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ESTIMATES OF OZONE RESPONSE TO VARIOUS COMBINATIONS OF NO$_x$ AND VOC EMISSION REDUCTIONS IN THE EASTERN UNITED STATES

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ABSTRACT

There is increasing recognition that controls on NO$_x$ emissions may be necessary, in addition to existing and future VOC controls, for the abatement of ozone (O$_3$) over portions of the United States. This study compares various combinations of anthropogenic NO$_x$ and VOC emission reductions through a series of model simulations. A total of 6 simulations were performed with the Regional Oxidant Model (ROM) for a 9-day period in July 1988. Each simulation reduced anthropogenic NO$_x$ and VOC emissions across-the-board by different amounts. Maximum O$_3$ concentrations for the period were compared between the simulations. Comparison of the simulations suggests that (1) NO$_x$ controls may be more effective than VOC controls in reducing peak O$_3$ over most of the eastern United States; (2) VOC controls are most effective in urban areas having large sources of emissions; (3) NO$_x$ controls may increase O$_3$ near large point sources; and (4) the benefit gained from increasing the amount of VOC controls may lessen as the amount of NO$_x$ control is increased.

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1. INTRODUCTION

Feasible solutions to the tropospheric ozone (O$_3$) problem have evaded scientists and policy makers for many years. Emissions which are precursors to O$_3$ formation, volatile organic compounds (VOC) and nitrogen oxides (NO$_x$), have to be controlled to reduce O$_3$ concentrations. The relationship between precursor controls and actual reductions in O$_3$ concentrations is highly nonlinear. Historically, efforts to reduce O$_3$ concentrations have focused primarily on VOC emissions. Recent modeling studies (Trainer et al., 1987; Sillman et al., 1990; McKeen et al., 1991) have characterized much of the eastern United States as NO$_x$-limited, suggesting that NO$_x$ controls may be more effective. However, certain areas such as New York City may be VOC-limited (EPA, 1991), where NO$_x$ emission controls may be counter-productive in reducing O$_3$ concentrations in the local area. Selection of VOC or NO$_x$ controls (or a combination of the two) will probably vary spatially, depending on the chemical regime of the location.

Previous studies, such as EPA (1991), have examined combinations of emission reductions and have provided valuable insight into the effectiveness of different controls. However, scientific understanding of the results of these studies has been hampered because of the spatial variability in emission reductions. There is a clear need for a systematic examination of NO$_x$ and VOC emission reductions to provide a basis for more effectively formulating future control strategies.

This study examines the response of maximum ozone concentrations to uniform spatial and temporal reductions in VOC and NO$_x$ emissions. The Regional Oxidant Model (ROM) (Lamb, 1983; Young et al., 1989) was used to simulate the July 2-10, 1988 period of high O$_3$.

2. PROCEDURE

A total of twenty-five simulations are planned for this study. Six have been completed, including simulations with the following sets of anthropogenic emissions (the same set of biogenic emissions were used for each simulation): (1) Base case; (2) 25% VOC and 25% NO$_x$ controls; (3) 75% VOC and 25% NO$_x$ controls; (4) 50% VOC and 50% NO$_x$ controls; (5) 25% VOC and 75% NO$_x$ controls; (6) 75% VOC and 75% NO$_x$ controls. Simulations were performed with ROM version 2.2. ROM simulates most of the physical and chemical processes responsible for the buildup of O$_3$ on regional scales ($\sim 10^6$ km^2). Detailed descriptions of the model can be found in Lamb (1983); Young et al. (1989); and Roselle et al. (1991). For this study, the modeling domain was chosen to cover the eastern United States, consisting of 16,128 grid cells within each vertical layer. The grid size was (1/4)° longitude × (1/6)° latitude, or approximately (18.5 km)^2. The model uses 3 dynamic vertical layers to represent the planetary boundary layer and capping inversion. During an entire simulation period, horizontal advection, diffusion and gas-phase chemistry for specified pollutants are modeled prognostically in the 3 vertical layers. Meteorological fields are derived by objective analysis of observed data from both surface and upper-air stations. The chemistry mechanism incorporated in the model is the Carbon-Bond Mechanism IV (Gery et al., 1989), which consists of 83 reactions and 33 individual species. In all simulations, relatively clean air ([O$_3$] = $\sim$ 30 ppb) was assumed for both the initial and lateral boundary conditions. The O$_3$ boundary condition for the top of the model was $\sim$ 40 ppb. It should be noted that there are limitations to the ROM, which include the inability to resolve sub-

grid processes, uncertainties in the input data, crude treatment of point sources, and relatively coarse horizontal and vertical resolution. Evaluations of the ROM (Schere and Wayland, 1989; Pierce et al., 1990a; Roselle, 1992) have shown that the model does well in positioning most of the major O_3 plumes, but tends to underpredict concentrations when observed values are above 70-80 ppb, and overpredict values below this level. For the current simulation, the model underpredicted maximum O_3 concentrations over the episode by 17 ppb (11%), on average.

Anthropogenic emissions were derived from the 1985 National Acid Precipitation Assessment Program (NAPAP) inventory (Saeger et al., 1989), which included emissions of VOC, NO_x, and CO (carbon monoxide). Major point, area, and mobile source emissions were considered in the inventory with day of the week (Saturday, Sunday, and weekday) and hourly variations on the emission rates. Additionally, mobile source evaporative emissions were adjusted for daily temperature variations using the empirical model MOBILE4.2 (EPA, 1989). For each emission reduction simulation, the emissions from all anthropogenic sources were reduced uniformly in space and time. Biogenic emissions were provided by the Biogenic Emissions Inventory System (BEIS) of Pierce et al. (1990b). Biogenic emissions were the same in each simulation.

3. DISCUSSION AND RESULTS

3.1 Base Case Simulation

Figure 1a presents the maximum hourly-averaged predicted O_3 concentrations for each grid in ROM's lowest vertical layer (100-300 m deep) for the 9-day period July 2-10, 1988. Maximum concentrations over the entire episode were chosen for this analysis because of their importance in determining if a location has exceeded the National Ambient Air Quality Standard (NAAQS) for ozone. In this plot, lower concentrations are denoted with lighter shades and higher concentrations with darker shades. High concentrations of ozone (>120 ppb) were predicted over a large area in association with a stagnating high pressure system. Anticyclonic circulation around the high pressure system dominated the Great Lakes, Ohio Valley, and mid-Atlantic States. The Northeast Corridor (the extended overlapping urban areas from Washington, D.C. through Boston), however, experienced flow from the southwest in association with a low pressure trough along the East Coast. High O_3 concentrations extended from central Georgia to Lake Huron, and from St. Louis, Missouri to the coast of Maine. Much of this area was predicted to have O_3 in excess of 120 ppb. Concentrations above 180 ppb were predicted near Chicago, Lake Michigan, Lake Erie, Washington, D.C., Philadelphia, and New York City. Concentrations above 150 ppb were predicted for Atlanta, Lake Ontario, Pittsburgh, Charleston (West Virginia), and along a small segment of the Ohio River. The highest predicted hourly-average O_3 concentration was 262 ppb, which occurred over Lake Michigan. The average peak O_3 concentration across the domain was 75 ppb. Predicted concentrations were relatively low (<60 ppb) over other areas, although smaller composite (over the episode) urban plumes were apparent in Texas, Oklahoma, and Louisiana.

3.2 Comparison of Simulations

25% VOC, 25% NOx Reductions. Compared to the base case (fig. 1b), predicted O_3 was reduced by 5%-15% across most of the Eastern United States for this control strategy. Ozone decreased by the largest amount (>15%) in an area over Lake Michigan, which had high concentrations in the base case. To the north and south of this area, the model predicted an increase (>5%) in O_3 for this control strategy. Areas with an increase in O_3 were widely-scattered across the domain and were primarily located near emission source regions. The spatial extent of these areas was limited to just a few grid cells.

75% VOC, 25% NOx Reductions. Figure 1c shows that for this strategy, O_3 was reduced by 15% to 30% in many areas of the Northeast, the Great Lakes, and over Texas and Louisiana. In composite urban plumes from New York City, Toronto, Montreal, Detroit, Cleveland, Chicago, and Houston, O_3 decreased by more than 30%. Other areas of the domain saw O_3 reductions on the order of 5%-15%. The largest reduction in O_3 was 56%, which occurred over Lake Michigan.

Comparing results from the simulations with 25% VOC, 25% NO_x controls and 75% VOC, 25% NO_x controls (fig. 1b and c) shows the relative benefit of adding more VOC controls to a NO_x control of 25%. Increasing VOC controls had the greatest impact on the Northeast, the Great Lakes area, and the western Gulf Coast states, as evidenced by the large areas with 15%-30% reductions in O_3. Additionally, with higher VOC controls, there were fewer grid cells showing an increase in O_3 over the base case simulation. Very little additional reduction in O_3 was predicted for the Southeast.

50% VOC, 50% NOx Reductions. In this simulation, O_3 decreased by 15%-30% over most of the model domain (fig. 1d). Over Lake Michigan and scattered over the Southeast, O_3 decreased by more than 30%. On average, peak O_3 concentrations decreased by 17%. There were a few locations with higher O_3 concentrations following the emissions reduction; the spatial extent of these regions was limited to only a few grid cells, located near large NO_x point sources.

In a similar modeling study, McKeen et al. (1991) also examined O_3 sensitivity to 50% reductions in both VOC and NO_x emissions. Their results are consistent with this study, showing a decrease in afternoon ozone concentrations of 12% to 16% across much of the eastern United States, but an increase in some grid cells which had high NO_x emissions.

25% VOC, 75% NOx Reductions. In this simulation, O_3 was reduced by more than 30% across much of the domain (fig. 1e). The controls reduced O_3 by 29% on average. Ozone decreased by more than 50% across the Southeast. There were a few grid cells near large NO_x sources which had higher O_3 in this simulation than in the base case. Some of the specific locations included Toronto, Chicago, Tampa, Houston, Baton Rouge, and New Orleans. The locations with an increase in O_3 were generally limited in spatial extent to the core urban area.

75% VOC, 75% NOx Reductions. This simulation compares well to the simulation with 25% VOC and 75% NO_x controls. Ozone decreased substantially with the emission reductions (fig. 1f); much of the East was predicted to have O_3 reduced by more than 30%. The Southeast had the largest reductions in O_3, however, the spatial extent of the area with >50% decrease in O_3 was smaller than in the simulation with 25% VOC and 75% NO_x reductions. Additionally, there were
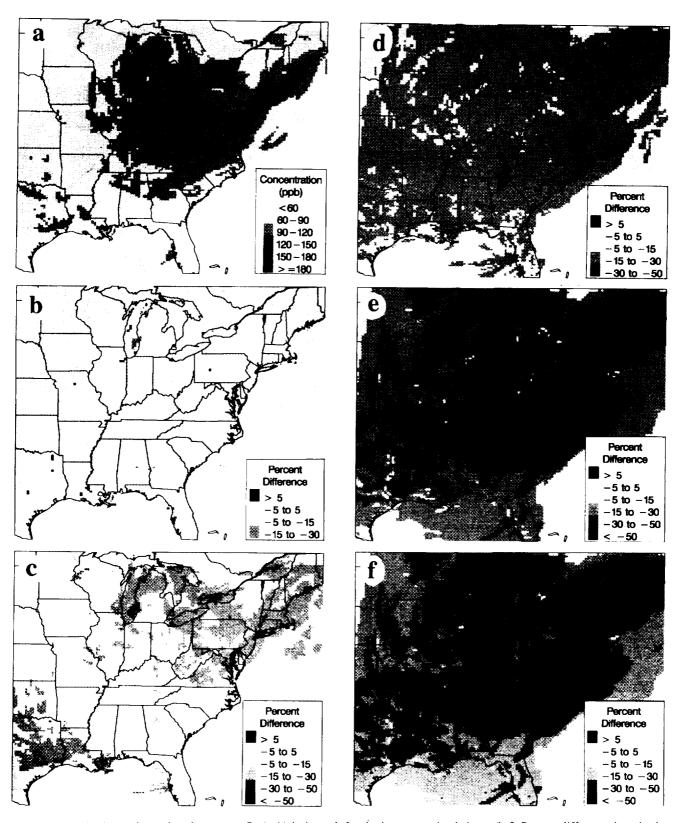
Fig. 1. (a) Episode maximum hourly-average O$_3$ (ppb) in layer 1 for the base case simulation. (b-f) Percent difference in episode maximum O$_3$ (layer 1) between the base case and simulations with anthropogenic emission reductions of: (b) 25% VOC, 25% NO$_x$, (c) 75% VOC, 25% NO$_x$, (d) 50% VOC, 50% NO$_x$, (e) 25% VOC, 75% NO$_x$, and (f) 75% VOC, 75% NO$_x$.

fewer grid cells in this simulations which had higher O₃ than the base case. The additional VOC controls examined in this simulation further improved O₃ in many of the major urban plumes, but hindered improvements in the Southeast where large NO_x controls were highly effective.

4. SUMMARY AND CONCLUSIONS

Simulations of the eastern United States were performed with the ROM for 6 different levels of anthropogenic emissions. The period of July 2-10, 1988 was studied because of the extremely high O₃ concentrations that occurred over the area. Initial results have been analyzed (summarized in Table 1) and have provided preliminary indications of the effects of different levels of emission controls. VOC controls benefit urban plumes near their sources, including the cities of Chicago, Detroit, Cleveland, Toronto, New York, Houston, and Dallas. NO_x controls produced widespread reductions in ozone. Controlling NO_x by 75% (with limited VOC control) made a much larger impact than 75% VOC control (with limited NO_x control). Reducing VOC emissions caused almost no increase in O₃, whereas NO_x controls did increase O₃ near large sources. The predicted behavior of O₃ suggests that much of the eastern United States is NO_x-limited, which is in agreement with modeling studies by Sillman et al. (1990) and McKeen et al. (1991).

As in most modeling studies, there are uncertainties that should be emphasized. In regards to scale, the ROM grid size may be too coarse to adequately resolve the fast chemical reactions which occur near large emission sources, and therefore the accuracy is reduced in model estimates of near-urban increases in O₃ due to NO_x controls. Uncertainty in model inputs is also a very important issue. It is generally accepted that anthropogenic VOC emissions are underestimated in the current inventories. For example, Fujita et al. (1992) suggest that mobile source VOC emissions are 2-3 times higher than the current estimates. Correction of the inventories for this underestimation of VOC's will only increase the preference for NO_x controls.

Further analysis will be performed on these simulations, and on the 19 simulations to be performed in the future. This paper examined only maximum predicted O₃ concentrations. The final objectives in this study are to develop response surfaces (isopleth plots) for various pollutants as a function of VOC and NO_x controls. Analysis of the response surfaces will then be performed on chemically-coherent subregions of the model domain. Upon completion, this research should provide

Table 1. Statistics on Episode Maximum Ozone[*]

Reduction Strategy	Mean Conc. (ppb)	Mean difference from base case	
		(ppb)	(%)
Base Case	75	-	-
25% NO_x 25% VOC	69	-6	-7
25% NO_x 75% VOC	66	-9	-10
50% NO_x 50% VOC	60	-15	-17
75% NO_x 25% VOC	49	-26	-29
75% NO_x 75% VOC	49	-26	-29

[*] Computed for model layer 1 over the entire domain (N = 16,128 grid cells).

policy makers with detailed information on the predicted chemical nature of the eastern U.S. and may provide additional guidance for developing control strategies.

REFERENCES

EPA, 1989: User's Guide to MOBILE4, EPA-AA-TEB-89-01, U.S. Environmental Protection Agency, Office of Mobile Sources, Ann Arbor, MI.

EPA, 1991: Regional Ozone Modeling for Northeast Transport (ROMNET), edited by N.C. Possiel, L. Milich, and B. Goodrich, EPA-450/4-91-002a, U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC.

Fujita, E.M., B.E. Croes, C.L. Bennett, et al., 1992: Comparison of emission inventory and ambient concentration ratios of CO, NMOG, and NO_x in California's South Coast Air Basin, J. Air & Waste Manag. Assoc., 42, 264-276.

Gery, M., G. Whitten, J. Killus, et al., 1989: A photochemical kinetics mechanism for urban and regional scale computer modeling, J. Geophys. Res., 94, 12,925-12,956.

Lamb, R.G., 1983: A regional scale (1000 km) model of photochemical air pollution: Part 1. Theoretical formulation, EPA-600/3-83-035, U.S. Environmental Protection Agency, Research Triangle Park, NC.

McKeen, S.A., E.-Y. Hsie, and S.C. Liu, 1991: A study of the dependence of rural ozone on ozone precursors in the eastern United States, J. Geophys. Res., 96, 15,377-15,394.

Pierce, T.E., K.L. Schere, D.C. Doll, et al., 1990a: Evaluation of the Regional Oxidant Model (version 2.1) using ambient and diagnostic simulations, EPA/600/3-90-046, U.S. Environmental Protection Agency, Research Triangle Park, NC.

Pierce, T.E., B.K. Lamb, and A.R. Van Meter, 1990b: Development of a biogenic emissions inventory system for regional scale air pollution models, in Proceedings of the 83rd Air and Waste Manag. Assoc. Annual Meeting, Pittsburgh, PA.

Roselle, S.J., T.E. Pierce, and K.L. Schere, 1991: The sensitivity of regional ozone modeling to biogenic hydrocarbons, J. Geophys. Res., 96: 7371-7394.

Roselle, S.J., 1992: Sensitivity of modeled ozone concentrations to uncertainties in biogenic emissions, EPA/600/R-92/067, U.S. Environmental Protection Agency, Research Triangle Park, NC.

Saeger, M., J. Langstaff, R. Walters, et al., 1989: The NAPAP emissions inventory (version 2): Development of the annual data and modeler's tapes, EPA-600/7-89-012a, U.S. Environmental Protection Agency, Research Triangle Park, NC.

Schere, K.L., and R.A. Wayland, 1989: EPA Regional Oxidant Model (ROM 2.0): Evaluation on the 1980 NEROS data bases, EPA-600/3-89-057, U.S. Environmental Protection Agency, Research Triangle Park, NC.

Sillman, S., J.A. Logan, and S.C. Wofsy, 1990: The sensitivity of ozone to nitrogen oxides and hydrocarbons in regional ozone episodes, J. Geophys. Res., 95, 1837-1851.

Trainer, M., E.J. Williams, D.D. Parrish, et al., 1987: Models and observations of the impact of natural hydrocarbons on rural ozone, Nature, 329, 705-707.

Young, J., M. Aissa, T. Boehm, et al., 1989: Development of the Regional Oxidant Model version 2.1, EPA-600/3-89-044, U.S. Environmental Protection Agency, Research Triangle Park, NC.