

**ASSESSING THE EFFECTS OF TRANSBOUNDARY
POLLUTION ON NEW YORK'S AIR QUALITY**

**FINAL REPORT 03-02
JANUARY 2003**

**NEW YORK STATE
ENERGY RESEARCH AND
DEVELOPMENT AUTHORITY**



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Prepared for the
**NEW YORK STATE
ENERGY RESEARCH AND
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Albany, NY
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PREFACE

The New York State Energy Research and Development Authority (NYSERDA) is pleased to publish, “Assessing the Effects of Transboundary Pollution on New York’s Air Quality.” The report was prepared by the New York State Department of Environmental Conservation.

The Principal Investigator was Dr. S.T. Rao, during his tenure at the New York State Department of Environmental Conservation. Dr. Rao is now the Director of the Atmospheric Modeling Division of the U.S. Environmental Protection Agency and has continued his research in air quality modeling and policy analysis.

As part of this project, a Memorandum of Understanding (MOU) was developed between the New York State Department of Environmental Conservation and the Ontario Ministry of the Environment to facilitate the exchange of scientific information between the State of New York and the Canadian Province of Ontario, and to develop approaches to deal with the problems relating to transboundary pollution.

This project was funded as part of the New York Energy SmartSM Environmental Monitoring, Evaluation, and Protection Program and represents one of several air quality modeling studies underway in New York.

Key Words: Transboundary pollution, ozone trends, air quality modeling, air quality data analysis, environmental policy-making

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NYSERDA appreciates the input of project reviewers, especially Dr. Praveen Amar, Director, Science and Policy, NESCAUM, and Dr. Daniel Jacob, Professor of Atmospheric Chemistry & Environmental Engineering, Division of Engineering and Applied Science, Harvard University.

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EXECUTIVE SUMMARY

Ozone pollution continues to be a major problem downwind of many metropolitan areas throughout the United States. Despite three decades of efforts to control this photochemically-produced pollutant, and despite some promising trends in ozone levels, many areas are still in nonattainment, especially from the perspective of the 8-hour ozone standard. While the inherent nonlinearities of ozone photochemistry make it one of the most difficult air pollutants to understand, the long-range transport of ozone and its precursors further complicate the picture. The goals of this project were to better understand the transport of ozone in the region covering the Canadian Province of Ontario and New York State, and how transboundary pollution would be impacted by different emission control strategies.

Project Background

Many atmospheric pollutants are transported over long distances, affecting air quality on the regional scale. For example, despite substantial reductions in the emissions of Volatile Organic Compounds (VOCs) and Nitrogen Oxides (NO_x) over New York State, long-range transport of ozone and its precursors from outside of the state continue to contribute substantially to air quality problems within the State. The transboundary pollution problem may be examined in two ways: using observations and models. Using observations, trends and spatial scales may be examined, which can be combined with atmospheric flow climatology to quantitatively assess regional effects. Trajectories define a potential region of influence; observations of pollutant concentrations can help delineate the trends and possible effects of changes in emission control policies. To estimate quantitatively the effects of atmospheric flow patterns and emissions, one needs meteorological and photochemical modeling as well. Air quality models can help us understand the ozone formation, transport and mixing processes.

The objectives of this project were as follows:

1. Assemble and analyze pertinent meteorological and air quality databases to determine trends in meteorologically-adjusted ozone and its precursors over the region covering the Canadian Province of Ontario and New York State, and examine the impact of implemented emission controls in reducing the effects of transboundary pollution on New York.

2. Apply state-of-the-art, regional-scale air quality modeling systems, namely, the MODELS3/CMAQ system - a 3rd generation regional model developed by EPA, and the UAM-V that was used by NYSDEC and EPA in the OTAG (Ozone Transport Assessment Group) process. An updated emission inventory was used to more accurately reflect the emissions from Ontario and the eastern United States. The ability of these modeling systems to simulate the observed ozone air quality for the base year of 1995 was evaluated on a seasonal basis.

3. Assess the efficacy of selected NO_x and VOC emission reductions, as predicted by the two air-quality models in reducing the effects of transboundary pollution, and develop methods for using the ozone modeling results in a regulatory setting with an increased degree of confidence. It should be noted that EPA considers the two models (CMAQ and UAM-V) as acceptable tools to address the new 8-hour ozone standard.

Results

While simple questions can be framed about the processes affecting ozone air quality, not all such questions have simple answers.

1. What can we learn from an examination of the pertinent meteorological and air quality databases?
 - Ozone precursors have been generally decreasing throughout Ontario and the New York metropolitan area, while ozone itself is decreasing in the New York metropolitan area and increasing throughout Ontario. A back-trajectory clustering technique illustrates that higher than average ozone concentrations in the Northeast are associated with winds from the Ohio River Valley and the industrial Midwest.

2. What is the uncertainty associated with the model-predicted ozone concentrations from the current state-of-the-art photochemical modeling systems?

- Modeled ozone concentrations from the current generation, regional-scale photochemical modeling systems have large uncertainties, stemming from meteorological, emissions, and other model input data. For example, different meteorological drivers applied to one photochemical model can produce ~20% uncertainty in the predicted peak O₃ concentrations. Also, different photochemical modeling systems driven with the same emissions can produce differences on the order of ~20 to 30% in the predicted peak O₃ concentrations. Analysis of meteorological and photochemical model outputs reveals that models are unable to simulate the intra-day (timescale of <10 hours) variability properly, yielding ~10% as the lowest bound for the modeling uncertainty even when the model and its input are perfect.

3. How should the modeling results be used in the regulatory setting?

- In light of the inherent uncertainties associated with episodic-type modeling with the grid-based photochemical models, model simulation periods need to cover longer time periods than just 2-3 episodic days. Further, it is important to consider averaging the model-predicted ozone concentrations over all simulation days, rather than predictions of peak ozone levels on individual days, for greater confidence in the use of models for regulatory purposes. In addition, model predictions need to be used in the probabilistic form, rather than in the deterministic form, in evaluating whether a selected emission control strategy could lead to compliance with the relevant air quality standards.

ASSESSING THE EFFECTS OF TRANSBOUNDARY POLLUTION ON NEW YORK'S AIR QUALITY

Section 1

INTRODUCTION

Many atmospheric pollutants are transported over long distances, affecting air quality on the regional scale. For example, despite substantial reductions in the emissions of Volatile Organic Compounds (VOCs) and Nitrogen Oxides (NO_x) over New York State, long-range transport of ozone and its precursors from outside of the state continue to contribute substantially to air quality problems within the state. We can examine the transboundary pollution problem in two ways: using observations and using models. Using observations, we can examine trends and spatial scales, which can be combined with atmospheric flow climatology to quantitatively assess regional effects. Trajectories define a potential region of influence; measurements can show trends and the possible effects of changes in emission control policies. To estimate quantitatively the effects of atmospheric flow patterns and emissions, one needs meteorological and photochemical modeling as well. Air quality models can help us understand the ozone formation, transport and mixing processes.

The goals of this project are to better understand the transport of ozone in the region covering the Canadian Province of Ontario and New York State, and how transboundary pollution would be impacted by different emission control strategies. The project objectives were as follows:

1. Assemble and analyze pertinent meteorological and air quality databases to determine trends in meteorologically-adjusted ozone and its precursors over the region covering the Canadian Province of Ontario and New York State, and examine the impact of implemented emission controls in reducing the effects of transboundary pollution on New York.
2. Apply state-of-the-art, regional-scale air quality modeling systems, namely, the MODELS3 system - a 3rd generation regional model developed by EPA, and the UAM-V that was used by NYSDEC in the OTAG (Ozone Transport Assessment Group) process. In this project, we use an updated emission inventory that accurately reflects emissions from Ontario and the Eastern US, and evaluate the ability of these modeling systems in simulating the observed ozone air quality for the base year of 1995 on a seasonal basis.
3. Assess the efficacy of selected NO_x and VOC emission reductions, as predicted by the two models (MODELS3, UAM-V) in reducing the effects of transboundary pollution, and develop methods for using the ozone modeling results in a regulatory setting with an increased degree of confidence. It should be noted that EPA considers the two models (MODELS3 and UAM-V) as acceptable tools to address the new 8-hour ozone standard.

We refer generally to the first objective as ‘data analyses’, the second as ‘model evaluation’, and the third as ‘efficacy and methods’ (modeling results).

With regard to Data Analyses, we examined long-term trends in ozone (O₃) and its precursors (NO₂ and total nonmethane organic carbon) for Ontario and the eastern US, and compared these for the Toronto and New York City metropolitan areas. NO₂ is generally decreasing throughout Ontario, while ozone is generally increasing. NO₂ and total nonmethane organic carbon (TNMOC) are decreasing in both the Toronto and New York City metropolitan areas while ozone is decreasing in the New York City metropolitan area and increasing in Toronto. The decreases of precursor concentrations in both Ontario and New York are consistent with the emission control programs implemented in both of these areas. The decrease of ozone concentrations in New York and the increase throughout

Ontario may both actually be the effects of these same control programs, owing to the nonlinear relationships which exist between ozone and its precursors. However, Ontario is surrounded by areas with upward ozone trends while the New York City metropolitan area is imbedded in a region of downward trends, and this in part may explain the difference between the ozone trends observed over the two cities.

With regard to Model Evaluation, we performed numerous analyses including: 1) developing a new model performance evaluation methodology, and using it to evaluate both the air quality models and their meteorological drivers (Hogrefe et al., 2001a and b; Biswas et al., 2001a), 2) examining the sensitivity of photochemical models to the meteorological input fields (Biswas et al., 2000, Biswas and Rao, 2001b), and 3) examining in detail model performance at rural monitors (Sistla et al., 2001a and b).

The new model evaluation methodology entails spectrally decomposing the time series of model output, and comparing each spectral component with the corresponding component of the observational data. For both the photochemical model and its meteorological driver(s), we find that the model performance is best on the longest (synoptic and longer forcings) time scales, and worst on the shortest (intraday and diurnal forcings) time scales. We have examined the sensitivity of the UAM-V to its meteorological input by performing the same simulation with two different meteorological drivers, RAMS3b and MM5; the difference in the UAM-V predicted ozone concentrations may be as high as 20%.

With regard to the third objective, Efficacy and Methods, we performed the following: In order to ascertain the spatial extent of the area(s) in which transboundary transport plays a role, we performed three-dimensional modeling with all anthropogenic emissions removed from the grid cells surrounding various urban centers. The spatial extent of the airshed for ozone is found to be on the order of 500 km, suggesting that transboundary transport will affect areas within 500 km of major urban centers (Civerolo et al., 2002).

We also developed a number of methodologies for utilizing the results of modeling in the

regulatory setting. Briefly, these include 1) averaging the model results over all days simulated (Rao et al., 2000), 2) simulating longer periods of time (Rao et al., 2000), and 3) using model predictions in the *relative* rather than *absolute* sense in designing emission control programs (USEPA, 1999). The last of these is not independent of the first two; in fact, in order to use the model predictions in the relative sense, they must be averaged over a time period sufficiently long to make the so-called Relative Reduction Factors (RRFs) stable. Using extreme value statistics and resampling techniques, we developed a method by which the probability of exceeding the NAAQS (National Ambient Air Quality Standard) may be estimated to build confidence in the use of models in the regulatory framework (Hogrefe and Rao, 2001, Rao and Hogrefe, 2001).

Numerous articles have been published in peer-reviewed literature based on the work performed under this contract. A list of these papers is presented in Appendix A.

List of Acronyms

AIRS	Aerometric Information and Retrieval System
CASTNet	Clean Air Status Trends Network
GPMP	Gaseous Pollutant Monitoring Program
HY-SPLIT	Hybrid Single Particle Lagrangian Integrated Trajectories
KZ filter	Kolmogorov-Zurbenko filter
MAQSIP	Multiscale Air Quality Simulation Platform
MCP	Multiple Comparison Procedure
MM5	Mesoscale Model Version 5
NAAQS	National Ambient Air Quality Standard
NAMS	National Aerometric Monitoring System
NGM	Nested Grid Model
OTAG	Ozone Transport Assessment Group
PAMS	Photochemical Assessment Monitoring Stations
RAMS	Regional Atmospheric Model System
RRF	Relative Reduction Factor
SAQM	San Joaquin Valley Air Quality Model
SIP	State Implementation Plan
SLAMS	State and Local Air Monitoring System
SMRAQ	Seasonal Model for Regional Air Quality
TNMOC	Total NonMethane Organic Carbon
UAM-V	Urban Airshed Model with Variable Grid
VOC	Volatile Organic Compound

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Section 2

OBJECTIVE 1: DATA ANALYSES

Assemble and analyze pertinent meteorological and air quality databases to determine trends in meteorologically-adjusted ozone and its precursors over the region covering the Canadian Province of Ontario and New York State and examine the impact of emission controls implemented in reducing the effects of transboundary pollution on New York.

We examined the long-term averages and trends in ozone, nitrogen oxides and hydrocarbons in the province of Ontario and the eastern United States, and performed a more detailed comparison of air pollution in the two major urban areas of Toronto and New York City. We used a back-trajectory clustering methodology to illustrate that higher than average ozone concentrations are associated with particular wind regimes. We attempted to analyze the data from as many different perspectives as possible.

DATA

NO₂ and ozone data for the province of Ontario were supplied by the Ontario Ministry of the Environment. Of the 55 NO₂ monitors and 74 ozone monitors in Ontario for which we have data, 44 are classified as “ambient” monitors. Of these, 28 which have collocated NO₂ and ozone instruments were chosen for further study, the selection being based on the length of time covered by the data. For the time period 1983 through 1997, all of these sites have at least 10 years of NO₂ and ozone data except Kitchener (7 years NO₂, 8 years O₃) and Hamilton (9 years NO₂, 13 years O₃). The monitoring sites chosen and their addresses are

listed in Table 1. Figure 1 displays the locations of these monitors; the inset shows the monitors along the western side of Lake Ontario, including the Toronto metropolitan area which runs from Mississauga north to Scarborough.

Speciated hydrocarbon data (24-hour canister samples) for Ontario were supplied by Environment Canada. These data consist of up to 164 different compounds. Also, hourly ozone and NO₂ data for the United States were obtained from the US Environmental Protection Agency's AIRS (Aerometric Information and Retrieval System). These data comprise the SLAMS (State and Local Air Monitoring System) and NAMS (National Aerometric Monitoring System) monitoring programs.

Total NonMethane Organic Carbon (TNMOC) data, collected by Radian Corporation under contract to the EPA, were also available for several sites in the New York City Metropolitan Area; these are 6 to 9 a.m. canister samples. In order to facilitate comparison with these TNMOC data, the speciated data from Ontario were summed. Because of potential differences between 6 to 9 a.m. and 24-hour averages, we confine our interest to trends in TNMOC concentrations in this study. Speciated hydrocarbon data are available from only one site in the New York City Metropolitan Area for a reasonably long period of time (Bronx Botanical Garden); these data have not been used in this study.

ANALYSIS METHODS

Daily averages represent the arithmetic average of the 24 hourly average concentrations which constitute the record of raw data for each day. The natural logarithm is applied to each daily average value to compute the "log-transformed" time series which is subsequently used as input for the KZ (Kolmogorov-Zurbenko) filter (Rao and Zurbenko, 1994). The KZ filter is a low-pass filter and is simply several repetitions of a moving average: the number of repetitions and window length of the moving average are chosen to select the cut-off frequency for the filter. Since we are interested here in the long-term behavior of the NO₂ and ozone time series, we have used three repetitions of a 365 day moving average.

Table 1. Locations of monitoring sites and trends in NO₂ and ozone in Ontario

ID	CITY	ADDRESS	NO ₂ trend %/yr	O ₃ trend %/yr
12008	Windsor	467 University Av W	-0.46±0.03	0.13±0.05
14064	Sarnia	Centennial Pk Front St/Cn Tracks	-0.96±0.06	-0.12±0.03
15001	London	Wrn Fair Grounds King/Rectory	-1.40±0.07	0.57±0.04
15013	Parkhill	Puc Building	-1.00±0.05	0.47±0.02
22071	Simcoe	Experimental Farm	-0.24±0.07	-0.14±0.02
22901	Long Point	Provincial Park	-0.75±0.07	-0.05±0.03
26060	Kitchener	West Ave/Homewood	-0.33±0.09	-0.67±0.06
27067	St Catherines	Argyle Crescent	-2.54±0.06	-1.15±0.05
29000	Hamilton	Elgin/Kelly	-2.15±0.05	0.52±0.04
29114	Hamilton	Vickers Rd/East 8 TH St.	-0.68±0.06	1.15±0.03
29118	Hamilton	Main St/W Hwy 403	0.55±0.04	1.36±0.03
31190	Toronto	CN Tower	-1.10±0.10	4.29±0.07
33003	Scarboro	Lawrence/Kennedy	1.25±0.06	1.16±0.03
34020	North York	Hendon Ave (Yonge St/Finch Ave)	-2.03±0.08	0.85±0.04
35003	Etobicoke	Elmcrest Rd (Centennial Pk)	0.55±0.03	0.31±0.02
35033	Etobicoke	Evans/Arnold Av.	-0.69±0.03	1.69±0.02
36030	York	Clearview Heights	-1.30±0.06	1.21±0.04
44008	Burlington	Hwy2/North Shore Blvd E.	-3.39±0.07	1.01±0.03
44015	Oakville	Bronte Rd/Woburn Cres.	0.57±0.03	0.82 ±0.03
45025	Oshawa	PS Ritson Rd/Olive Av	-0.88±0.04	1.57±0.04
46110	Mississauga	Queensway	-1.89±0.04	1.24±0.03
48002	Stouffville	Hwy47/E of Hwy48	-1.80±0.10	1.46±0.05
49010	Dorset	Hwy117/Paint Lake Road	-1.40±0.03	1.00±0.03
51001	Ottawa	MCD GDS Rideau/ Wurtenburg St.	-0.14±0.07	0.63±0.04
56051	Cornwall	Memorial Pk Bedford/Third Sts.	-1.82±0.02	-1.07±0.04
63200	Thunder Bay	MTC 615 James St S	-1.19±0.04	2.37±0.06
71068	Sault Ste Marie	Wm. Merrifield School	-3.10±0.03	0.21±0.02
77203	Sudbury	Science North	-2.13±0.07	0.35±0.03

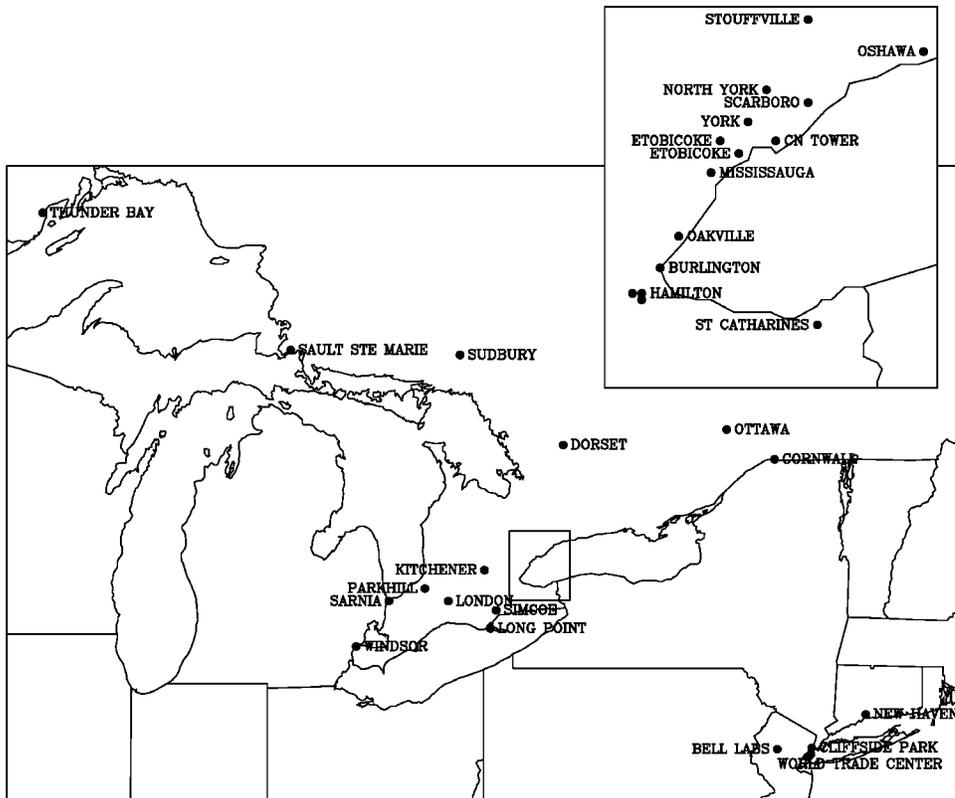


Figure 1. Locations of monitoring sites

This will remove the annual cycle and fluctuations with periods less than approximately 630 days, passing what is here referred to as the “long-term component” (Porter et al., 2001).

The long-term average is formed by averaging the long-term component; the long-term trend is determined as the slope of the least squares linear fit to the long-term component. The KZ filter provides a very good visualization of the long-term behavior of the time series without destroying the information on important interannual variations, i.e., El Nino and other intra-decadal variations. We have computed long-term trends and averages for NO₂ and ozone.

To compare the effects of the synoptic-scale atmospheric transport patterns on ozone concentration levels observed at two elevated sites (the CN tower in Toronto and the former World Trade Center in New York City), we used the trajectory-clustering methodology described by Brankov et al. (1998). The approach entails calculating a large number of back-trajectories from the observational site over a long period of time, and subjecting them to cluster analyses. We used the Hybrid Single Particle Lagrangian Integrated Trajectories (HY-SPLIT3) model (Draxler, 1992) to calculate 24-hour back-trajectories for every summer day (June, July and August) over a period of 7 years (1989-1995). Since we were interested in the overnight transport, each trajectory’s starting time was 6 a.m. local time. The trajectory starting heights were correspondent with ozone measurement heights: 444m for the CN tower and 457m for the World Trade Center (WTC). The trajectory calculations were based on wind fields from the National Meteorological Center’s Nested Grid Model (NGM) (Rolph, 1992). The NGM database contains three dimensional wind vectors with a horizontal resolution of 180 km and ten vertical layers up to seven kilometers. The data are archived at 2-hour time steps.

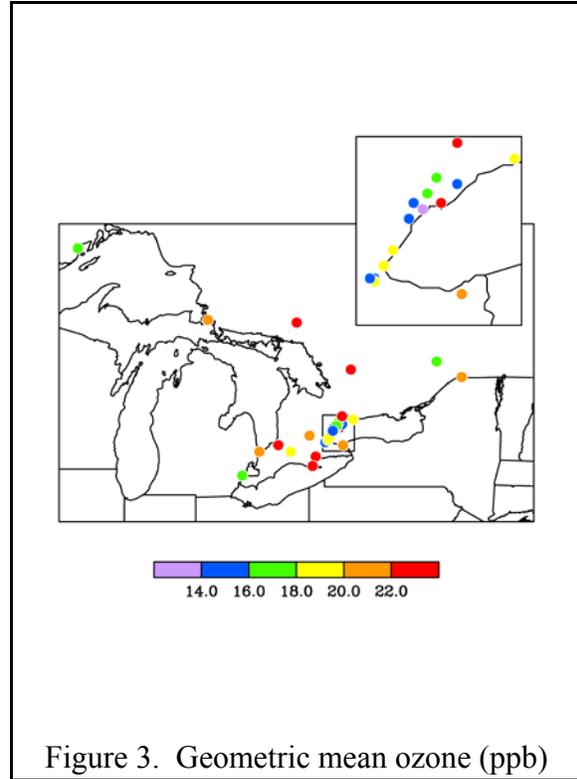
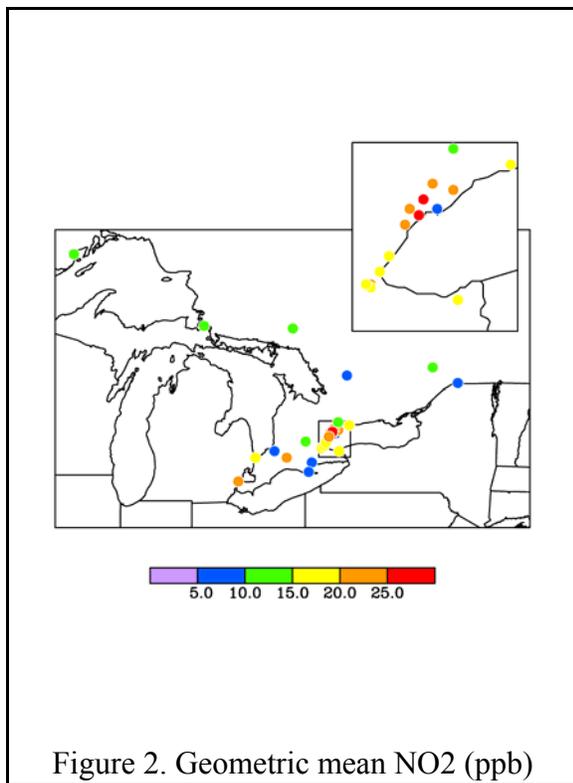
The trajectory-clustering technique was applied to group together trajectories closest to each other and with similar directions. This way, the large number of trajectories is reduced to a manageable number of trajectory groups/clusters and the many different air flow regimes to a finite number of synoptic conditions. The observed ozone time series is first de-seasonalized and de-trended and only the short-term component (weather-related variations) is segregated according to the back-trajectory clusters. To be consistent with the trajectory arrival times,

we used time series of ozone concentrations measured at 6 a.m. Following Brankov et al. (1999), we used a non-parametric rank-type Multiple Comparison Procedure (MCP) to test for statistically significant differences in the chemical composition of the clusters. This methodology enabled us to identify distinct atmospheric transport patterns associated with high levels of ozone concentrations, study the effects of transboundary pollution exchange, and identify potential source regions of this pollutant.

RESULTS

Long-term Averages

Maps of the average values of the long-term components of NO₂ and ozone are given for all 28 monitors in Figures 2 and 3 respectively. Figure 3a shows the long-term average of ozone for the summer season only. The lowest NO₂ concentrations occur in a band of rural monitors located south and west of Toronto which includes Parkhill, Simcoe, and Long Point, while the highest occur in the urbanized areas of Toronto (excluding the elevated CN Tower), Windsor, and Ottawa. In contrast, the lowest ozone concentrations generally occur in these same urban



locations while the highest occur at the rural sites. While rural monitors recorded the lowest concentrations of NO_2 , they have some of the highest ozone concentrations.

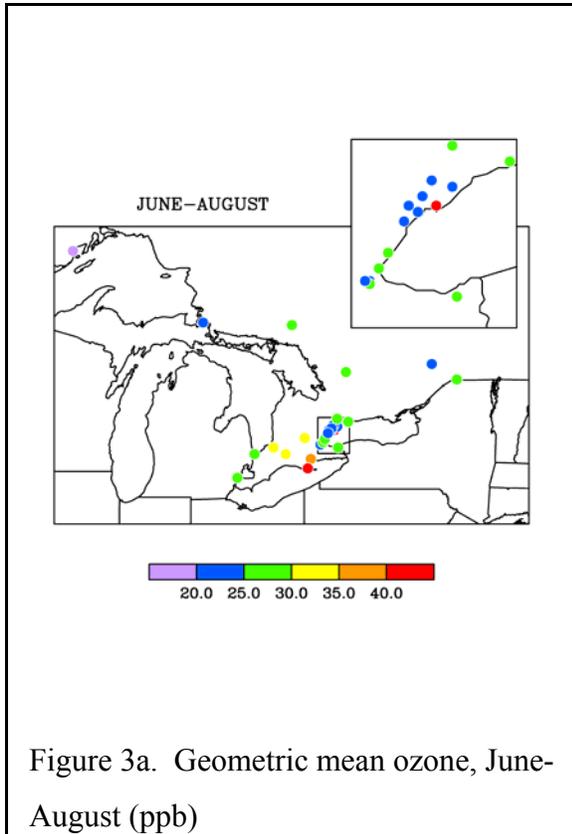


Figure 3a. Geometric mean ozone, June-August (ppb)

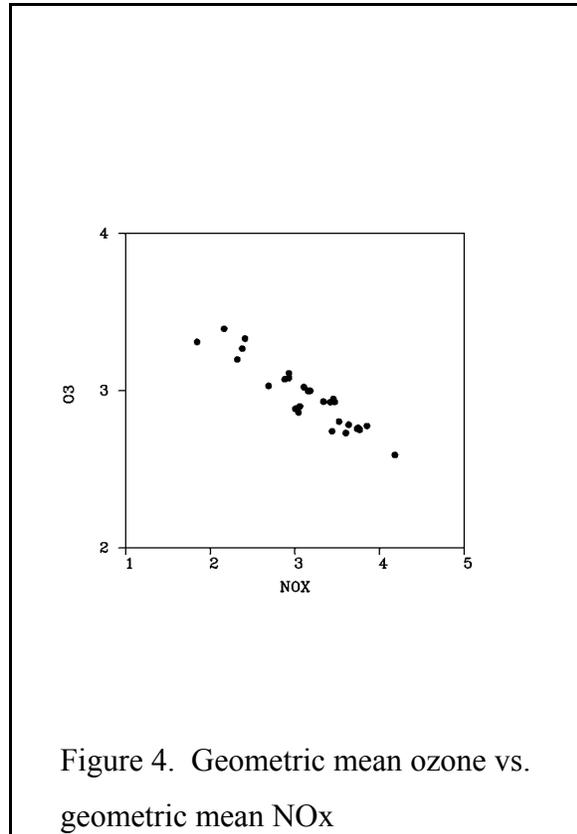


Figure 4. Geometric mean ozone vs. geometric mean NO_x

Figure 4 displays the average long-term concentrations of daily average ozone versus those for NO_x . An inverse relationship is clearly evident in Figure 4, indicating that the titration of ozone by NO is very important in determining the mean concentration of ozone. This effect is very pronounced in the wintertime, and, in fact, it is the wintertime data that are responsible for the very high correlation seen in the data; a plot of ozone versus NO_x for the summer season only does not exhibit nearly as high a correlation.

In order to facilitate a comparison between the ozone situation in Ontario and New York, we chose a few “representative” sites in the Toronto metropolitan area and the New York City metropolitan area. Since the primary interest from an air quality management point of view has generally been in urban areas, this is justified. Site selection was based on the length of

the data record and the “representativeness” of the site as one of either “upwind”, “urban” or “downwind”.

We considered Simcoe, a rural site upwind of the Toronto Metropolitan area, the CN Tower, an elevated site (elevation 444 m), Etobicoke, a site representative of urban Toronto, and Oshawa, a downwind site. We used the following ozone and NO₂ sites around New York City: Bell Labs (upwind), Cliffside Park (urban), New Haven (downwind), and the World Trade Center (elevation 457 m).

The long-term components of NO₂ and ozone are presented in Figures 5 and 6 for the monitoring sites considered here. For both Toronto and New York City, the figures are arranged with the upwind site on the left, the downwind site on the right, and the urban site in the center. The elevated sites are shown in Figure 7. For both Toronto and New York City, NO₂ concentrations increase as one moves from upwind locations to the urban area and remain high at downwind sites. On the other hand, ozone concentrations are highest upwind and decrease as one moves to the urban and downwind areas, most likely due to NO_x scavenging (titration). Pollutant concentration levels at the elevated sites are comparable to levels at the corresponding upwind sites (Figure 7).

Hydrocarbons

The following Canadian hydrocarbon sites were investigated: Toronto, Hamilton, and Stouffville. The following US sites were investigated: Plainfield and Newark, New Jersey, and Eisenhower Park, New York. Long-term components of TNMOC are depicted in Figures 8 and 9. For TNMOC, categorization of a monitor as “upwind”, “urban”, or “downwind” is not as obvious as for ozone and NO₂. However, we may note that the average TNMOC concentrations are not surprisingly higher in urban areas than suburban areas.

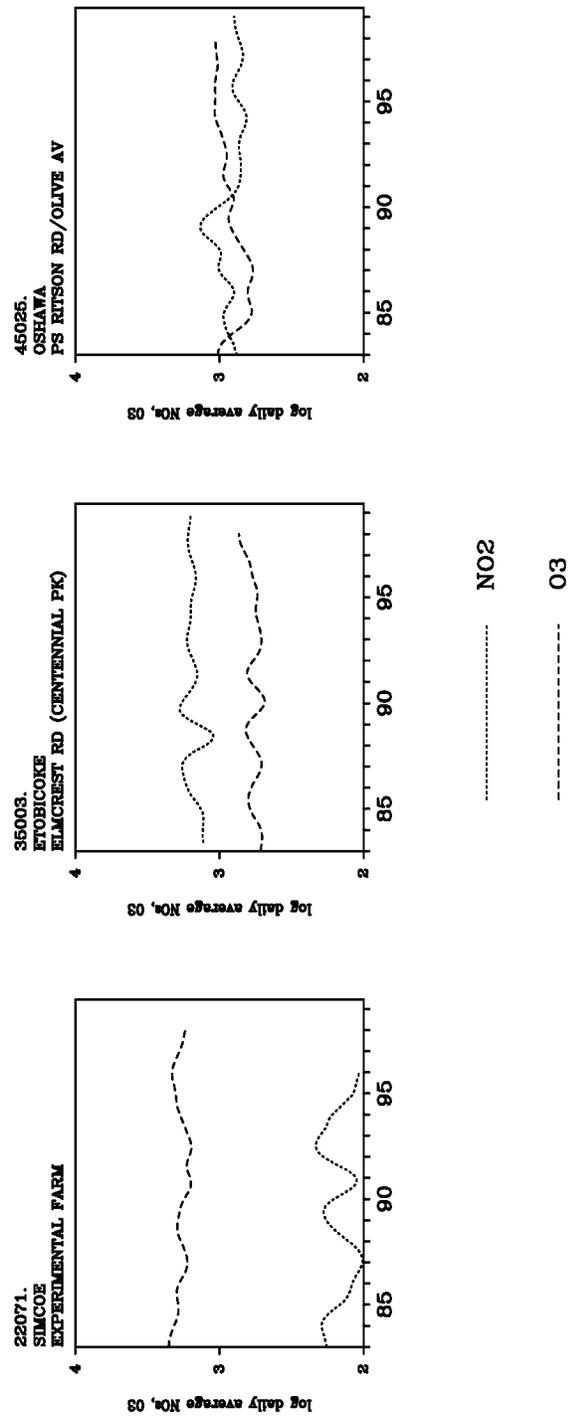


Figure 5. Long-term components of NO₂ and O₃ for upwind, urban, and downwind sites of Toronto

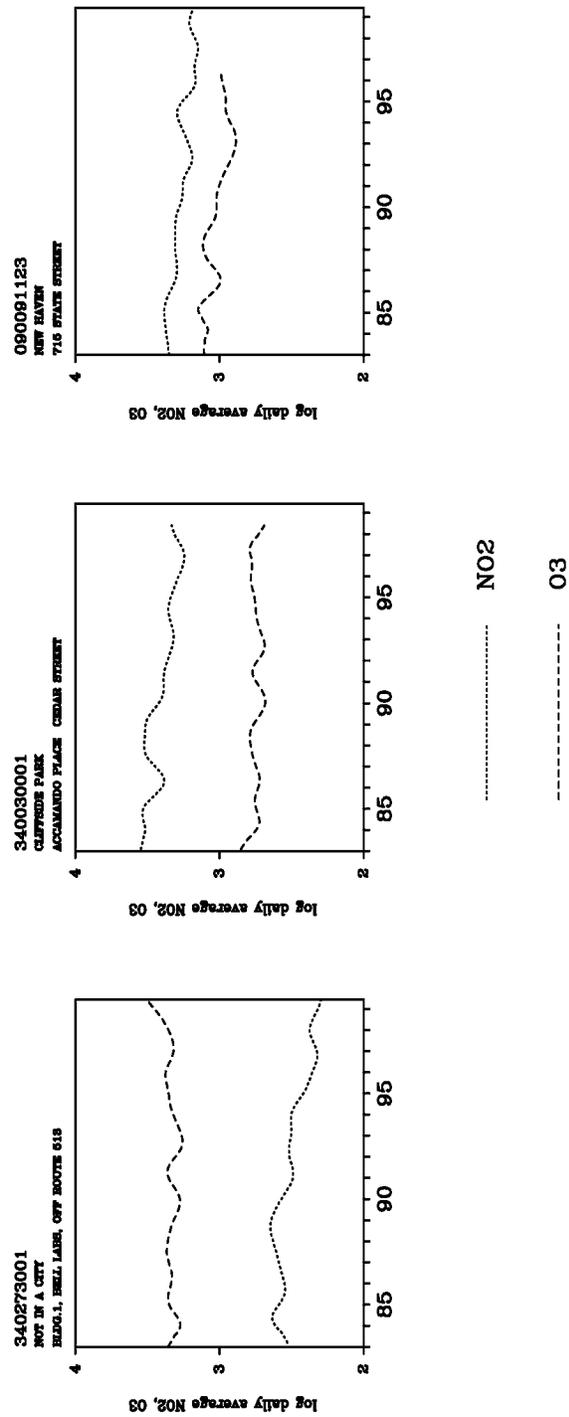


Figure 6. Long-term components of NO2 and O3 for upwind, urban and downwind sites of New York

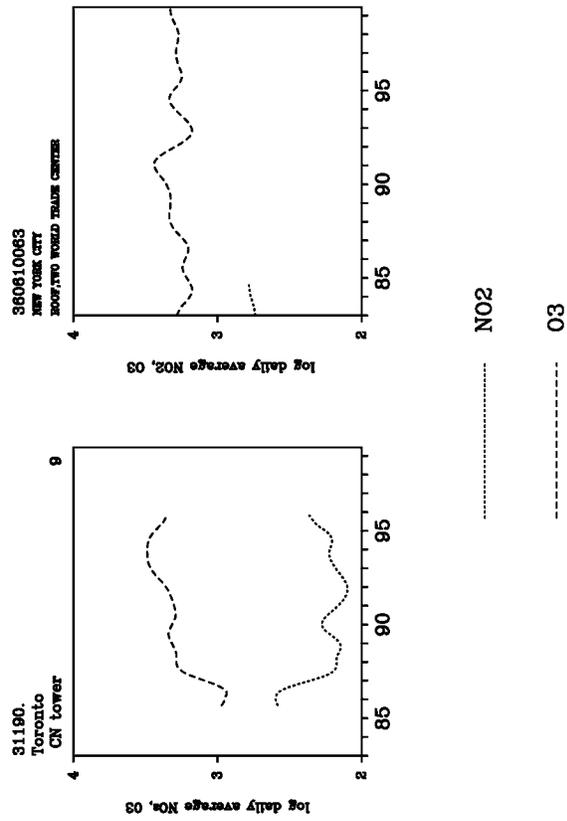


Figure 7. Long-term components of NO2 and O3 for CN Tower and World Trade Center

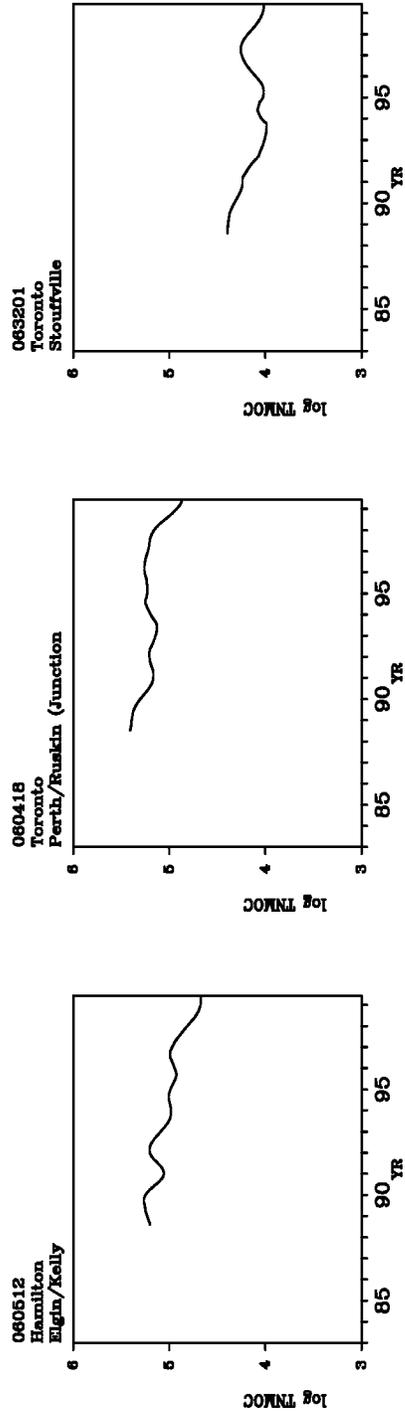


Figure 8. Long-term components of TNMOC for upwind, urban and downwind sites of Toronto

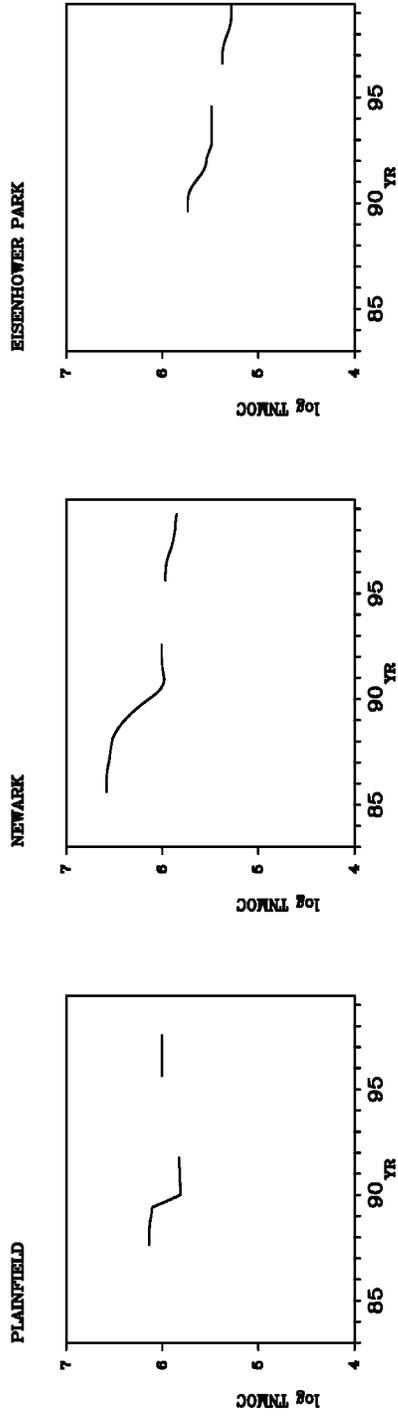


Figure 9. Long-term components of TNMOC for upwind, urban and downwind sites of New York

Long-term Trends

Trends (i.e., the slopes of linear regressions) in the long-term components have been determined for all NO₂ and ozone data available between 1983 and 1998 inclusive and are also shown in Table 1, along with their 95% confidence intervals. The confidence intervals are all seen to be much smaller than the magnitude of the slopes, so that all trends are significantly different from zero. NO₂ has been decreasing at all stations except Hamilton, Scarborough, Etobicoke, and Oakville; ozone is increasing at all sites except Sarnia, Simcoe, Long Point, Kitchener, St. Catherines, and Cornwall. The largest decrease in NO₂ is at Sault Ste. Marie; the largest decrease in ozone is at St. Catherines, while the largest ozone increase is at the CN Tower. Figures 10 and 11 display maps of the long-term trends in NO₂ and ozone respectively. While NO₂ concentrations are generally decreasing as ozone concentrations are generally increasing, there is no simple linear relationship between the trends as was found for the long-term average concentrations.

Long-term linear trends and 95% confidence intervals for the slopes in NO₂ and ozone for the time periods indicated are shown for both the Toronto and New York City Metropolitan Areas in Table 2. Similarly, trends and 95% confidence intervals for TNMOC are shown in Table 3. For both the Toronto and New York City Metropolitan Areas, both NO₂ and TNMOC are decreasing. In the Toronto Metropolitan Area, ozone is increasing while in the NYC Metropolitan Area, ozone is decreasing at the sites selected. Long-term trends in TNMOC concentrations are decreasing at both the three Ontario Metropolitan Area monitoring sites and the three New York City Metropolitan Area sites considered here. The slopes and 95% confidence intervals for them are shown in Table 3. Emission controls have apparently been effective in controlling VOCs in both urban areas.

NO₂ trends are generally decreasing one or two percent per year at the sites examined here, consistent with U.S. national decreases of 11% over the period 1991-2000, or 14% over the period 1981-2000 (USEPA, 2001).

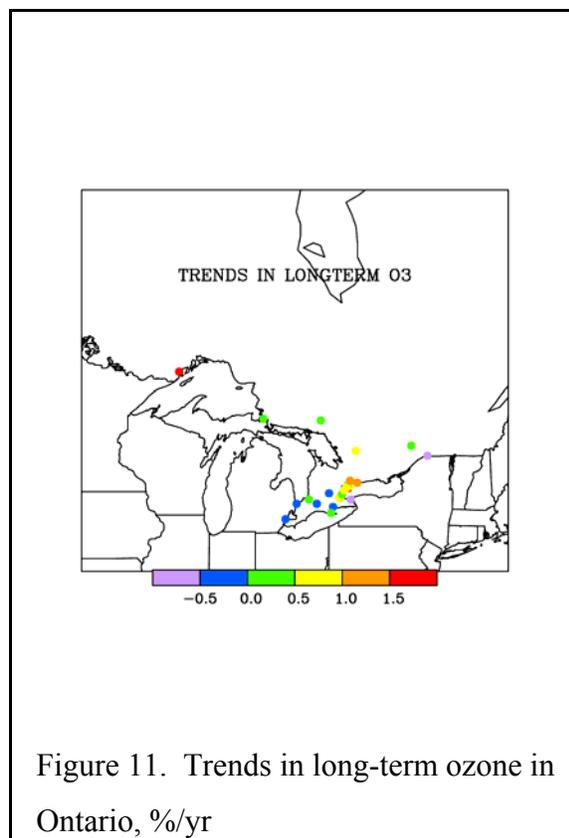
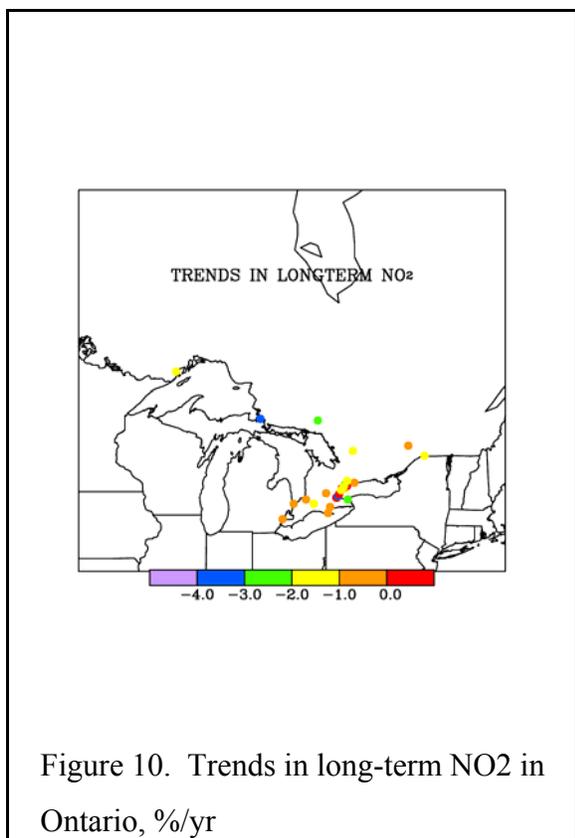


Table 2. Trends in NO₂ and ozone for the Toronto and NYC metropolitan areas

Site ID	Location	NO ₂ trend, %/yr	O ₃ trend, %/yr
22071	Simcoe, ONT	-0.24±0.07 (1983-1998)	-0.14±0.02 (1993-1998)
31190	CN Tower, ONT	-1.09±0.10 (1985-1995)	4.29±0.07 (1985-1995)
35003	Etobicoke, ONT	0.55±0.03 (1983-1997)	0.31±0.02 (1993-1997)
45025	Oshawa, ONT	-0.88±0.04 (1983-1997)	1.57±0.04 (1993-1997)
090091123	New Haven, CT	-1.32±0.04 (1983-1999)	-0.52±0.05 (1983-1996)
340030001	Cliffside Park, NJ	-1.86±0.02 (1983-1998)	-0.03±0.02 (1983-1999)
340273001	Bell Labs, NJ	-1.94± 0.04 (1983-1999)	0.00± 0.04 (1983-1999)
360610063	World Trade Center, NYC	insufficient data	-2.3 ±0.15 (1985-1999)

Table 3. Trends in Total NonMethane Organic Carbon

	TNMOC trend, %/yr	95% CI
Toronto, Ontario	-2.27 (1989-99)	0.08
Hamilton, Ontario	-4.87 (1989-99)	0.058
Stouffville, Ontario	-2.07 (1989-99)	0.11
Eisenhower Park, NY	-3.83 (1990-98)	0.10
Newark, NJ	-5.84 (1986-98)	0.07
Plainfield, NY	-0.37 (1988-96)	0.09

While both NO₂ and TNMOC appear to be decreasing in the Toronto and New York City metropolitan areas, ozone appears to be increasing throughout Ontario, and in Toronto in particular, while it has been decreasing in the New York City metropolitan area. This difference may be due to the fact that Ontario is surrounded by a large region of upward trends, while ozone in areas surrounding New York is decreasing. It may also be that the two cities are in different VOC/NO_x regimes. Unfortunately, the differences (24-hour speciated hydrocarbons in Toronto vs 6-9 a.m. total nonmethane hydrocarbons in NYC) in the precursor data from the two areas preclude further investigation of this hypothesis. While in the future, hourly data from PAMS (Photochemical Assessment Monitoring Stations) data may be useful in a study such as this, currently it has not been collected for a sufficient length of time to enable reliable calculations of trends. Another factor which needs to be investigated is the contribution of the transport of both ozone and its precursors. It is possible that the amount of ozone transported from source regions upwind of Toronto is increasing while the transported component from source regions upwind of New York City is decreasing. Figure 12 displays linear trends in ozone concentration calculated for the ozone season (mid-April to mid-October) for Ontario and Eastern U.S. sites. Only trends significant at the 95% confidence level are shown. Ozone is decreasing in New York State and at most sites downwind (east) of it, but increases are found at many locations west and south of it. Ontario is surrounded by areas with upward ozone trends while the New York City area is imbedded in a region of downward trends. This, in part explains the difference in ozone trends in the two cities. Increases in the long-term average concentrations of ozone were also found by Lin et al. (2001) at many locations in the United States.

US Monitors: 1985-1998 (Ozone Season Data)
Canadian Monitors: 1985-1997 (Ozone Season Data)

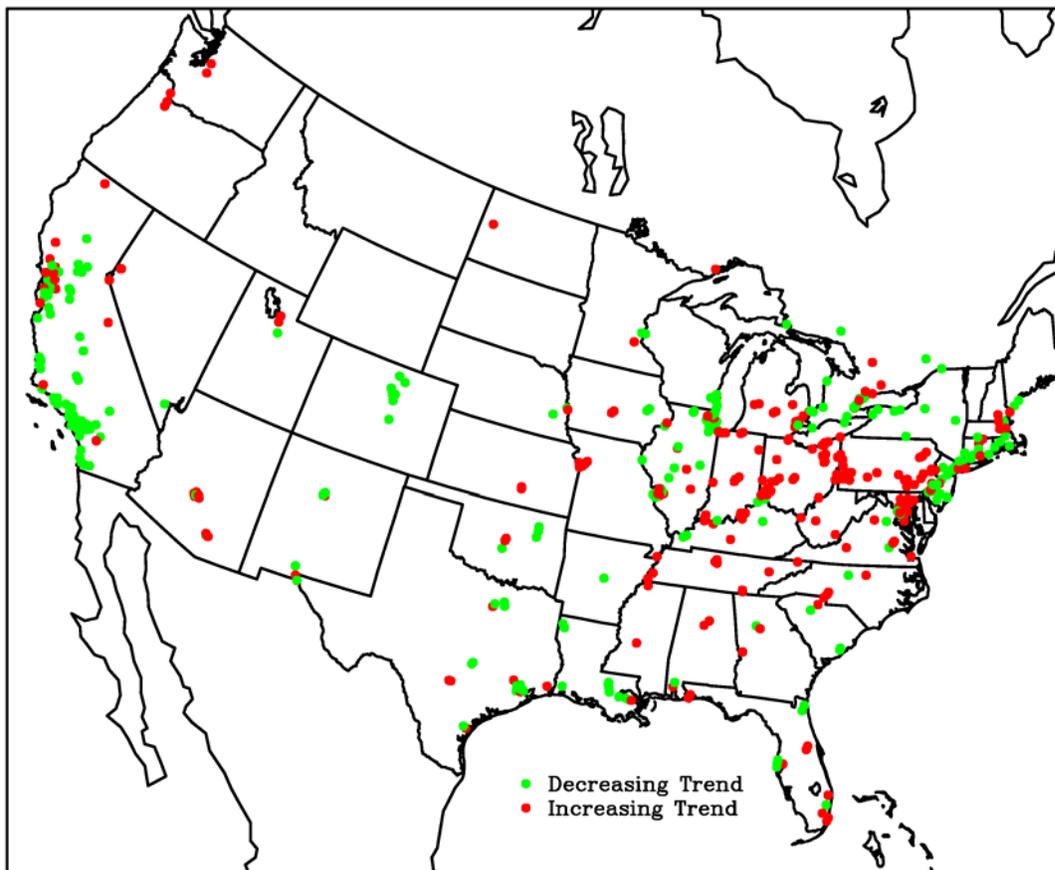


Figure 12. Linear trends in raw ozone

Ozone transport to the CN Tower Site

The back-trajectory clustering methodology applied on CN tower back-trajectories resulted in 8 clusters of trajectories whose average trajectories are shown in Figure 13. For further reference, each cluster is named according to its general direction: N, NE, SE etc. The letter L stands for “local” because trajectories in this cluster are short and associated with local, slow-moving air masses. The percentage of all trajectories belonging to each cluster is also shown in the figure. For the 7 summers (months of June, July and August of years 1989-1996), 54% of all the trajectories arriving to the CN tower are associated with air masses traveling over Canada (clusters NE, N, NW and L), while the remaining 46% of the airflow regimes are bringing air from the U.S. (clusters W, SW, S and SE).

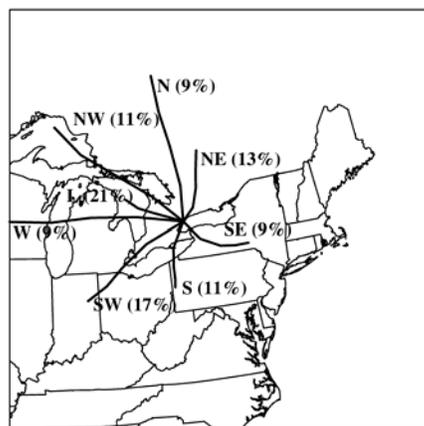


Figure 13. Average back-trajectories for 8 trajectory clusters at CN Tower, Toronto

Figure 14 shows box-whisker plots of “ozone clusters” obtained by segregating short-term ozone concentration data according to clusters in Figure 13. Each box-whisker displays 5 percentiles (10th, 25th, 50th, 75th, and 90th) as well as the minimum and maximum concentrations of ozone concentrations assigned to one particular cluster. Since the non-clustered short-term ozone is a zero-mean process, positive/negative median deviations seen in Figure 14 indicate positive/negative forcing associated with a particular wind direction. Positive forcing, in turn, indicates the existence of a potential pollution source region in that direction.

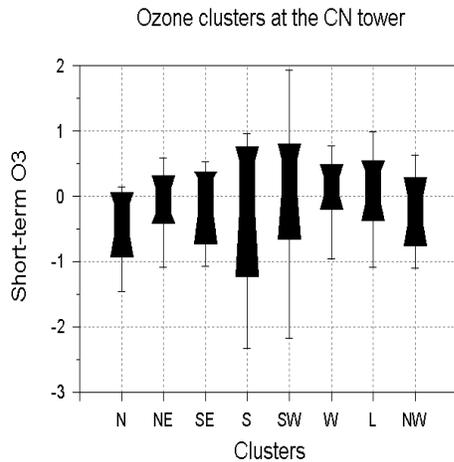


Figure 14.

The results from the multiple comparison of ozone clusters displayed in Figure 14 are presented in Table 4. The first column contains cluster names, the second indicates the number of ozone sampling days assigned to each cluster (i.e. sample size). The clusters are sorted according to their average ranks given in column 4. The average rank is an indicator of the magnitude of the concentrations assigned to that cluster: the higher the concentration levels in the

cluster, the higher the rank. For the period examined (7 summers: 1989-1995), the highest ranking clusters were SW and S, and then W and L. The first three of these are associated with trans-boundary pollution transport: air flows coming from the Ohio River Valley and the industrial Midwest (37% of all trajectories), and 21% of the very polluted air is associated with local air masses (cluster L) slowly moving east over Ontario (see Figure 13 for cluster directions). The lowest ranking clusters (associated with lowest pollutant concentration levels) are N and NW. These two clusters contain about 20% of all trajectories.

Columns under the “homogeneous groups” in Table 4 show the MCP results. Cells containing asterisks (*) in the same column indicate concentration clusters that are not significantly different from each other. When interpreting the MCP results, it is important to remember that, as with any significance test, equality of the groups is not demonstrated when the statistical test fails to reject the null hypothesis (which is in this case: ozone clusters are not significantly different from each other). Also, statistical inferences from the real-world data should be assisted by an understanding of the underlying problem. For example, although Table 4 indicates “no significant difference” between ozone concentration levels in SE and NW clusters that does not imply that their pollution levels are equal, and it certainly doesn’t imply pollution transport from the same source region (see Figure 13). However, we

Table 4. Results of Multiple Comparison Procedure at CN tower. Ozone clusters in this table are corresponding to the trajectory clusters in Figure 13. Cells containing a star in the same column indicate clusters that are not found significantly different from each other.

cluster name	# of days	average rank	homogeneous groups			
SW	91	365				*
S	68	333			*	*
W	47	328			*	*
L	119	316			*	*
SE	52	262		*	*	
NE	72	234		*		
NW	63	201	*	*		
N	50	125	*			

can say that the MCP led to the identification of three significantly different ozone concentration levels at the CN tower: 1) southerly, southwesterly, westerly, and local transport scenarios (clusters S, SW, W and L) which result in significantly higher ozone concentrations at this site, 2) northwesterly and northerly flows (NW and N clusters) which result in significantly lower ozone concentrations, and 3) northeasterly and southeasterly flows (NE and SE clusters) which are related to intermediate levels of ozone concentrations at the CN tower.

The average ozone concentration for each summer in the 1989-1995 period, calculated in each individual cluster revealed weak downward trends in clusters SW, N and NE and no significant trends in the remaining 5 clusters.

Ozone transport to the World Trade Center Site

Application of the back-trajectory clustering methodology at the 24-hour long back trajectories at the WTC resulted in cluster-average trajectories shown in Figure 15. Only N and NW clusters (containing 24% of all trajectories) are associated with airflows from Canada. Cluster “L” in Figure 15 is associated with local stagnation, and the average trajectory is very short, remaining within the state of New Jersey. Results of MCP of ozone clusters associated with trajectory clusters in Figure 15 are shown in Table 5. Clusters are again sorted according to their ranks in column 3. Asterisks under the “homogeneous groups” clearly identify two significantly different groups of clusters: those with high ozone concentrations (SW, W, L, S and NW) and those with low ozone concentrations (N, NE and SE). While the “high ozone group” represents 70% of all examined days, the “low ozone group” represents 30% of them. Similar to the situation at the CN tower, the highest ranking clusters (SW and W) are associated with pollution transport from the Ohio River Valley. Other highly ranked clusters are L (associated with local ozone production), S (associated with pollution transport from the urban east coast) and NW (associated with Canadian air masses transported from southern Ontario). Just like the CN tower, the cleanest air masses are transported to the WTC with northerly flows (cluster N).

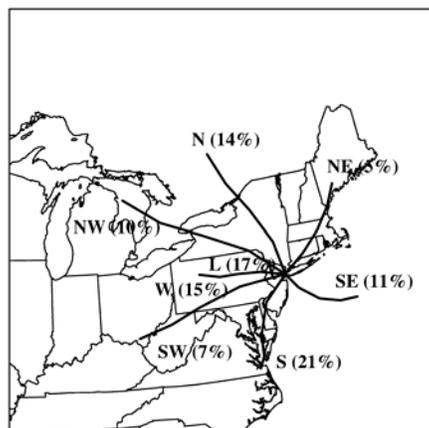


Figure 15. Average back-trajectories for 8 trajectory clusters at World Trade Center, New York

Table 5. Results of Multiple Comparison Procedure at WTC. Ozone clusters in this table are corresponding to the trajectory clusters in Figure 15. Cells containing a star in the same column indicate clusters that are not found significantly different from each other.

cluster name	# of days	average rank	homogeneous groups	
SW	43	386		*
W	100	375		*
L	103	357		*
S	130	354		*
NW	63	331		*
SE	67	235	*	
N	91	234	*	
NE	34	140	*	

ASSESSING THE EFFECTS OF TRANSBOUNDARY POLLUTION
ON NEW YORK'S AIR QUALITY

Section 3

OBJECTIVE 2: MODEL EVALUATION

Apply state-of-the-science regional-scale air quality modeling systems, namely, the MODELS3 system - a 3rd generation regional model developed by EPA, and the UAM-V that was used by NYSDEC in the OTAG process. In this project, we used an updated emission inventory that accurately reflects emissions from Ontario and the eastern US, and evaluated the ability of these modeling systems in simulating the observed ozone air quality for the base year of 1995 on a seasonal basis.

We performed several analyses toward the goal of model performance evaluation: For example, we

- developed a new model performance evaluation methodology, and used it to evaluate both air quality models and their meteorological drivers. Basically, the model output is spectrally decomposed at each grid cell, and each spectral component is individually compared with the corresponding spectral component of the observational data.
- examined the sensitivity of a photochemical model (UAM-V) to the meteorological input fields generated by two different meteorological drivers (RAMS and MM5).
- examined in detail the performance of UAM-V/RAMS3b at rural monitors.
- performed a detailed comparison of two photochemical modeling systems, namely, RAMS/UAM-V and MM5/SAQM.

Rather than rely on traditional model evaluation statistics (Hanna, 1994; Lyons et al., 1995; Olerud and Wheeler, 1997), we used scale analysis to evaluate two state-of-the-art meteorological models, namely MM5 and RAMS3b, which are currently being used to drive

regional-scale air quality models (Hogrefe et al., 2001a). In this study, seasonal time series of observations and predictions for temperature, water vapor, and wind speed were spectrally decomposed into fluctuations operating on the intra-day, diurnal, synoptic and longer-term time scales. Traditional model evaluation statistics were also used to examine how the method of spectral decomposition can help provide additional insight into the models' performance. It was found that both meteorological models under-represent the variance of fluctuations on the intra-day time scale. Correlations between model predictions and observations for temperature and wind speed were found to be insignificant on the intra-day timescale, high for the diurnal component because of the inherent diurnal cycle but low for the amplitude of the diurnal component, and highest for the synoptic and longer-term components. The poor performance on the intra-day time scale is most likely due to the relatively coarse grid resolution used in the model. The better model performance on the longer time scales suggests that current regional-scale models are most skillful for characterizing average patterns over extended periods, rather than on 1 to 2 day episodic events. When these meteorological models are used to drive air quality simulations, air quality predictions cannot be expected to be accurate for scales which are not captured by all the input fields used in the photochemical model.

Similarly, scale analysis was also used to examine ozone predictions from two regional-scale air quality models (Hogrefe et al., 2001b); seasonal time series of observations and predictions from the RAMS3b/UAM-V and MM5/MAQSIP (SMRAQ) modeling systems for ozone were spectrally decomposed into fluctuations operating on the same time scales as for the meteorological drivers, i.e., the intra-day, diurnal, synoptic and longer-term time scales. In the past, photochemical models were applied for the duration of one or a few historical ozone episodes, and model evaluation was generally limited to the comparison of ozone predictions and measurements through a set of statistical performance measures (Tesche et al., 1990; USEPA, 1991, 1994, and 1999, Tesche et al., 1996). Traditional model evaluation statistics were again used to examine how scale analysis can help improve our understanding of the models' performance. The UAM-V underestimates the total variance (energy) of the ozone time series when compared with observations, but shows a higher mean value than the

observations; MAQSIP, on the other hand, is better able to reproduce the average energy and mean concentration of the observations. However, neither modeling system captures the amount of variability present on the intra-day time scale, due most likely to the grid resolution used in the models and the poor performance of the meteorological drivers on this time scale. For both, the correlations between predictions and observations are insignificant for the intra-day component, high for the diurnal component (because of the inherent diurnal cycle), but low for the amplitude of the diurnal component, and highest for the synoptic and baseline components. This better model performance on the longer time scales suggests again that current regional-scale models are most skillful in characterizing average patterns over extended periods rather than in predicting concentrations at specific locations at specific times during 1 to 2 day episodic events. This longer-term averaging is also most relevant to regulatory policies that are aimed at meeting and maintaining the regulatory standards.

We have also examined the sensitivity of UAM-V to the meteorological input fields by comparing results of the same UAM-V simulation derived from inputs from different meteorological models, namely RAMS and MM5 (Biswas et al., 2000; Biswas and Rao, 2001b). Model-simulated meteorological fields are subject to uncertainty from sources such as model initialization, prescribed physical parameterizations, and data assimilation methods (Seaman and Michelson, 2000; Shafran et al., 2000). Because a number of prognostic meteorological models are now being used in photochemical modeling analysis, a question that arises is whether there might be significant differences in the modeled ozone concentrations, and in the efficacy of an emission control strategy if different meteorological drivers are used for the same photochemical model. The uncertainty/variability in predicting the peak O₃ concentration is on the order of 20% for the UAM-V driven by RAMS versus MM5.

The performance of two commonly used regional-scale Eulerian photochemical modeling systems, namely, RAMS/UAM-V and MM5/SAQM, has been examined from a regulatory or operational perspective (Sistla et al., 2001b). While the Urban Airshed Model with Variable Grid (UAM-V) is driven with the meteorological fields derived from the Regional Atmospheric Model System (RAMS), the San Joaquin Valley Air Quality Model (SAQM)

used the meteorological fields derived from the Pennsylvania State University - National Center for Atmospheric Research Mesoscale Model Version 5 (MM5). Model performance in reproducing the observed ozone air quality over the eastern United States was evaluated for three typical high-ozone episodic events that occurred during 16-20 June, 12-16 July, and 30 July-2 August, 1995. The prevailing meteorological conditions associated with these three episodes are characterized by a slow eastward-moving high pressure system, westerly and southwesterly low-level jets, stable boundary layers, and the Appalachian lee-side trough. The results suggest that the abilities of the RAMS/UAM-V system and of the MM5/SAQM system to reproduce the observed ozone concentrations are comparable when model outputs are averaged over all simulated days. For different emission reduction options, the response of both modeling systems was directionally similar in terms of changes in ozone levels, but the magnitude of ozone improvement differed for individual episode days at individual grid cells.

The recent regulatory actions towards the longer-term (i.e., 8-hr) average ozone standard have brought forth the potential for many rural areas to be in non-compliance. However, since most rural areas have generally few sources of anthropogenic emissions, the measured ozone levels primarily reflect the effects of the transport of ozone and its precursor pollutants, and natural emissions. While photochemical grid models have been applied to urban areas to develop ozone mitigation measures, these efforts have been limited to high ozone episode events only and do not adequately cover rural regions. In order to examine the predictive ability of the modeling system at rural monitoring stations that are part of the Clean Air Status Trends Network (CASTNet) and the Gaseous Pollutant Monitoring Program (GPMP), we applied a photochemical modeling system, RAMS/UAM-V to the eastern United States for the period of June 1 through August 31, 1995. The measured daily 1-hr ozone maxima and seasonal average of the daily 1-hr ozone maxima were found to be in better agreement with the predictions of the modeling system than were the daily 8-hr ozone maxima. In addition, the response of the modeling system is poor in reproducing the measured ozone levels over the diurnal cycle, consistent with the finding that the models fail to accurately simulate the intra-day variations present in the observed data. These findings

suggest the need for improvements of the modeling system if it is to be used to address the 8-hr ozone standard (Sistla et al., 2001a).

The major findings of this section, i.e., a net modeling uncertainty of ~20%, and the inability of the models to simulate intraday/diurnal variability, call into question the basis of episodic modeling for regulatory applications and indicate the need to switch from event-based to longer term modeling.

ASSESSING THE EFFECTS OF TRANSBOUNDARY POLLUTION
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Section 4

OBJECTIVE 3: EFFICACY AND METHODS (MODELING RESULTS)

Assess the efficacy of selected NOx and VOC emission reductions as predicted by the two models (MODELS3, UAM-V) in reducing the effects of transboundary pollution and develop methods for using the ozone modeling results in a regulatory setting with an increased degree of confidence. It should be noted that EPA considers the two models (MODELS3 and UAM-V) as acceptable tools to address the new 8-hr ozone standard.

SPATIAL INFLUENCE OF EMISSIONS

In order to quantitatively describe the airshed for ozone, PM, and respective precursors, one needs to perform 3-dimensional modeling. By removing the anthropogenic emissions from the area of interest, and examining the resulting reductions in predicted regional ozone concentrations, one can estimate the spatial influence of a given source region. We have performed a seasonal simulation utilizing the RAMS/UAM-V modeling system under the following two scenarios: the elimination of all anthropogenic emissions in New York State, and the elimination of all

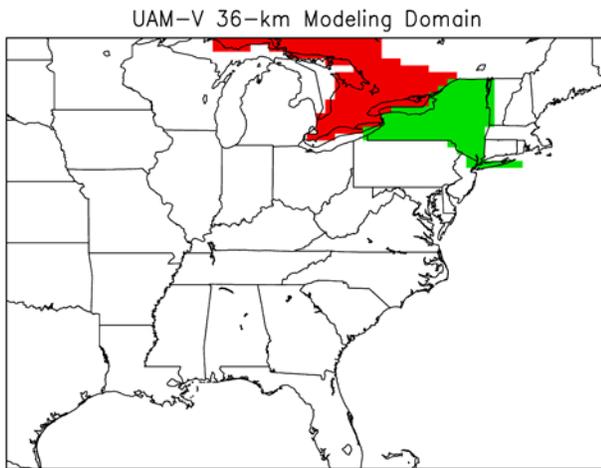
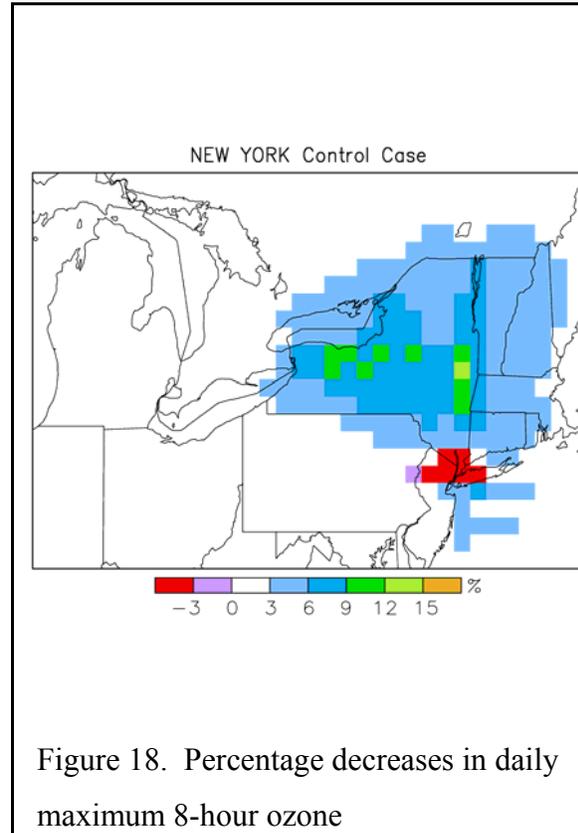
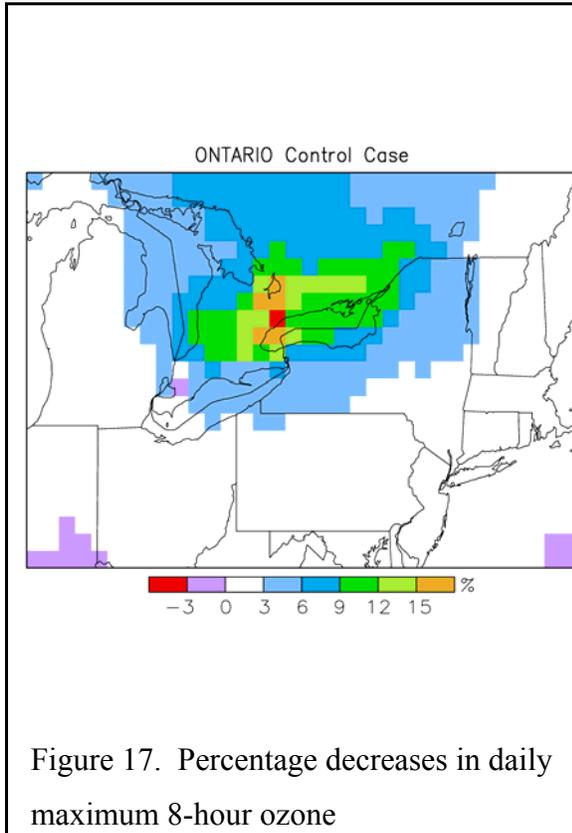


Figure 16. UAM-V 36-km modeling domain

anthropogenic emissions in the Canadian Province of Ontario. This enables us to quantify the effects of emissions in one region on the other. Figure 16 shows the 36-km UAM-V domain; the grid cells in red indicate the Ontario emissions region, while the green areas indicate the New York domain.



In the Ontario emissions reduction case, improvements of 15% or greater are evident in the near-field (see Figure 17). Along the New York boundary, the emissions reductions in Ontario led to peak ozone concentration decreases in excess of 6% throughout most of New York State. The dramatic NO_x reductions near Toronto actually led to increased ozone. The situation was similar in the New York emissions reduction case (see Figure 18), where ozone improvements within New York ranged from about 3% to 15%. Even along southern Ontario, the ozone decreased by up to about 6%. It should be emphasized that these percentage reductions are seasonal averages; the percentage reduction at a grid cell on any one day may be quite large. In addition, the sign of the change may vary from day to day,

depending upon the prevailing meteorological conditions.

USING MODELING RESULTS IN THE REGULATORY SETTING

We have examined episodic simulations performed for three high-ozone events over the eastern U.S. with several state-of-the-art photochemical modeling systems. There are significant differences between the observed and simulated diurnal cycles for ozone (both in terms of ozone accumulation for the day and hour-to-hour changes), which vary among modeling systems. This is consistent with the discussion in the previous section regarding the poor correlations found for both the amplitude of the diurnal component, i.e., the daily maximum concentration, and the intra-day component, which determines the shape of the diurnal oscillation, in particular its amplitude. These differences on the diurnal time scale introduce a large amount of uncertainty in the predicted daily maximum ozone concentrations on individual days; predictions of the daily maximum ozone concentrations with the current-generation photochemical modeling systems during an individual ozone episode can vary as much as 40 ppb. This uncertainty can be reduced when the predicted daily maximum ozone concentrations are averaged over all episode days simulated (Rao et al., 2000).

Since photochemical modeling systems display significant model-to-model differences when applied to only episodic days, their use in traditional ozone attainment demonstrations is highly uncertain. Atmospheric processes operating on time scales ranging from several days to several weeks are essential contributors to the days of high ozone concentrations at both urban and rural locations. In the two seasonal simulations with two different modeling systems, the highest model-to-observation as well as model-to-model correlations are present on the synoptic and baseline time scales, suggesting that, in the regulatory setting, the current generation modeling systems should be applied to simulation periods longer than a single episode. Furthermore, the model-predicted efficacies of a particular emission control strategy are highly variable for short simulation periods. The shift from 1-hr to the 8-hr ozone standard requires that we move away from modeling over small domains for shorter time periods to modeling over larger domains covering longer time periods (Rao et al., 2001).

The original guidance for the regulatory use of photochemical models uses the model

predicted ozone concentrations in a deterministic absolute sense: the model output for each grid cell for the emission control case must be below the NAAQS (USEPA, 1991). This “Pass/Fail” mode of application is not a robust way of using the model predicted concentrations since they are not reliable enough to be used this way (see discussion in section 3 - Model Evaluation). Later EPA guidance, (USEPA, 1999), while still deterministic, uses the model output in a relative sense by means of a site-specific Relative Reduction Factor (RRF). The RRF is the ratio of the mean model-predicted daily ozone maxima for the emission control- and base-cases. To determine whether a given control scenario would lead to compliance at a given location, the design value for that location is multiplied by the RRF for that particular control strategy. We showed that longer modeling periods need to be considered when estimating the RRF (Hogrefe et al. 2000; Rao et al. 2001). We have developed a methodology for utilizing the model-predicted concentrations in both a relative and a probabilistic sense.

We have also developed an integrated observational-modeling approach to transform the deterministic nature of attainment demonstrations of the National Ambient Air Quality Standard (NAAQS) into the probabilistic framework (Hogrefe and Rao, 2001; Rao and Hogrefe, 2001), focusing on the application to the 1-hr and 8-hr NAAQS for ozone. Model-to-model differences in predicted relative responses to emission reductions are shown to be smaller than model-to-model differences in predicted absolute ozone concentrations, supporting the EPA’s new draft guidance for the use of models in attainment demonstrations (USEPA, 1999). In addition, we used extreme value statistics and resampling techniques to estimate the probability of exceeding the NAAQS for both 1-hr and 8-hr ozone concentrations. The emission reductions stemming from the emission control strategies proposed in the EPA’s SIP call (USEPA, 1998) substantially reduce the probability of exceeding the NAAQS over large areas, especially for the 8-hr average ozone concentrations.

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APPENDIX A:

PUBLICATIONS COMPLETED UNDER THIS CONTRACT

- 1) Biswas, Jhumoor, S. Trivikrama Rao, Kesu Zhang, and Nelson Seaman, 2000, Sensitivity of the regional ozone modeling results to different physical processes within a meteorological model, 11th Joint Conference on the Applications of Air Pollution Meteorology with the AWMA, 9-14 January 2000, Long Beach, California, American Meteorological Society
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