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Chapter 2.15

Integrated observational and modeling approaches for evaluating the effectiveness of ozone control policies ☆

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Abstract

Although there has been much progress in improving ambient air quality, the concentrations of ozone continue to exceed their acceptable levels in the United States and many parts of the world. Because of the regional nature of this pollutant, the U.S. Environmental Protection Agency (EPA) required the States to undertake large reductions in nitrogen oxides (NO_x) emissions by 2004 to address long-range transport of ozone and its precursors as part of their State Implementation Plans (SIPs). As a result of this control program, referred to as the "NO_x SIP Call", 21 states in the eastern United States have achieved substantial NO_x reductions from the utility sector by 2004. This emission control strategy should greatly reduce ambient ground-level ozone in the eastern United States. A systematic tracking of air quality achievements is necessary to properly evaluate U.S. policy and program results in protecting public health and the environment. Consequently, EPA has undertaken a comprehensive assessment of the effectiveness of this control program to meet its objectives. In this paper, integrated observational and modeling methodologies are described in order to assess the impact of NO_x emission reductions on maximum ozone concentrations. In particular, the emphasis in this paper is on selected results from the photochemical modeling effort. A set of modeling scenarios was performed that included point source NO_x emission measurements from extended summer periods before and after implementation of the emission reductions. The model results revealed discernable decreases in daily maximum 8-h ozone in

☆This work constitutes a contribution to the NOAA Air Quality Program. Although it has been reviewed by EPA and NOAA and approved for publication, it does not necessarily reflect their policies or views.

downwind areas of a major point source emission region exhibiting substantial NO_x emission reductions, and the impacts were found to be greater at higher ozone values, which is a desirable benefit of the control program.

1. Introduction

Numerous emission control programs have been implemented by the U.S. Environmental Protection Agency (EPA, 2005) in an effort to reduce ground-level ozone concentrations by reducing emissions of VOCs or NO_x, the key precursor species involved in the photochemical production of ozone. Although there has been a downward trend in ozone levels, daily maximum 8-h ozone concentrations continue to exceed acceptable limits in widespread areas of the eastern United States. The findings of an extensive data analyses and modeling effort performed under the auspices of the OTAG (Ozone Transport and Assessment Group) program revealed that regional-scale transport of ozone and its precursors contributed to elevated ozone concentrations in downwind areas within the eastern states. Consequently, the EPA responded with the development and implementation of an emission control policy known as the NO_x State Implementation Plan (SIP) Call. The NO_x SIP Call rule was designed to reduce the interstate transport of ozone and its precursor species by requiring substantial NO_x emission reductions from major point sources in 22 eastern states with full implementation of controls before the summer 2004 ozone season.

It is important to assess whether a major emission control program achieves its objectives and provides a discernable improvement in air quality. Consequently, a research effort has been underway using a combination of observation-based and model-based methods to investigate the effectiveness of this emission control program in decreasing peak ozone concentrations. The modeling approach applied in this effort involves photochemical simulations with the Community Multiscale Air Quality (CMAQ) model. Accurate point source NO_x emission measurements provide a reliable emissions data set for the model simulations. CMAQ model simulations were performed for 3-month summer periods in 2002 and 2004, which corresponded to ozone seasons before and after implementation of the NO_x emission controls, respectively. A set of modeling scenarios was designed to allow for an examination of the separate impacts of the emission changes and meteorological differences on ozone concentrations. The observation-based approach employs a statistical filter/regression technique in an attempt to analyze multi-year time series of ozone and meteorological parameter measurements at an extensive monitoring network. Attributing an ozone trend to particular emission changes is confounded by the strong influence of weather conditions on ozone levels and the year-to-year variability in the meteorological conditions, as was the case during these two summer seasons. However, after accounting for the influence of meteorological variables, differences in meteorologically adjusted maximum ozone concentrations from separate summer seasons can be attributed to the effect of a change in total emissions. The methods and procedures employed with these two approaches are described. Selected results from the photochemical modeling effort are emphasized herein to demonstrate the impact on ozone and NO_x concentrations from the point source emission changes in an attempt to provide evidence of the benefits of this emission reduction program.

2. Data sets and methods

2.1. Observational data sets

The ozone and meteorological measurements used in this study were collected at monitoring sites of the Clean Air Status and Trends Network (CASTNet; http://www.epa.gov/castnet), which are situated in primarily rural locations of the United States. For this study, data from nearly 50 monitoring sites in the eastern U.S. were analyzed to determine the daily maximum 8-h ozone concentrations from hourly measurements at each site.

Major U.S. point sources, particularly electrical generating units at fossil-fuel power plants and large industrial sources, have been equipped with Continuous Emissions Monitoring systems (CEMS) in order to provide direct, hourly emission measurements of NO_x and SO_2. The hourly CEMS data for the summer seasons of 2002 and 2004 were available for use in this analysis and modeling effort.

2.2. Observation-based analysis approach

The statistical filter/regression methods described by Milanchus et al. (1998) were applied to account for the meteorological influence on maximum ozone concentrations at each CASTNet site over a measurement period spanning 1988–2004. Briefly, the methodology involves the spectral decomposition of the time series of ozone and meteorological variables into fluctuations occurring on various temporal scales before quantifying the relationship between two observed variables. By the judicious application of an iterative moving average KZ filter algorithm (Rao and Zurbenko, 1994), separation of the variations was achieved at frequencies related to the shorter term, synoptic-scale from the seasonal or baseline time scales. Subsequently, multiple regression analysis was performed among the baseline components of ozone, total daily solar radiation, and specific humidity to derive the portion of the ozone baseline fluctuation explained by these meteorological variables. Likewise, the regression technique was applied to the synoptic-scale components of ozone along with the total daily solar radiation and dew point depression to identify the ozone variability governed by these meteorological variables. Finally, meteorologically-adjusted ozone values were derived as the sum of the baseline and synoptic-scale residuals, which represent the contribution of ozone fluctuations not explained by the meteorological variables. Further details about the application of this statistical filter/ regression approach and results are provided in Gego et al. (2007).

2.3. Photochemical modeling approach

The CMAQ modeling system (version 4.5) was applied in this modeling study. CMAQ is a comprehensive Eulerian air quality grid model that is capable of simultaneously treating a wide variety of atmospheric pollutants (e.g., oxidants, aerosols, air toxics, and mercury species). For the photochemical simulations performed in this study, the Carbon-Bond 4 (CB4 version 4.2) gas-phase chemical mechanism was used in conjunction with the Euler backward interactive (EBI) chemistry solver. CMAQ contains state-of-science algorithms to solve the relevant atmospheric processes. The CMAQ model was configured with the standard science options to treat particular physical processes as described in Byun and Schere (2006).

Meteorological fields were generated by the Penn State/NCAR Mesoscale Model (MM5 version 3.6.3). The MM5 model simulations included four-dimensional data assimilation (FDDA) of observed wind, temperature, and moisture data to provide more accurate 3-D modeled fields, and an improved land-surface scheme to improve model response to varying soil moisture and vegetation conditions over the summer season. The CMAQ Meteorology-Chemistry Interface Processor (MCIP v3.1) program was exercised to extract/reformat MM5 output into data sets containing the hourly 2-D and 3-D meteorological fields required by CMAQ.

The 3-D emission data sets were generated by the comprehensive Sparse Matrix Operator Kernel Emissions (SMOKE version 2.2; http://www.smoke-model.org) processing system. Anthropogenic emissions from the EPA 2001 National Emissions Inventory (NEI version 3) were used to generate surface and elevated non-CEMS point source emissions. Natural surface emissions of NO_x, isoprene and other biogenic VOC species were computed by the Biogenic Emissions Inventory System (BEIS version 3.13). The MOBILE6 model was applied to use projections of vehicle-miles-traveled (VMT) and fleet factors to develop gridded motor vehicle emissions for the 2002 and 2004 periods. The SMOKE system also computed plume rise using stack parameters and meteorological fields in order to allocate all point source emissions into the proper vertical layers.

2.3.1. Model setup

The CMAQ modeling domain encompassed the eastern half of the United States and southeastern Canada with 205 × 199 horizontal grid cells with a 12-km grid dimension. The vertical structure consisted of 14 layers extending from the surface to over 15 km on a sigma-pressure, terrain-following coordinate system. The initial conditions and lateral boundary concentrations for each modeling scenario were defined to be the same set of time-invariant tropospheric background values. The modeling period was from May 28 through August 31 for both summer seasons with the first four days considered as a model spin-up period. Consequently, the model results for 92 days starting on June 1 of each summer were used in the analyses.

2.3.2. Modeling scenarios

Table 1 lists the model scenarios designed to allow for investigation of the impacts of emission change and different meteorological conditions on ozone concentration levels. Of particular interest, comparisons of base case results (M02E02, M04E04) with those from the M02E04 and M04E02 modeling scenarios permit an assessment of emission effects on ozone levels during the summer 2002 and 2004 periods, respectively. Additionally, comparative analysis of M02E02 and M04E04 results

Table 1. CMAQ model simulation scenarios

	Summer meteorology	
	2002	2004
Emissions		
2002	M02E02	M04E02
2004	M02E04	M04E04

provides information on the combined effects of meteorology and emissions on ozone levels between the two summer periods. Since weather conditions during the summer of 2002 were more favorable for ozone formation than the cool and wet summer of 2004, results from the model scenarios highlighted in Table 1, which applied the 2002 meteorology using emissions reflecting pre-control and post-control CEM data, are the focus of this paper.

2.4. Trajectory modeling

The trajectory method employed in this effort was the Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT version 4.7, http://www.arl.noaa.gov/ready/hysplit4.html). The HYSPLIT model was applied to generate back trajectories originating from each CASTNet site location for each day to identify cases when the airflow at each monitoring site had previously passed through the Ohio River Valley (ORV) region, since this area contained numerous point sources exhibiting large NO_x emission reductions. Forward trajectories in time were also generated starting from the specific location and altitude of selected point source plume emissions. Hourly trajectory coordinates were converted into CMAQ grid cell indices in order to extract the pollutant concentrations from the CMAQ 3-D concentration files. For this trajectory analysis, a new interface utility program (MCIP2ARL) was developed and applied to retrieve the MCIP hourly meteorological parameter fields, the same meteorology applied in the CMAQ simulations, to generate input date sets compatible for use in the HYSPLIT trajectory model calculations.

3. Results and discussion

3.1. Point source NO_x emission changes

The locations of major point sources and the percentage change in their NO_x emissions due to the NO_x SIP Call program are depicted in Fig. 1. Substantial reductions in NO_x emissions of greater than 80% are evident at numerous individual sources, particularly in the largest NO_x-emitting point sources within the Ohio River Valley (ORV) region. Figure 2 shows that most major point source emissions are injected into model layers at heights from 200 m to over 600 m above ground over the diurnal period, which allows for considerable transport of elevated plumes during the nocturnal period. A significant decrease is evident in NO_x emissions

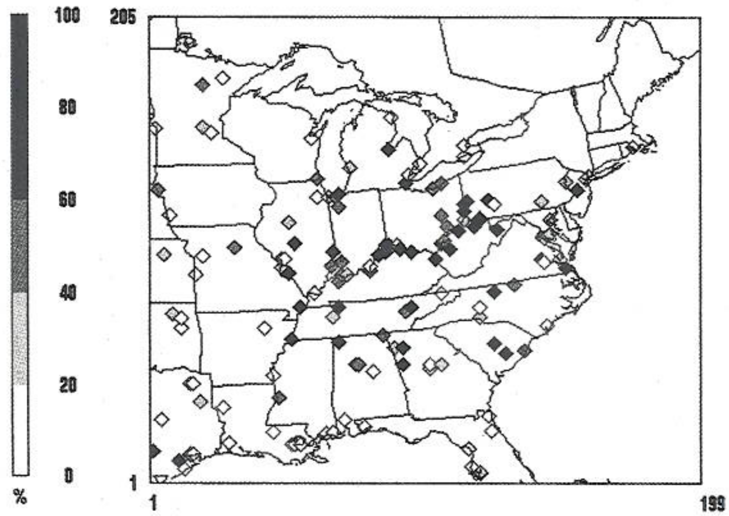

Figure 1. Locations of major NO_x point sources in the CMAQ modeling domain and percentage emission reduction between 2002 and 2004.

between the base and the post-control cases indicating less NO_x is available for transport due to the control program. Results of analysis of the hourly CEMS measurements also indicated a substantial decrease in NO_x emissions at all hours, even during the daytime peak-demand period. There was a 36% drop in NO_x emissions from the CEMS point sources from 2002 to 2004; however, this contributed to a rather modest 8% decrease in total domain-wide NO_x emissions between the M02E02 and M02E04 modeling scenarios.

3.2. Results of photochemical modeling

A variety of weather conditions and flow patterns were simulated during the summer periods being modeled. Of particular interest are the cases when the synoptic pattern exhibits a high-pressure area in the eastern or southeastern U.S., which sets up a generally southwesterly wind flow across the ORV emission region. Elevated ozone levels in the eastern U.S. are associated with this flow pattern as horizontal pollutant transport is more pronounced toward the northeastern states. During the summer 2002 period, 19 cases were identified when a predominately southwestern flow occurred across the ORV area. The impact on total nitrogen species (NO_y) is depicted in Fig. 3, which displays the average percent difference between the M02E02 and M02E04 results in layer 5 (~400 m AGL) at a

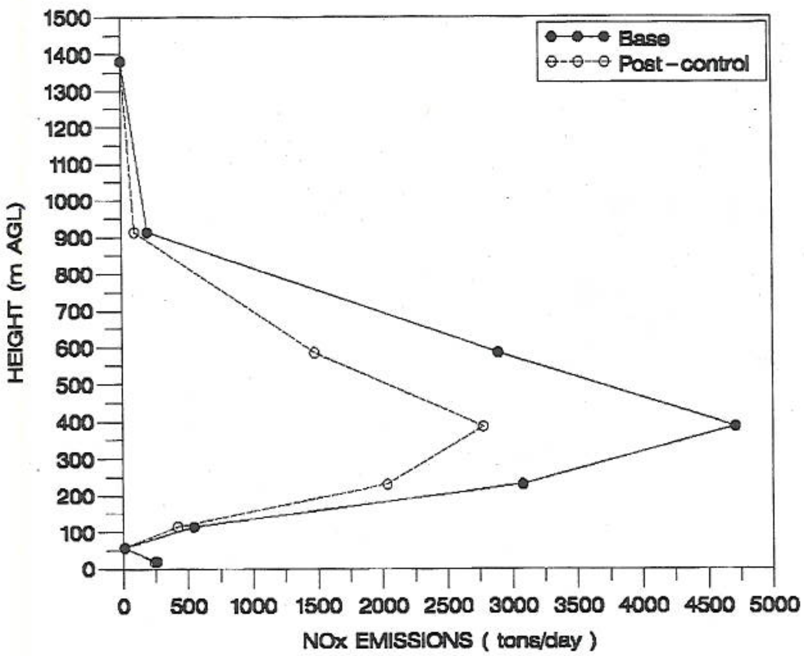

Figure 2. Vertical distribution of daily NO_x emissions from CEMS sources for 2002 base and post-control cases.

late afternoon hour (2100 UTC) from these cases. Although the largest decreases in NO_y ($NO_y = NO_x +$ secondary nitrogen species) are clearly evident close to the point source locations due to much lower NO_x levels from the emission reductions, the notable decreases in NO_y found at further downwind distances from the ORV in north-central Pennsylvania, central New York state, southern Canada, and even off the east coast are due to decreases in secondary species (i.e., HNO_3). Figure 4 reveals a broad area of lower ozone at the surface. However, a downwind area with a more pronounced decrease in maximum 8-h ozone exhibits a southwest/northeast orientation due to the alignment of the many point sources in the ORV with the wind flow pattern in these cases.

Trajectory analysis also provided valuable information about the impact of the NO_x point source reductions from these model simulations. Species concentrations generated from these modeling scenarios along a forward trajectory downwind from selected point sources in individual cases were examined to investigate differences between the modeling scenario results, since average results for the cases shown above tends to dampen the impact with distance because of flow variations among the

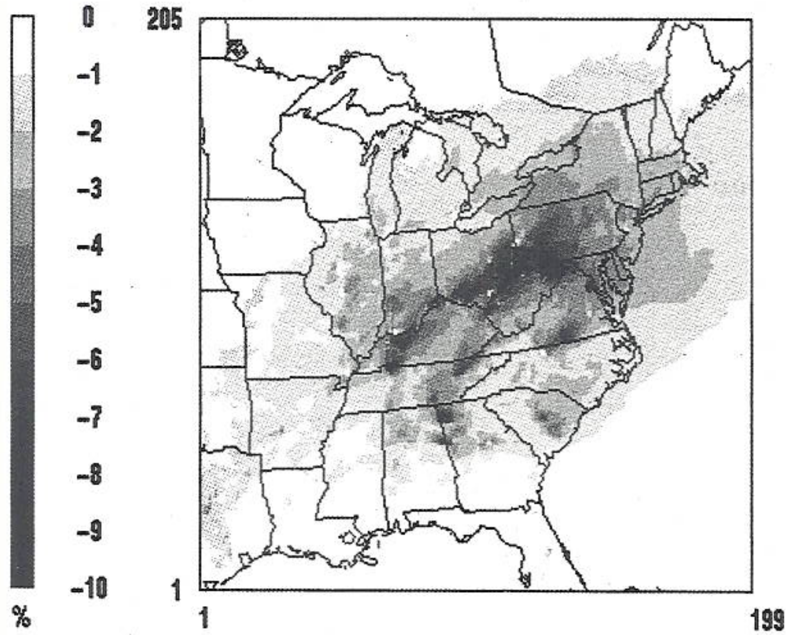

Figure 4. Reduction (%) in maximum 8-h ozone between the base M02E02 and M02E04 scenarios for SW flows.

greater reductions in peak ozone values tended to occur at higher ozone levels, a desirable benefit of the control program. Although the standard deviation bars reveal variability in results among the network sites, a similar trend was found at individual sites with differences in the slope apparent across the monitoring network.

4. Summary

Selected modeling results revealed discernable decreases in daily maximum 8-h ozone and in NO_y concentrations downwind of a major point source region, which experienced substantial NO_x emission reductions after implementation of the NO_x SIP Call program. Results from CMAQ modeling scenarios from the summer 2002 period coupled with trajectory analysis are highlighted to show that point source NO_x emission reductions produced notable decreases in NO_x concentrations at considerable distances downwind from an elevated plume release at night. It is evident that a broad region of the eastern U.S. and southeastern Canada benefited by lower maximum 8-h ozone values due to the NO_x control program. Additional model simulations currently being pursued will

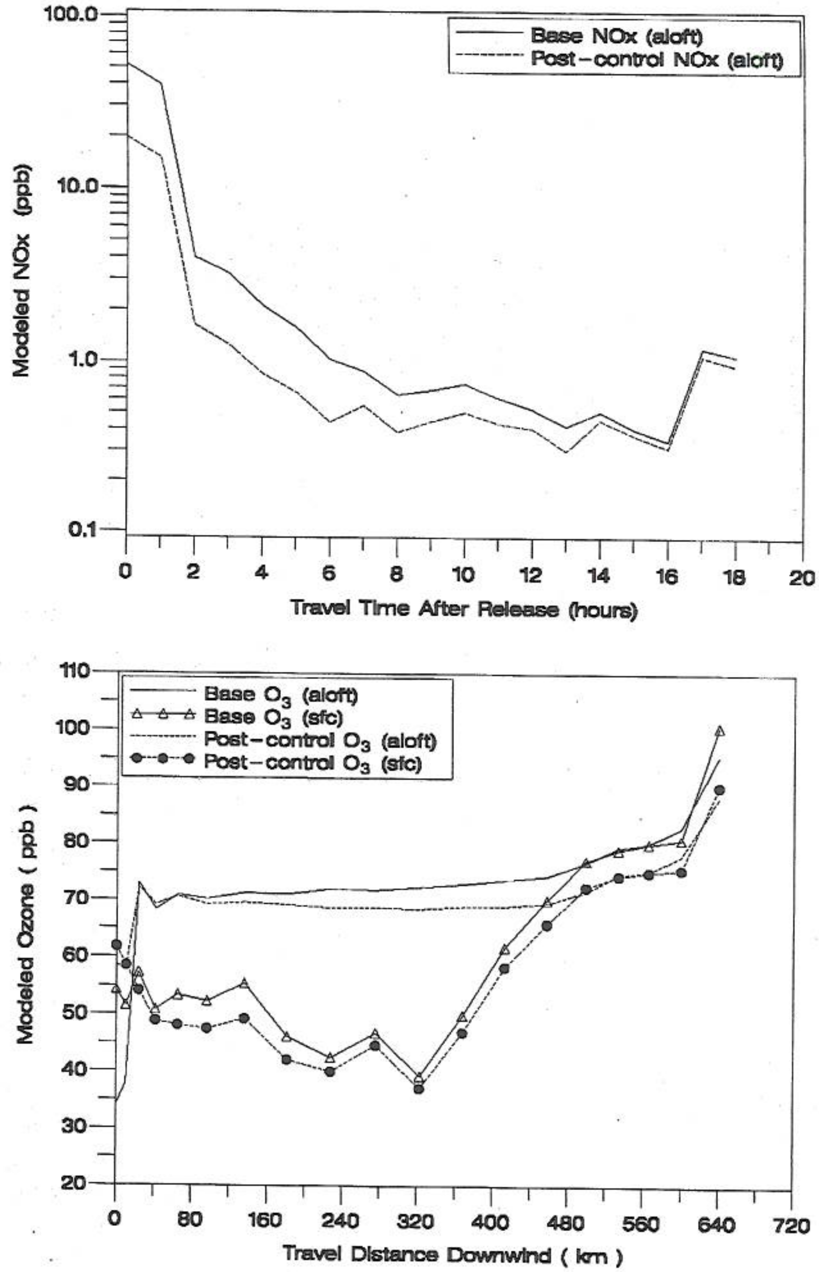

Figure 5. Modeled concentrations from the M02E02 and M02E04 scenarios for NO_x (left) and ozone aloft and at the surface (right) along a trajectory initiated from an elevated point source emission release at 0200 UTC on 11 June 2002. Note the different scales for the *X*-axes are time (left) and distance (right).

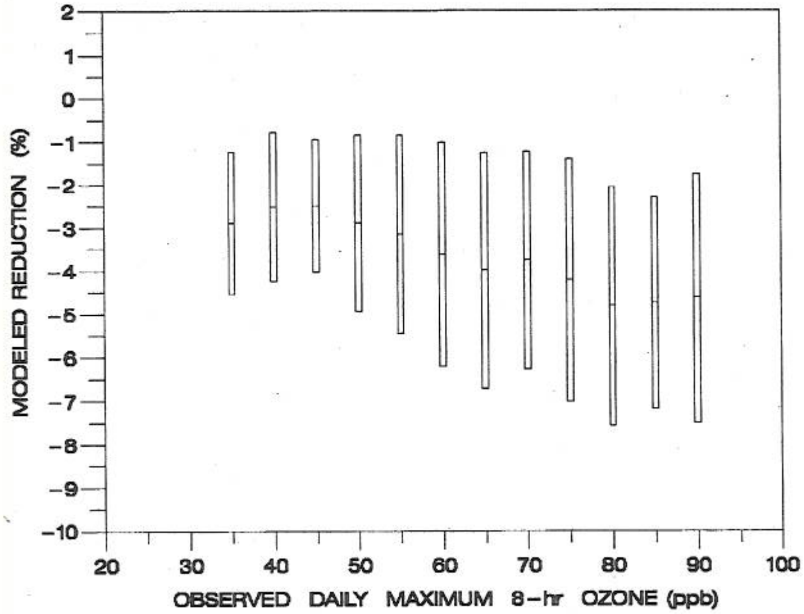

Figure 6. CMAQ modeled reduction in maximum 8-h ozone between the M02E02 and M02E04 scenarios results compared to observed maximum 8-h ozone concentrations from all CASTNet sites during downwind cases from the ORV.

include emission changes in mobile sources in addition to the NO_x point source reductions employed in the M02E04 scenario, as well as boundary conditions from a recent CMAQ model run on a continental domain, which would provide a better estimate of the full impact from a total emission change over the two-year period.

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REFERENCES

Byun, D., Schere, K.L., 2006. Review of the governing equations, computational algorithms, and other components of the Models-3 Community Multiscale Air Quality (CMAQ) modeling system. Appl. Mech. Rev. 59, 51–77.

Environmental Protection Agency, 2005. Evaluating ozone control programs in the eastern United States: Focus on the NO_x budget trading program, EPA454-K-05-001, http://www.epa.gov/airtrends/2005

Gego, E., Porter, P.S., Gilliland, A., Rao, S.T., 2007. Observation-based assessment of the effectiveness of nitrogen-oxide emission reductions on ozone air quality over the eastern United States. In press —J. Appl. Meteorol. Climatol.

Milanchus, M., Rao, S.T., Zurbenko, I.G., 1998. Evaluating the effectiveness of ozone management efforts in the presence of meteorological variability. J. Air Waste Manage. Assoc. 48, 201–215.

Rao, S.T., Zurbenko, I.G., 1994. Detecting and tracking changes in ozone air quality. J. of Air Waste Manage. Assoc. 44, 1089–1092.