O_3 concentration than emissions from any other individual source state, there are 20 states for which at least one neighboring state's NO_x emissions make a larger contribution to mean all-hour O_3 concentrations than the state's own emissions. These states include 10 northeastern states (CT, DE, MD, MA, NH, NJ, NY, PA, RI, and VT), four midwestern states (AR, IN, MS, SD), two western states (UT and NV), and two southeastern states (SC and WV). Furthermore, in the remaining 28 continental states, a neighboring state frequently contributes a comparable quantity of surface O_3 as that resulting from intrastate emissions. For instance, California NO_x emissions contribute an increase of 7.5 ppbv to mean all-hour surface O_3 concentrations in Arizona, only slightly less than the 7.8 ppbv O_3 resulting from Arizona's own NO_x emissions.

We calculate the largest contribution to monthly mean peak 8 h O_3 concentrations over a grid cell within each receptor state resulting from each state's NO_x emissions during July 1996 and present it as a SRM in SI Table S1. Table S1 shows that localized short-term impacts of interstate transport can be far larger than state-level monthly means. The complexity of a full substate analysis on the national scale, however, is beyond the scope of this study.

3.4. Analysis of the Clean Air Interstate Rule. We use our SRMs to examine the efficacy of the geographic range of states included under the Clean Air Interstate Rule (CAIR). Figure 3 shows the relative contributions of NO_x emissions from CAIR-regulated states to monthly mean 8 h peak O_3 concentrations in each CAIR state. In determining which states to include under the CAIR for NO_x , EPA also removed emissions from individual states in order to model the effects of upwind emissions on specific locations in downwind states (10). While the EPA study focused on the effects on nonattainment areas in downwind states, we calculate here the area-weighted impacts over entire states.

Among the 26 states participating in the CAIR O_3 program 90% or more of monthly mean peak 8 h surface O_3 in 17 states resulted from NO_x emissions from states within the CAIR O_3 region. Texas was included in the CAIR program for

particulate matter only. Including an additional summer cap on NO_x emissions from Texas would likely assist several downwind states achieve the O_3 NAAQS (see Table S1).

3.5. Relative Importance of Local Emissions and Interstate Transport. Figure 4 shows the contribution of intrastate NO_x emissions and interstate transport to monthly mean 8 h peak O_3 concentrations in (a) ppbv and (b) percent for each state. Figure 4a shows that the total contribution to state-level monthly mean 8 h peak O_3 resulting from NO_x emissions in the U.S. ranges from about 5–34 ppbv. For individual states, intrastate NO_x emissions contribute between 1 and 22 ppbv, whereas out-of-state NO_x emissions contribute between 2 and 30 ppbv to monthly mean peak 8 h O_3 . Background O_3 from intercontinental transport and the stratosphere makes up the rest. Figure 4b shows that for receptor states located upwind of most other states and/or having large NO_x emissions themselves (e.g., CA, TX, WA, and FL), the contribution of interstate transport is less than 20% of the total peak 8 h surface O_3 concentrations derived from U.S. NO_x emissions. However, out-of state NO_x emissions are responsible for over 90% of the surface O_3 derived from domestic NO_x emissions in states with small NO_x emissions downwind of states with large NO_x emissions. Of O_3 produced from domestic NO_x emissions, interstate transport contributes between 75 and 97% of peak 8 h O_3 in 13 continental states, between 55 and 75% in 24 states, and between 8 and 45% in the 11 states least affected by interstate transport. For 80% of the contiguous states, we find interstate transport to be more important than local emissions for peak O_3 concentrations.

4. Discussion of Uncertainties and Results

The relationships described in our SRMs are derived from model calculations with uncertainties associated with emissions, transport and photochemistry. Our SRMs were derived using 1996 emissions. Between 1996 and 2006, domestic regulations reduced NO_x emissions by 25% nationally (approximately 40% from large stationary sources and 22% from on-road mobile sources) and anthropogenic VOC emissions by 15% (22), although such emission trends are subject to

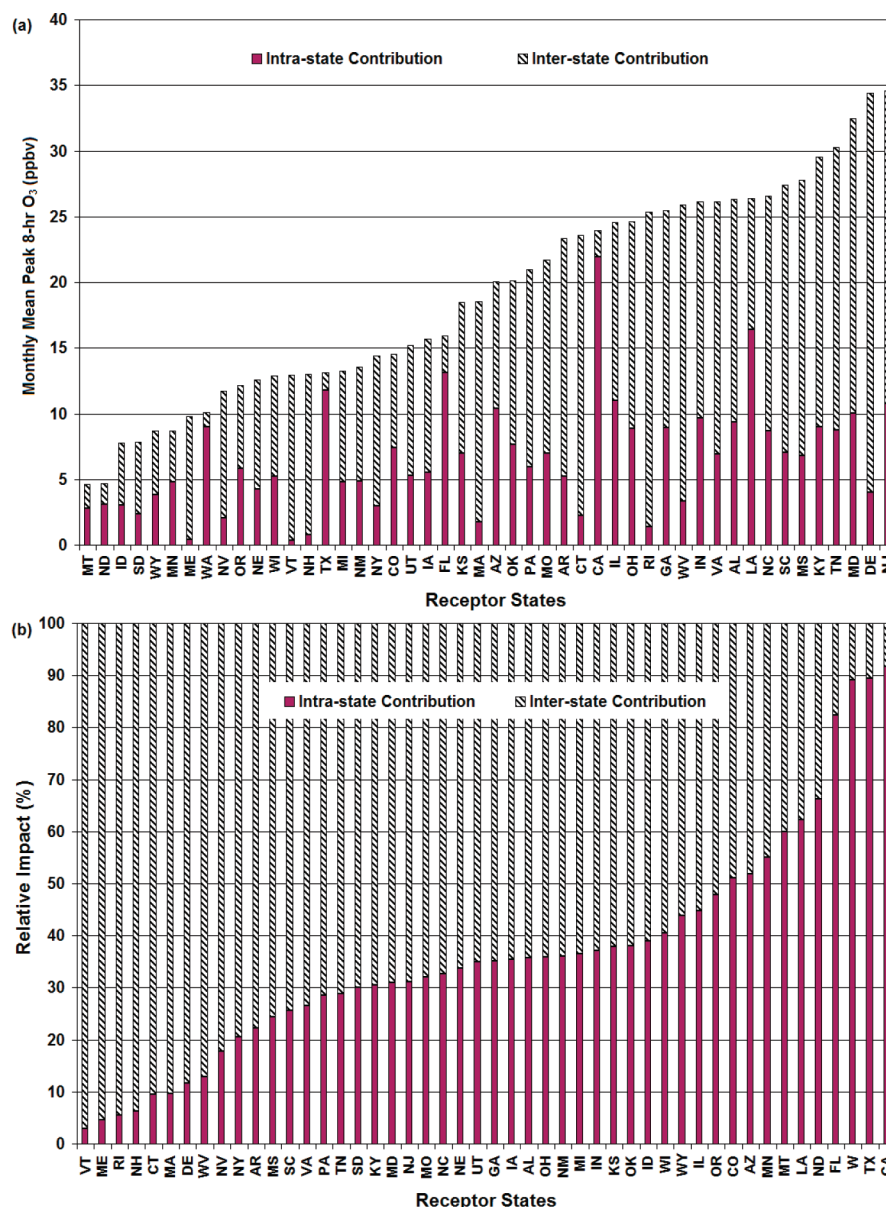


FIGURE 4. Contributions from intrastate and interstate NO_x emissions to monthly mean peak 8 h surface O₃ concentrations in (a) ppbv; (b) percent.

debate (23). A similar analysis to ours using current emissions would be valuable.

Meteorology varies over short time-scales and our presentation of monthly mean S–R relationships will underestimate peak impacts over shorter intervals. Extrapolation of the S–R values to other months or years will be affected by seasonal and inter-annual variability in meteorology and emissions. Our SRMs represent mean state-level impacts of each state's NO_x emissions in July 1996 on monthly mean peak 8 h and all-hour surface O₃ concentrations. Actual changes at a particular location within a receptor state or at a specific time vary and will frequently be different from the state-level monthly average (SI Table S1).

Although 36 km resolution is high for a set of national-scale simulations, it results in rapid dilution of power-plant and urban plumes which could result in excess O₃ formation. However, recent work found that first-order sensitivities to NO_x emissions are consistent across domains using 4, 12, and 36 km resolutions when fine scale results are aggregated to the coarsest cell size (21).

Our objective is to characterize the full effect of a state's NO_x emissions on surface O₃ over the continental U.S.

However, the results we obtain by removing each state's NO_x emissions are influenced by nonlinearity in photochemistry. The effect of removing 100% of emissions can not be directly interpolated to provide a quantitative result for a more feasible reduction in anthropogenic NO_x emissions.

Elevated concentrations of ground-level O₃ are a regional-scale problem. Although local emission controls are usually beneficial, individual states can find it impossible to meet the NAAQS for O₃ by limiting only their own NO_x emissions, as interstate transport of O₃ and its precursors can contribute substantially to state-level surface O₃ concentrations. Our SRMs show that intrastate NO_x emissions change state-level all-hour mean O₃ concentrations by –1.4 to 15.1 ppbv and monthly mean 8 h peak O₃ concentrations by 0.4 to 22.0 ppbv. For 16 (20) states a single neighboring state's NO_x emissions made a larger contribution to monthly mean maximum 8 h (all-hour) O₃ concentrations than the state's own emissions. For 80% of the contiguous states, interstate transport is more important than local emissions for peak O₃ concentrations.

EPA recognized the importance of interstate transport when developing the Clean Air Interstate Rule (CAIR). Based

on our SRMs, we found the geographical range of the CAIR program to be sufficient to address interstate transport of O₃ in most participating states. However, including TX, the state with the largest NO_x emissions, future NO_x control programs would reduce O₃ concentrations in at least a dozen states that received more than 1 ppbv of monthly mean daily maximum 8 hour O₃ concentrations from TX in July 1996.

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Supporting Information Available

A description of the CMAQ model configuration and evaluation, plus important figures and tables describing additional model results is included. This information is available free of charge via the Internet at <http://pubs.acs.org>.

Literature Cited

- (1) National Research Council (NRC). *Air Quality Management in the United States*; National Research Council of the National Academies: Washington DC, 2004.
- (2) Bell, M. L.; Dominici, F.; Samet, J. M. A meta-analysis of time-series studies of ozone and mortality with comparison to the national morbidity, mortality, and air pollution study. *Epidemiology* **2005**, *16* (4), 436–445.
- (3) U.S. EPA. *Air Quality Criteria for Ozone and Related Photochemical Oxidants (Final)*, EPA/600/R-05/004aF-cF; U.S. Environmental Protection Agency: Washington, DC, 2006.
- (4) Heck, W. W.; Taylor, O. C.; Adams, R.; Bingham, G.; Miller, J.; Preston, E.; Weinstein, L. Assessment of crop loss from ozone. *J. Air Pollut. Control Assoc.* **1982**, *32*, 353–361.
- (5) Mauzerall, D. L.; Wang, X. Protecting Agricultural crops from the effects of tropospheric ozone exposure: Reconciling science and standard setting in the United States, Europe, and Asia. *Annu. Rev. Energy Environ.* **2001**, *26*, 237–68.
- (6) Findings of significant contribution and rulemaking for certain States in the Ozone Transport Assessment Group region for purposes of reducing regional transport of ozone; US EPA, OAQSP, Rule. RIN: 2060-AH10, 63 *Fed. Regist.* **1998**, Vol 63, 57356.
- (7) National Air Quality and Emissions Trends Report, EPA-454-/F-00-002; U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards: Research Triangle Park, NC, 2000.
- (8) Lin, C.-Y. C.; Jacob, D. J.; Fiore, A. M. Trends in exceedances of the ozone air quality standard in the continental United States, 1980–1998. *Atmos. Environ.* **2001**, *35*, 3217–3228.
- (9) U.S. Environmental Protection Agency (US EPA). Green Book - 8-h Ozone Nonattainment Areas, 2008. <http://www.epa.gov/air/oaqps/greenbk/gntc.html> (accessed March 12, 2008).
- (10) U.S. Environmental Protection Agency (US EPA). Clean Air Interstate Rule - Basic Information, 2005. <http://www.epa.gov/cair/basic.html> (accessed January 16, 2007).
- (11) Lin, X.; Trainer, M.; Liu, S. C. On the nonlinearity of tropospheric ozone production. *J. Geophys. Res.* **1988**, *93*, 15879–15888.
- (12) Mauzerall, D. L.; Sultan, B.; Kim, N.; Bradford, D. NO_x Emissions: Variability in Ozone Production, Resulting Health Damages and Economic Costs. *Atmos. Environ.* **2005**, *39*, 2851–2866.
- (13) Tong, D. Q.; Muller, N. Z.; Mauzerall, D. L.; Mendelsohn, R. O. Integrated Assessment of the Spatial Variability of Ozone Impacts from Emissions of Nitrogen Oxides. *Environ. Sci. Technol.* **2006**, *40* (5), 1395–1400.
- (14) Bergin, M. S.; Shih, J. S.; Krupnick, A. J.; Boylan, J. W.; Wilkinson, J. G.; Odman, M. T.; Russell, A. G. Regional Air Quality: Local and Interstate Impacts of NO_x and SO₂ Emissions on Ozone and Fine Particulate Matter in the Eastern United States. *Environ. Sci. Technol.* **2007**, *41* (13), 4677–4689.
- (15) Byun, D. W.; Schere, L. K. Review of the governing equations, computational algorithms, and other components of the Models-3 community multiscale air quality (CMAQ) modeling system. *Appl. Mech. Rev.* **2006**, *59* (2), 51–77.
- (16) Tong, D. Q.; Mauzerall, D. L. Spatial variability of summertime tropospheric ozone over the continental United States: Implications of an evaluation of the CMAQ model. *Atmos. Environ.* **2006**, *40* (17), 3041–3056.
- (17) Horowitz, L. W.; Walters, S.; Mauzerall, D. L.; Emmons, L. K.; Rasch, P. J.; Granier, C.; Tie, X. X.; Lamarque, J. F.; Schultz, M.; Tyndall, G. S.; Orlando, J. J.; Brasseur, G. P. A global simulation of tropospheric ozone and related tracers: Description and evaluation of MOZART, version 2. *J. Geophys. Res.* **2003**, *108* D24 4784, doi: 10.1029/2002JD002853.
- (18) Vukovich, F. M. Regional-scale boundary layer ozone variations in the eastern United States and their association with meteorological variations. *Atmos. Environ.* **1995**, *29* (17), 2259–2273.
- (19) Fiore, A. M.; Jacob, D. J.; Bey, I.; Yantosca, R. M.; Field, B. D.; Fusco, A. C. Background ozone over the United States in summer: Origin, trend, and contribution to pollution episodes. *J. Geophys. Res.* **2002**, *107* (D15), 10–1029.
- (20) Eder, B. K.; Davis, J. M.; Bloomfield, P. A characterization of the spatiotemporal variability of non-urban ozone concentrations over the eastern United States. *Atmos. Environ.* **1993**, *27A*, 2645–2668.
- (21) Cohan, D. S.; Hu, Y.; Russell, A. G. Dependence of ozone sensitivity analysis on grid resolution. *Atmos. Environ.* **2006**, *40* (1), 126–135.
- (22) U.S. Environmental Protection Agency. National Emissions Inventory (NEI) Air Pollutant Emissions Trends, Office of Air Quality Planning and Standards (OAQPS), Research Triangle Park, NC. <http://www.epa.gov/ttn/chief/trends/trends06/nationaltier1upto2006basedon2002finalv2.1.xls> (accessed on October 11, 2007).
- (23) Parrish, D. D. Critical evaluation of US on-road vehicle emission inventories. *Atmos. Environ.* **2006**, *40* (13), 2288–2300.
- (24) United States Court of Appeals for the District of Columbia Circuit, No. 05-1244, State of North Carolina vs. Environmental Protection Agency, July 11, 2008.

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