

## **Source-Receptor Relationships for Ozone and Fine Particulates in the Eastern United States**

Jhih-Shyang Shih, Alan J. Krupnick, Michelle S. Bergin and Armistead G. Russell

May 2004 • Discussion Paper 04–25



**RESOURCES**  
FOR THE FUTURE

Resources for the Future  
1616 P Street, NW  
Washington, D.C. 20036  
Telephone: 202–328–5000  
Fax: 202–939–3460  
Internet: <http://www.rff.org>

© 2004 Resources for the Future. All rights reserved. No portion of this paper may be reproduced without permission of the authors.

Discussion papers are research materials circulated by their authors for purposes of information and discussion. They have not necessarily undergone formal peer review or editorial treatment.

## **Source-Receptor Relationships for Ozone and Fine Particulates in the Eastern United States**

Jhih-Shyang Shih, Alan J. Krupnick, Michelle S. Bergin, and Armistead G. Russell

### **Abstract**

A key question in developing effective mitigation strategies for ozone and particulate matter is identifying which source regions contribute to concentrations in receptor regions. Using a direct approach with a regional, multiscale three-dimensional model, we derive multiple source-receptor matrices (S-Rs) to show inter- and intrastate impacts of emissions on both ozone and PM<sub>2.5</sub> over the eastern United States. Our results show that local (in-state) emissions generally account for about 23% of both local ozone concentrations and PM<sub>2.5</sub> concentrations, while neighboring states contribute much of the rest. The relative impact of each state on others varies dramatically between episodes. In reducing fine particulate concentrations, we find that reducing SO<sub>2</sub> emissions can be 10 times as effective as reducing NO<sub>x</sub> emissions. SO<sub>2</sub> reductions can lead to some increase in nitrates, but this is relatively small. NO<sub>x</sub> reductions, however, lead to both ozone reductions and some reduction in nitrate and sulfate particulate matter.

**Keywords:** source-receptor, ozone, particulate matter, sensitivity analysis, air quality simulation, National Ambient Air Quality Standards

**JEL Classification Numbers:** Q2, Q25

## Contents

<b>Introduction.....</b>	<b>1</b>
<b>Modeling Approach .....</b>	<b>3</b>
Model Description .....	3
Application Description .....	4
Model Performance.....	5
<b>Results and Discussion.....</b>	<b>7</b>
Sensitivity Results.....	7
Derivation of Multiple Pollutant S-R Matrices.....	7
Ozone Sensitivity and Control Effects.....	9
PM <sub>2.5</sub> Sensitivity and Control Effects .....	11
Comparison of Long-Range Response of Ozone and PM <sub>2.5</sub> Sensitivities .....	13
Issues of Nonlinearity and Uncertainty.....	14
<b>Conclusions.....</b>	<b>14</b>
<b>References.....</b>	<b>16</b>

# Source-Receptor Relationships for Ozone and Fine Particulates in the Eastern United States

Jhih-Shyang Shih, Alan J. Krupnick, Michelle Bergin, and Armistead G. Russell\*

## Introduction

Controversy over the extent to which emissions in one state affect air quality in other states has been a central feature of the debate over air pollution control policy since 1990. This controversy culminated in a series of lawsuits brought by East Coast states against Midwestern states, charging that the latter were making attainment of the National Ambient Air Quality Standard for ozone much more difficult for the former. Additionally, given their influence in creating fine particulate concentrations (PM<sub>2.5</sub>: particles with a diameter less than 2.5 microns), the temporal and spatial contribution of sulfur dioxide (SO<sub>2</sub>) and oxides of nitrogen (NO<sub>x</sub>) emissions and the effect of NO<sub>x</sub> emissions reductions on ozone must be understood if effective air pollution policies are to be crafted.

Ozone and PM<sub>2.5</sub> share common sources and formation routes in the atmosphere. Ozone, a gas, is formed secondarily in the atmosphere by reactions between NO<sub>x</sub> and volatile organic compounds (VOCs) in the presence of sunlight. Fine particulate matter is composed of many different chemical species, including (but not limited to) ammonium sulfate, ammonium nitrate, and organic and elemental carbon. Depending on the chemical composition, PM<sub>2.5</sub> can be directly emitted (e.g., from diesel vehicles, dust, and biomass burning) and/or formed secondarily from reactions of sulfur dioxide, NO<sub>x</sub>, ammonia (NH<sub>3</sub>), and VOCs. Controls placed

---

\* Jhih-Shyang Shih and Alan J. Krupnick are Fellow and Senior Fellow, respectively, at Resources for the Future. Michelle Bergin and Armistead G. Russell are Ph.D. candidate and professor, respectively, at Georgia Institute of Technology. We thank Yueh-Jiun Yang, Jim Wilkinson, Jim Boylan, Allen Basala, Sheau-Rong Lou, Dick Morgenstern and Kris Wernstedt for valuable discussions. We are grateful to Lisa Crooks and Mike Batz for research assistance. This study is partly supported by a partnership grant from the USEPA and NSF and through a USEPA STAR graduate fellowship. Note that superscripts refer to numbered references on pages 16–17.

to reduce one form of particulate may affect concentrations of the other, though not necessarily proportionally or even in the same direction. For example, reducing SO<sub>2</sub> emissions will generally lead to a reduction in sulfate particulate matter, though less than proportionally.<sup>1</sup> Further, the reduction in sulfate leads to more ammonia being available to form particulate ammonium nitrate—i.e., there is a “rebound” in the amount of particulate formed. The two effects mitigate the total PM reduction that might be first expected. Also investigated is the “bounce-back” effect, where NO<sub>x</sub> reductions can lead to localized increases in ozone due to reduced scavenging of ozone and ozone precursors. Typically, this happens within a few tens of kilometers of the source region studied, with reductions found downwind.

Previous studies have explored the responses of ozone and of PM<sub>2.5</sub> to various controls.<sup>2-5</sup> Control strategies exist that lower both PM<sub>2.5</sub> and ozone concurrently, or lower one or the other. Thus, from an air quality management perspective, it is desirable to understand the relative efficacy of NO<sub>x</sub> and SO<sub>2</sub> emissions reductions in controlling these air pollutants.

To address these issues, we developed a new modeling approach to derive multiple source-receptor sensitivity matrices (S-Rs).<sup>6</sup> Independence, additivity, and linearity are assumed, and have been demonstrated to be reasonably accurate.<sup>1</sup> We compare the relative effectiveness of regional (interstate) vs. local (intrastate) controls on NO<sub>x</sub> and SO<sub>2</sub> emissions as they affect ozone and PM<sub>2.5</sub> concentrations, the effectiveness of controlling NO<sub>x</sub> from elevated sources (i.e., electric utilities and large industrial plants), the relative effectiveness of reducing NO<sub>x</sub> vs. SO<sub>2</sub> emissions to reduce PM<sub>2.5</sub>, and the sensitivity of these results to different weather patterns. We study two very different meteorological episodes (in July and May 1995), each lasting approximately 10 days. The 20 days allow us to investigate the impact of meteorological variability on pollutant transport and local production. We also investigate the extent of the “bounce-back” effect and the “rebound” effect, reducing part of the expected benefit from controls. Finally, we investigate to what degree the efficacy of controls depends on how one quantifies the response—e.g., looking at a spatial (area) weighting or considering exposure by using a population-based approach.

## Modeling Approach

### Model Description

In this study, the Urban-to-Regional Multiscale (URM) One Atmosphere Model (URM-1ATM)<sup>7-8</sup> and the Regional Atmospheric Modeling System (RAMS) are used to account for the processes significantly affecting ozone and PM<sub>2.5</sub> concentrations in the atmosphere, including atmospheric physics, gas and aerosol phase chemistry, cloud and precipitation processes, and wet and dry deposition. RAMS is used to recreate the physics of an historical period of time, providing details and spatial coverage unavailable from observations. URM-1ATM solves the atmospheric diffusion equation (ADE) for the change in concentration,  $c$ , of species  $i$  with time,

$$\frac{\partial c_i}{\partial t} + \nabla \cdot (\mathbf{u}c_i) = \nabla \cdot (\mathbf{K}\nabla c_i) + f_i + S_i \quad (1)$$

where  $\mathbf{u}$  is a velocity field,  $\mathbf{K}$  is the diffusivity tensor,  $f_i$  represents the production by chemical reaction of species  $i$ , and  $S_i$  represents sources and sinks of species  $i$ . As used here, a direct sensitivity capability using the Direct Decoupled Method in Three Dimensions (DDM-3D)<sup>1-2</sup> is employed to calculate the local sensitivities of specified model outputs simultaneously with concentrations. As shown in Equation 2, the sensitivity,  $S_{ij}$ , of a model output,  $C_i$  (such as pollutant concentration of species  $i$ ) to specified model inputs or parameters,  $P_j$  (e.g., elevated NO<sub>x</sub> emissions) is calculated as the ratio of the change in output  $C_i$  to an incremental change of input or parameter  $P_j$ .

$$S_{ij} = \frac{\partial C_i}{\partial P_j} \quad (2)$$

This leads to an additional set of equations that are solved concurrently with the concentrations, but the structure of those equations is very similar to the original (Equation 1), and is solved efficiently. This sensitivity is a local derivative, so a linear assumption is then made to extrapolate the result to a non-zero perturbation. This assumption has been well tested for the

variables of interest for this study.<sup>1</sup> A more detailed description of the model is available elsewhere.<sup>8-9</sup>

### **Application Description**

We use a multiscale grid structure encompassing the eastern United States (Figure 1). The finest grids are placed over major source regions such as the Ohio River Valley, where many power plants and large industries are located, and over highly populated regions such as the East Coast corridor. This approach allows evaluation of potential population exposure to pollutants and captures high-population-related sources such as automobile exhaust, fast food restaurants, and so forth. Here, the term “potential exposure” denotes that we do not take into account micro-environmental variations in concentrations or variations in individual activities. The vertical grid has seven nonuniform layers, with the thinnest layer near the ground and the layers thickening with an increase in altitude. This vertical scheme allows detailed treatment of ground-level sources and best represents ground-level mixing and dry deposition processes, allowing for diurnal changes in mixing depths, and capturing aloft multiday transport events. A comparison of the effects of different grid scales and spatial allocation on model performance is presented elsewhere.<sup>10</sup>

URM-1ATM is applied to two well-studied episodes occurring in July 9-19, 1995 and May 22-29, 1995. These base episodes were selected because high-quality and complete data was available, was previously modeled using a different multiscale grid definition but with the same simulation system,<sup>8</sup> and because the data covered large meteorological variation with moderate to high pollution formation. Meteorological information is developed using the Regional Atmospheric Modeling System (RAMS)<sup>11</sup> in a nonhydrostatic mode, including cloud and rain microphysics. Prevailing winds in these episodes are toward the East Coast from the Midwest in July, and toward the Southeast from the Mid-southern states in May. These are common wind patterns for the summer and spring seasons, respectively.

Emissions were generated using the Emissions Modeling System (EMS-95).<sup>12</sup> Because 1995 emissions are not relevant for future policy evaluation, 2010 day-specific emissions are estimated under conditions that are anticipated with changes in population growth, vehicle turnover, emissions control technologies, and anticipated emissions regulations. These future scenarios are used to evaluate potential control strategies. Meteorological and initial and boundary conditions are hourly and day-specific, and are held constant for base and future years. Table 1a presents the estimated average day emissions for the 1995 basecases and the state-specific average day 2010 (future case) emissions. The emissions differences between 1995 basecases and the 2010 future case are due to implementation of the Tier 2 automotive emissions regulations, the “NO<sub>x</sub> SIP Call,” acid deposition controls from the 1990 Clean Air Act Amendments (CAAA), and other controls. Additional information on these model inputs is described elsewhere.<sup>8-9</sup>

### Model Performance

Both of the 1995 basecase episodes have been used to evaluate model performance against ambient measurements. Excluding model ramp-up days, nine and six days of simulation are available for evaluation in the July and May episodes, respectively. Data from the U.S. Environmental Protection Agency’s Aerometric Information Retrieval System (AIRS)<sup>13</sup> is used to evaluate model performance for ozone prediction. EPA guidelines for urban scale ozone modeling are +/- 15% for Normalized Mean Bias (NMB), and +/- 35% for Normalized Mean Error (NME). This regional-scale application resulted in an average NMB for ozone of 3.2% for the May episode and of 1.6% for the July episode; the NME was 16.8% for May and 21.4% for July. All these results were well within the stated guidelines. These values are calculated from measurements at sites coinciding with either 24km<sup>2</sup> or 48km<sup>2</sup> cells. Data was available from nearly 500 sites for each episode.

The Interagency Monitoring of Protected Visual Environments (IMPROVE) network<sup>14</sup> provides 24-hour averaged speciated aerosol data taken on Wednesday and Saturday of each week in our episodes, and this data is used to evaluate model performance for aerosols. Three



days of data was available during the July episode (July 12, 15, and 19), with measurements from 18 sites in total, 12 of which coincided with 24km<sup>2</sup> or 48km<sup>2</sup> cells. These sites resulted in an average NMB for PM<sub>2.5</sub> of -24.6% and an average NME of 31.67%. Two days of data was available for the May episode (May 24 and 27), with measurements from 17 sites in total, 11 of which coincided with 24km<sup>2</sup> or 48km<sup>2</sup> cells. These sites resulted in an average NMB for PM<sub>2.5</sub> of -9.4% and an average NME of 28.30%. There are no current guidelines to indicate acceptable model performance for aerosols. A detailed description of ozone and speciated aerosol model performance for this application is presented elsewhere.<sup>10</sup>

Figures 2a and 3a show sample results for 2010 ozone and PM<sub>2.5</sub> concentrations for the July episode. The predicted one-hour daily maximum ozone concentrations for the entire domain for the July episode are between 84.5 ppb and 104 ppb, well below the peak levels simulated using the 1995 emissions (which is 120 ppb), showing the impact of predicted future controls. The ozone concentration reaches its peak on the fourth day of the episode. The predicted 24-hour average daily maximum PM<sub>2.5</sub> concentrations over the entire domain for the July episode are mostly between 46.7µg/m<sup>3</sup> and 70 µg/m<sup>3</sup>. The concentrations start exceeding 59 µg/m<sup>3</sup> after the third day, peak on July 14, and decrease again after July 15.

May's lower temperature than July explains its lower ozone and PM<sub>2.5</sub> concentrations. The ozone concentration for the May episode is between 61.6 ppb and 89.5 ppb, about 20% lower, on average, than the concentration during the July episode. The 24-hour average PM<sub>2.5</sub> concentration during the May episode is between 25.4 µg/m<sup>3</sup> and 65.5 µg/m<sup>3</sup>, about 22% lower than the concentration during the July episode.

## Results and Discussion

### Sensitivity Results

After ensuring adequate model performance and developing future case simulations, the 2010 episodes are used to examine the sensitivity and control effects of ozone and aerosol concentrations to reductions in specific emissions source types in different states. Here, S-R coefficients are derived based on 30% emissions reductions. Nineteen states (or state combinations) are evaluated as both sources and receptors. Results from reductions in two major source types are presented: elevated  $\text{NO}_x$ , such as from power plants and large industrial sites; and total  $\text{SO}_2$  emissions, largely elevated emissions from coal-fired power plants. As examples, Figures 2b and 3b show the “sensitivity” of ozone and  $\text{PM}_{2.5}$ , respectively, resulting from emissions reductions in Tennessee during the July 2010 episode. Figures 2b(1-3) show the change in concentration of one-hour daily maximum ozone plumes at three different times of the day when elevated  $\text{NO}_x$  emissions are reduced by 30%. The impact on ozone concentrations in other states is obvious (Figures 2b1 and 2b3). As is discussed later, some states, particularly those near the Atlantic coast, are clearly receptors of many states’ emissions during the episodes simulated (Figures 4-5).

Figure 3a shows the baseline results for  $\text{PM}_{2.5}$  concentrations during July 17, 2010, and Figures 3b(1-3) show  $\text{PM}_{2.5}$  sensitivity to 30%  $\text{SO}_2$  emissions reductions from OH, KY, and IN. The results clearly show the interstate impact of emissions reductions from these states.

### Derivation of Multiple Pollutant S-R Matrices

The receptor states/regions of interest typically cover multiple simulation grid cells. Therefore, to derive source-receptor matrices (S-Rs), we aggregated individual grid cell sensitivity values to a single receptor site value. These sensitivity values represent the marginal reduction of emissions from the source region to ozone or  $\text{PM}_{2.5}$  reduction at a receptor site. The sensitivity used for aggregation is the change of pollutant concentration at the peak of the specified time scale (one-hour or eight-hour daily maximum for ozone, and 24-hour average for

PM<sub>2.5</sub>). This source-receptor aggregation is performed on both a population-weighted and an area-weighted basis. Population-weighted S-Rs are needed for estimating potential health benefits from application of source controls, and also give a better proxy for health effects than do area-weighted measures. From a regulatory perspective, the population-weighted S-Rs are also more useful because they better apply to the urban areas (of course, under the Clean Air Act, people living in rural areas are accorded the same level of protection as people in urban areas.) The area-weighted S-Rs are useful to see the pure spatial and temporal effects of emissions on concentrations.

Over the number of episode days,  $D$ , for the number of grid cells,  $i$ , covering receptor site  $r$ ,  $1HDM_{sr}$  is the area- or population-weighted one-hour daily maximum ozone source receptor coefficient in change of ppb at receptor  $r$  per 1,000 tons/day of NO<sub>x</sub> emissions reduction from source  $s$ .

$$1HDM_{sr} = \frac{\sum_{i=1}^I \sum_{d=1}^D [\Delta O3_{id}] p_i}{\Delta E_s * D * \sum_{i=1}^I p_i} * 1000 \quad (3)$$

Here,  $\Delta O3_{id}$  is the daily maximum one-hour ozone concentration change in cell  $i$  and day  $d$ . The daily average is calculated for each grid cell. For population weighting, this value is multiplied by the cell population ( $p_i$ ) and divided by the total population, or  $p_i$  is set to 1 for area weighting. The value is then divided by the source precursor's average daily emissions reduction in tons per day, ( $\Delta E_s$ ). This value is multiplied by 1,000 to convert it to ppb per 1,000 tons/day reduction. This large factor is used for normalizing the sensitivities to avoid errors from very small numbers.

While a per ton sensitivity comparison is useful for evaluating equivalent emissions reductions between states, states have very large differences in what they emit (see Table 1b), and therefore in how much they are able to reduce emissions. To account for this discrepancy, we also define and present what we term “control effects” to quantify the pollution reduction in receptor states that would be achieved by reducing emissions from the 19 states/regions

combined. Control effects are calculated by multiplying the sensitivity matrix by a vector of 30% emissions reductions from each of the source states. We use population-weighted sensitivities in these control effects calculations.

### **Ozone Sensitivity and Control Effects**

There are four types of results combining the May and July episodes with area-weighted and population-weighted sensitivities and control effects. We start with the July episode and area-weighted sensitivities, and then compare these effects to those for May.

*July Sensitivities.* For the July episode and area-weighted S-Rs, states generally have only a limited ability to reduce their own ozone concentrations by reducing elevated  $\text{NO}_x$  emissions (Table 2), although their own share of ozone reductions per unit  $\text{NO}_x$  reduction is the single largest for 53% of states (Figure 4). Results show that the average contribution of elevated  $\text{NO}_x$  control to local one-hour ozone concentrations is about 23%. The range of local control contribution for elevated  $\text{NO}_x$  is between 12% (MO) and 67% (WI). Note that emissions from states outside of the 19 evaluated for sensitivities are not considered, and hence are not contributing to this analysis. For this reason, a state such as Wisconsin may appear to be contributing more to its own pollution than it actually is. The top five states contributing to ozone concentrations in their own and other states (on a per unit emissions basis) are TN, KY, VA, WV, and NC. The aggregate contributions to ozone per unit  $\text{NO}_x$  reductions from these five states to the rest of the domain are 12.4%, 8.7%, 8.5%, 7.3%, and 6.3%, respectively. In general, downwind states (given our July conditions), such as MACTRI (MA, CT, and RI, combined), MI, NY, MO, and NJ, contribute less to other states' pollution.

We also calculate the sensitivities of eight-hour daily maximum ozone concentrations consistent with the new U.S. ozone standard. The new standard is based on the fourth highest eight-hour running average concentration of ozone in a year, while the historical standard is based on the one-hour daily maximum. In general, the sensitivities of eight-hour daily maximum

ozone concentrations to NO<sub>x</sub> changes are slightly smaller than those for the one-hour daily maximum ozone concentrations—2.5% smaller on average for elevated source NO<sub>x</sub> reductions.

To calculate health benefits obtained from emissions reductions, one would need to know the change in population exposure. Since population is not uniformly distributed within individual receptor state/regions, population-weighted sensitivities should be used in such calculations instead of area-weighted sensitivities. For July, our population-weighted one-hour NO<sub>x</sub> to ozone sensitivities are about 17.7% larger on average than area-weighted sensitivities, with more than 74% of the local population-weighted sensitivities exceeding the corresponding area-weighted sensitivities. Some of the differences can be quite striking. For example, for the NJ receptor site, the one-hour ozone population-weighted sensitivity for NJ elevated NO<sub>x</sub> is almost 40% bigger than that of its area-weighted counterpart (4.7 ppb vs. 3.5 ppb).

*July Control Effects.* Because the sensitivities were calculated based on a 30% reduction in emissions, we estimate control effects for this same percentage change. This change, while substantial, is lower than reductions proposed by the EPA Interstate Air Quality Rule, which imply reductions in NO<sub>x</sub> and SO<sub>2</sub> around 75% by 2015, on top of planned reductions already in our 2010 baseline.

We find that the aggregate maximum one-hour ozone reduction ranges from 0.18 ppb (WI) to 1.54 ppb (DE+MD) when elevated source emissions are reduced. These reductions may be compared to baseline estimated ozone concentrations of 67.3 ppb (WI) and 77.2 ppb (DE+MD). These reductions appear to be a small fraction of baseline ozone concentrations, in part because emissions outside of the 19 states (other states and the domain boundaries) are not reduced, contributing to elevated ozone levels.

*July vs. May.* Lower May temperatures and other differences in meteorological conditions between May and July should result in lower predicted ozone concentrations in May, lower ozone sensitivities to NO<sub>x</sub> in May, and different distributions of ozone contributions across states (as seen in Figure 3). Indeed, the spatial and temporal average predicted one-hour daily maximum ozone concentration in May is 53.9 ppb, while in July the average is 61.7 ppb. Also,

the average area-weighted one-hour daily maximum ozone sensitivity with respect to unit point source NO<sub>x</sub> reductions for all sources and receptors for the May episode is 0.39 ppb, while it is 0.53 ppb for July, about 36% larger.

Less predictably, the local ozone sensitivity to elevated NO<sub>x</sub> reductions is higher for the May episode (26% vs. 23% for July, on average). This implies that local control is more effective in May compared to July. Because the winds come predominantly from the South in May and from the Midwest in July, the top contributing states to other states' ozone (as measured by ozone sensitivities) tend to be from the South in May versus the Midwest in July (Figure 3). It can also be seen that the variation among states' contributions is larger in May.

### **PM<sub>2.5</sub> Sensitivity and Control Effects**

Fine particulate matter (considered here to be PM<sub>2.5</sub>) contains a mix of primary and secondary components. In this paper, our discussion focuses on the sensitivity and control effects of 24-hour averages of secondary PM<sub>2.5</sub> only, starting with the July area-weighted sensitivities with respect to sulfur dioxide and elevated source NO<sub>x</sub> emissions. The choice of a 24-hour concentration averaging period is largely driven by the availability of measured data that was available for model evaluation, and by the U.S. air quality standard, which is based on 24-hour and one-year averages.

*July Sensitivities.* PM<sub>2.5</sub> area-weighted sensitivities with respect to SO<sub>2</sub> are far greater than those with respect to NO<sub>x</sub>. For all sources and receptors during the July episode, the PM<sub>2.5</sub> average sensitivity with respect to total SO<sub>2</sub> emissions is about 10 times the sensitivity with respect to elevated point NO<sub>x</sub>. This is calculated from the average of the elemental ratio of two source-receptor matrices, namely the PM<sub>2.5</sub> source-receptor matrix with respect to total SO<sub>2</sub> emissions divided by the PM<sub>2.5</sub> source-receptor matrix with respect to elevated point NO<sub>x</sub>. As a specific example, OH contributes 0.57 μg/m<sup>3</sup> of PM<sub>2.5</sub> to PA per 1,000 tons of SO<sub>2</sub> emissions (on average), but NO<sub>x</sub> emissions from OH elevated point sources contribute only 0.047 μg/m<sup>3</sup> to PA

(per 1,000 tons). Thus, reducing a ton of total SO<sub>2</sub> emissions in OH will have about 12 times the impact on PM<sub>2.5</sub> concentrations in PA as would reducing a ton of OH point NO<sub>x</sub> emissions.

As expected, we found that decreasing SO<sub>2</sub> always decreases sulfates and increases nitrates, with the former change being about 28 times the latter for the July episode. Our PM<sub>2.5</sub> sensitivities represent the net effect of SO<sub>2</sub> emissions control (including sulfate aerosol reduction and slight nitrate aerosol increases). West et al.<sup>15</sup> studied marginal PM<sub>2.5</sub> under an equilibrium model to estimate the conditions for nonlinear response to changes in sulfate concentrations. Their study considered local effects (without pollutant transport). They found that reductions in SO<sub>2</sub> emissions may cause an increase in aerosol nitrate (the “bounce-back” effect) because nitric acid converts to nitrate. Such conditions are found to be common and significant in winter—perhaps three to four times as prevalent as in summer. As West et al.<sup>15</sup> pointed out, the bounce-back effect can be so large that PM<sub>2.5</sub> concentrations can actually increase. In our study (as in others),<sup>8</sup> we found that reducing SO<sub>2</sub> emissions reduces PM<sub>2.5</sub> concentrations, and that the bounce-back effect is small. Consideration of winter periods might find a larger formation of nitrate aerosol.

*July Control Effects.* By reducing elevated NO<sub>x</sub> emissions 30% from each of the 19 evaluated states/regions, the aggregate PM<sub>2.5</sub> reduction in these states ranges from 0.006 μg/m<sup>3</sup> (MO) to 0.071 μg/m<sup>3</sup> (KY), and are derived from small reductions in particulate nitrate, sulfate, and ammonium. These reductions are small largely because nitrate levels during the summer in this region are small. SO<sub>2</sub> sensitivities are much larger. By reducing total SO<sub>2</sub> emissions 30% from each of these states, the aggregate reduction ranges from 0.13 μg/m<sup>3</sup> (WI) to 2.55 μg/m<sup>3</sup> (VA). The reduction contributed by source states is not equally distributed, depending on the emissions reduction and S-R coefficients between sources and receptor states. Virtually all of the reduction is due to decreases in sulfate and ammonium, and there is usually a concurrent increase in nitrate. These results may be compared to the annual PM<sub>2.5</sub> standard of 15 μg/m<sup>3</sup> and the daily standard of 65 μg/m<sup>3</sup> (the average over three consecutive years of the 98<sup>th</sup> percentile of the daily value of PM<sub>2.5</sub>). Also, note that states such as WI and MO are on the upwind side of the region being

studied, so are affected by emissions not accounted for in this analysis. For the July episode, our 24-hour daily average PM<sub>2.5</sub> concentrations range from 4.7 µg/m<sup>3</sup> to 32.7 µg/m<sup>3</sup>.

*July versus May.* We find that predicted PM<sub>2.5</sub> concentrations in May are low, averaging 12.0 µg/m<sup>3</sup>, while in July they average 16.5 µg/m<sup>3</sup>. Also, the average 24-hour area-weighted PM<sub>2.5</sub> sensitivities with respect to SO<sub>2</sub> reductions for all sources and receptors for the May episode are about one-fifth the average for July: 0.032 µg/m<sup>3</sup> versus 0.17 µg/m<sup>3</sup>. This is because, compared to the July episode, the May episode had faster winds, lower temperatures, more rain, and lower concentrations of biogenic VOCs and hydroxyl radicals.

On the other hand, the fraction of local contributions to PM<sub>2.5</sub> reductions is higher for the May episode (29.0% vs. 23%, on average). In addition, because the winds come predominantly from the South in May and from the Midwest in July, the top contributing states to other states' PM<sub>2.5</sub> tend to be from the South in May versus the Midwest in July. As with ozone, the variation among states' contributions is larger in May (Figure 5).

### Comparison of Long-Range Response of Ozone and PM<sub>2.5</sub> Sensitivities

In this section, we examine how distance affects ozone and PM<sub>2.5</sub> sensitivities. A regression analysis was performed to explain ozone and PM<sub>2.5</sub> sensitivities in terms of distance from the source-state centroid to the receptor-state centroid. The dependent variable,  $r_{ij}$ , is the remote sensitivity normalized by the source-state local sensitivity.

$$r_{ij} = \beta_0 + \beta_1 * d_{ij} + \varepsilon \quad (4)$$

The right hand side,  $d_{ij}$ , is the distance between the  $i$ th source-state centroid and the  $j$ th receptor-state centroid.  $\beta_0$  is the constant term of the regression equation. The slope ( $\beta_1$ ) of this simple regression model can be interpreted as the decreasing sensitivity per unit of distance.  $\varepsilon$  is the stochastic error term. On average for the July episode, the PM<sub>2.5</sub> sensitivity to elevated source NO<sub>x</sub> emissions reductions is significant, decreasing about 0.053% per km; the ozone sensitivity decreases about 0.040 % per km. For the May episode, the PM<sub>2.5</sub> sensitivity reduction is 0.041%



per km and the ozone sensitivity reduction is about 0.037 % per km. Results of this regression are shown in Table 3.

### Issues of Nonlinearity and Uncertainty

There are a number of important caveats that apply to the results of this study. First, we assume that linearity holds within the 30% range of emissions changes and note that this assumption was tested and found to be acceptable.<sup>1,6</sup> Nevertheless, if the control measures were to lead to emissions reductions much larger than 30%, the assumed linearity of response to controls may no longer be satisfactory. In such a case, a second calculation at or near the reduced emissions level may be necessary to account for the nonlinearity in pollutant response. Second, we did not consider other sources of uncertainty and variability. For example, we only considered two meteorological episodes (a total of 15 simulation days for use), and did not extensively study the effect of meteorological input variation. While we have captured a high and moderate pollutant episode, variation in wind direction, temperature, and other parameters may significantly affect sensitivity results.

### Conclusions

While the absolute effects of NO<sub>x</sub> and SO<sub>2</sub> control on ozone and PM<sub>2.5</sub> concentrations are sensitive to meteorological conditions, reducing elevated NO<sub>x</sub> emissions from our 19-state study area by 30% from 2010 levels will have, at most, a modest impact on ozone concentrations and PM<sub>2.5</sub> concentrations in most of those states. Note, however, that boundary emissions and emissions from other states in the domain were not eliminated. Comparatively, reducing SO<sub>2</sub> emissions can have significant effects on PM<sub>2.5</sub> concentrations. While “rebound” and “bounce-back” effects are observed, they are generally not significant enough to result in perverse effects on ozone during these episodes. The rebound effect on PM<sub>2.5</sub> of SO<sub>2</sub> reductions is generally small (in terms of nitrate increases).

Long-range transport of precursors is an important issue for both pollutants, although perhaps less so than is commonly thought. Local contributions to air pollution problems account for about 23% of total ozone concentrations and PM<sub>2.5</sub> concentrations, and neighbouring states contribute much of the rest. The ozone sensitivity results are comparable with others in the literature based on simpler models.<sup>16-18</sup> The PM sensitivity coefficients, however, are smaller.<sup>18</sup> This result may be due, in part, to the episodes chosen for study.

Area and population-weighted sensitivity matrices tell very different stories. We found that population-weighted sensitivities exceed area-weighted sensitivities by as much as six times in some regions. When evaluating health damages, one should use population-weighted sensitivities to account for human exposure.

**References**

1. Odman, M.T., J.W. Boylan, J.G. Wilkinson, and A.G. Russell. Integrated Modeling for Air Quality Assessment: The Southern Appalachian Mountains Initiative Project. *J. Phys. IV*. **2002**, 12 (PR10): 211-234
2. Russell, A.G., K.F. McCue, and G.R. Cass. Mathematical Modeling of the Formation of Nitrogen-Containing Pollutants—2. Evaluation of the Effects of Emission Controls. *Environ. Sci. Technol.* **1998**, 22, 1336-1347.
3. Russell, A.G., K.F. McCue, and G.R. Cass. Mathematical Modeling of the Formation of Nitrogen-Containing Air Pollutants—1. Evaluation of an Eulerian Photochemical Model. *Environ. Sci. Technol.* **1998**, 22, 263-271.
4. Meng, Z., D. Dabdub, and J.H. Seinfeld. Chemical Coupling Between Atmospheric Ozone and Particulate Matter. *Science* **1997**, 277 (5322), 116-119.
5. Stockwell, W.R., J.B. Milford, G.J. McRae, P. Middleton, and J.S. Chang. Nonlinear Coupling in the NO<sub>x</sub>-SO<sub>x</sub>-Reactive Organic System. *Atmos. Environ.* **1988**, 22, 2481-2490.
6. Yang, Y.J., J.W. Wilkinson, and A.G. Russell. Fast, Direct Sensitivity Analysis of Multidimensional Photochemical Models. *Environ. Sci. Technol.* **1997**, 31, 2859-2868.
7. Kumar, N., M.T. Odman, and A.G. Russell. Multiscale Air Quality Modeling: Application to Southern California. *J. Geophys. Res.* **1994**, 99, 5385-5397.
8. Boylan, J.W., M.T. Odman, J.G. Wilkinson, A.G. Russell, K. Doty, W. Norris, and R. McNider. Development of a Comprehensive, Multiscale “One Atmosphere” Modeling System: Application to the Southern Appalachian Mountains. *Atmos. Environ.* **2002**, 36, 3721-3734.
9. Bergin, M.S, J-S Shih, J.W. Boylan, J.G. Wilkinson, A.J. Krupnick, and A.G. Russell. Inter- and Intra-State Impacts of NO<sub>x</sub> and SO<sub>2</sub> Emissions on Ozone and Fine Particulate Matter. To be submitted to *Environ. Sci. Technol* (**2004**).

10. Bergin, M.S, J.W. Boylan, J.G. Wilkinson, J-S Shih, A.J. Krupnick, and A.G. Russell. Effects of Multiscale Grid Resolution and Spatial Distribution on 3D Model Performance for Ozone and Aerosols. To be submitted to *Environ. Sci. Technol* (2004).
11. Pielke, R.A., W.R. Cotton, R.L. Walko, C.J. Tremback, W.A. Lyons, L.D. Grasso, M.E. Nicholls, M.D. Moran, D.A. Wesley, T.J. Lee, and J. H. Copeland, A Comprehensive Meteorological Modeling System - RAMS. *Meteor. Atmos. Phys.* **1992**, *49*, 69-91.
12. Wilkinson, J.G., C.F. Loomis, D.E. McNally, R.A. Emigh, and T.W. Tesche. Technical Formulation Document: SARMAP/LMOS Emissions Modeling System (EMS-95). AG-90/TS26 & AG-90/TS27. Alpine Geophysics, Pittsburgh, PA (1994).
13. USEPA EPA AIRS Data. U.S. Environmental Protection Agency, Office of Air Quality Planning & Standards, Information Transfer & Program Integration Division, Information Transfer Group. [www.epa.gov/airsdata](http://www.epa.gov/airsdata) (2001).
14. NPS. National Park Service, Air Quality Research Division, Fort Collins. Anonymous ftp at [ftp://alta\\_vista.cira.colostate.edu](ftp://alta_vista.cira.colostate.edu) in /data/improve (2000).
15. West, J.J., A.S. Ansari, and N.P. Spyros. Marginal PM<sub>2.5</sub>: Nonlinear Aerosol Mass Response to Sulfate Reductions in the Eastern United States. *J. Air & Waste Manage. Assoc.* **1999**, *49*, 1415-1424.
16. Rao, S.T.; Mount, T.D.; Dorris, G. Least Cost Control Strategies to Reduce Ozone in the Northeastern Urban Corridor. The NY State Department of Environmental Conservation, Division of Air Resources, Albany, NY (1999).
17. Krupnick, Alan J., Virginia D. McConnell, David H. Austin, Matthew Cannon, Terrell Stoessell, and Brian Morton. The Chesapeake Bay and the Control of NO<sub>x</sub> Emissions: A Policy Analysis, RFF discussion paper 98-46, August, Washington, DC. (1998).
18. Krupnick, A., V. McConnell, M. Cannon, T. Stoessell, and M. Batz. Cost-Effective NO<sub>x</sub> Control in the Eastern United States, RFF discussion paper 00-18, August, Washington, DC. (2000).

**Table 1(a). Basecase domain-wide 1995 average day emissions (tons/day)**

Emission	Source Type	July 11-19	May 24-29
NO <sub>x</sub>	Area	3,008	5,573
	Biogenic	5,284	3,024
	Power plants	22,248	17,063
	Non-road Mobile	7,319	5,932
	On-road Mobile	12,509	12,855
	Elevated Point	8,110	7,425
SO <sub>2</sub>	Power plants	34,140	26,364
	Other	20,094	20,259
Organic Gases	Biogenic	182,381	76,315
	Anthropogenic	46,014	43,467

**Table 1(b). State-specific 2010 average day emissions (tons/day)**

State	July 11-19					May 24-29				
	NO <sub>x</sub>		SO <sub>2</sub>	Organic Gases		NO <sub>x</sub>		SO <sub>2</sub>	Organic Gases	
	Elevated	Ground level	Total	Biogenic	Anthro <sup>a</sup>	Elevated	Ground level	Total	Biogenic	Anthro <sup>a</sup>
AL	343	644	1,487	11,457	1,051	338	710	1,493	6,275	1,152
DC	1	48	26	7	30	1	50	25	3	30
GA	340	649	2,043	9,412	1,162	335	969	1,977	6,017	1,494
IL	667	1,489	2,268	2,870	1,988	661	1,342	2,442	563	1,897
IN	516	1,033	2,514	2,239	1,394	505	887	2,495	555	1,319
KY	292	653	1,762	5,491	785	288	591	1,761	2,097	767
MA + CT +RI	120	609	841	3,083	1,090	116	526	875	1,798	934
MD + DE	172	406	852	1,285	604	164	430	928	497	605
MI	544	902	1,343	3,541	1,409	546	988	1,399	842	1,454
MO	234	682	1,066	12,545	1,082	231	532	1,033	2,718	1,073
NC	317	606	1,972	6,961	1,461	311	739	1,936	3,475	1,590
NJ	90	461	748	901	844	90	578	951	359	862
NY	249	795	1,559	2,130	1,558	231	857	2,164	841	1,549
OH	497	1,033	2,732	2,537	1,542	483	989	2,650	737	1,449
PA	549	738	1,388	4,852	1,231	533	870	2,078	1,465	1,179
SC	246	423	942	4,420	1,050	242	438	931	2,690	1,072
TN	216	891	1,098	7,124	1,619	212	859	1,092	3,131	1,587
VA	186	748	1,376	7,014	1,250	180	737	1,401	2,425	1,254
WI	186	601	1,159	5,283	891	181	548	1,224	1,077	873
WV	331	273	1,550	3,916	415	324	285	1,543	1,341	401

<sup>a</sup> Anthro: Anthropogenic VOC

**Table 2. Summary of state contributions (%) to local air quality per unit emissions reduction (July episode, area-weighted)**

Receptor State	1-hr Ozone	8-hr Ozone	Episode average 24-hr PM <sub>2.5</sub>	
	Reduced Elevated NO <sub>x</sub>	Reduced Elevated NO <sub>x</sub>	Reduced Elevated NO <sub>x</sub>	Reduced Total SO <sub>2</sub>
AL	36	34	35	36
DE+MD	21	20	12	14
GA	24	23	24	27
IL	13	10	11	19
IN	19	17	18	22
KY	25	25	22	23
MA+CT+RI	16	20	19	21
MI	12	13	16	15
MO	12	14	21	12
NC	22	21	19	22
NJ	21	21	10	15
NY	14	13	16	20
OH	15	15	6	15
PA	16	15	17	21
SC	32	32	33	42
TN	41	39	38	35
VA	24	24	21	16
WI	67	67	69	40
WV	17	17	10	25
<b>Average</b>	23	23	22	23

Local contribution to reductions in one-hour daily maximum ozone and eight-hour daily maximum ozone from unit reductions in elevated NO<sub>x</sub> and reductions in 24-hour PM<sub>2.5</sub> concentrations from unit reductions in elevated NO<sub>x</sub> and total SO<sub>2</sub> emissions. For example, if each state reduced elevated NO<sub>x</sub> emissions by one ton, then 36% of the resulting ozone reductions in Alabama would be attributed to reductions of NO<sub>x</sub> within Alabama.

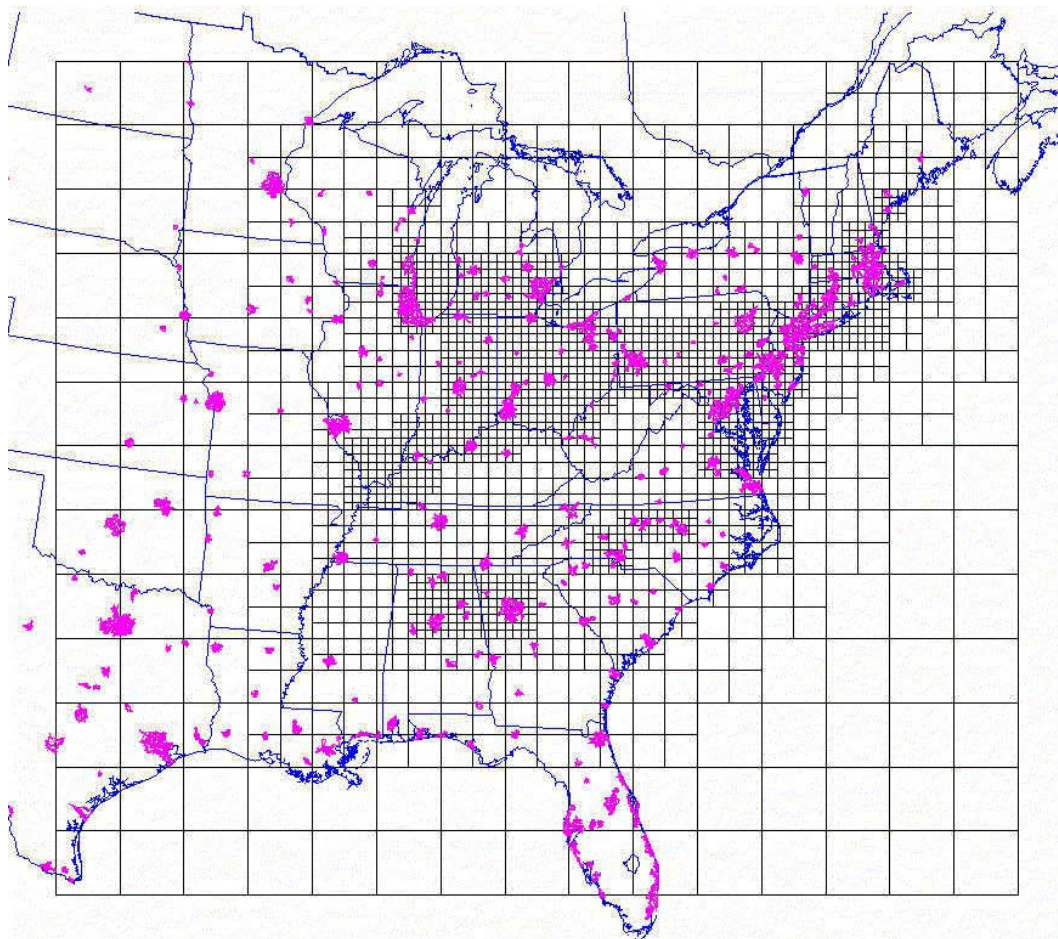
**Table 3. Results of long-range response of ozone and PM<sub>2.5</sub> sensitivities with respect to elevated point NO<sub>x</sub> reduction for July and May episodes.**

Regression	July ozone	July PM <sub>2.5</sub>	May ozone	May PM <sub>2.5</sub>
$\beta_0$	0.72 (13.95)	0.95 (10.63)	0.63 (17.86)	0.64 (21.13)
$\beta_1$	-0.00040 (-8.56)	-0.00053 (-6.56)	-0.00037 (-11.67)	-0.00041 (-14.92)
Adj R-squared	0.167	0.105	0.273	0.381

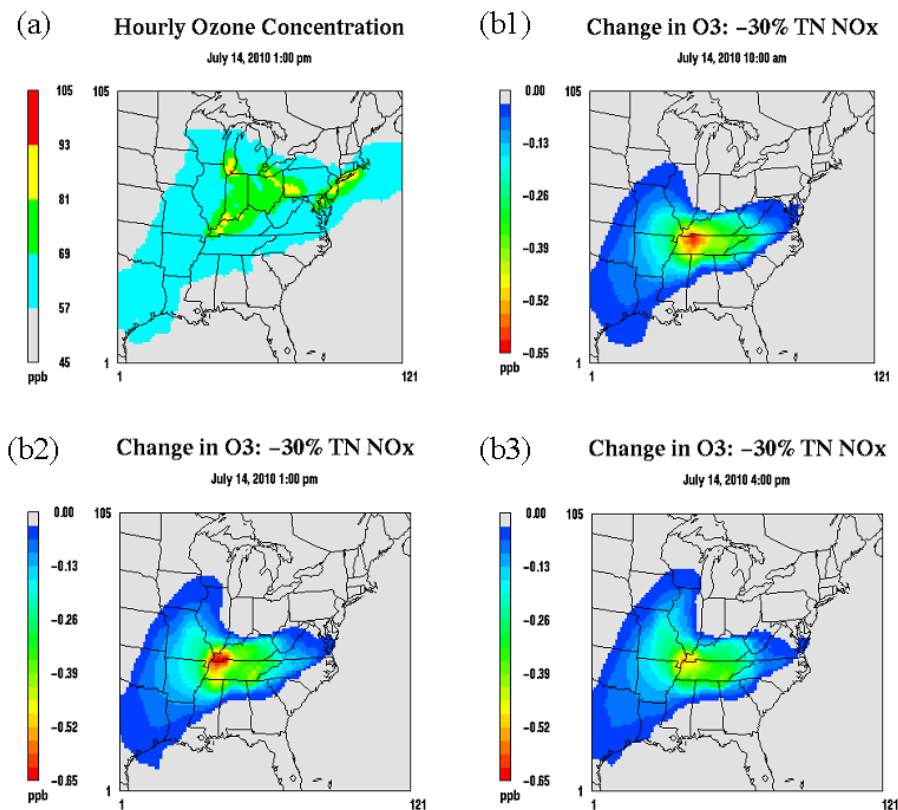
Numbers in parentheses are t-statistics. All slope coefficients are significant at the 95% level or better.



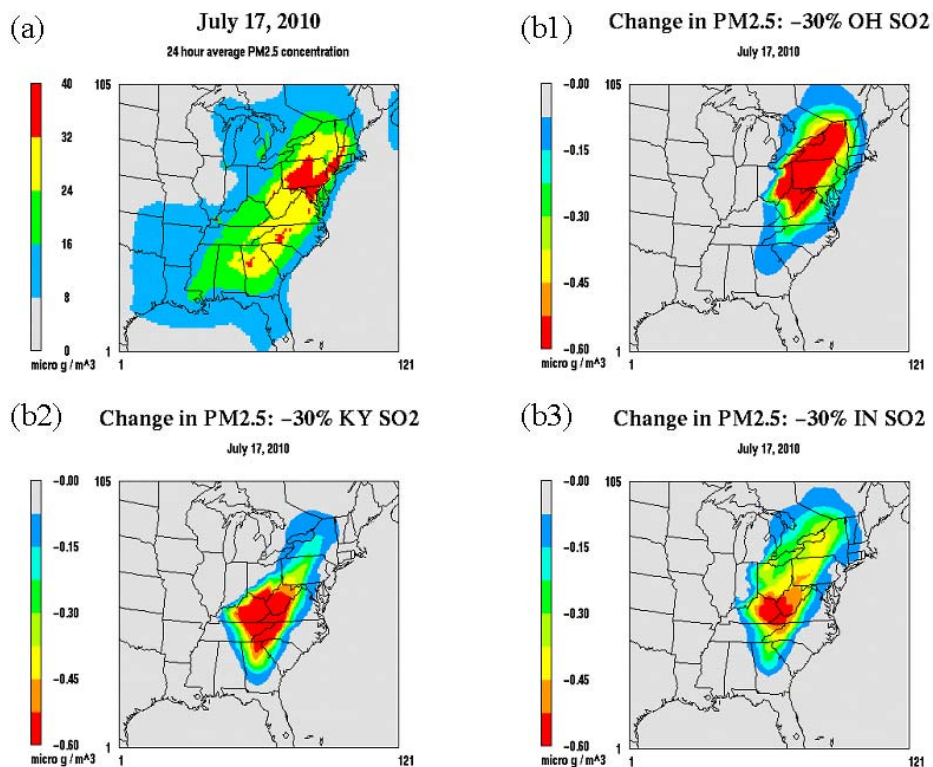
**Figure 1.** Multiscale grid used to model changes in ozone and particulate species from changes in  $\text{NO}_x$  and  $\text{SO}_2$ . The finest resolution has horizontal grids of 24km per side, and the other cells are 48km, 96km, and 192km per side. The shaded areas represent high population densities (urban areas). Fine-scale cells are placed over areas of high industrial or population densities.



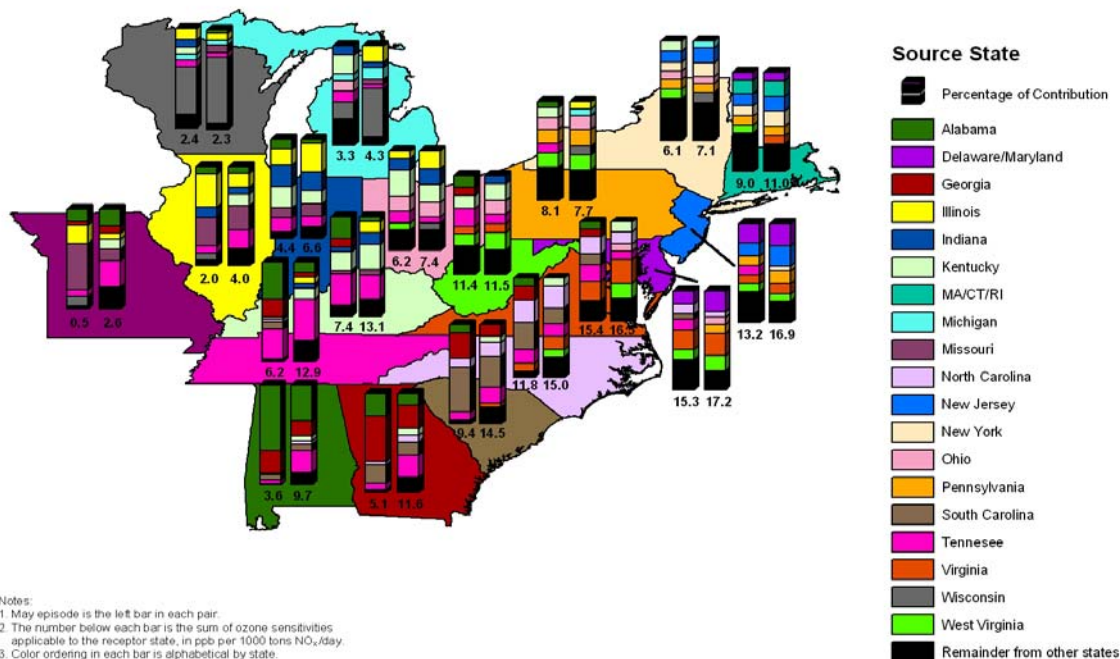
**Figure 2.** Ozone and its sensitivity to a reduction of elevated  $\text{NO}_x$  emissions. (a) Concentration of ozone simulated for July 14, 2010 at 1:00 p.m. (b) Change in ozone concentration at (b1) 10:00 a.m., (b2) 1:00 p.m., and (b3) 4:00 p.m. caused by a 30% reduction in elevated  $\text{NO}_x$  emissions from Tennessee.



**Figure 3.** PM<sub>2.5</sub> 24-hour average concentration and sensitivity to total SO<sub>2</sub>. (a) 24-hour average concentration of PM<sub>2.5</sub> simulated for July 17, 2010; (b) 24-hour average change in PM<sub>2.5</sub> concentration caused by a 30% reduction in total SO<sub>2</sub> emissions from (b1) Ohio, (b2) Kentucky, and (b3) Indiana.



**Figure 4.** Relative area-weighted one-hour maximum average ozone sensitivities with respect to a unit reduction in elevated NO<sub>x</sub> (ppb per 1,000 tons/day) for both the May and July episodes. For each state, the top six contributing state/regions are shown, with the rest of the evaluated states combined into an “other” category. For each receptor, the left column represents the results for May and the right the results for July. The height of an individual component of a column represents the percentage of ozone change caused by a unit NO<sub>x</sub> reduction. The color of the component indicates the source (state/region) of that emission. For example, for the state of Maryland during the July episode, the column indicates that the top six contributors to Maryland ozone concentrations are Maryland, West Virginia, Virginia, Pennsylvania, Ohio, and North Carolina. During the May episode, however, Maryland’s own contribution is lower, and Tennessee and Kentucky become major contributors.



**Figure 5.** Relative area-weighted 24-hour average PM<sub>2.5</sub> sensitivities with respect to a unit reduction in total SO<sub>2</sub> emissions (µg/m<sup>3</sup> per 1,000 tons/day) for the May and July episodes. The top six contributing state/regions and “all other states” are presented.

