DEVELOPMENT AND APPLICATION OF MADRID: A NEW AEROSOL MODULE IN MODELS-3/CMAQ

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1. INTRODUCTION

A new aerosol model, the Model of Aerosol Dynamics, Reaction, Ionization and Dissolution (MADRID) [1, 2] has been developed to simulate atmospheric particulate matter (PM). MADRID was developed based on (1) a comprehensive review of existing modules of aerosol dynamics, thermodynamics and chemistry, (2) the selection of modules that offered the best compromise between numerical accuracy and computational efficiency and (3) the integration of these modules to form a coherent model. MADRID and the CMU bulk aqueous-phase chemistry[3] have been incorporated into the 3-D Models-3/Community Multiscale Air Quality model (CMAQ) [4]. The resulting model, CMAQ-MADRID, was applied to simulate the 27-28 August 1987 episode of the Southern California Air Quality Study (SCAQS) in the Los Angeles (LA) basin. Its performance was evaluated for O_3, $PM_{2.5}$, PM_{10} and PM chemical components, and compared with results from other PM models. Sensitivity studies were conducted to evaluate the sensitivity and the sources of uncertainties in model predictions. CMAQ-MADRID was provided to EPA for public utilization in October 2002.

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2.0 MODEL DESCRIPTION

MADRID uses ISORROPIA[5] to simulate the thermodynamics of inorganic PM species. SOA formation is treated using two modules: an empirical representation (referred to as MADRID 1) that is based on a reversible absorption theory and data obtained in smog chamber experiments[6, 7] and a mechanistic representation (referred to as MADRID 2) that simulates an external mixture of hydrophilic and hydrophobic particles[8]. The moving-center technique of Jacobson[9] is used to simulate particle growth by condensation or shrinkage by volatilization, which is the most accurate approach among four existing algorithms that we tested[10]. The formation of new sulfate particles is simulated using the approach of McMurry and Friedlander[11], following the recommendation of Zhang et al.[10] Gas/particle mass transfer is simulated with two algorithms: the CIT bulk equilibrium approach that assumes full equilibrium between gas and particulate phases but allocates transferred mass to different size sections based on condensational growth law[12], and the CMU hybrid approach that treats mass transfer explicitly for coarse particles and assumes full equilibrium for fine particles[13]. The CMU cloud chemistry[3] includes 34 gas-aqueous and aqueous equilibria and 99 aqueous-phase kinetic reactions. It simulates the formation of sulfate and nitrate and reactions for carbonate, chlorine, organic and oxygen species that are involved in sulfate and nitrate formation. Heterogeneous reactions are simulated following Jacob[14].

3.0 APPLICATION OF CMAQ-MADRID

CMAQ-MADRID was applied to simulate the 27-28 August 1987 SCAQS episode in the LA basin. The meteorological fields were simulated using the meteorological mesoscale model MM5 with four-dimensional data assimilation. A post-processing step was added to develop a diagnostic 4-D field of mixing heights using aloft data available throughout the basin. Emissions of gases and PM generally followed Pai et al.[15] The total PM mass emissions and chemical speciation were obtained from Meng et al.[12] Both 2- and 8-section representations of the particle size distribution were used. For 2-section representation, PM emissions were assigned to sections according to the default size distribution of CMAQ[4]. Initial and boundary conditions for gases were based on Pai et al.[15]

Figure 1 shows the observed and simulated 24-hr average concentrations for $PM_{2.5}$ and its chemical compositions on August 27 and 28 at three sites: Hawthorne (HAWT), Los Angeles (CELA), and Riverside (RIVR). They represent an upwind coastal site in the western basin, a downtown area with high motor vehicle emissions, and a downwind urban site in the eastern basin, respectively. The observed concentrations at the three sites show the evolution of $PM_{2.5}$ and PM_{10} across the basin from the coast to the inland, with relatively low concentrations near the coast but significantly higher concentrations as the air mass was transported across the basin. PM reached its highest level among all monitoring sites at RIVR on both days. The simulations with both size resolutions reproduce well the observed evolution of PM and its composition, although the model tends to overpredict PM concentrations at HAWT and CELA on August 27 but underpredict those on August 28 at all sites, particularly at RIVR. This can be partially attributed to the overpredictions on August 27 and the underpredictions on August 28 in the concentrations of precursors for secondary PM as a result of mispredicted wind speeds and vertical mixing. Other factors (e.g., uncertainties in emissions of primary PM species and precursors of secondary PM species) also contributed, to some extent, to the inaccuracies in the model predictions.

Figure 2 compares the predicted PM mass size distribution on August 28, 1987 with the measured PM mass size distribution obtained by Hering et al.[16] at Claremont (CLAR) and RIVR. We discuss here the results obtained with the moving-center scheme for particle growth. Results with a finite-difference scheme will be discussed in section 4. The model correctly predicts a unimodal distribution for accumulation mode PM that is typical at RIVR during the summer time but it fails to reproduce the typical bimodal distribution of accumulation mode PM at CLAR (A bimodal distribution was however, predicted for some periods at CLAR and other sites). The predicted peak value at RIVR is higher than that at CLAR and both peak values occur in the same size range, consistent with observations. However, the PM mass concentration peaks in the size section of 0.215-0.464 μm at both sites, which is somewhat off from the diameter of the observed peak values (i.e., 0.52 μm). The peak values are underpredicted by 30% at Claremont and 16% at Riverside. In addition to the aforementioned factors that contribute to differences between observations and model results, a finer size resolution (> 8 sections) is needed to accurately simulate PM size distribution.

The model performance is evaluated following the existing guidance[17]. Our evaluation focuses on the mean normalized gross error (MNGE) and mean normalized bias (MNB) in the O_3 and PM predictions at the sampling sites. The MNGE and MNB in O_3 predictions at 38 sites with a cut off value of 40 ppb are 34% and 9% for August 27-28. The predicted mean $PM_{2.5}$ and PM_{10} concentrations averaged over 8 PM sites are 51.8 and 91.7 μg m^{-3} on August 27 and 43.4 and 81.3 μg m^{-3} on August 28, which overpredict the observed values of 42.3 and 75.9 μg m^{-3} by 21-22% on August 27 but compare well with the observed values of 48.1 and 85.3 μg m^{-3} on August 28. The MNGE and MNB in the predicted $PM_{2.5}$ concentrations are 42% and 30% on August 27 and 47% and -2% on August 28. The model performance statistics for $PM_{2.5}$, PM_{10} and their compositions are consistent with those obtained with the other PM models[12, 18, 19].

4.0 SENSITIVITY STUDY

We have studied the sensitivity of the model predictions to various model formulations including heterogeneous reactions, SOA formation, particle growth due to condensation (or shrinkage due to volatilization) and aqueous-phase chemistry as well as gas/particle mass transfer. As an example, Figure 2 demonstrates the model sensitivity to two different particle growth schemes: a simple finite-difference scheme and the moving-center scheme at CLAR and RIVR on August 28. The finite-difference scheme moves mass from section to section during each time

step, causing significant numerical diffusion. Whereas, the moving-center scheme only moves mass from section to section when the section center grows out of the section, therefore minimizing the numerical diffusion. Another fundamental difference between the moving-center and the finite-difference schemes is that the former predicts both PM mass and number concentrations, whereas the latter predicts only the PM mass concentrations and diagnoses the PM number concentrations from the predicted PM mass and the fixed PM mean diameters. Compared to the observed size distributions, the finite-difference scheme tends to predict a diffusive type of distribution for $PM_{2.5}$, with high concentrations in size sections 1 and 2 (0.0215-0.0464 μm and 0.0464-0.1 μm, respectively) at both sites, resulting in an underprediction in PM mass concentration of accumulation mode. In contrast, the moving-center scheme predicts a size distribution that is closer to the observations in terms of the magnitudes and general shape of size-resolved PM composition, although the peak PM mass is somewhat underpredicted and is off the observed size for peak PM mass.

5.0 CONCLUSION

A new aerosol model, CMAQ-MADRID, was applied to simulate an air pollution episode in the LA basin. Model performance for both O_3 and PM predictions was consistent with existing guidance. The sensitivity of model predictions was evaluated with respect to several major areas of uncertainties in PM modeling including the treatment of heterogeneous reactions, and different modules/algorithms for SOA formation, particle growth due to condensation and aqueous-phase chemistry as well as gas/particle mass transfer. Our study has identified several factors that are critical to the accuracy of the model predictions. These include accurate model inputs (e.g., emissions and meteorology), realistic representations of various atmospheric processes (e.g., SOA formation), appropriate numerical algorithms for PM dynamics (e.g., condensational growth and gas/particle mass transfer) and fine particle size resolution (e.g., > 8 size sections).

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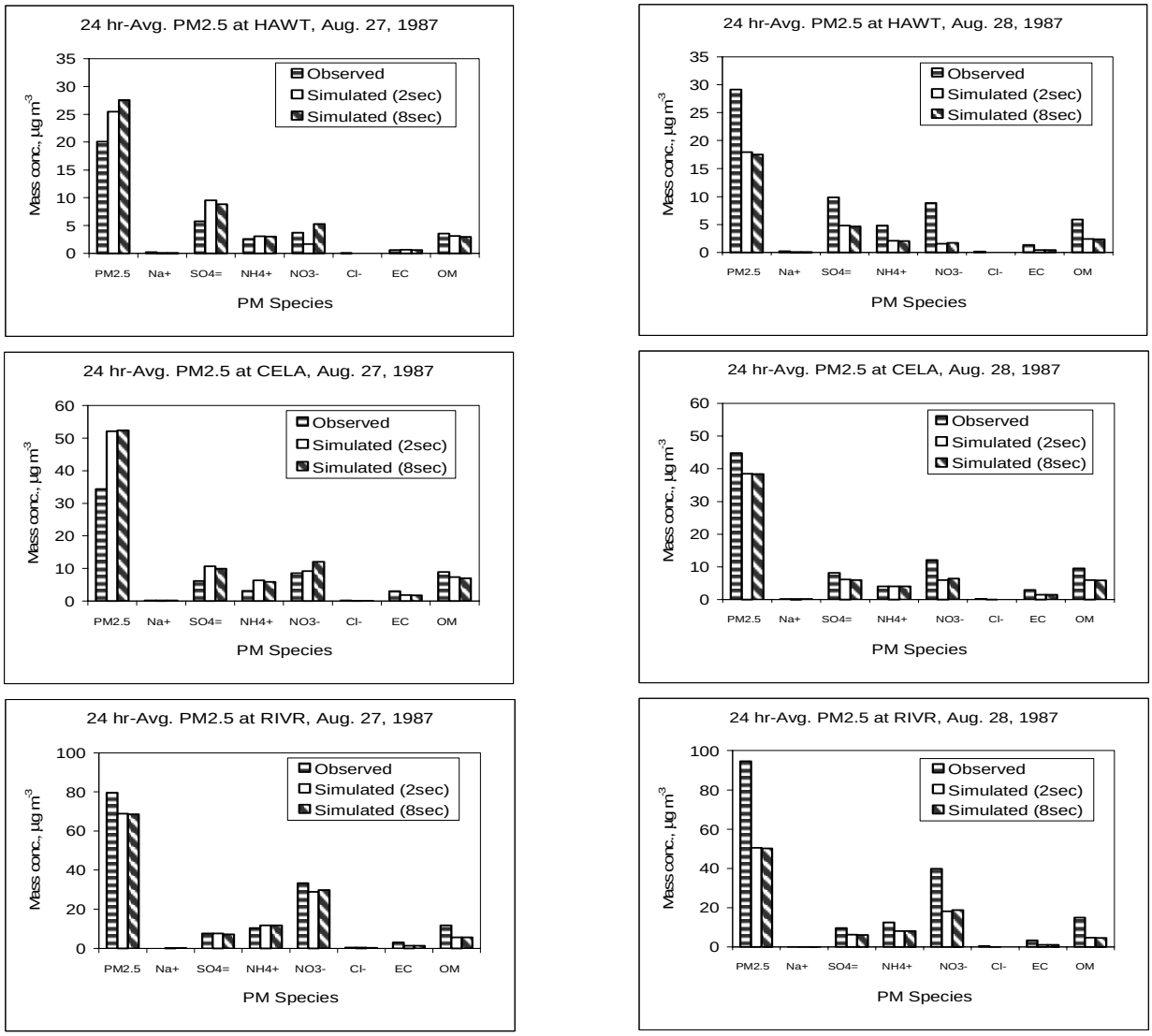

Fig. 1. Observed and predicted 24-hr average concentrations for $PM_{2.5}$ and its chemical compositions on August 27-28, 1987 at Hawthorne [HAWT], Central LA (CELA), and Riverside (RIVR), CA.

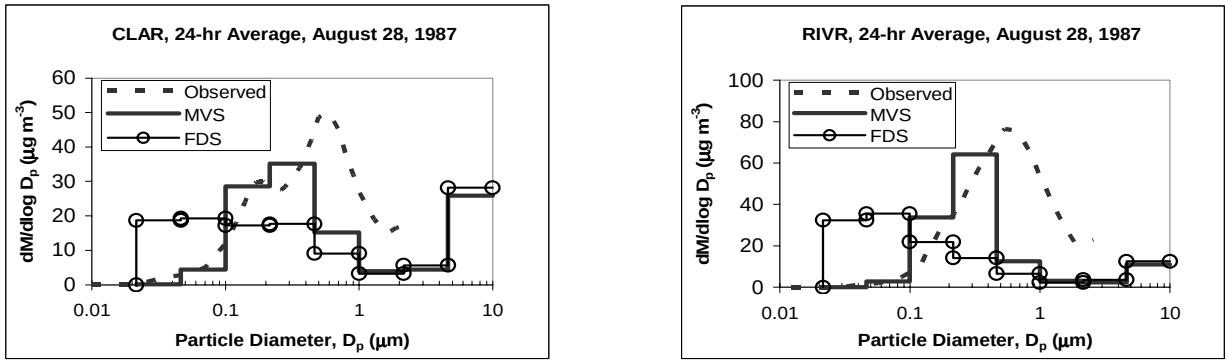

Fig. 2. Observed vs. simulated average PM size distribution at CLAR and RIVR on August 28, 1987 with the moving-center scheme (MVS) or the finite-difference scheme (FDS) for particle growth.