NITROGEN OXIDES, REGIONAL TRANSPORT, AND OZONE AIR QUALITY: RESULTS OF A REGIONAL-SCALE MODEL FOR THE MIDWESTERN UNITED STATES

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Abstract. An overview of the role of NO_x in the formation of rural O_3 , regional transport and its potential impact on urban air quality is presented. An analysis of a specific O_3 excursion in southeast Michigan (8-2-90) is performed based on a combined urban and regional-scale model. The regional component of the model represents transport and photochemistry from sources as far away as Texas. Results suggest that rural O_3 and regional transport sensitive to NO_x emissions and relatively insensitive to changes in volatile organic carbon (VOC) emissions. This differs from the situation in urban areas, where O_3 is sensitive to both NO_x and VOC. Regional transport and upwind NO_x emissions have a significant impact on peak O_3 in Detroit. Implications for urban and regional-scale abatement strategies are discussed.

1. Introduction

It has been apparent for many years that elevated O₃ in eastern North America is a regional-scale problem with dimensions that transcend state and even national boundaries. As illustrated in Figure 1, elevated O₃ concentrations in eastern North America have been observed to extend over distances of 1500 km or more and to affect rural air masses as well as urban air [1-4]. Despite the regional nature of the problem, strategies for O₃ abatement in the U. S. have been based on models (e.g. EKMA and UAM) which represent a single metropolitan area without explicit regard for the influence of changing upwind emissions. The limited scope of existing air quality models coupled with the failure of past efforts to control O₃ [5] suggest the need for a more extensive analysis that includes the impact of regional transport.

This paper presents overview of the factors that affect large-scale regional transport of O_3 in eastern North America, and presents a regional-scale analysis of a specific O_3 episode in the midwestern U.S. The overview is designed to acquaint readers familiar with the dynamics of O_3 , nitrogen oxides (NO + NO₂, abbreviated as NO_x) and volatile organic carbon (VOC) in urban locations with the dynamics that affect rural O_3 and regional transport. It will be argued that the relation between precursor emissions (NO_x and VOC) and regional transport of O_3 is fundamentally different from the relation between O_3 and precursors in urban centers. Consequently a regional O_3 control strategy must involve more than just a sum of localized urban strategies. The analysis of a midwestern O_3 episode is based on the Plumes Model, developed at Harvard University for the investigation of regional and global scale

O₃ production, in combination with a nested urban grid model for the Detroit area. The selected episode occurred on August 2, 1988, and is being used by the Michigan Department of Natural Resources (DNR) as the design day for evaluation of abatement strategies in southeast Michigan. Results of the regional-scale model will illustrate the difference between urban and regional control regimes and the potential impact of regional transport on urban air quality.

2. Background Rural Ozone

The impact of regional transport on urban air quality can best be understood through the concept of 'background rural ozone'. Background rural O₃ refers to the average concentration of O₃ over a rural air mass with spatial extent of 450 km or more. The distinguishing factor in background rural O₃ is that it is not associated with export from a specific urban source region (as would occur, for example, in rural Connecticut or Long Island under the influence of the urban plume from New York City). Background O₃ represents a uniform concentration over a wide area, and consequently has a high likelihood of being found *upwind* of major urban centers. Consequently background O₃ frequently has an impact on urban air quality as transport of background O₃ into the urban center adds to local photochemical production.

The importance of background rural O_3 is demonstrated by observations (e.g. Figure 1) which show O_3 concentrations in the 0.08 to 0.10 ppm (part per million)

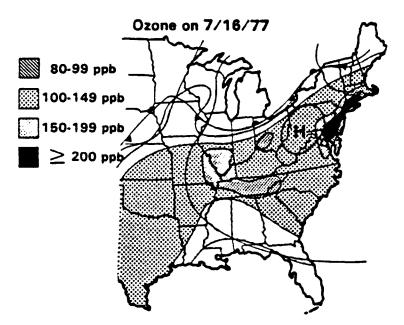


Fig. 1. Observed O₃ concentrations at sites in eastern North America on July 16, 1977, from Wolff and Lioy [3].

range at rural sites extending over a wide area [3], and by high spatial correlation for O₃ concentration at widely separated rural sites [4]. Direct evidence for the extent of high background O₃ was provided by an aircraft study by Clarke and Ching [6], in which O₃ concentrations were measured along airplane transects through an eastward-moving air mass in Ohio, Pennsylvania and New York. The final transect extended along a ~450 km path from Fredrick, MD, to Kingston, NY. O₃ concentrations ranged from 0.07 to 0.11 ppm along this transect, with an average concentration of 0.09 ppm. Observed wind fields made it likely that this air would be transported into the New York, Philadelphia and Washington D.C. metropolitan areas.

Observations [3, 4, 6] suggest that background O_3 in eastern North America frequently reaches 0.08 to 0.10 ppm, but rarely exceeds 0.12 ppm, the level on which the National Ambient Air Quality Standard (NAAQS) for O_3 is based.* However there is a strong likelihood that transport of O_3 at concentrations of 0.08–0.10 ppm can contribute to NAAQS violations in downwind urban areas. Observations in southeast Michigan (Figure 2) show a strong correlation between observed peak O_3 in the Detroit-Port Huron corridor and peak O_3 at Saline, a rural site to the southwest. Exceedences of the NAAQS standard in Detroit are most commonly associated with peak $O_3 > 0.09$ ppm at Saline.

Samson and Shi [7] have identified meteorological circulation (in term of 24-hour back-trajectories) associated with elevated O₃ in cites throughout the eastern United States. Results for Boston, MA (Figure 3) show that elevated O₃ is associated with transport from the North American continent, but not with transport from individual urban centers. Air quality violations in Boston are as likely associated with transport from upstate New York and Ontario as with transport from the northeast corridor. Air quality violations in Boston were never observed when winds were southerly off the ocean. These results suggest that background rural O₃, rather than transported urban plumes, has a significant impact on air quality in Boston. Similar results were obtained for other American cities, although interurban transport was also detected in some northeast corridor cities.

3. Relation between background O₃ and Nitrogen Oxides

The relation between O_3 and precursors in urban areas has been the subject of numerous studies and is familiar to air quality analysts [8]. Urban O_3 may be sensitive to either (NO_x or anthropogenic VOC emission, and sensitivity may vary for different cities, [9, 10] or for different locations within the same city [11]. While the processes that contribute to background rural O_3 have many similarities to the urban case, there are some fundamental differences between the urban and rural cases. Background rural O_3 is most likely sensitive to NO_x emissions and

^{*} The NAAQS for O_3 is 0.12 ppm hourly average not to be exceeded in any more than three days in any three year period.

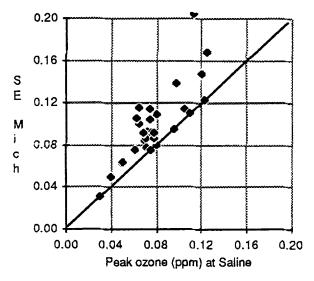


Fig. 2. Diurnal peak hourly O₃ (ppm) over ten sites in the Detroit-Port Huron corridor versus diurnal peak hourly O₃ observed at Saline, Michigan for July 1-31, 1988. The solid line represents equal x-and y-axis values.

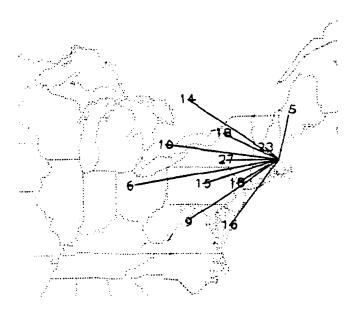


Fig. 3. Mean 24 hr trajectory displacement for wind flow categories associated with O₃ concentrations greater than 0.08 ppm for Boston, MA. Numbers refer to the number of events associated with each trajectory displacement category, based on a total of 168 cases during the period 1983-1985. Violations of the 0.12 ppm NAAQS standard were associated with each category roughly in proportion to the number of >0.08 ppm events. From Samson and Shi [7].

insensitive to VOC [12, 13]. Reasons for NO_x dependence of rural O_3 relate to photochemical processes and are worth summarizing.

Two major factors contribute to background rural O_3 in eastern North America: export of O_3 photochemically produced in urban and power plant plumes, and rural photochemical production of O_3 . The latter is associated with a combination of transport of O_3 precursors (NO_x and VOC) from urban centers and with precursor emissions from 'distributed' sources, small cities and towns scattered throughout the region. Biogenic VOC emissions also contribute significantly to rural O_3 production [12]. Recent calculations suggest that significant O_3 production can occur with NO_x concentrations as low as 1 ppb [13]. Observations suggest that daytime NO_x concentrations of 1 ppb are commonly found at rural sites throughout eastern North America [12, 14].

Photochemical production of O_3 may occur either in a VOC-sensitive photochemical regime or in a NO_x -sensitive regime. For urban conditions the divide between VOC-sensitive and NO_x -sensitive regimes depends on the relative amounts of VOC and NO_x , with the VOC-sensitive regime occurring when VOC/NO_x ratios are relatively low and the NO_x -sensitive regime occurring when VOC/NO_x ratios are high. For NO_x concentrations typical of rural areas, photochemistry is likely to be in the NO_x -sensitive regime even at low VOC/NO_x ratios. This is illustrated in Figure 4 (from Sillman *et al.* [13] which shows calculated O_3 versus noon NO_x

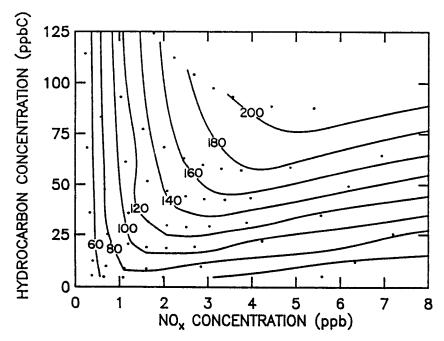


Fig. 4. O₃ (in ppb) as a function of the concentrations of NO_x and VOC (in ppbC) at 4 p.m., based on a calculations representing photochemical evolution during a 4-day stagnation with continuous NO_x and VOC emissions. Dots represent VOC and NO_x concentrations for individual simulations used to derive the O₃ contours. Biogenic emissions are zero. From Sillman *et al.* [13].

and VOC concentrations following a 4-day stagnation. Photochemistry is in a NO_x -sensitive regime for NO_x concentrations of 1 ppb or lower, regardless of VOC. The calculation assumes zero biogenic emissions; inclusion of isoprene would extend the NO_x sensitive regime further. For daytime NO_x and VOC concentrations observed in rural Pennsylvania (1 ppb NO_x , 20 ppbC VOC [13]) the calculated O_3 is 0.09 ppm (0.095 ppm if biogenics are included), close to the observed background O_3 during region-wide stagnation events.

The existence of a NO_x -sensitive photochemical regime at low NO_x concentrations is associated with processes that govern the concentration of OH and odd hydrogen radicals [13]. Odd hydrogen (defined here as the sum of OH, OH_2 and RO_2 radicals such as CH_3 O_2 , $C_2H_3O_2$, etc.) is removed from the system by one of three processes: formation of peroxides, formation of HNO₃, or formation of peroxyacetylnitrate (PAN). When formation of peroxides is the dominant sink for odd hydrogen, the concentration of HO_2 is relatively invariant and the rate of O_3 production is limited by the reaction

$$HO_2 + NO \rightarrow OH + NO_2$$
 (R1)

followed by photolysis of NO_2 to produce O_3 . The rate of this reaction increases as NO increases, corresponding to the NO_x -sensitive photochemical regime. When HNO_3 formation is the dominant sink, then the concentration of both OH and HO_2 decrease with increasing NO_x , causing O_3 formation to slow. The rate-limiting step for O_3 formation is now

$$VOC + OH \rightarrow RO_2 \tag{R2}$$

or

$$CO + OH \longrightarrow HO_2 + CO_2$$
 (with O_2) (R3)

and the rate of O_3 formation increases with increasing VOC concentration and decrease with increasing NO_x . This is the VOC-limited regime. For urban conditions the important reactions are all VOC reactions; inorganic reactions (such as R3 and the formation of H_2O_2) are relatively minor. Since the rate of peroxide formation increases with VOC and HNO_3 formation increases with NO_x , the NO_x -sensitive and VOC-sensitive photochemical regimes correspond to high and low VOC/NO_x ratios respectively. But at low NO_x the inorganic reactions have a major impact and formation of H_2O_2 becomes the dominant sink for odd hydrogen. For these conditions the system will be in a NO_x -sensitive regimes regardless of the VOC concentration.

The relation between rural O_3 and NO_x emissions can be identified in theory based on photochemical mechanisms. Empirical evidence linking rural O_3 with NO_x is more difficult to obtain since NO_x and O_3 have very different lifetimes within the atmospheric boundary layer (\sim 3-day lifetime for O_3 , \sim 3-hr lifetime for NO_x). Some empirical evidence may be derived by examining correlations between O_3 and NO_y , defined as the sum of NO_x and its chemical by-products (PAN, HNO₃,

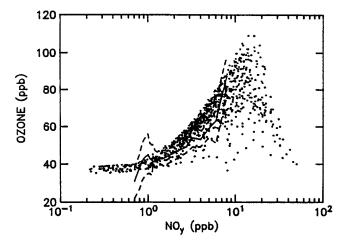


Fig. 5. O_3 vs NO_y concentrations (in ppb) at 6 p.m. predicted by a 20×20 km Eulerian grid model for Indiana, Ohio and western Pennsylvania. The lines show the average and standard deviation of observed O_3 concentrations as a function of observed NO_y at Niwot Ridge, Colorado. From Sillman et al. [13] with Niwot Ridge data from Fahey et al. [15].

alkyl nitrates, etc.). NO_y may be used as a surrogate for total NO_x emissions, since all NO_y species were originally emitted as NO_x . Model calculations (Figure 5) predict a strong correlation between O_3 and NO_y in rural locations. A correlation between O_3 and NO_y has been observed at a rural site in Colorado [15] which corresponds to model predictions. A similar correlation between O_3 and NO_y has recently been observed at rural eastern locations [12, 14].

Based on the above analysis, we suggest that analyses based on current photochemical mechanisms will all predict a strong relation between NO_x emissions and background rural O_3 . In contrast with the urban situation, analyses for background rural O_3 are less likely to be sensitive to model assumptions (emission rates, wind fields, etc.). We emphasize that this conclusion does not apply to rural locations influenced by direct urban plumes (e.g. southern Connecticut, Lake Michigan shoreline), but only to the 0.08 to 0.10 ppm 'ozone soup' that blankets the eastern U.S. during regional stagnation events.

4. The O₃ Exceedence of August 2, 1988

Violations of air quality standards were recorded on August 2, 1988 at three locations in southeastern Michigan: Osborn High School (0.137 ppm), in northeast Detroit; New Haven (0.144 ppm), 60 km northeast of downtown Detroit; and Port Huron (0.127 ppm), 90 km northeast of downtown Detroit. At all sites the exceedence was short-lived and was followed by a sharp drop in O₃. Maximum O₃ occurred from 1 to 2 pm EST in Detroit, 2 to 3 at New Haven and at 3 to 4 Port Huron, suggesting that ozone-laden air progressed downwind from Detroit. Although the 5 other monitoring sites in southeast Michigan did not register an exceedence, O₃

levels of 0.100 ppm or higher were recorded everywhere except at Saline (0.085 ppm maximum) and Ann Arbor (0.08 ppm).

In addition, violations of air quality standards also occurred at locations in Illinois, Wisconsin, Indiana, Ohio and Kentucky on both August 1 and August 2. Although most available observations represent urban sites, there are indications that O_3 was higher than normal in rural locations as well. Nearly all sites in the 6-state region recorded O_3 values of >0.070 ppm, considerably higher than clean-air O_3 [16].

The meteorology on August 1 and 2 was marked by southwesterly air flow throughout the midwest with temperatures in excess of 35 °C (95 °F). The afternoon mixed layer height was determined to be 1830 meters, based on rawinsonde observations at Flint, MI. Back-trajectory analysis [17] indicated that air located in southeastern Michigan on August 2 had been in Missouri July 30 and in northern Texas on July 29. Surface winds in the Detroit were light (1–2 m sec⁻¹) during the morning of August 2. Wind speeds increased to 4 m s⁻¹ after 1 p.m. [18] possibly contributing to lower O₃ concentrations in the late afternoon.

5. Simulation Methods

Regional-scale simulations were performed using the Plumes Model, which is described in detail elsewhere [19]. Because the Plumes Model represents an unfamiliar approach to the task of air quality modeling a brief summary will be presented here. In comparison with other models, the Plumes Model uses an extremely coarse horizontal grid, 400×480 km. Ordinarilly, use of such a large grid would lead to errors in the representation of photochemical processes, since no distinction is made between urban, suburban and rural locations. To avoid this, the Plumes Model uses a sub-grid structure within each 400×480 km horizontal grid box. The sub-grid structure, illustrated in Figure 6, divides each grid box into regions representing (a) the evolution of an urban plume, including both the urban center and downwind diffusion; (b) the evolution of a power plant plume from source to diffuse downwind plume; and (c) the evolution of the remainder of the rural air mass. The plume sub-grid boxes export O_3 , NO_x and VOC into the rural sub-grid box, as happens in the real-world situation.

The Plumes Model has been extensively tested in comparison with an equivalent model with a 20×20 km grid. Results [19] show that Plumes Model calculations for average O_3 , NO_x , PAN, HNO₃ and OH agree with 20×20 km grid calculations to within a few percent. Limited tests have also been made against rural O_3 observations. In contrast, standard grid models with resolution of 80×80 km or larger may make significant errors in comparison with a 20×20 km model, overestimating O_3 and other secondary species and underestimating NO_x . Use of the Plumes Model involves some disadvantages in that (a) the model accurately provides region-wide average concentrations only, and (b) variations in rural O_3 concentrations on a spatial scale smaller than 400×480 km is represented crudely.

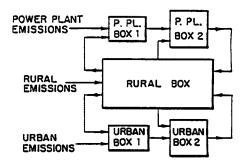


Fig. 6. Diagram of the Plumes Model subgrid structure, with boxes representing evolving urban and power plant plumes and rural area.

However it offers a large savings in computation time, and can be used more easily to explore alternative scenarios or processes that occur on a large spatial scale.

For urban applications the Plumes Model has been combined with a 5×5 km urban grid model (UGM), the latter providing an urban 'window' within the Plumes Model structure to represent local processes in a specific urban region. In the combined model the rural subsection of the Plumes Model is used to provide boundary conditions for the UGM. This combined approach is successful only when species concentrations upwind of the UGM can be represented by the 400×480 km rural average concentrations calculated by the Plumes Model. The size of the UGM must be chosen to avoid the possibility of ignoring transport from an adjacent urban area. Both the Plumes Model and the UGM use the photochemical mechanism of Lurmann et al. (complete form) [20] with photolysis rates from Logan et al. [21]. Horizontal advection is by second-order moments [22] in the Plumes Model and by the method of Smolarkiewicz [23] in the UGM. The models include two vertical layers, a time-varying mixed layer and an entrainment layer, with the diurnal variation of the mixed layer derived from van Ulden and Holtslag [24]. Deposition velocities are as follows: HNO_3 , 2.5 cm s⁻¹, O_3 and NO_2 , 0.6 cm s⁻¹; NO, 0.1 cm s⁻¹; H₂O₂ and ROOH, 1.0 cm s⁻¹; and PAN, 0.25 cm s⁻¹. Initial and boundary conditions are 40 ppb O₃, 5 ppbC VOC and 0.2 ppb NO_x. Anthropogenic emissions are from the NAPAP Version 5.2 inventory [25]. Biogenic emissions of isoprene were derived from Lamb et al. [26] based on land used data from matthews [27]. Other biogenic species were not included.

In order to represent the Detroit O₃ excursion of August 2, the combined model was exercised for a 4-day period (July 30-August 2). The Plumes Model was used to represent photochemical evolution for a region bounded roughly by Detroit, Atlanta, GA, Dallas, and Omaha, NB (Figure 7). The UGM grid covered the corridor from Detroit to Port Huron. Both model domains were chosen based on observed wind fields during the period. Regional transport in the Plumes Model was calculated based on average wind speeds between 0 and 1500 m. Transport in the UGM was based on surface winds, which were considerably lighter than winds

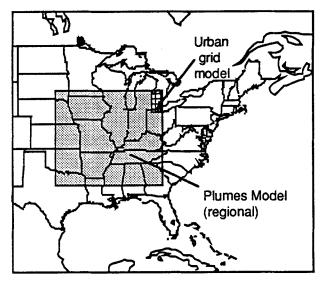


Fig. 7. Domain for the combined urban/regional simulation for July 30-August 2, 1988.

aloft. Isoprene emissions within the Detroit-Port Huron corridor were included, assuming 20% forest cover (the same percentage used for rural lower Michigan). The resulting emission rate for isoprene, adjusted for the abnormally warm temperatures of the period, was 3.15×10^{11} molecules cm² sec⁻¹ (daytime). At this rate, isoprene would account for 16% of daytime VOC emissions in the Detroit-Port Huron corridor (4% in the city of Detroit), and more than 50% of daytime VOC emissions over the regional model domain.

6. Results

The simulation predicted rural O₃ concentrations of 0.075 ppm in Michigan on August 2, with concentrations of 0.085–0.090 ppm in locations affected by diffuse urban or power plant plumes. Predicted rural O₃ compared well with observations at Saline (0.080 to 0.085 ppm) and Ann Arbor (0.075–0.080 ppm). Predicted O₃ also shows good agreement with observations at twelve rural and small urban locations throughout the midwest (Table 1). The model significantly underestimates O₃ at four rural sites (Madison, WI, Moline, IL, Decatur, IL and Paducah, KY), possibly due to the influence of local emission sources. The model overestimates O₃ at one site (Mammoth Cave, KY). The overall comparison suggests that the model accurately characterizes O₃ concentrations throughout the air mass, though it may underestimate O₃ in northwestern Illinois and Wisconsin.

Simulated urban O₃ (Table 1 and Figure 8) showed generally good agreement with observed values at sites throughout the Detroit area but had some major discrepancies. Predicted peak O₃ for August 2 (0.159 ppm) is significantly higher than the observed peak at New Haven (0.144 ppm). As shown in Figure 8 the

TABLE I
Simulated vs observed peak O₃ concentrations (ppm) for Detroit and rural Midwestern sites

	August 1		August 2	
	Simulated	Observed	Simulated	Observed
Rural				73
Ann ARbor, MI	0.072	0.080	0.076	0.080
Lima, OH	0.072	0.066	0.076	0.066
Columbus, OH	0.073	0.071	0.075	0.078
Madison, WI	0.061	0.073	0.060	0.072
Moline, IL	0.061	0.077	0.060	0.074
Quincy, IL	0.058	0.061	0.057	0.056
Decatur, IL	0.058	0.073	0.057	0.081
Terre Haute, IN	0.065	0.062	0.066	0.066
Paducah, KY	0.055	0.069	0.054	0.073
Mammoth Cave, KY	0.069	0.053	0.068	0.058
Shepherdsville, KY	0.069	0.088	0.068	0074
Livingston, KY	0.074	0.080	0;073	0.081
Urban				
Detroit, MI	0.095	0.110	0.128	0.137
Warren, MI	0.102	0.102	0.129	0.111
Southfield, MI	0.090	0.098	0.108	0.102
New Haven, MI	0.113	0.109	0.159	0.144
Port Huron, MI	0.114	0.126	0.149	0.127

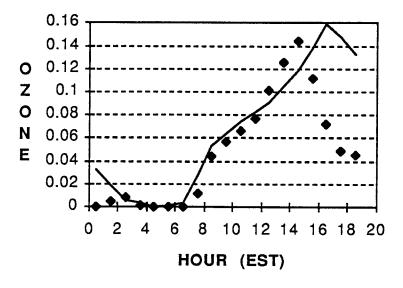


Fig. 8. Simulated O₃ at New Haven, Michigan, (line) on August 2, 1988, compared with observations (dots).

major discrepancy occurs in late afternoon, when simulated O_3 reaches its peak and observed O_3 drops sharply. This pattern suggests that the error may be associated with inaccuracies in the wind fields used in the model. The predicted geographical variation also is in close agreement with observations, with peak O_3 occurring at Port Huron on August 1 and New Heaven on August 2.

Concentrations of NO_x and VOC on August 2 are not available, but limited comparisons can be made with observations on other days. Kelly *et al.* [28] report average VOC/NO_x ratios of 4.8:1 based on 6-9 a.m. observations on 90 summer days in 1981, with slightly higher ratios (5.6:1) on days with high O_3 . The predicted VOC/NO_x ratio (4.5:1 on August 1, 6.5:1 on August 2) is consistent with these observations, although higher ratios (7:1 and 9:1) are predicted for 10 km north of Detroit. Simulated rural concentrations of VOC (~20 ppbC at noon), isoprene (0.3 ppb) and NO_x (0.5 ppb) are also broadly consistent with observations at rural sites in eastern North America [14, 29, 30].

One important aspect of the August 1–22 episode has not been represented: the unusually high O_3 concentrations in the Lake Michigan airshed. During August 1–2 high O_3 (>.15 ppm) was observed at numerous sites bordering Lake Michigan: Kenosha, Racine and Manitowoc, WI, and Muskegon, MI. A modification of the UGM is currently being planned to represent the complicated circulation pattern in the vicinity of Lake Michigan. Since surface winds throughout the Lake Michigan airshed were from the south and southwest on both August 1 and August 2, these processes were unlikely to affect the Detroit area.

7. Impact of Regional Emissions on O₃ in Detroit

The combined urban/regional model was exercised for a variety of changed VOC and NO_x emission scenarios. The specific goal of the emission scenarios was to identify the potential impact of emissions from distant upwind sources on peak O_3 in the Detroit-Port Huron corridor. We are especially interested to identify differences between abatement strategies from an analysis limited to emissions in the Detroit-Port Huron corridor, and strategies derived based on a regional-scale analysis. Consequently the model exercises included scenarios with changed emissions in the Detroit-Port Huron corridor only, and scenarios with changed emissions throughout the area covered by the regional grid.

The effect of reduced VOC emissions is shown in Figure 9. Reduced VOC emissions are predicted to lower peak O₃ within the Detroit-Port Huron corridor. However VOC reductions at upwind locations are predicted to have virtually no impact on O₃ in the Detroit area. As shown in Figure 9, the difference between the urbanonly and regional emission reduction scenarios is less than 0.005 ppm, even when region-wide reductions amount to 60%. Reduced VOC emissions also have no impact on Saline, a rural site upwind of Detroit with O₃ equivalent to the rural background. Regional VOC reductions have little impact because background rural O₃ is sensitive to NO_x rather than to VOC throughout the model domain. Direct transport of

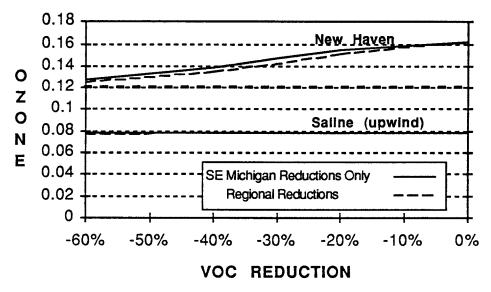


Fig. 9. Predicted peak O₃ (ppm) with reduced VOC emissions in southeast Michigan (solid lines) and with reduced VOC emissions throughout the model domain (dashed lines).

VOC into the Detroit area is insignificant in comparison to local emissions for the meteorology of this particular event.

Reduced NO_x emissions also cause a drop in Detroit O_3 , but in contrast to VOC, NO_x has a larger impact on regional transport. As shown in Figure 10, NO_x emissions have a substantial impact on rural O_3 concentrations upwind of Detroit. O_3 at New Haven drops partly in response to local NO_x reductions and partly due to regional NO_x reductions upwind of Detroit. NO_x emissions from as far away as Illinois appear to have an impact on Detroit O_3 .

The relationship between NO_x emissions and regional transport shown here is consistent with the theoretical analysis presented above and in agreement with previous studies [11, 13] linking O_3 production in rural areas to NO_x rather than to anthropogenic VOC. The impact of upwind NO_x reductions (roughly equal in size to the impact of reduced local NO_x) occurs because upwind NO_x reductions reduce background rural O_3 , which in turn reduces transport of O_3 into the Detroit area.

Although it is dangerous to make policy choices based on results of a single model for a single event, the results shown in Figures 9 and 10 have implications pertaining to the choice between VOC and NO_x controls that are likely to be repeated in subsequent analyses. First, VOC controls appear more effective than NO_x controls if only relatively modest (20%) emission reductions are made. NO_x controls appear to be comparatively more effective relative to VOC controls as the percent reduction is increased, so that if a 50% reduction is contemplated, NO_x controls appear more effective than VOC controls. This difference between the NO_x/VOC choice with a 20% reduction, in contrast to the choice with a 50% reduction, is consistent

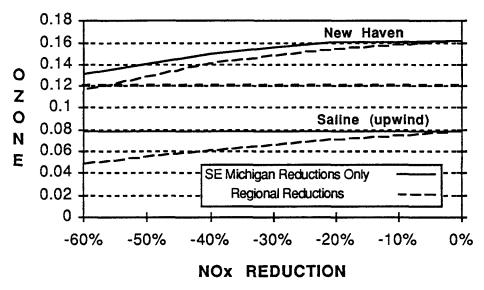


Fig. 10. Predicted peak O₃ (ppm) with reduced NO_x emissions in southeast Michigan (solid lines) and with reduced NO_x emissions throughout the model domain (dashed lines).

with the view of photochemical processes that emerged from Possiel [10]. NO_x controls appear to be more effective in reducing *moderate* O_3 , while VOC controls appear more effective in reducing *severe* O_3 . Second, the inclusion of a regional scale analysis makes the option of NO_x controls appear more attractive than it would if an urban-only analysis were used. A 50% NO_x reduction would appear equivalent to a 50% VOC reduction, if the analysis were limited to the Detroit area, but the inclusion of region-wide reductions makes a 50% NO_x reduction appear significantly more effective than a 50% VOC reduction.

8. Conclusion

A regional-scale simulation has been used to investigate control strategies for O_3 in the Detroit metropolitan area. The model combined a detailed urban grid for Detroit with a regional simulation that accounted for precursor emissions from as far away as Texas. Simulated O_3 agrees with observed concentrations at locations throughout the Detroit area and at rural sites throughout the midwest. A more extensive comparison between model results and observed urban and rural VOC and NO_x concentrations is needed in order to insure that the simulated processes correspond to atmospheric conditions.

The focus of the exercise has been to contrast the results of a regional-scale analysis for O_3 with an analysis limited to a single metropolitan area. VOC controls have little regional impact, but NO_x controls have a significant impact on regional transport of O_3 . Consequently analyses that do not include regional transport may underestimate the impact of NO_x .

If the results presented here are correct, they would seriously stress existing policy-making procedures. Under current practice in the U.S. emission controls are applied on a state-by-state basis except in locations where major metropolitan areas lie close to state boundaries (i.e. the northeast corridor). The results presented here suggest that regional transport has a significant impact even in a location (Detroit) where upwind sources lie hundreds of miles away. The results also suggest that the best VOC-reduction strategy may have no impact on regional transport. To express this at its (exaggerated) worst, the results suggest that individual metropolitan areas will benefit by reducing their own VOC emissions, and by having their upwind neighbors reduce NO_x emissions. New regulatory mechanisms may be needed to cope with this difficult situation.

References

- [1] Samson, P. J. and Ragland, K.: 1977, 'Ozone and Visibility Reduction in the Midwest: Evidence for Large-Scale Transport. J. Appl. Meteor. 16, 1101.
- [2] Hov, O., Hesstvedt, E., and Isaksen, I. S. A.: 1978, 'Long-Range Transport of Tropospheric Ozone', Nature 273, 341.
- [3] Wolff, G. T. and Lioy, P. J.: 1980, 'Development of an Ozone River Associated with Synoptic Scale Episodes in the Eastern United States', *Environ. Sci. Technol.* 14, 1257.
- [4] Logan, J. A.: 1989, 'Ozone in Rural Areas of the United States', J. Geophys. Res. 94, 8511.
- [5] Walker, H. M.: 1985, 'Ten-year Ozone Trends in California and Texas', J. Air Pollut. Control Assoc. 35, 903.
- [6] Clarke, J. F. and Ching, J. K. S.: 1983, 'Aircraft Observations of Regional Transport of Ozone in the Northeastern United States', Atmos. Environ. 17, 1703.
- [7] Samson, P. J. and Shi, B.: 1988, 'A Meteorological Investigation of High Ozone Values in American Cities', Prepared for U.S. Congress, Office of Technology Assessment, Washington, D. C., August, 1988.
- [8] Seinfeld, J. H.: 1989, 'Urban Air Pollution: State of the Science', Science 243, 745.
- [9] Scheffe, R.: 1990, The Five Cities Study, Presented at the Symposium on the Role and Importance of Nitrogen Oxides Emission Controls in Ozone Attainment Strategies for Eastern North America. Northeast States for Coordinated Air Use Management (NESCAUM), Boston, MA, May 30-June 1, 1990.
- [10] Possiel, N.: 1990, The Role of Nitrogen Oxide Emissions in Oxidant Production as Predicted by the Regional Oxidant Model (ROM), Presented at the Symposium on the Role and Importance of Nitrogen Oxides Emission Controls in Ozone Attainment Strategies for Eastern North America. Northeast States for Coordinated Air Use Management (NESCAUM), Boston, MA, May 30-June 1, 1990.
- [11] Milford, J., Russell, A. G., and McRae, G. J.: 1989, 'A New Approach to Photochemical Pollution Control: Implications of Spatial Patterns in Pollutant Responses to Reductions in Nitrogen Oxides and Reactive Organic Gas Emissions, *Environ. Sci. Tech.* 23, 1290.
- [12] Trainer, M., Williams, E. J., Parrish, D. D., Buhr, M. P., Allwine, E. J., Westberg, H. H., Fehsenfeld, F. C., and Liu, S. C.: 1987, 'Models and Observations of the Impact of Natural Hydrocarbons on Rural Ozone', *Nature* 329, 705.
- [13] Sillman, S., Logan, J. A., and Wofsy, S. C.: 1990, 'The Sensitivity of Ozone to Nitrogen Oxides and Hydrocarbons in Regional Ozone Episodes, *J. Geophys. Res.* 95, 1837.
- [14] Demerjian, K.: 1990, 'Spatial and Temporal Patterns in Ambient Concentrations of Oxidants, Reactive Organic Compounds and Nitrogenous Compounds', Presented at the Symposium on the Role and Importance of Nitrogen Oxides Emission Controls in Ozone Attainment Strategies for Eastern North America. Northeast States for Coordinated Air Use Management (NESCAUM), Boston, MA, May 30-June 1, 1990.

- [15] Fahey, D. W., Hubler, G., Parrish, D. D., Williams, E. J., Norton, R. B., Ridley, B. A., Singh, H. B., Liu, S. C., and Fehsenfeld, F. C.: 1986, Reactive nitrogen species in the troposphere: Measurements of NO, NO₂, HNO₃, particulate nitrate, peroxyacetylnitrate (PAN), O₃ and total reactive odd nitrogen (NO₂) at Niwot Ridge, Cologado, J. Geophys. Res. 91, 9781.
- [16] Michigan Department of Natural Resources: 1988, ozone monitoring sites.
- [17] Heffter, J. L.: 1981, 'Air Resources Laboratories Atmosopheric Transport and Dispersion Model (ARL-ATAD), NOAA Tech. Memo. ERL-ARL-81, Air Resources Laboratories, Silver Spring, MD, 17 p.
- [18] Foltman, R.: 1988, Detroit Edison Company, based in Detroit Edison mesoscale wind measurements network.
- [19] Sillman, S., Logan, J. A., and Wofsy, S. C.: 1990, 'A Regional-Scale Model for Ozone in the United States with a Sub-Grid Representation of Urban and Power Plant Plumes', J. Geoph. Res. 95, 5731.
- [20] Lurmann, F. W., Lloyd, A. C., and Atkinson, R.: 1986, 'A Chemical Mechanism for Use in Long-Range Transport/Acid Deposition Computer Modeling', J. Geophys. Res. 91, 10905.
- [21] Logan, J. A., Prather, M. J., Wofsy, S. C., and McElroy, M. B.: 1981, 'Tropospheric Chemistry: A Global Perspective', J. Geophys. Res. 86, 7210.
- [22] Prather, M. J.: 1986, 'Numerical Advection by Conservation of Second-Order Moments', J. Geophys. Res. 91, 6671.
- [23] Smolarkiewicz, P. K.: 1983, 'A Simple Positive Definite Advection Scheme with Small Implicit Diffusion', Mon. Wea. Rev. 11, 479.
- [24] Ulden, A. P. van, and Holtslag, A. A. M.: 1985, 'Estimation of Atmospheric Boundary-Layer Parameters for Diffusion Applications', J. Climate Apppl. Meteor. 24, 1196.
- [25] U.S. Environment Protection Agency: 1986, 'Development of the 1980 NAPAP emissions inventory', EPA-600/7-86-057a, Environmental Protection Agency, Research Triangle Park, NC.
- [26] Lamb, B., Westberg, H., Allwine, G., and Quarles, T.: 1985, 'Biogenic Hydrocarbon Emissions from Deciduous and Coniferous Trees in the United States', J. Geophys. Res. 90, 2380.
- [27] Matthews, E.: 1983, 'Global Vegetation and Land Use: New High-Resolution Data Bases for Climate Studies', J. Climate Appl. Meteorol. 22, 474.
- [28] Kelly, N. A., Ferman, M. A., and Wolff, G. T.: 1986, 'The Chemical and Meteorological Conditions Associated with High and Low Ozone Concentrations in Southeastern Michigan and Nearby Areas of Ontario', J. Air Pollut. Control Assoc. 36, 150.
- [29] Arnts, R. A. and Meeks, S. A.: 1981, 'Biogenic Hydrocarbon Contribution to Ambient Air in Selected Areas', Atmos. Environ. 15, 1643.
- [30] Sexton, KJ. and Westberg, H.: 1984, 'Non-Methane Hydrocarbon Composition of Urban and Rural Atmospheres', *Atmos. Environ.* 18, 1125.