SIMULATIING AEROSOL MASS AND SIZE DISTRIBUTIONS OVER CALIFORNIA USING CMAQ-MADRID

Xiao-Huan Liu[1,3], Ping Liu[1], Yang Zhang[1*], Betty Pun[2], and Wen-Xing Wang[3]

[1]Department of Marine, Earth, and Atmospheric Sciences,
North Carolina State University, Raleigh, NC
[2]Atmospheric and Environmental Research, Inc., San Ramon, CA
[3]Environment Research Institute, Shandong University, Jinan, Shandong Province, P.R China

1. INTRODUCTION

Central California (CA) is a region with the highest ambient particulate matter (PM) concentrations in the United States (U.S.). San Joaquin Valley (SJV) located in the central CA is the main contributor to the high PM concentrations (Chow et al., 2006; Herner et al., 2006; Herner et al., 2005; Ying and Kleeman, 2006). SJV is surrounded by the coastal mountain ranges on the west and by the Sierra Nevada ranges on the east. The San Francisco Bay Delta is the only outlet to the ocean. During winter, a high-pressure system named the Great Basin High often stays over this region for several days to weeks, resulting in elevated temperatures aloft, low wind speeds, and low pressure gradients (Herner et al., 2005, 2006). Under such a condition, pollutants emitted are trapped in the area, leading to high PM concentrations. PM has significant effects on human health, visibility, and climate. Current regulations exist for PM with an aerodynamic diameter less than or equal to 2.5 or 10 µm ($PM_{2.5}$ or PM_{10}, respectively), but research suggests that PM with diameter smaller than 0.1 µm ($PM_{0.1}$) may have equally serious health effects to human beings. The peak concentrations of $PM_{0.1}$ in central CA are among the highest in the U.S. (Herner et al., 2005). It is therefore important to simulate PM mass and size distributions for the development of effective emission control strategies for PM attainment in this area.

In this work, the Model-3 Community Multiscale Air Quality Modeling system with the Model of Aerosol Dynamics, Reaction, Ionization, and Dissolution (CMAQ-MADRID 1) (Zhang et al., 2004; Pun et al., 2006) is applied to the central and northern CA to simulate air pollutants for the period of December 25-31, 2000 from the California Regional PM_{10}/$PM_{2.5}$ Air Quality Study (CRPAQS). CRPAQS was conducted over SJV and surrounding air basins (i.e., Sacramento Valley and San Francisco Bay) during December 2, 1999 through February 3, 2001 to understand the causes of high PM concentrations for PM attainment in central CA (Chow et al., 2006). A model evaluation against CRPAQS monitoring data (e.g., PM mass concentration and size distribution) is conducted to evaluate the model performance and analyze likely causes of the model biases. The objectives of this study are to simulate PM mass and number concentrations as well as size distribution in central CA and to understand formation mechanism underlying the high PM concentrations to support $PM_{2.5}$ attainment effort in this area.

2. MODEL SIMULATION SETUP

CMAQ-MADRID 1 simulations are conducted at a 4-km horizontal grid spacing (185×185 horizontal grid cells) over a domain that covers the central and northern CA, including the entire SJV and a portion of Nevada, as shown in Fig. 1. The vertical resolution includes 15 layers from surface to tropopause. The meteorological fields and chemical inputs such as emissions and initial and boundary conditions are provided by AER, Inc. More detailed description on model configurations along with inputs can be found in Pun et al. (2008).

12 and 24 sections are used in separate simulations to represent PM size distribution from 0.001 to 10 µm. Coagulation is simulated in the baseline simulations. Sensitivity simulations are also conducted without coagulation to investigate the impacts of coagulation on the simulated particle number concentrations and size distributions. All simulations use the Statewide Air Pollution Research Center (SAPRC99) gas-phase chemical mechanism and the Carnegie-Mellon University (CMU) aqueous-phase chemical mechanism.

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*Corresponding author: Yang Zhang, Department of Marine, Earth, and Atmospheric Sciences, Campus Box 8208, NCSU, Raleigh, NC 27695; e-mail: yang_zhang@ncsu.edu

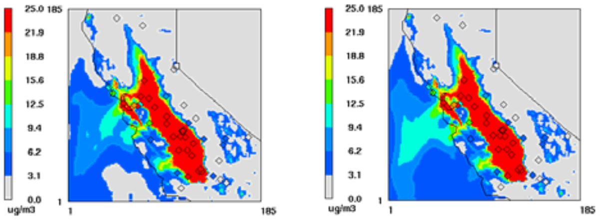
Fig. 1. CMAQ-MADRID 1 modeling domain.

3. MODEL RESULTS

3.1 Comparison between 12- and 24-Section Simulations

3.1.1 Spatial distribution and performance statistics of $PM_{2.5}$

Fig. 2 shows the spatial distributions of observed and simulated weekly-mean $PM_{2.5}$ mass concentrations. Both 12- and 24-section simulations predict extremely-high $PM_{2.5}$ concentrations in the SJV area. Observed high $PM_{2.5}$ is mainly caused by the strong stagnation event occurred from December 16, 2000 to January 7, 2001 (Herner et al., 2005). Simulated and observed mass concentrations of $PM_{2.5}$ generally agree well at most sites except a few sites located to the southeast of SJV (i.e., Death Valley Park, Mojave Station), with Normalized Mean Bias (NMBs) of -2.7% and 3.6% for the 12- and 24-section simulations, respectively.



(a)12-section (b) 24-section

Fig. 2. Observed and simulated spatial distributions of weekly-mean $PM_{2.5}$ mass concentrations from simulations with (a) 12 and (b) 24 PM size sections.

Table 1 gives the performance statistics of CMAQ-MADRID 1 for $PM_{2.5}$ and its components. CMAQ-MADRID 1 slightly underpredicts $PM_{2.5}$ mass concentrations by 2.7% using 12 sections and slightly overpredicts it by 3.6% using 24 sections, indicating a good agreement between observed and simulated values. Both simulations underpredict sulfate (SO_4^{2-}, with NMBs of -17.1% and -6.2%), organic matter (OM, with NMBs of -61.0%), and black carbon (BC, with NMBs of 32.2% and 31.8%), and overpredict nitrate (NO_3^-, with NMBs of 33.5% and 38.6%) and ammonium (NH_4^+, with NMBs of 18.8% and 22.1%), indicating larger biases for predictions of PM species, particularly OM.

Table 1. Performance statistics for $PM_{2.5}$ and its components.

Species		Obs. ($\mu g\ m^{-3}$)	Sim. ($\mu g\ m^{-3}$)	Corr.[2]	NMBs (%)
$PM_{2.5}$	12-sec[1]	32.28	31.42	0.85	-2.7
	24-sec		33.43	0.85	3.6
SO_4^{2-}	12-sec	1.44	1.19	0.44	-17.1
	24-sec		1.35	0.38	-6.2
NO_3^-	12-sec	12.34	16.47	0.68	33.5
	24-sec		17.10	0.68	38.6
NH_4^+	12-sec	4.23	5.03	0.69	18.8
	24-sec		5.17	0.69	22.1
OM	12-sec	14.69	5.72	0.73	-61.0
	24-sec		5.73	0.73	-61.0
EC	12-sec	3.83	2.60	0.70	-32.2
	24-sec		2.61	0.70	-31.8

[1] sec - section
[2] Corr. - Correlation Coefficient

3.1.2 Size distribution of $PM_{2.5}$

Fig. 3 shows particle size distributions predicted by 12- and 24-section simulations at Angiola and Fresno on December 27, 2000. The 24-section simulation provides higher mass concentration peak than the 12-section simulation at both sites, with the peak values of 24-section simulation closer to the observations. Both simulations overpredict the mass concentrations of PM with diameter smaller than 0.18 µm and consequently underpredict those in the range of 0.32-2.5 µm. The uncertainties in the initial size distribution assumed and insufficient growth simulated with a full equilibrium gas/particle mass transfer approach may help explain these discrepancies.

3.1.3 Temporal variation of $PM_{2.5}$

Fig. 4 gives the $PM_{2.5}$ temporal variations at 4 sites in central California. Bakersfield and Fresno sites represent major urban areas in SJV, Angiola represents an area with regional transport, and Altamont Pass is a rural site. As expected,

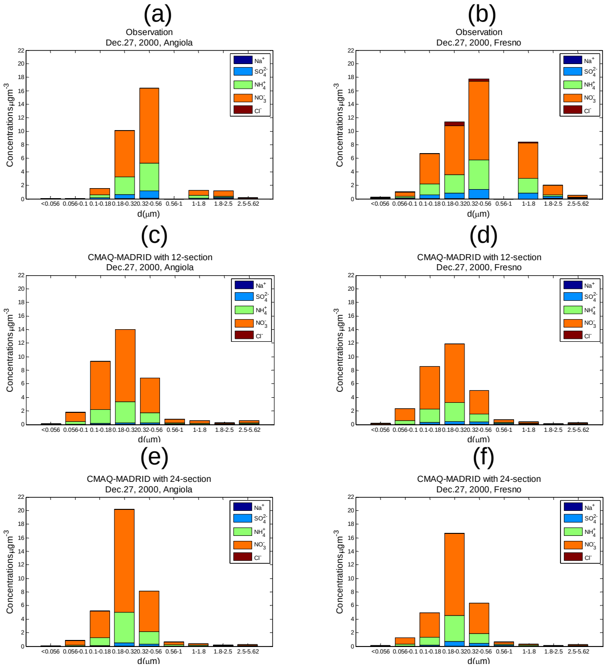


Fig. 3. Particle size distributions at Angiola and Fresno, from observations ((a) and (b)) and model simulations with 12-section ((c) and (d)) and 24-section ((e) and (f)) on December 27, 2000.

Bakersfield and Fresno have significantly higher $PM_{2.5}$ concentrations than Altamont Pass. Air quality at Angiola, located between Bakersfield and Fresno and surrounded by farm fields, is mainly affected by regional transport of pollutants. Simulated PM mass concentrations at Angiola are similar to those in Bakersfield and Fresno.

At all sites, 12- and 24-section simulations give similar $PM_{2.5}$ temporal variation trends, although simulated trends are not in good agreement with observations. The biases in meteorological predictions over complex terrain under a strong stagnation event and uncertainties in inputs and model formulation, such as emissions and aerosol processes, may contribute to the difference between simulated and observed temporal variations of $PM_{2.5}$.

3.2 Sensitivity Simulations

For both 12- and 24-section simulations, coagulation process significantly reduces number concentrations of particles with diameter smaller than 0.21 μm, and slightly increases particles number concentration between 0.21-0.46 μm through particle growth. Over the entire domain, simulations with coagulation reduce $PM_{2.5}$ number concentrations by up to 83-91%. In addition to changes in number concentrations, the mass concentrations of PM with diameter smaller than 0.1 μm are reduced and those in 0.1-0.46 μm are increased, but the total PM mass concentrations over the entire size range remain unchanged. Fig. 5 gives the predicted size distributions of aerosol number and mass concentrations by 12 sections with and without coagulation process at Fresno on December 27, 2000. Comparison between Fig. 5 (b) and (d) also indicates that the PM mass size distributions predicted by simulations with coagulation process are in better agreement with observations shown in Fig. 3 (b), because more PM masses grow into larger particles (0.1 μm) by coagulation.

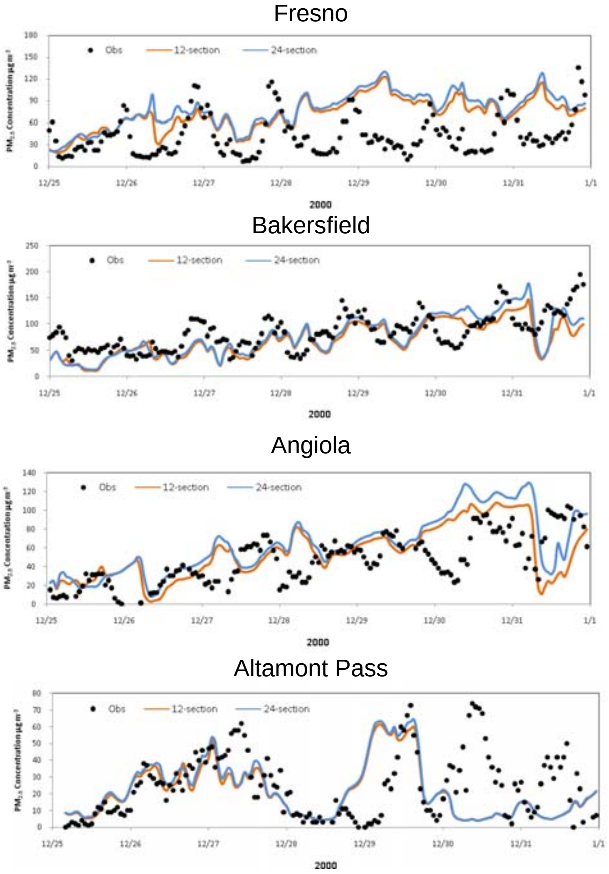
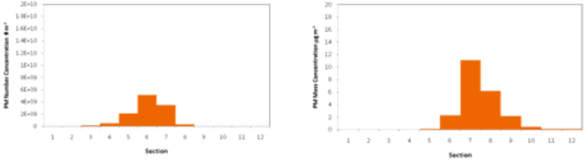


Fig. 4. Temporal variations of $PM_{2.5}$ at 4 sites in central California.

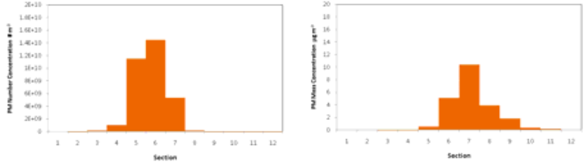
4. SUMMARY

CMAQ-MADRID 1 with 12 and 24 size sections simulations are conducted to simulate aerosol mass and size distributions in central CA during 25-31 December, 2000. Both 12- and 24-section simulations reproduce well the high mass concentrations of $PM_{2.5}$ in the Central Valley,



Number Concentration
(a) coagulation

Mass Concentration
(b)coagulation



(c) Non-coagulation

(d) Non-coagulation

Fig. 5. Size distributions of PM number and mass concentrations with 12-section at Fresno, with and without coagulation on December 27, 2000.

but some biases exist in simulated PM species concentrations. While NO_3^- and NH_4^+, main $PM_{2.5}$ components in this area, are overpredicted by 38.6% and 22.1%, respectively, OM is underpredicted by 61%. 24-section simulation produces a size distribution with peak concentrations closer to observations, although both simulations overpredict the mass concentrations of PM with diameter smaller than 0.18 µm and underpredict those in the range of 0.32 to 2.5 µm. While 12- and 24-section simulations reproduce $PM_{2.5}$ mass, they fail to capture the temporal variations of $PM_{2.5}$ at nearly all the sites. Model difficulties in simulating meteorology over complex terrain and incapabilities of meteorological schemes in capturing local scale variations are possible reasons. Sensitivity simulation results show that coagulation process leads to 83-91% reduction of simulated $PM_{2.5}$ number concentrations, and also change mass concentrations in different size bins. Simulated PM mass size distributions with coagulation are in better agreement with observations, indicating the importance of coagulation in accurately simulating PM size distribution.

6. ACKNOWLEDGEMENTS

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