TRANSBOUNDARY POLLUTION:
OZONE AND FINE PARTICULATE MATTER IN THE NORTHEAST

Prepared for

NEW YORK STATE ENERGY
RESEARCH AND DEVELOPMENT AUTHORITY

by

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INTRODUCTION

Atmospheric concentrations of ozone (O_3) and fine particulate matter (PM-2.5) in parts of New York State and other regions of the northeastern United States continue to reach or exceed levels set in the National Ambient Air Quality Standards (NAAQS). The challenge to policy makers and state regulatory agencies is designing optimum control measures to reduce ambient concentrations of O_3 and PM-2.5 to levels below the NAAQS. To enhance the scientific basis upon which such measures can be based, NYSERDA has recently supported two major studies, "Analysis of Ozone and Fine Particulate Matter in the Northeastern United States" and "Assessing the Effects of Transboundary Pollution on New York's Air Quality," under its Environmental Monitoring, Evaluation and Protection (EMEP) Program. These studies were directed by Dr. S. T. Rao and conducted by researchers from the University at Albany, the New York State Department of Environmental Conservation, and the Ontario Ministry of the Environment, Toronto. Besides addressing a range of pertinent scientific and policy-relevant questions, the studies fostered collaboration and partnerships among the participating institutions as well as other entities, such as the U.S. Environmental Protection Agency (EPA).

This summary document provides an overview of the two research projects and considers the implications of their findings for regulatory policy making and for future research. Readers interested in specific details of both projects are referred to the complete final project reports (NYSERDA publications 03-02 and 03-04, see **www.nyserda.org/environment/emepreports.html**) and the 28 peer-reviewed articles published during the course of these projects, which are referenced in the final reports.

POLICY IMPLICATIONS

This research has confirmed the regional nature of ozone and fine particulate matter pollution in the northeastern United States and southern Canada, and underscores that no state in the Northeast alone can adequately address pollutant problems until region-wide control strategies for O_3 and PM-2.5 are implemented.

In an effort to assist policy makers and state regulatory agencies in designing optimum control measures to reduce ambient concentrations of O_3 and PM-2.5, this research has proposed methods for integrating both observations and model predictions into an overall framework that draws on the strengths of both approaches while also providing a measure of the uncertainty.

The results have already helped shape recent EPA guidelines for procedures to demonstrate attainment and maintenance of the 8-hour ozone and PM-2.5 National Ambient Air Quality Standards (NAAQS), and the methods described will allow decision makers to assess the probability of attaining the NAAQS under simulated emissions control strategies. This tool is also valuable for comparing emissions control options and their associated costs.

ANALYSIS OF OBSERVED POLLUTANT CONCENTRATIONS

To improve our understanding of the problem of long-range transport of air pollution and source-receptor relationships for trace-level toxic air contaminants, spatiotemporal features of trace metal observations were analyzed through a combination of statistical tools and back-trajectory analysis. Back trajectories, which trace the history of an air mass arriving at a receptor location, have been used successfully to identify potential pollution source regions and atmospheric patterns associated with the long-range transport of trace-level air contaminants. When trajectories are grouped in clusters according to large-scale atmospheric flow patterns, comparisons of pollutant concentrations associated with each cluster can identify the potential source regions of the pollution measured at the station. For example, if a pollution source is south of a monitor, the monitor will likely display higher-than-average concentrations of this pollutant on days for which back trajectories indicate southerly flow. Air masses arriving at receptor sites in the Adirondacks can be clustered into eight groups (Figure 1), and back trajectories show that relatively high concentrations of most anthropogenic trace elements in the Adirondacks are associated with southwesterly winds from the Midwestern United States and northwesterly winds from nearby Ontario (Figure 2).
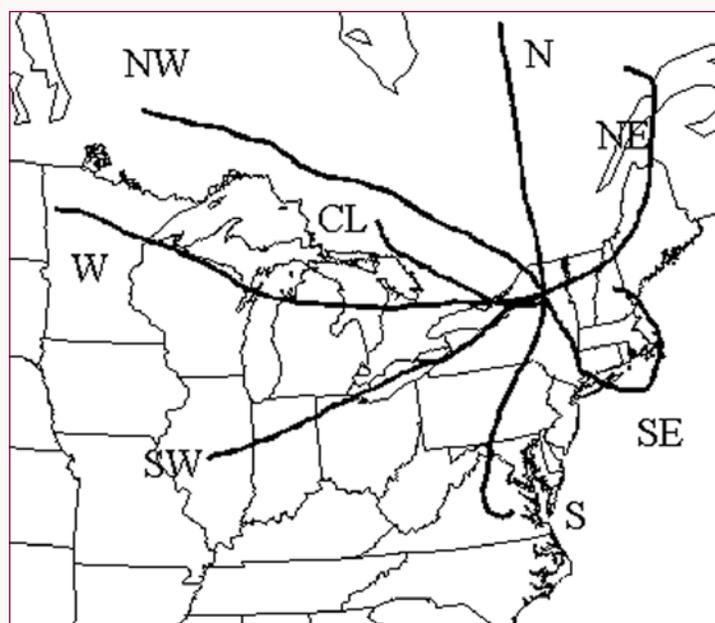

Figure 1: Eight clusters of average back trajectories for receptor sites near Whiteface Mountain, New York, are labeled according to the origin of the air mass: northwest (NW), north (N), northeast (NE), southeast (SE), south (S), southwest (SW), west (W), and close regions toward the northwest (CL). Adapted from the Brankov et al., "Identifying Pollutant Source Regions Using Multiply Censored Data," *Env. Sc. Tech.* 33: 2273-77, 1999.

Although trace metal concentrations are mostly determined by emissions, atmospheric transport, and deposition, concentrations of O_3 and PM-2.5 are determined by a complex interplay of emissions (both anthropogenic and natural), chemical transformations (such as formation of ozone from hydrocarbons and nitrogen oxides in the presence of sunlight), and physical processes (such as atmospheric transport and removal from the atmosphere by wet and dry deposition). Therefore, concentrations measured at an individual monitor usually cannot be unequivocally attributed to one specific source, complicating the task of devising an emissions control strategy aimed at meeting and maintaining the NAAQS. To address this problem, the two research projects employed the concept of an "airshed" for O_3 and PM-2.5 in an effort to identify the spatial scale for these pollution problems and to delineate regions exhibiting similarities in their pollution characteristics. Various analytical techniques, briefly outlined below, were employed to estimate the spatial extent of such airsheds.
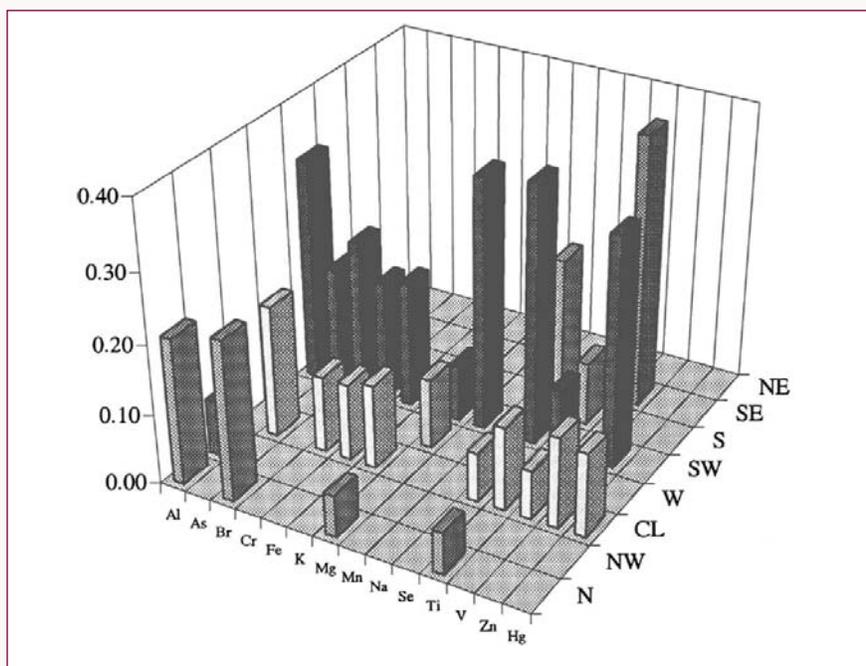

Figure 2: Strength of the positive forcing of several trace elements associated with each of the eight trajectory clusters shown in Figure 1 for sampling sites in the Adirondacks, New York. Most trace elements show elevated levels for the SW and CL back-trajectory clusters. Reproduced from Athanassiadis et al., "Spatial and Temporal Variations in the Trace Elemental Data over the Northeastern United States," *Env. Poll.* 123: 439-49, 2003.

One method for determining the spatial scales for the air pollutants utilizes the correlation coefficients between same-day measurements of a particular species at pairs of stations. The decay of the correlation with distance is approximated by an exponential function, from which the characteristic spatial scale for the pollutant can be inferred. Previous studies show that the characteristic spatial scale for ozone is on the order of 600 km along the direction of a uniform prevailing wind. A similar analysis was performed as part of the two NYSERDA research projects, using both daily average PM-2.5 mass and daily maximum 8-hour ozone concentrations from locations throughout the eastern United States. The results indicate that the spatial extent for the synoptic forcing in these time series is on the order of 500 to 600 km, consistent with the previous studies.

The spatial scale for ozone was further confirmed by assessing the characteristic 1- to 2-day t ransport distance from the weather-induced variations embedded in the observed ozone time series. This time-lagged correlation analysis was performed for Pittsburgh and all other monitors in the Northeast. Each station's time series was lagged with respect to that of Pittsburgh by 0±2 days, and the number of lags needed to maximize the correlation was determined. Results from this analysis suggest that ozone levels in the region from Virginia to Maine can potentially be affected by emissions in the Pittsburgh area within 1 day. Similarly, air masses from as far away as Michigan or the Carolinas have the potential to affect pollutant levels at Pittsburgh.

The back-trajectory clustering methodology discussed above for trace metal analysis was also applied to ozone measurements on top of the World Trade Center in New York City and the Canadian National (CN) Tower in Toronto. For the World Trade Center site, the trajectory clusters associated with the highest ozone concentrations were indicative of atmospheric transport from the Ohio River Valley, local recirculation, transport from the urban east coast, and Canadian air masses transported from southern Ontario. The cleanest air masses were associated with northerly flows. For the CN Tower site, the clusters with the highest ozone concentrations were associated with southerly and westerly flows from the Ohio River Valley and the industrial Midwest as well as locally meandering trajectories. The clusters with the lowest ozone concentrations were associated with northerly and northwesterly flows.

The complexity and regional nature of ozone air pollution were further illustrated through long-term trends in ozone and its precursors (nitrogen dioxide, (NO_2), and total nonmethane organic compounds, TNMOC) for Ontario and the eastern United States, and comparisons of these trends for the Toronto and New York metropolitan areas. NO_2 has been generally decreasing throughout Ontario, but ozone has been generally increasing. Both NO_2 and TNMOC have been decreasing in both the Toronto and the New York metropolitan areas, but ozone is decreasing in the New York metropolitan area and increasing in Toronto. The decreases of precursor concentrations in both Ontario and New York are consistent with the emissions control programs implemented in both areas. The decrease of ozone concentrations in New York and the increase throughout Ontario may actually result from the same control programs because of the nonlinear relationships between ozone and its precursors. However, Ontario is surrounded by areas with upward ozone trends, whereas New York City is in a region of downward trends, and this difference may in part explain the ozone trends.

In summary, all the observation-based analyses performed as part of these NYSERDA-funded research projects consistently corroborate the notion that O_3 and PM-2.5 pollution is a regional, multistate, and even international problem. As higher spatial and temporal resolution and better speciation on PM-2.5 become available, this analysis should be repeated to investigate whether different spatial scales exist for different components of PM-2.5.

## APPLICATION AND EVALUATION OF NUMERICAL MODELS

Although analysis of observed pollutant concentrations as described above formed the basis of the two projects, researchers also used computer models designed to simulate the complex interplay between meteorology and atmospheric chemistry. These models are based on the numerical representation of our current understanding of the physical and chemical processes affecting pollutant concentrations and can be used to predict the spatial and temporal distribution of pollutant concentrations. In one of the applications of such models during the research projects, the "atmospheric region of influence" for anthropogenic emissions from two source regions—Ontario and New York State—was examined by performing model simulations in which emissions from these regions had been removed.

In the Ontario emissions reduction case, peak ozone improvements of 15% or greater are evident in the near-field (Figure 3a). The emissions reductions in Ontario also led to peak ozone concentration decreases in excess of 6% throughout most of New York State. The situation was similar in the New York emissions reductions case (Figure 3b), where ozone improvements within New York ranged from 3% to 15%. Even in southern Ontario, the ozone decreased as much as 6%. These percentage reductions are seasonal averages; they are not representative of the ozone changes on any particular day.

Such modeling simulations reaffirmed the results of the analysis of observational data presented in Section 2—that the O_3 and PM-2.5 problem crosses state and international boundaries. This finding implies that one cannot determine the out-of-state versus in-state contributions of reactive pollutants like O_3 and PM-2.5 from just a few monitoring sites because of the pollutant interdependence among several states within the 600 km airshed. For example, emissions from Baltimore and Washington, D.C., have the potential to affect Philadelphia, which in turn can affect pollution levels downwind throughout New York State and New England. Further, the results imply that no state in the Northeast alone can adequately address pollutant problems until region-wide control strategies for O_3 and PM-2.5 are implemented.

Researchers supported by the two projects also worked on improving the ability of atmospheric models to simulate crucial physical processes. Specifically, the model improvement efforts were directed at the simulation of land-atmosphere interactions and at ensuring a consistent treatment of vertical mixing processes through all model components.

a)

New York Control Case

b)

Ontario Control Case
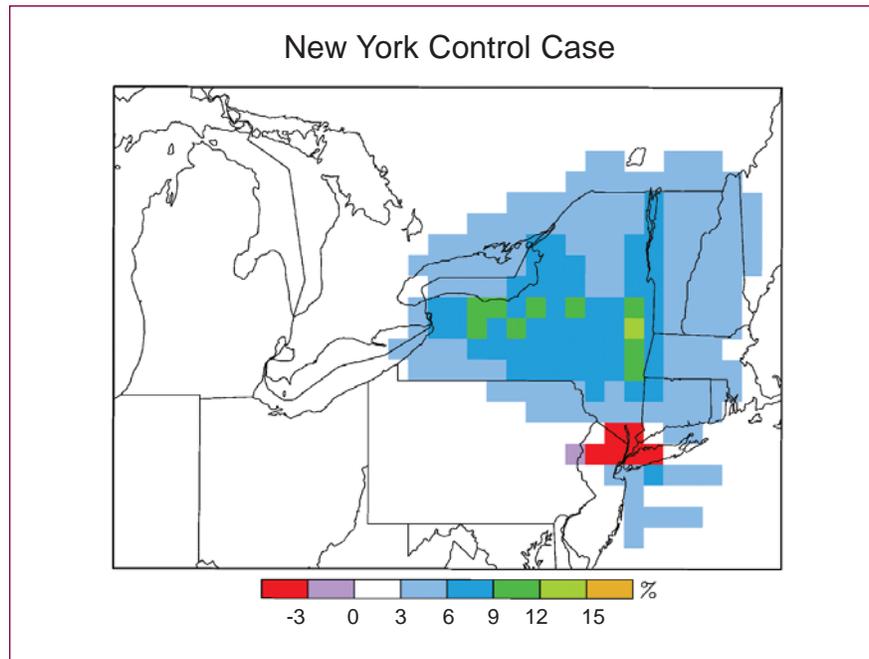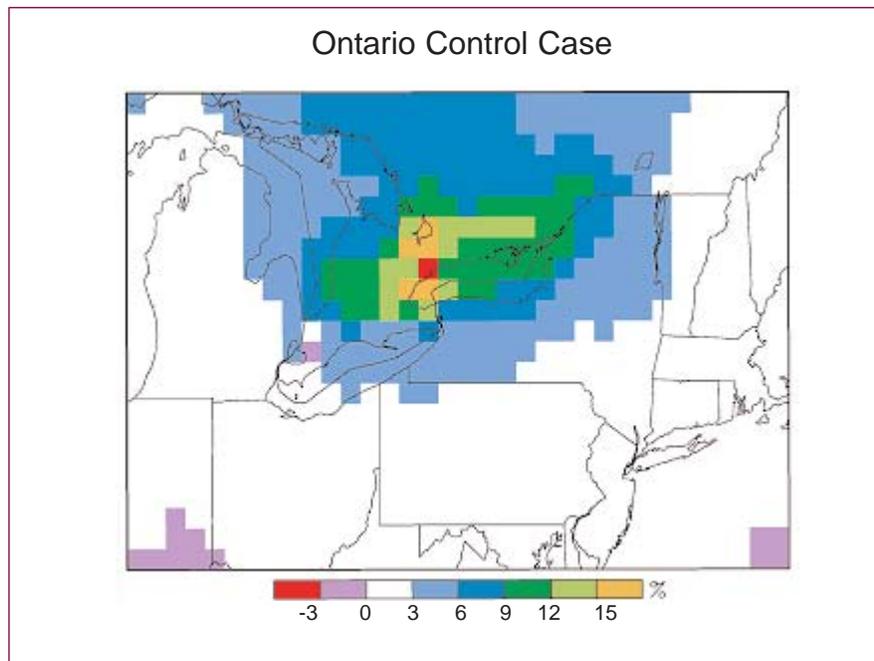
Figures 3a-b: Model-predicted percentage reduction in summertime average daily maximum 8-hour ozone concentrations resulting from the elimination of all anthropogenic emissions in a) New York State and b) Ontario. These percentage reductions are seasonal averages; the percentage reduction at a grid cell on any one day may be significantly larger.

Coupled meteorological and photochemical modeling systems are being used to design emissions control strategies aimed at meeting and maintaining the NAAQS. Meaningful and thorough evaluations of these meteorological and air quality modeling systems are therefore critical. Researchers working on the two NYSERDA-funded projects have performed some of the most comprehensive model evaluations published in the scientific literature; references to these studies are listed in the final reports of both projects. One of the most important findings was that current-generation modeling systems perform best at predicting ozone fluctuations on time scales longer than one day. In addition, the uncertainty in predicting the daily peak ozone concentration is on the order of 20% for the models studied.

The analysis of model simulations summarized above has two significant policy implications. First, the results confirm the conclusion derived from the analysis of observational data—that O_3 and PM-2.5 pollution in the northeastern United States is a regional problem which requires mitigation collaboration among local, state, and federal agencies. Second, the findings essentially invalidate the previous regulatory practice of basing emissions management decisions on a small number of model predictions of the daily maximum concentrations for individual days. Accordingly, EPA recently revised its guidance on using modeling results for regulatory purposes and now calls for averaging the predicted daily maximum ozone concentrations over all days simulated, as opposed to using the peak predicted ozone concentration during an episode.

COMBINED USE OF OBSERVATIONS AND MODEL OUTPUTS IN THE REGULATORY SETTING

What are the implications of the findings discussed above for meeting and maintaining the NAAQS? Clearly, information to enhance our understanding of the air pollution problem and improve regulatory practices for its mitigation can be gleaned from both analysis of observed variables and the application of computer models. The two approaches should be viewed as complementary: observations present "ground truth" but are limited in space and time, and model predictions provide detailed information in space and time and can simulate future emissions scenarios, but also suffer from uncertainties in our scientific understanding and its implementation in numerical models and from limitations of model input. Thus, both projects proposed methods for integrating the observations and model predictions into an overall framework that would draw on the strengths of both approaches while also providing a measure of the uncertainty associated with the final answer.

Because the modeling uncertainties in predicting the daily maximum ozone concentrations on individual days are on the order of 20%, the use of the model results in an absolute sense—as done in the previous attainment demonstrations in the state implementation plan (SIP) process—is questionable. Taking into account findings from the NYSERDA-supported research as well as its own analysis, EPA revised its guidance to call for using model results in a relative sense in upcoming attainment demonstrations for the 8-hour O_3 and the PM-2.5 NAAQS. According to these draft EPA guidelines, model predictions of simulation-averaged daily ozone maxima are to be derived from two air quality model runs: one with present-day emission (base case) and one with emissions reflecting a future emissions control policy (control case). The relative reduction factor (RRF) at a given location is defined as the ratio of the mean of the daily maximum ozone concentrations during the simulation period for the emissions control case and the base case. These RRF values are then used to scale the observed design values to determine future attainment of the NAAQS at each monitoring location.

This new approach does take into account the better model performance on longer time scales since the model-predictions are averaged over all simulation days, and it also combines the strength of observations (using the observed design value to reflect ambient conditions) with the strength of models (to predict the response of the atmosphere to a reduction in pollutant emissions). However, this approach yields information about future attainment of the NAAQS only in a pass-fail mode (it gives policy makers no confidence estimates about the efficacy of a control strategy), and information is limited to the location of existing monitors (it disregards the wealth of spatial information available from model predictions). In other words, although the researchers of the NYSERDA-funded projects believe that the new EPA guidance is a step in the right direction because it takes into account their findings about models' performance, new methodologies would further improve the new approach.

In one effort, researchers used a combination of extreme value statistics and bootstrap resampling techniques to estimate the temporal uncertainty of predicting future-year design values. Their method led to the creation of probability density functions, which would allow decision makers to assess the probability of attaining the NAAQS under the simulated emissions control strategies. This tool would be especially valuable for comparing emissions control options and their associated costs. For example, although two emissions control options with widely different costs might yield different attainment answers when considered in the pass-fail mode, their attainment probabilities might differ by only a few percentage points—information that could be of interest in selecting between the two options.

Building upon that analysis, a subsequent study supported by NYSERDA developed methodologies to create spatial maps of current and future design values and NAAQS attainment probabilities. Because observations made at nearby locations are more correlated than those far apart, two statistical techniques were used to estimate spatial maps and the uncertainty associated with their creation from the original monitoring data. When combined with the RRF methodology outlined above, both techniques can then be used to generate maps of the probability of exceeding the NAAQS.

To illustrate the usefulness of the mapping approach to decision makers., Figures 4a-b display the 8-hour ozone NAAQS exceedance probabilities over the northeastern United States for both the 1998-2000 base period and the 2007 SIP call scenario for emissions of nitrogen oxides (NO_x). In the control case simulations, emissions of NO_x and volatile organic compounds (VOC) were reduced by an average of 16% to 21% across all anthropogenic sources and all states, relative to the base case.

a)
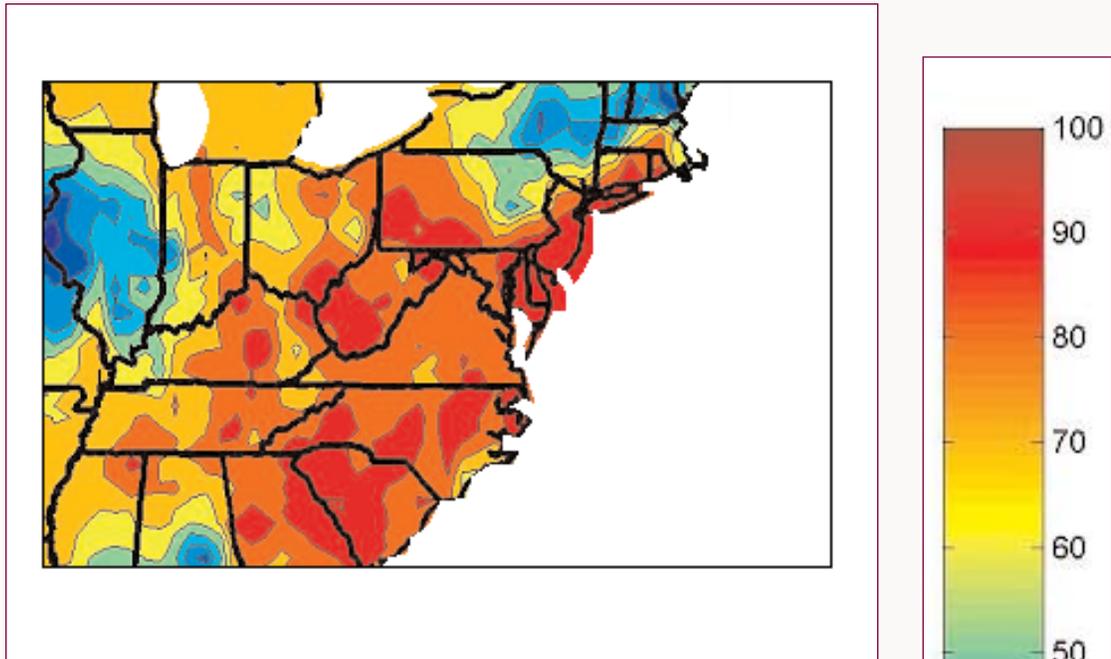
b)
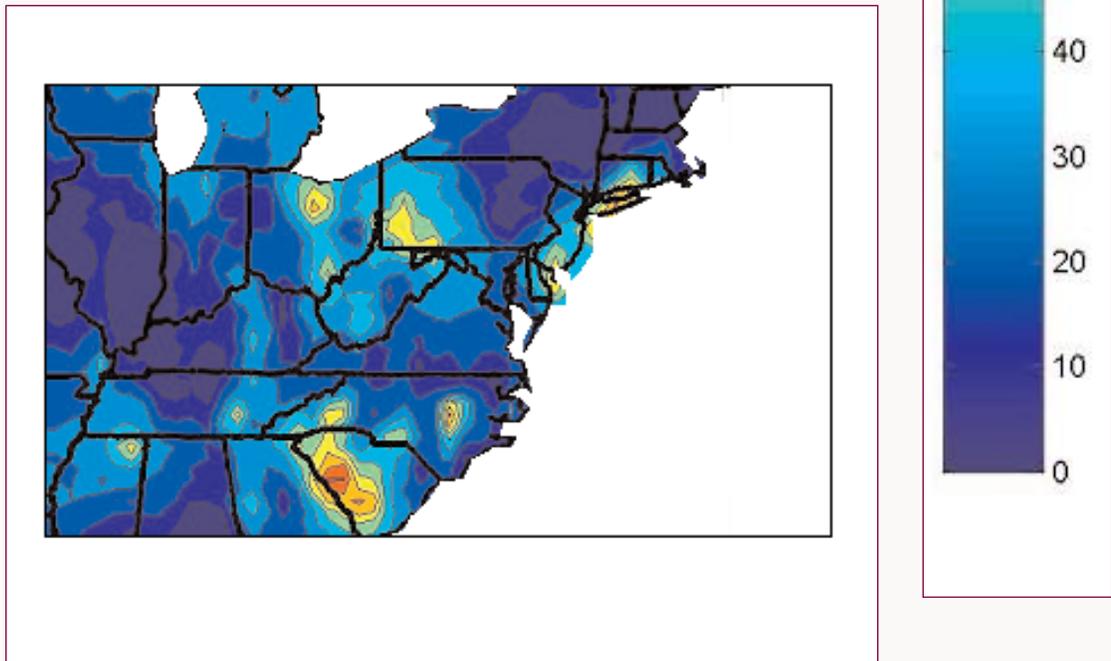
Figures 4a-b: The combined use of observed design values, geostatistical simulations, and model-predicted responses to an emissions control scenario to estimate the probability of exceeding the 8-hour ozone NAAQS. Exceedance probabilities (in percent) are shown for a) the 1998-2000 base period and b) the emissions control case. Adapted from Gego et al., "Probabilistic Assessment of Regional Scale Ozone Pollution in the Eastern United States", in *NATO Science Series IV: Earth and Environmental Sciences, Vol. 30, Air Pollution Processes in Regional Scale*, D. Melas and D. Syrakov (eds.), Kluwer Academic Publishers, Dordrecht, 2003.

The base period exceedance probabilities for New York State (Figure 4a) range from less than 20% in central upstate New York to more than 75% near Buffalo and more than 90% on Long Island. In contrast, the NAAQS exceedance probabilities associated with the NO_x SIP call emissions control strategy are less than 20% for most portions of New York State but about 75% on Long Island. Using this information, policy makers can conclude that the NO_x SIP call emissions control strategy would considerably boost New York State's ability to meet the 8-hour ozone NAAQS, even though additional measures would be necessary to further reduce the exceedance probabilities in the remaining hotspots. By extension, these proposed techniques would be of particular interest to other states that are preparing SIPs for the 8-hour O_3 and PM-2.5 standards, and they also provide a scientific basis for utilizing both observations and model predictions of concentrations in a probabilistic framework.

SUMMARY

Through the extensive analysis of observation and model predictions, the two studies supported by NYSERDA have confirmed the regional nature of ozone and fine particulate matter pollution in the northeastern United States and southern Canada. The findings underscore that no state in the Northeast alone can adequately address pollutant problems until region-wide control strategies are implemented for O_3 and PM-2.5. As part of the NYSERDA-funded projects, partnerships between academic institutions and state and federal agencies have been built, and the results from the research projects have helped shape recent EPA guidelines for procedures to demonstrate attainment and maintenance of the 8-hour O_3 and PM-2.5 NAAQS. A more comprehensive analysis of PM-2.5 was hindered by the temporal and spatial sparsity of observational data; however, more data have become available in recent years, as have models that simulate the interactions among multiple pollutants over multiple scales. As still more information becomes available, including speciated PM-2.5 data, observational and modeling analyses should be extended accordingly. Finally, the probabilistic framework proposed in these studies—aimed at integrating the spatiotemporal information of observations and model predictions and applied to ozone concentrations for demonstration purposes—should be expanded to address multipollutant problems within the "one atmosphere" approach.

EMEP Program Overview

NYSERDA's Environmental Monitoring, Evaluation and Protection (EMEP) Program is funded through the System Benefits Charge (SBC) under the **New York Energy \$mart**[SM] Program. The primary mission of EMEP is to support research to address environmental issues related to the generation of electricity. Since its inception in 1998, the EMEP Program has provided objective and policy-relevant research to:

- Improve the scientific understanding of electricity-related pollutants in the environment;

- Assess the environmental impact of electricity generation relative to other sources of pollution;

- Help develop approaches to mitigate impacts of electricity generation and improve environmental quality.

EMEP has also supported development of advanced environmental instrumentation.

The EMEP Program currently supports research in four critical regional environmental issues related to electricity generation: ozone, fine particles, acid deposition, and mercury. Program Opportunity Notices (PONs) are issued periodically to seek proposals which address targeted research areas. Projects are reviewed and selected through this competitive process. The program is guided by a steering committee comprised of representatives from the New York State Departments of Environmental Conservation (DEC), Health (DOH), and Public Service (DPS); the U.S. Environmental Protection Agency (U.S. EPA); the National Oceanic and Atmospheric Administration (NOAA); a utility association; and three environmental/public interest groups. Also, a science advisory committee provides program support and periodic review in critical disciplines.

Under EMEP, NYSERDA sponsors conferences and workshops for policy-makers and scientists to share information. They cover a wide range of topics, from asthma in New York City to mercury in remote regions of the Adirondacks. NYSERDA also plans to commission papers to "translate" scientific results into a form useful for policy-makers. As research reports become available, NYSERDA and its research partners will post information on-line (see **www.nyserda.org**). Program Opportunity Notices and information about ongoing projects may also be found on the website.