J3.3 IMPLEMENTATION AND TESTING OF A NEW AEROSOL MODULE IN WRF/CHEM

Xiaoming Hu and Yang Zhang*
Department of Marine, Earth and Atmospheric Sciences, North Carolina State University, Raleigh, NC

1. INTRODUCTION

Atmospheric aerosols have adverse effects on human health and visibility and play an important role in climate changes. Understanding of their chemical composition, ambient concentrations, as well as dynamic processes is essential for the development of effective control strategies to reduce their adverse impacts. Air quality models (AQMs) for particulate matter (PM) provide a fundamental tool to investigate the formation and fate of atmospheric aerosols in both retrospective and forecasting modes. 3-D Eulerian grid-based AQMs are suited for simulation of aerosols over large domains (Seigneur, 2001). Accurately simulating atmospheric aerosols requires model treatments of all major processes including emission, formation of condensable species, nucleation, coagulation, condensation, gas/particle mass transfer, transport, deposition, and cloud-processing of aerosols. Large uncertainties exist in these treatments in AQMs. Several 3-D AQMs have a modular structure that enables alternative aerosol modules within the same model framework, which facilitates the testing of different aerosol treatments in one 3-D AQM.

The Weather Research and Forecasting/Chemistry Model (WRF/Chem) is a 3-D AQM that simulates trace gases and aerosols simultaneously with meteorological fields within the framework of WRF, which is the next generation meteorological model. Several agencies are working collaboratively to develop WRF/Chem. There are two existing aerosol modules in the Pacific Northwest National Laboratory (PNNL) version of the WRF/Chem (Grell et al., 2005; Fast et al., 2005). One is the Modal Aerosol Dynamics Model for Europe (MADE) (Ackermann et al., 1998) with the secondary organic aerosol model (SORGAM) (Schell et al., 2001) (referred to as MADE/SORGAM). MADE/SORGAM uses a modal approach to represent the aerosol size distribution. It uses the modified model for an aerosol reacting system (MARS-A) for thermodynamics of inorganic species. The other is the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) (Zaveri et al., 2005a). MOSAIC employs a sectional treatment of the aerosol size distribution. It uses multi-component equilibrium solver for aerosols, multi-component Taylor expansion model (MESA-MTEM) for thermodynamics of inorganic species. Secondary organic aerosol (SOA) is currently not treated in this version of MOSAIC. In this study, a detailed aerosol model, the Model of Aerosol Dynamics, Reaction, Ionization, and Dissolution (MADRID) (Zhang et al., 2004), has been incorporated into the PNNL's version of WRF/Chem (version 2.0.3) (referred to as WRF/Chem-MADRID). MADRID differs from the previous two aerosol modules in terms of size representation used, chemical species treated, assumptions, and numerical algorithms used. MADRID is based on a sectional representation of the particle size distribution and includes a detailed treatment of aerosol dynamics and secondary organic aerosol (SOA) formation. MADRID uses ISORROPIA (Nenes et al., 1998) for thermodynamics of inorganic species. As an initial implementation, MADRID is coupled to the PNNL's version of Carbon-Bond Mechanism (i.e., CBM-Z). The SOA module in WRF/Chem-MADRID is not activated in the simulation presented in this paper because it requires a large memory that the current version of WRF/Chem does not support.

2. APPLICATION OF WRF/Chem-MADRID

2.1 Testbed and Model Setup

A 5-day episode (August 28 to September 2) from 2000 Texas Air Quality Study (TexAQS-2000) in the southern U.S. is used to test the WRF/Chem-MADRID. TexAQS-2000 is an intensive field study for ozone (O_3) and other pollutant issues in the Houston/Galveston area where severe pollution of O_3 and volatile organic compounds (VOCs) occurs most frequently. The model inputs are setup for a region of 1056 × 1056 km^2 with a 12-km horizontal grid spacing and 56 layers vertically from surface to 16 km. This episode has also been used for study of sensitivity of WRF/Chem predictions to various meteorological schemes in Misenis et al. (2005),

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

which presents detailed meteorology predictions and evaluation. This paper focuses on the incorporation and evaluation of WRF/Chem-MADRID.

Parameterizations used for the WRF/Chem-MADRID simulation include the Goddard shortwave radiation scheme, the rapid and accurate radiative transfer model (RRTM) for longwave radiation, the Yonsei University (YSU) boundary layer scheme, and the Noah land-surface scheme. The simulation is conducted with an equilibrium approach for gas/particle mass transfer. Eight size sections are used to represent the aerosol size distribution.

The predictions of WRF/Chem-MADRID are evaluated against in situ observations for gas-phase species (e.g., O_3, SO_2, NO_2, and NO), $PM_{2.5}$, and its composition.

2.2 Preliminary Results

An evaluation of predictions of gas-phase species concentrations show the normalized mean biases (NMBs) of 19.8% for O_3, 305% for SO_2, 24.6% for NO_2, and -60.2% for NO. The temporal distributions show that the discrepancies between simulations and observations for these species do not increase with time distinctly. Overpredictions of O_3 mixing ratios occur primarily at night. This can be attributed partially to the underpredictions of NO mixing ratios at night, which results in a reduced titration of O_3 by NO. Inaccuracy of the predicted nocturnal planetary boundary layer (PBL) height could also account partially for the inaccurate prediction of O_3 at night (Misenis et al., 2005).

Figure 1 shows the spatial distribution of the predicted 24-hr average $PM_{2.5}$ concentrations and the 24-hr average wind field on 29 August (central daylight time (CDT)), 2000. The predicted $PM_{2.5}$ distribution is consistent with the pattern of the wind filed. Houston, Galveston, New Orleans, and Baton Rouge are the main sources of the plumes. The main stream of the plumes occurs at the downwind area of those sources. The emissions of primary $PM_{2.5}$ species such as black carbon (BC) and other unknown inorganic $PM_{2.5}$ are relatively high in Houston, the emissions of SO_2 are relatively high in Baton Rouge and the emissions of CO and NO_x are relatively high in Dallas. Correspondingly, the high primary $PM_{2.5}$ concentrations result in relatively high $PM_{2.5}$ concentrations in Houston and its vicinity area and high SO_2 mixing ratios result in relatively high secondary sulfate concentrations in Baton Rouge and its vicinity area. The predicted unknown inorganic species account for 35%~65% of the total $PM_{2.5}$ concentration in the simulation domain. The highest ratio of unknown inorganic species to total $PM_{2.5}$ (65%) occurs in Houston.

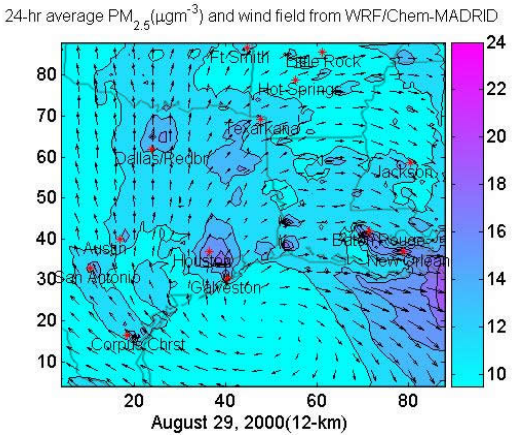

Figure 1. The spatial distribution of the 24-hr average $PM_{2.5}$ concentrations and the 24-hr average wind field predicted by WRF/Chem-MADRID on 29 August 2000.

Figure 2 shows 5-day time series plots of observed and predicted $PM_{2.5}$ concentrations at two sites: Galveston Airport (GALC) and LaPorte (H08H). WRF/Chem-MADRID captures the diurnal variation of concentration of $PM_{2.5}$ relatively better at GALC than at LaPorte. This can be attributed to better predictions of both large- and local-scale wind patterns at GALC. Figure 3 shows the observed and simulated diurnal variation of wind fields at GALC and LaPorte**.** The wind field at GALC is dominated by an onshore flow on 28-29 August, and a shore-parallel or offshore large-scale flow and the stagnant or near-stagnant conditions during noontime (~12:00 CDT) on the remaining days. The change of the wind direction from northwesterlies in the morning to southerlies or southwesterlies in the afternoon and the stagnant flow provide good indications of the impact of the land-sea breeze at GALC, particularly on 30-31 August. The northwesterlies in the morning carry the pollutants from the Houston area to GALC, the near-stagnant condition during noontime helps the pollutants to accumulate at GCLC, resulting in the highest $PM_{2.5}$ concentrations that typically occur 1 or 2 hours lag behind the stagnant wind condition. The peak values of $PM_{2.5}$ occurring at ~13:00 CDT on 30-31 August at GALC are well captured by WRF/Chem-MADRID. The discrepancies of the predicted and observed $PM_{2.5}$ concentration increase slightly after the first two days during this episode.

Unlike GALC, WRF/Chem-MADRID predicts two peaks (at ~7:00 and 19:00 CDT) in one day at LaPorte (also at Deer Park and Baytown, figures are not shown here). The morning peaks are higher than the afternoon peaks (except on August 28). The large disagreement between the predictions and observations at LaPorte may be partially attributed to the failure of the model in capturing the small-scale features. There are significant inconsistencies between the observed and simulated diurnal variations of wind vectors of LaPorte, as shown in Figure 3. LaPorte is located near the Galveston gulf, where the coastal line is rather inhomogeneous. The wind flow is likely affected by complex land-bay breezes. The transport and accumulation of air pollutants at LaPorte are thus more complex than GALC. A finer horizontal grid resolution may be needed for WRF/Chem-MADRID to capture the small-scale flow circulation pattern and its influence on the transport of air pollutants. Although WRF/Chem-MADRID does not predict well the observed diurnal variation of $PM_{2.5}$ concentrations at LaPorte, the predicted two-peak pattern of the $PM_{2.5}$ concentration (i.e., a strong morning peak and a weaker peak in late afternoon) is often observed in Southeastern Texas (Russell et al., 2004). While the model does not reproduce the local variation due probably to the use of a coarse grid resolution in the model simulation, it captures well the general observed region-wide average feature. Some hypotheses have been proposed to explain the "two peak" pattern in this region (Allen, 2005). Strong traffic sources, low mixing heights, and bursts of photochemical activity associated with sunrise may explain the morning peak. The peak in the afternoon may reflect a contribution from biogenic secondary organic aerosol (SOA), which generally lags behind the peak of the emission of key SOA precursors e.g., isoprene, monoterpene at noontime).

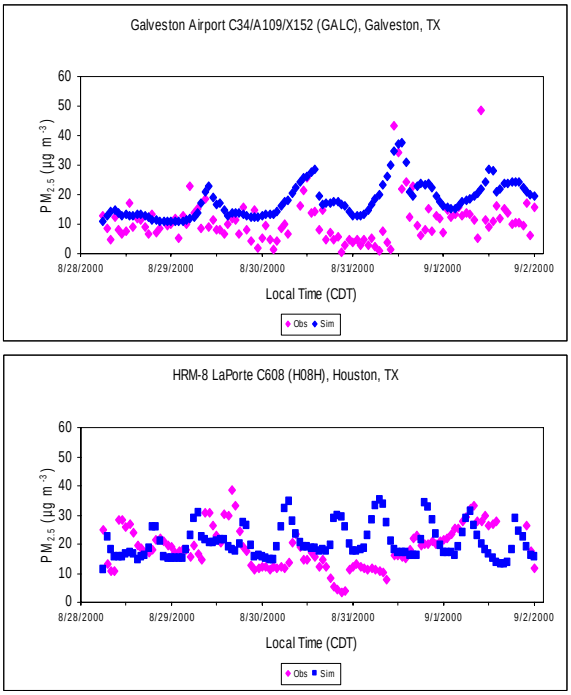

Figure 2. The time series plots of observed and predicted hourly $PM_{2.5}$ concentrations at (a) Galveston Airport (GALC); (b) LaPorte (H08H).

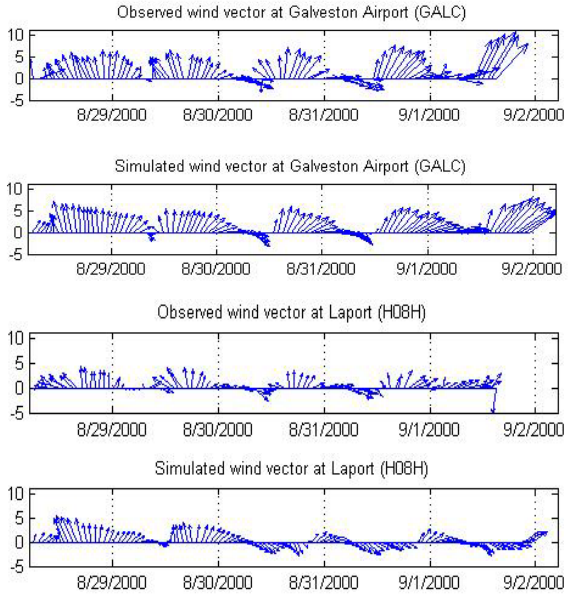

Figure 3. The observed and simulated wind vectors at Galveston Airport (GALC) and LaPorte (H08H) during 6 CDT, 28 August through 0 CDT, 2 September 2000.

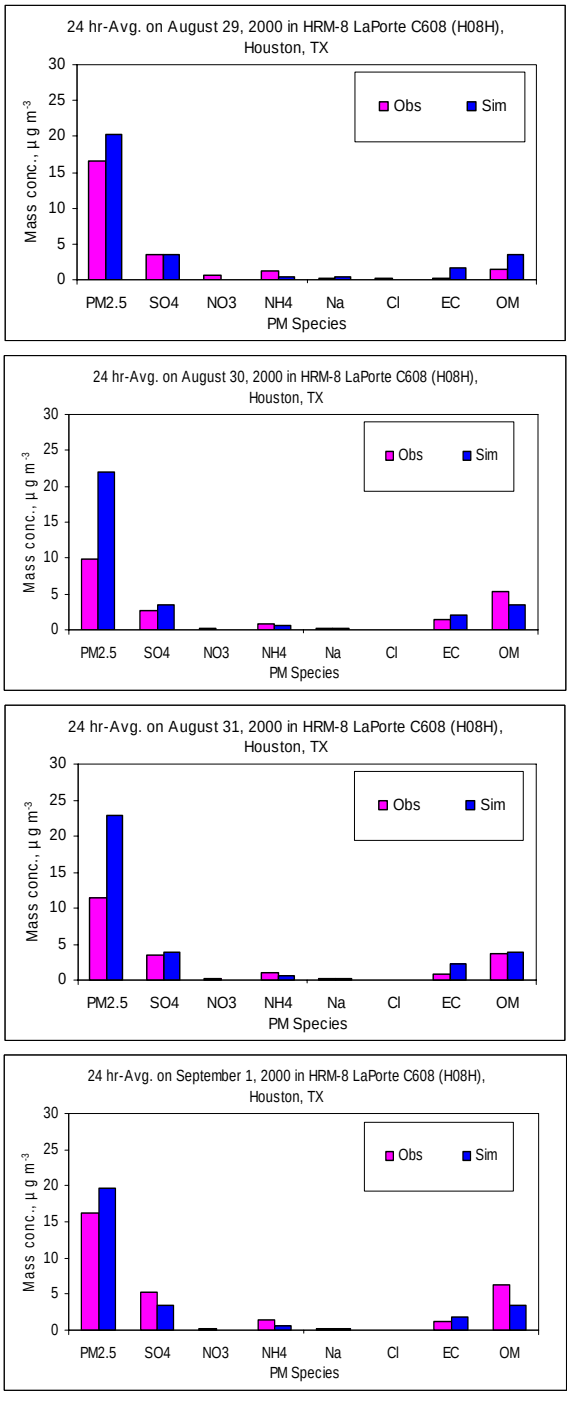

Figure 4. The observed and predicted 24-hr average concentrations of $PM_{2.5}$ and its chemical composition on 29 August through 1 September 2000 at LaPorte (H08H), TX.

The NMB of the hourly $PM_{2.5}$ predictions is 41.7%, indicating a moderate-to-large overprediction. The observed PM composition is only available at two super sites: LaPorte and Williams Tower. Figure 4 shows the observed and simulated 24-hr average concentrations of $PM_{2.5}$ and its chemical composition from 29 August to 1 September at LaPorte, during which the total $PM_{2.5}$ concentrations at LaPorte are overpredicted by 23.3%, 123.9%, 101.3%, 21% respectively. The NMBs of the predicted SO_4^{2-} concentrations are between -34% and 29%. The concentrations of NH_4^+ are underpredicted up to 62%. Those of BC are overpredicted up to 646%. The NMBs of the predictions of organic matter (OM) are between -45% and 128% (note that the observed OM is obtained by multiplying the observed organic carbon (OC) by a factor of 1.4). The observed contributions of NO_3^-, Na^+, and Cl^- to the total $PM_{2.5}$ are quite small at LaPorte. Other unknown inorganic species (OIN) contribute significantly to the total $PM_{2.5}$ concentrations on all days. The large discrepancies between observed and simulated total $PM_{2.5}$ concentrations can be attributed mostly to the inaccurate OIN predictions during 30-31 August and 1 September. The overpredictions in BC concentrations indicate possible overestimations of BC emissions. The inaccurate predictions of OM concentrations may be caused by inaccurate emissions of primary OM or no predictions of SOA by the WRF/Chem-MADRID simulation presented here, or both. The predicted OC to BC ratios range from 1.1 to 1.6, which are much lower than the observed OC to BC ratios of 2.6 to 5.2**.** Analyses of observed OC/BC ratios in primary emissions and at downwind locations suggest that SOA may be an important constituent of OM in Southeast Texas in summer (Russell et al., 2004), which is due to the relatively higher hydrocarbon reactivities during this time period (Daum, 2002). The underestimation in OC/BC ratios in this study may be attributed to several factors including no SOA predictions, inaccurate primary OC and BC emissions, or a combination of these factors.

3. SUMMARY AND FUTURE WORK

The MADRID aerosol module has been incorporated into WRF/Chem. WRF/Chem-MADRID has been tested and evaluated with a 5-day episode from the TexAQS-2000. The simulation using an equilibrium approach for gas-particle mass transfer overpredicts both O_3 and $PM_{2.5}$ by 19.8% and 41.7%, respectively. The results shown here are quite preliminary, in particular, the WRF/Chem-MADRID simulation conducted here does not include SOA. Once SOA module is incorporated into WRF/Chem-MADRID, a more rigorous evaluation will be conducted. Diagnostic and sensitivity simulations will also be

conducted to identify likely causes for inaccurate model predictions.

In addition to the equilibrium approach, MADRID includes dynamic and hybrid approaches for simulating gas/particle mass transfer (Zhang et al., 2005; Hu et al., 2005). Two different condensational algorithms (i.e., Bott (Bott, 1989) and Trajectory-Grid (Chock et al., 2000)) are used in the dynamic approach. These approaches will be tested in WRF/Chem-MADRID. The simulations using the equilibrium approach for gas-particle mass transfer were conducted with eight processors on the NCSU's IBM blade center Linux clusters. The simulation using dynamic and hybrid approaches may increase the computational time. In box model tests, the dynamic approach costs more CPU time than the equilibrium approach by at least an order of magnitude. The foreseeable increase ratio of computation to communication will likely increase the scalability of the whole model system (Grell et al., 2002). The gas/particle mass transfer approach and the condensational algorithm that provide the best compromise between numerical accuracy and computational efficiency will be tested and recommended for real-time forecasting applications with WRF/Chem-MADRID.

4. ACKNOWLEDGEMENTS

This work is performed under the National Science Foundation Career Award No. Atm-0348819, and the Memorandum of Understanding between the U.S. Environmental Protection Agency (EPA) and the U.S. Department of Commerce's National Oceanic and Atmospheric Administration (NOAA) and under agreement number DW13921548. The authors thank J. D. Fast, W. I. Gustafson Jr., R. C. Easter of PNNL for providing the PNNL's version of WRF/Chem, the source code for the conversion of FORTRAN 77/90 fixed format to FORTRAN 90 free format, and helpful discussions during the incorporation of MADRID into WRF/Chem and WRF/Chem-MADRID simualtions; M. Estes of Texas Commission on Environmental Quality (TCEQ) and J. D. Fast of PNNL for providing some observational data; Shaocai Yu of the U.S. NOAA/EPA for providing the Fortran code for statistical calculations; and D. Hamilton and S. Krishnan of NCSU for their assistance in model and observational data processing.

5. REFERENCES

Ackermann, I. J., H. Hass, M. Memmesheimer, A. Ebel, F. S. Binkowski, and U. Shankar, 1998, Modal aerosol dynamics model for Europe: Development and first applications, *Atmos. Environ.*, 32, No.17, 2981-2999.

Allen, D., 2005, Gulf coast aerosol research and Characterization program (Houston Supersite), Univ. of Texas at Austin, TX.

Bott, A., 1989, A positive definite advection scheme obtained by nonlinear renormalization of the advective fluxes, *Mon. Weather Rev.,* 117, 1006-1015.

Chock, D. P., and S. L. Winkler, 2000, A trajectory-grid approach for solving the condensation and evaporation equations of aerosols, *Atmos. Environ.,* 34, 2957-2973.

Daum, P., 2002, An update on Analysis of TexAQS 2000 Data, presented at the DOE Atmospheric Sciences Program (ASP) Meeting 19-21 March, Albuquerque, NM.

Fast, J. D., W. I. Gustafson Jr., R. C. Easter, R. A. Zaveri, J. C. Barnard, E. G. Chapman, and G. A. Grell, 2005, Evolution of ozone, particulates, and aerosol direct forcing in the vicinity of Houston using a fully coupled meteorology-chemistry-aerosol model, submitted to *J. Geophys. Res.*

Grell, G. A., 2002, Real-time simultaneous prediction of air pollution and weather during the Houston 2000 Field experiment, presented at the 82nd Annual AMS Conference, 13-17 January, Orlando, FL.

Grell, G. A., S. E. Peckham, R. Schmitz, and S. A. McKeen, G. Frost, W. C. Skamarock, and B. Eder, 2005, Fully coupled "online" chemistry within the WRF model. *Atmos. Environ.,* in press,

Hu, X.-M., Y. Zhang, and M. Z. Jacobson, 2005, Evaluation of the trajectory-grid and the Bott schemes for solving aerosol condensation and evaporation equations, presented at the 4th Annual CMAS Models-3 User's Conference, September 26-28, Chapel Hill, NC.

Misenis, C., X.-M. Hu, S. Krishnan, and Y. Zhang, and J. D. Fast, 2006, Sensitivity of WRF/Chem predictions to meteorological schemes, to be presented at the 86th Annual AMS Conference/the 14th Joint Conference on the Applications of Air Pollution Meteorology with the A&WMA, 27 January–3 February, Atlanta, GA.

Nenes, A., S. N. Pandis, and C. Pilinis, 1998, ISORROPIA: A new thermodynamic equilibrium model for multiphase multicomponent inorganic aerosols, *Aquatic Geochemistry*, *4*, 123-152.

Russell, M. M., D. T. Allen, D. R. Collins and M. P. Fraser, 2004, Daily, seasonal and spatial trends in $PM_{2.5}$ mass and composition in southeast Texas, *Aerosol Sci. and Techno.*, 38 (S1) 14-26.

Seigneur, C., 2001, Current status of air quality models for particulate matter, *J. Air & Waste Management Assoc.* 51, 1508-1521.

Zaveri, R. A., R. C. Easter, J. D. Fast, and L. K. Peters, 2005 a, Model for simulating aerosol interactions and chemistry. To be submitted to *J. Geophys. Res.*

Zaveri, R. A., R. C. Easter, and L. K. Peters, 2005 b, A computationally efficient multi-component equilibrium solver for aerosols (MESA), *J. Geophys. Res*., accepted.

Zhang, Y., B. Pun, K. Vijayaraghavan, S.-Y. Wu, C. Seigneur, S. Pandis, M. Jacobson, A. Nenes, and J. H. Seinfeld, 2004, Development and application of the model of aerosol dynamics, reaction, ionization and dissolution (MADRID), *J. Geophys. Res.*, 109, D01202, doi:10.1029/2003JD003501.

Zhang, Y., X.-M. Hu, G. W. Howell, E. Sills, J. D. Fast, W. I. Gustafson Jr., R. A. Zaveri, G. A. Grell, S. E. Peckham, and S. A. McKeen, 2005, Modeling Atmospheric Aerosols in WRF/CHEM, *Proceeding of the 2005 Joint WRF/MM5 User's Workshop*, June, 27-30, Boulder, CO.