

# A COMPARISON OF PM 2.5 SIMULATIONS OVER THE EASTERN UNITED STATES USING CB-IV AND RADM2 CHEMICAL MECHANISMS

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## 1. INTRODUCTION

Fine particulate matter (PM<sub>2.5</sub>) in the atmosphere has been a great concern in recent years, affecting human health, visibility (regional haze), and climate change. Unlike ozone being treated as a single pollutant, PM<sub>2.5</sub> consists of mixtures of hundreds of components over a wide size distribution resulting from various physical and chemical processes that make PM<sub>2.5</sub> a difficult pollutant to model. PM modeling evaluation is currently difficult, due to the lack of adequate data for comparison. Majority of the studies to date used sparse 24-hour PM<sub>2.5</sub> measurements taken either on a daily basis or even twice a week, resulting in very limited information to evaluate the performance of the model. However, several hourly TEOM measurements for PM<sub>2.5</sub> are becoming available that could be used in the assessment of PM models.

The objectives of this study were to assess the MODELS-3/CMAQ (Byun and Ching, 1999), in simulating PM as well as the sensitivity of the chemistry in the model in terms of PM formation. In this study, we applied CMAQ for the period from July 8 to 18, 1999 over the eastern United States with both CB-IV and RADM2 chemistry. The simulation results will be compared with hourly TEOM measured data. We also compared the sensitivity of the chemical mechanisms of the model response to the emission reductions. Two emission reduction scenarios were simulated. The first scenario was a reduction of elevated SO<sub>x</sub> emissions by 50% and the second scenario was a combination of 50% reduction in SO<sub>x</sub> and 50% reduction in NO<sub>x</sub> from those elevated emission sources. Preliminary analysis of the simulations indicated that the response from RADM2 chemical mechanism is more sensitive to the emission reduction than CB-IV chemistry.

## 2. MODEL SIMULATIONS

The CMAQ horizontal grid size was set to 36 km with 79 cells along the east-west direction and 73 cells in the north-south direction covering a large portion of the eastern United States. There are 16 layers in the vertical

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direction, with the first 12 layers (below 3124 m), being identical to the setup of MM5 simulations (Ku et al, 2001). The initial conditions were set to background concentrations starting July 8. Since there were not sufficient observational data available, the hourly boundary conditions were extracted from the previous day's predictions in order to avoid using constant boundary conditions for the entire simulations. This approach had been applied to the base case as well as emission reduction simulations.

MM5 (Grell et al, 1994) was used to generate meteorological inputs for CMAQ. The details of the MM5 simulations can be found in Zhang et al (2001). The emission data is based on 1995 aerosol data provided by US EPA. There were two emission data sets corresponding to CB-IV and RADM2 chemistry, respectively. For both set of emission data, the aerosol emissions include only PM<sub>2.5</sub> and PM<sub>10</sub> and do not contain information about size distributions or chemical speciation.

## 3. MEASUREMENT DATASET

Hourly PM<sub>2.5</sub> measured with Tapered Element Oscillating Microbalance (TEOM) instruments, collected by routine monitoring networks in NY, NJ, NC, were retrieved through the EPA Aerometric Information Retrieval System (AIRS). In addition to the state-level monitoring data, we also obtained hourly TEOM measurements from the SouthEastern Aerosol Research and Characterization (SEARCH) network (<http://www.atmospheric-research.com>) and the TEOM measurements by the Department of Energy at sites Lawrenceville and Holbrook, PA. There are a total of 15 TEOM sites used in this study.

## 4.0 RESULTS AND DISCUSSIONS

### 4.1 Spatial Patterns of Daily Averaged Aerosol Concentrations

Figure 1 shows the daily averaged PM<sub>2.5</sub> mass concentrations on July 15, 1999 from both CB-IV and RADM2 chemistry. Both show a similar pattern with high concentrations extending from Indiana down to Alabama in the center portion of the modeling domain. However, magnitude wise, the RADM2 predicted PM<sub>2.5</sub> concentrations almost twice as higher than CB-IV

in the high concentration areas. Recognizing in recent studies indicated that the fine aerosol are dominated by sulfate in the Eastern of United States (Blanchard et al, and 2003, Mebust et al, 2003). It is of interesting to compare the differences in PM speciation that is due to the different chemistries. We calculated the percentage contribution of sulfate and nitrate to the total PM mass for the two simulations. The CB-IV based simulation showed relatively lower sulfate and higher nitrate contributions to the total PM<sub>2.5</sub> mass compared to those of the RADM2. In the high PM prediction areas, the contribution from sulfate is about 25 to 50 percent based on CB-IV chemistry, while it reaches about 70 to 85 percent based on RADM2 chemistry.

#### 4.2 Compare with TEOM hourly measured data

Figure 2 is the scatter plot between predicted and observed hourly PM concentrations over 15 TEOM measurements covering the period from July 13 through July 18, 1999. The one-to-one line and the best fit regression lines were also included in the plot. The CB-IV underpredicted the slope (0.19), however the large value of intercept (21.06) would compensate the underpredictions. The RADM2 showed almost a one-to-one slope fit (0.91), along with large value of intercept (26.90), which led to overpredict TEOM data by RADM2. The high intercept value of both CB-IV and RADM2 may indicate the systematically high bias of the model comparing with TEOM data. However, studies have indicated that TEOM measurements are bias low comparing with Filter Reference Method (FRM) measurements. Statistically, both CB-IV and RADM2 predictions were higher than the hourly TEOM measured data, with a bias of 1.43  $\mu\text{g m}^{-3}$  for CB-IV and 24.69  $\mu\text{g m}^{-3}$  for RADM2. The standard deviations are 21.54  $\mu\text{g m}^{-3}$  for CB-IV and 23.55  $\mu\text{g m}^{-3}$  for RADM2, respectively. The correlation coefficients are higher for RADM2 (0.56) than CB-IV (0.20).

#### 4.3 Emission change simulations

One of the important applications of modeling is to examine the effects of emission changes to achieve a reduction in predicted concentrations. To investigate if the different chemical mechanism (CB-IV and RADM2), may respond differently, we performed two emission reduction simulations; (1) 50% across-the-board SOx emissions reduction from elevated point sources, (2) 50% across-the-board SOx emissions reduction plus 50% across-the-board NOx emissions reduction from elevated point sources, for both CB-IV and RADM2. The intent is to compare the sensitivity of the chemical

mechanism to the change in emissions of SOx and NOx. Figure 3 displays the percentage concentration reduction of daily averaged PM<sub>2.5</sub> concentrations for July 15, 1999 to illustrate the difference in the emission reduction response of the chemistry mechanism. Shown in the top two panels (A and B) are the comparisons of the 50% SOx emission reduction, indicating a greater sensitivity in changing concentrations from RADM2 than CB-IV. When reducing the elevated SOx point sources by 50%, the RADM2 predicted about a 25% decrease in PM<sub>2.5</sub> concentrations, compared to less than about 10% decrease from CB-IV. The additional 50% NOx reduction (see panels C and D), both chemistry mechanisms show no significant change in the PM<sub>2.5</sub> concentrations reduction from 50% SOx reduction only. The CB-IV shows increase areas of impact in the Great Lake areas. When reducing SOx both RADM2 and CB-IV show an increase in nitrate, with the RADM2 predicting a 30% increase, but only about 3% from CB-IV. This finding is in agreement with the current thinking on SO<sub>2</sub>-NO<sub>x</sub>-NH<sub>3</sub> chemistry, as decreasing SOx emission may make more NH<sub>3</sub> available and increase particulate NH<sub>4</sub>NO<sub>3</sub> concentrations (Blanchard et al, 2003).

#### 5. SUMMARY

In this paper, we examined the uncertainties in CMAQ simulating PM concentrations stemming from the use of CB-IV and RADM2 chemical mechanisms. In general, RADM2 chemistry predicted higher PM<sub>2.5</sub> concentrations than CB-IV. Both CB-IV and RADM2 show fair comparisons with hourly TEOM PM<sub>2.5</sub> data at several sites over the eastern United States. The difference in the treatment of PM<sub>2.5</sub> compositions among CB-IV and RADM2 chemistry caused the difference in the response to the emission change. It is important to do further simulations to evaluate which chemical mechanism might be more robust to simulating the formation of PM<sub>2.5</sub>. Finally, the lack of continuous PM and corresponding speciation are some of the limiting factors in the pursuit of multi-pollutant one atmosphere modeling to address the emerging air quality issues.

#### 6. REFERENCES

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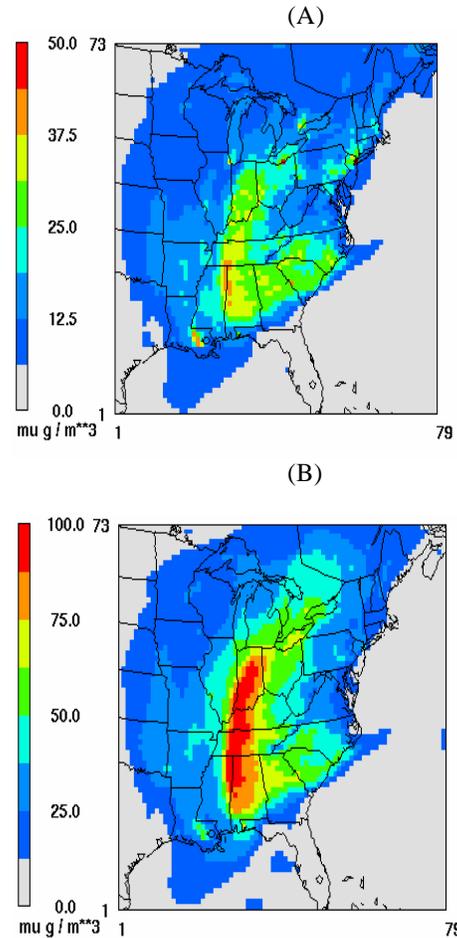


Figure 1. The daily averaged PM<sub>2.5</sub> concentrations on July 15, 1999 of (A) CB-IV, and (B) RADM2

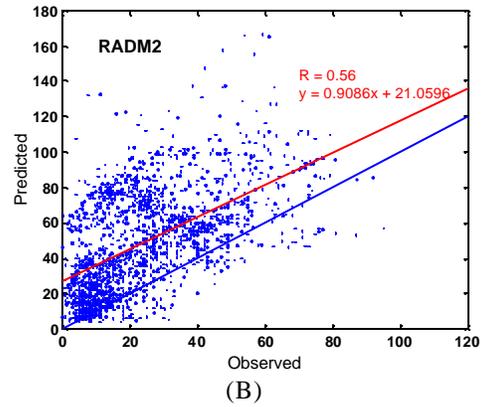
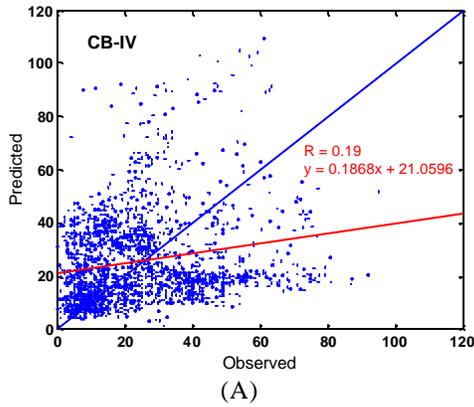
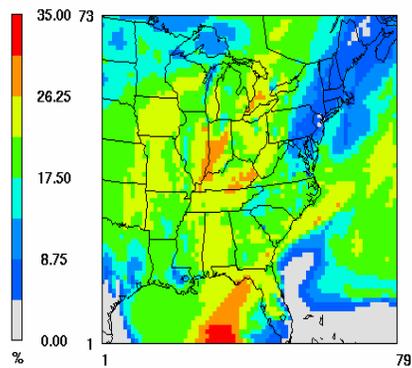
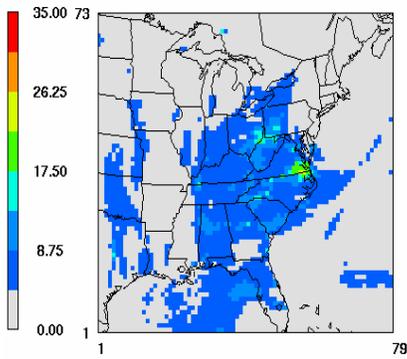


Figure 2: Scatter plots between observed and predicted hourly PM concentrations over 15 TEOM measurements covering from July 13 to 18, 1999 for (A) CB-IV and (B) RADM2.

(A)

(B)



(C)

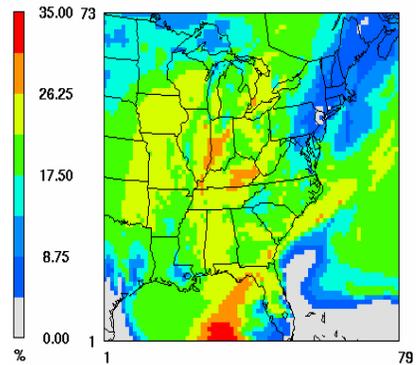
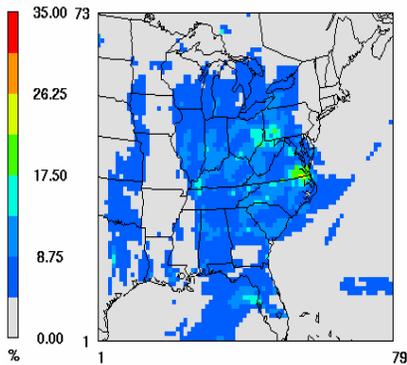


Figure 3: The percentage concentration reduction of daily averaged PM<sub>2.5</sub> concentration of July 15, 1999. (A) CB-IV and (B) RADM2 for the case of 50% SO<sub>x</sub> reduction; (C) CB-IV and (D) RADM2 for the case of 50% SO<sub>x</sub> and 50% NO<sub>x</sub> reduction.

