EFFECTS OF EMISSION ADJUSTMENTS ON PEAK GROUND-LEVEL OZONE CONCENTRATION IN SOUTHEAST TEXAS

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1. INTRODUCTION

Emission inventory input has long been recognized as the most predominant factor for air quality problems including ground-level O_3 formation aside from meteorological parameters (Jiang *et al*., 1997; Hanna *et al*., 1998; Bergin *et al*., 1999). The evaluation of the impact of varying emission inventory on ground-level ozone formation is usually accomplished through comprehensive modeling of the dynamic processes of emission, transport and chemistry, since the interactions between the emitted pollutants and atmospheric processes are complex and usually generate nonlinear responses.

The objective of this study is to assess the contribution of different emission sources in relation to the formation of ground-level ozone in the two southeast Texas urban areas -- Houston-Galveston and Beaumont-Port Arthur -- during the episode on August 25, 2000. The EPA's Community Multiscale Air Quality (CMAQ) modeling system is employed for conducting a series of sensitivity analyses to determine how the ground-level O_3 concentration responds to a set of designed emission inventory adjustments. In the sensitivity study, the peak O_3 concentration for each scenario is compared to that of an observation-verified, base-case simulation using the EPA's 1999 National Emission Inventory (NEI99) as the emission inventory input. Special emphasis was focused on the impact of anthropogenic VOCs and NO_x from point, mobile and area emission sources.

2. METHODS

2.1 Domain Grids and Episode

The domain is comprised of 87 x 87 Lambert Conformal grids encompassing an area of 1,089,936 km^2 with a 12-km resolution centered at 31.1N and 94.5W. The horizontal grid specification of the domain follows that of the 12-km MM5 meteorological grids, except that the outer five horizontal layers were removed to reduce the boundary effects. The selected simulation period is from August 22 to August 25, 2000. There was a Rapid Ozone Formation Event (ROFE) reported in the Houston-Galveston airshed on August 25, 2000 with ozone peak up to 194 ppbv.

2.2 Input Data and Models

The meteorological fields for the gridded emission processing and chemical transport simulations were generated during Texas Air Quality Study 2000 (TexAQS 2000) by Nielsen-Gammon (2001; 2002a; 2002b) using MM5 Release 3.4. The MM5 output files for the study domain were post-processed by EPA's Models-3 Meteorology-Chemistry Interface Processor Version 2.2 (MCIP2). During the processing, we performed a vertical layer collapsing to reduce the vertical layers from 43 sigma levels (42 vertical layers) to 22 sigma levels (21 vertical layers) for alleviating the computational costs associated with using a larger number of vertical layers.

The emission inventory data used in this study are EPA's NEI99 final version 2. The downloaded raw NEI99 data were converted into the Inventory Data Analyzer (IDA) format before being used as the input for gridded emission inventory processing. The detailed treatment and QA/QC routine for the raw NEI99 data has been presented elsewhere (Lin *et al*., 2003). The emission inventory and air quality models used in this study are the Sparse Matrix Operator Kernel Emissions (SMOKE) Modeling System Version 1.5 β and CMAQ Version 4.2.1, respectively. Both models were built on Linux platform using PGI compilers. In our CMAQ simulations, the updated SAPRC99 chemical mechanism (Carter, 1990; 1995) was employed for the chemical reactions of pollutants.

2.3 Sensitivity Analysis

A base-case simulation using the NEI99 emission inventory estimate and the MM5 meteorology was carried out to compare with the results of a number of different scenarios of emission adjustments. In the base case, simulation was accomplished based on scaling up the Texas portion of VOC emission from point sources in the NEI99 emission inventory to match the field ozone observations since previous study confirmed that the point source VOC emission had been consistently

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underestimated in the region (Daum *et al*., 2002). Since the primary objective of this study is to investigate the contribution of the precursor emission from different sources on ozone formation, the VOCs, NO_x and CO from each emission source (i.e., point, mobile, area and biogenic, etc.) were selectively reduced or removed to evaluate the difference in peak O_3 production using the same meteorological input and chemical transport schemes. Table 1 shows the sensitivity cases for various emission adjustments used in this study.

Table 1. The sensitivity cases for the emission adjustments in this study

Case No.	Emission Adjustment
1	Remove Texas point VOCs
2	Remove Texas mobile VOCs
3	Remove Texas area and non-road VOCs
4	Remove Texas point NO_x
5	Remove Texas mobile NO_x
6	Remove Texas area and non-road NO_x
7	Remove biogenic VOCs & NO_x in domain
8	Remove Texas mobile CO
9	Reduce 50% manmade VOCs & NO_x in domain
10	Reduce 50% manmade NO_x in domain
11	Reduce 50% manmade VOCs in domain
12	Remove VOCs & NO_x in non-Texas region

3. RESULTS AND DISCUSSIONS

3.1 Base-case Simulation

The base case simulation was carried out for the period from hour 00 August 22 to hour 00 August 26, 2000 UTC time. Because the ROFE occurred on August 25, 2000, we focus our base-case verification only on that particular day. The base-case simulation excellently depicts the rapid ozone formation event in Houston-Galveston area on August 25, 2000. Figure 1 shows the ozone evolution on that day. The ground-level O_3 started to form at significant rate from hour 10 local time at Galveston Bay. The O_3 plume gradually moved northwest toward inland to Houston urban area, resulting in high O_3 concentration (ROFE) then gradually dissipated as time progressed. This plume movement is consistent with field observations made by Texas Commission on Environmental Quality (TCEQ).

Figure 2 shows the scattered plots of the modeled and measured O_3 in Houston-Galveston and Beaumont-Port Arthur areas. Compared to the field observations made by TCEQ air monitoring network, the modeled time series and peak levels of O_3 agree excellently with the measured O_3 concentrations (Figure 2). The biases of the model for both urban areas are generally small. In Houston-Galveston airshed, a negative mean bias of 2.72 ppbv is observed between the modeled and measured O_3 concentrations using the data from 17 monitoring stations. For Beaumont - Port Arthur area, a

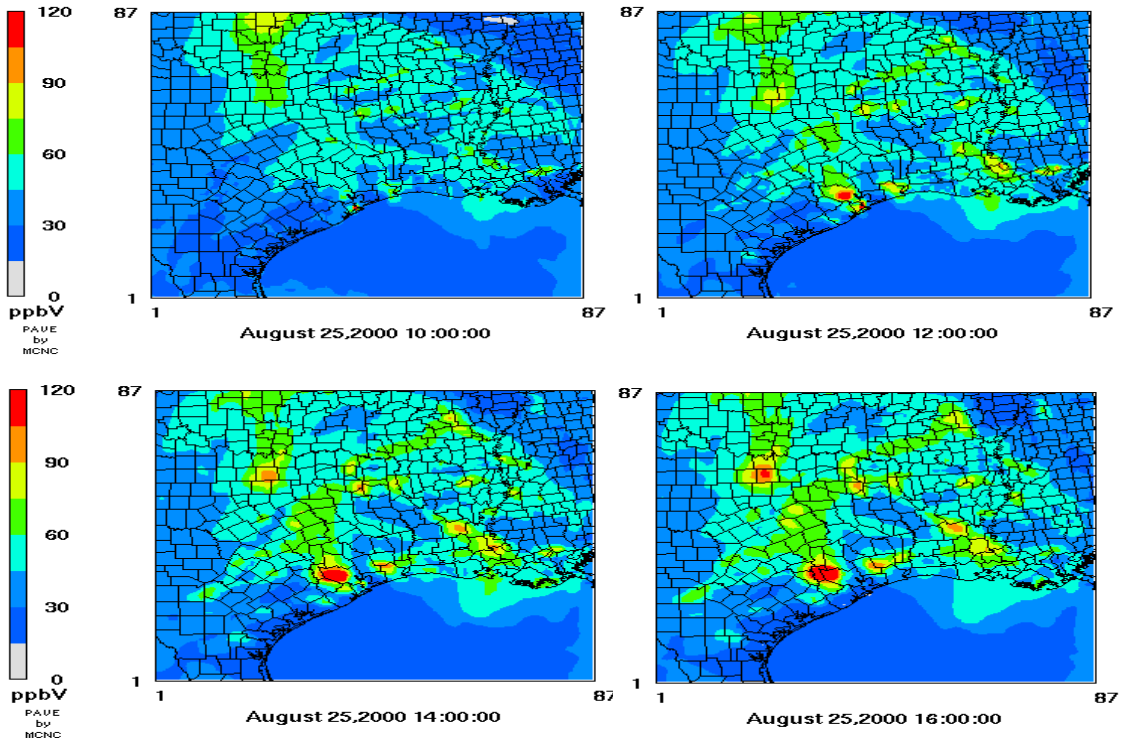

Figure 1. Modeled ozone plume for the base-case simulation from hours 10 to 16 local time on August 25, 2000. It can be clearly seen that the plume started at the Galveston Bay and moving northwest toward inland.

positive bias of 4.08 ppbv is observed using the data from 4 monitoring stations. The O_3 peak level differences are only 2 ppbv (modeled 196 ppbv and measured 194 ppbv) in Houston-Galveston and 6 ppbv (modeled 93 ppbv and measured 87 ppbv) in Beaumont-Port Arthur. This base-case simulation results are used for subsequent data comparison with the sensitivity cases.

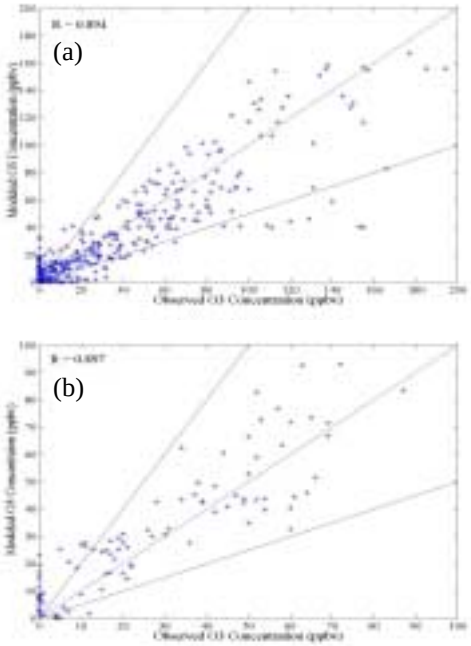

Figure 2. Scattered plot for the modeled and observed O_3 concentrations in (a) Houston-Galveston area, and (b) Beaumont-Port Arthur Area.

3.2 Sensitivity Analysis Results

Figure 3 shows the reduction in peak-hour O_3 concentration caused by the emission adjustments for the sensitivity cases shown in Table 1.

After removing VOC emission from Texas point source (Case 1), the peak hourly ozone concentration in Houston and Beaumont urban areas decreases as much as 128 and 27 ppbv, respectively, even though this emission adjustment only accounts for about 50 % of total VOC emission in Houston area. It should be noted, however, that the impact of this industrial VOC emission only affects a small number of grids in Houston-Galveston airshed for a short period of time. As winds dilute the VOC plume, the high potential of O_3 formation decreases rapidly as well. Removing Texas mobile source VOC emission (Case 2) from the emission inventory only reduces the peak O_3 concentration in Houston by 14 ppbv, although the area of impact is much greater than that of removing point source VOC emission. The removal of mobile source VOC does not seem to have significant impact on the ozone levels in Beaumont, only causing a peak O_3 reduction of 1 ppbv. Removing Texas area and non-road source VOC emission (Case 3) has similar impact as that of Case 2, resulting in 12 and 1 ppbv O_3 reduction in Houston and Beaumont, respectively. The cause for the small impact of mobile and area VOC is that mobile and area/non-road emissions are much more diffused emission sources compared to industrial emissions. Although such diffused emission can affect a greater area, the concentration of O_3 precursors from these emission sources is relatively diluted compared to those of point sources, resulting in smaller impact. Removing 50 % of anthropogenic (point, mobile, area and non-road sources combined) VOC emission for the entire domain (Case 11) leads to a peak O_3 reduction of 68 and 24 ppbv in Houston and Beaumont urban areas, respectively.

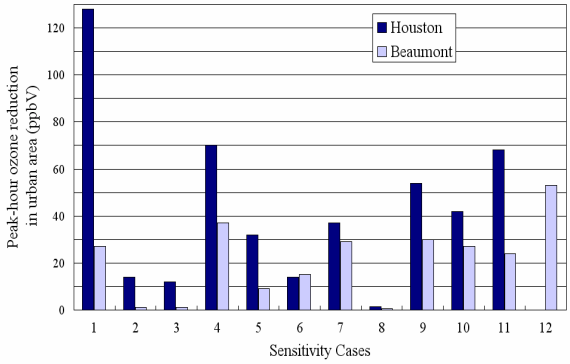

Figure 3. Reduction in peak hourly ozone concentration after the emission adjustments as shown in Table 1.

After removing Texas point source NO_x emission from the emission inventory (Case 4), the peak hourly O_3 concentration decreases by 70 and 37 ppbv in Houston and Beaumont, respectively. Compared to the base case, we also noticed that nighttime ozone increases up to 32 and 17 ppbv due to the NO_x emission adjustment. This is caused by the less degree of O_3 titration by NO in the absence of sunlight during nighttime. By removing Texas mobile source NO_x emission (Case 5), we found a decrease of O_3 concentration in a large area along the major Texas roadways. The decrease in peak O_3 level is up to 32 and 9 ppbv in Houston and Beaumont, respectively. The nighttime O_3 level is also increased by this mobile NO_x adjustment, but not as significant as in Case 4. This is mainly due to the more diffused nature of the mobile source emission compared to point source emission. Removing the area and non-road source NO_x (Case 6) results in a reduction of O_3 concentration over a large area. However, the magnitude is not as significant as in Cases 4 and 5. The greatest peak O_3 reduction was noted at Galveston at 25 ppbv. The reduction in Houston and Beaumont are 14 and 15 ppbv, respectively. Reducing the anthropogenic NO_x emission by 50 % for the entire domain (Case 10) results in a

decrease of peak ozone level of 42 and 27 ppbv in Houston and Beaumont, respectively.

Case 7 illustrates the impact of biogenic emission on ozone production in the urban areas. Biogenic emission contributes a small fraction of (< 10 %) of NO_x and large fraction (> 75 %) of VOC in the model domain (Allen and Durrenberger, 2002; Lin *et al*., 2003). However, most of the biogenic emission is located outside of the urban areas of Houston and Beaumont (Allen and Durrenberger, 2002). After removing biogenic emission of NO_x and VOCs for the entire domain (Case 7), we found a decrease of peak ozone concentration of 37 and 29 ppbv at the northwest side of Houston and the north side of Beaumont, respectively. The decreased O_3 level occurs only outside of the urban areas, indicating that the biogenic emission may not contribute to the urban non-attainment in Houston-Galveston and Beaumont-Port Arthur. Instead, the contribution is much more important in the region where high density of vegetation is located. In this sensitivity case, we also found that there is a large reduction of O_3 plume size in the east Louisiana region, with peak O_3 reduction at 37 ppbv. Reducing the anthropogenic VOC and NO_x emission by 50 % for the entire domain (Case 9) results in the decrease of the peak hourly O_3 concentration of 54 and 30 ppbv in Houston and Beaumont, respectively. In Houston area, such impact is less significant compared to Case 11 where only VOC emission adjustment (50 % reduction) is made. This is due to the VOC-limited condition in Houston urban area after the removal of 50 % of VOC from the emission inventory. Under such condition, reduction of NO_x may lead to greater O_3 production due to less daytime ozone titration by NO (Seinfeld and Pandis, 1998). Removing the anthropogenic emission of NO_x and VOCs from non-Texas region (Case 12) does not affect the O_3 formation scenario in Houston-Galveston area. However, the peak hourly O_3 concentration in north Beaumont decreases by up to 53 ppbv. This indicates that the O_3 formation in Beaumont can be affected by the transport of precursors from Louisiana due to its proximity to the state border. Reducing CO emission does not cause significant reduction in ozone levels in both urban areas (Case 8).

4. ACKNOWLEDGEMENT

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5. REFERENCES CITED

Allen D. T. and Durrenberger C. (2002) Accelerated science evaluation of ozone formation in the Houston-Galveston area: emission inventories, *Texas Conservation on Environmental Quality Science Synthesis Committee Report*, Austin, Texas US.

Bergin M. S., Noblet G. S., Petrini K., Dhieux J. R., Milford J. B. and Harley R. A. (1999) Formal uncertainty analysis of a Lagrangian photochemical air pollution model. *Environmental Science and Technology* **33**, 1116-1126.

Carter W. P. L. (1990) A detailed mechanism for the gas-phase atmospheric reactions of organic compounds. *Atmospheric Environment* **24 A**, 481-453.

Carter W. P. L. (1995) Computer modeling of environmental chamber measurements of maximum incremental reactivities of volatile organic compounds. *Atmospheric Environment* **29**, 2513-2527.

Daum P., Meagher J., Allen D. T. and Durrenberger C. (2002) Accelerated science evaluation of ozone formation in the Houston-Galveston area: summary, *Texas Conservation on Environmental Quality Science Synthesis Committee Report*, Austin, Texas US.

Hanna S. R., Chang J. C. and Fernau M. E. (1998) Monte Carlo estimates of uncertainties in predictions by photochemical grid model (UAM-IV) due to uncertainties in input variables. *Atmospheric Environment* **32**, 3619-3628.

Jiang, W., Singleton, D. L., Hedley, M. and Mclaren, R. (1997) Sensitivity of ozone concentrations to VOC and NO_x emissions in the Canadian lower Fraser valley. *Atmospheric Environment* **31**, 627-638.

Lin C.-J., Ho T. C., Chu H., Yang H., Mojica M. J., Krishnarajanagar N., Chiou P. and Hopper J. R. (2003) Comparison of EPA 1996 and 1999 national emission inventories: application in the West Gulf Coast Region, *Journal of Environmental Informatics*, in press.