

Evaluation of New Trace Gas and Aerosol Modules in WRF-chem using Measurements from TexAQ5 2000

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1.0 INTRODUCTION

Much progress has been made in modeling trace gases such as ozone and ozone precursors; however, relatively large uncertainties are still associated with urban, regional, and global-scale model predictions of aerosols (mass, size distribution, composition, physical characteristics, and optical properties) and aerosol direct and indirect forcing. A key step in improving the representation of particulates in global climate models is to first improve the performance of urban to regional scale models that can resolve many of the sub-grid scale processes a global climate model cannot.

We have used the PNNL Eulerian Gas Phase and Aerosol Scalable Unified System, PEGASUS (Fast et al., 2002), over the past several years to investigate how local to regional-scale meteorological processes influence the evolution of oxidants and particulates. However, PEGASUS employs “off-line” coupling of meteorology with chemistry and aerosols so that feedback mechanisms cannot be simulated. In anticipation of the emerging scientific issues associated with the interaction of mega-city pollutant emissions, regional air-quality, and climate change, we have begun using the Weather Research and Forecasting (WRF) community model as a framework to develop, evaluate, and improve aerosol parameterizations. In this study, we describe our on-going model development activities as well as preliminary results of coupled meteorology-chemistry-aerosol predictions for an urban area.

2.0 MODEL DESCRIPTION

As described by Grell et al. (2004), WRF-chem is a version of WRF that also simulates trace gases and particulates simultaneously with the meteorological fields. The initial version of WRF-chem includes the RADM2 photochemical mechanism and the MADE / SORGAM aerosol mechanism (Ackermann et al., 1998; Schell et al., 2001). This version of WRF-chem will be used by NOAA/FSL to produce operational air-quality forecasts (Penkham et al. 2004); therefore, the trace gas and aerosol mechanisms need to be computationally efficient. Our objective is to adapt the more complex treatments of gas-phase and aerosol chemistry from PEGASUS into WRF-chem and to implement aerosol-chemistry-cloud-radiation feedback processes associated with aerosol direct and indirect forcing.

2.1 Gas-phase chemistry

The gas-phase chemistry is modeled with the CBM-Z photochemical mechanism (Zaveri and Peters, 1999) that contains 55 prognostic species and 134 reactions. CBM-Z uses the lumped-structure approach for condensing organic species and reactions, and is based on the widely used Carbon Bond Mechanism (CBM-IV). CBM-Z extends the original CBM-IV to include reactive long-lived species and their intermediates, which may become important under conditions of transport over synoptic spatial scales. The major differences with CBM-IV include revised inorganic chemistry, explicit treatment of lesser reactive paraffins such as methane and ethane, revised treatments of reactive paraffin, olefin, and aromatic reactions, inclusion of alkyl and acyl peroxy radical interactions and their reactions with NO₃, inclusion of longer lived organic nitrates and hydroperoxides, and revised isoprene chemistry. CBM-Z is implemented using a regime-dependent approach in which the kinetics are partitioned into background, anthropogenic, and biogenic sub-mechanisms to reduce the overall computational time.

2.2 Aerosol chemistry

Aerosols are simulated using a sectional approach. The aerosol size distribution is divided into discrete size bins (currently 8), and each bin is assumed to be internally-mixed (all particles within a bin are assumed to have the same chemical composition). Both mass and number are simulated for each bin, and either the moving-center (Jacobson, 1997) or two-moment approach (Tzivion, 1989) is employed to solve the dynamic equations that determine the aerosol size and composition. Aerosol composition is made up of sulfate, nitrate, chloride, carbonate, ammonium, sodium, calcium, other inorganics, organic carbon, and elemental carbon. 112 prognostic species are required for 14 aerosol parameters and 8 size bins. Aerosol dry deposition is based on Binkowski and Shankar (1995).

Aerosol inorganic chemistry is simulated by the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) (Zaveri et al., 2004a). Condensation and evaporation of volatile species are modeled dynamically for each bin using highly efficient numerical techniques, and are driven by a thermodynamic module (Zaveri et al., 2004b) to compute the aerosol surface vapor concentrations of HNO₃, HCl and NH₃, as well as deliquescence and liquid/solid partitioning of various salts of sodium, calcium, ammonium, sulfate, nitrate, chloride and carbonates. MOSAIC employs a new mixing rule to estimate the mean activity coefficient of an electrolyte in a multi-component solution that is

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significantly more accurate and computationally efficient than other methods (Zaveri et al., 2004c).

2.3 Radiative properties

Photolysis rates will be computed using the FAST-J radiation scheme (Wild et al., 2000) and the predicted ozone, aerosol, and cloud profiles. The aerosol optical depth, single scattering albedo, and phase function expansion coefficients are a function of the refractive indices and size distribution obtained from the predicted aerosol mass and composition. FAST-J predictions have been evaluated using observations from Phoenix, Arizona, and Houston, Texas (Barnard et al., 2004) and were found to agree within the uncertainty of the measurements for both clear and cloudy conditions.

2.4 Status and future plans

A 'beta' version of WRF-chem that includes CBM-Z, MOSAIC, and direct radiative forcing has already been completed. During the remainder of 2004, testing of these modules, including FAST-J, will be completed and ported into version 2 of WRF, cloud chemistry and wet removal processes from PEGASUS will be adapted for WRF cumulus and microphysical parameterizations, and a more rigorous treatment of direct radiative forcing will be included. We will implement coagulation, nucleation, and a secondary organic aerosol mechanism compatible with CBM-Z and MOSAIC during 2005. The cloud microphysics will also be modified to include a prognostic treatment of cloud droplet number (Ghan et al., 1997) for the aerosol indirect effect.

In the near future, we will employ WRF-chem to evaluate and improve the performance of aerosol parameterizations using data from upcoming U.S. DOE field campaigns and to determine the urban to regional scale variations in aerosol concentrations and properties and their effect on direct and indirect forcing.

3. EXPERIMENTAL METHOD

3.1 Description of the 2000 TexAQS

Metropolitan Houston has a population of over 4 million people. Traffic and other human activities lead to high emissions of ozone precursors and particulates. In contrast with other large cities in the U.S., numerous industrial petrochemical facilities that emit large amounts of volatile organic chemicals are located a few kilometers east of Houston. Under favorable meteorological conditions of high temperatures, high humidity, and weak winds that frequently occur during the summer, extremely high ozone levels can be produced from the complex distribution of anthropogenic and biogenic emission sources. Houston has a network of air quality monitors that continuously measure ozone, NO_x, CO, SO₂, and particulate matter. Ozone often exceeds the 1-h standard of 120 ppb and the highest hourly concentration during 2000 was 199 ppb.

Extensive measurements were collected at the surface and aloft around Houston during August and September of 2000 as part of the Texas Air Quality Study (TexAQS) to better understand the processes associated with the evolution of ozone and its precursors. Scientists from the Texas Commission on

Environmental Quality (TCEQ), various government agencies including NOAA, DOE, National NASA, EPA, and NCAR, and several universities participated in the field campaign.

3.2 Configuration of WRF-chem

WRF-chem is currently based on version 1.3 of WRF that does not have nesting capability. Therefore, one grid was employed that encompassed the south-central U.S. with a grid spacing of 16 km. 60 grid points were used in the vertical with a grid spacing of 15 m adjacent to the surface. We intentionally chose a coarse horizontal grid spacing so that the computationally demanding chemistry and aerosol components of the model could be tested in a timely manner and compared with predictions from an off-line PEGASUS simulation that employed the same spatial resolution. Results from a higher resolution domain will be presented at the conference.

A simulation period between 12 UTC 28 August and 12 UTC 2 September was chosen when high ozone concentrations were observed around Houston. The meteorological conditions during this period were not favorable for long-range transport into the region so that the observed pollution was largely the result of local anthropogenic and biogenic emission sources.

Table 1 lists some of the parameterization options used in this study. Meteorological initial and boundary conditions were based on NCEP's AVN model analyses. Horizontally homogenous initial conditions were employed for trace gases and particulates; these values were also used for inflow boundary conditions. Trace gas emission rates were obtained from the TCEQ (Jiang and Fast, 2004) and emission rates of particulates were adapted from EPA's NET-96 inventory.

Test simulations indicated that predictions of wind, temperature, humidity, and boundary layer depth using the simpler land-surface model and boundary layer schemes (Table 1) were significantly better than those that used the OSU land-surface model and Mellor-Yamada-Janjic TKE boundary layer scheme.

Table 1. WRF-chem configuration options.

Longwave radiation	RRTM
Shortwave radiation	Dudhia
Surface layer	Monin-Obukhov
Land-surface model	thermal diffusion
Boundary layer scheme	MRF
Microphysics	none
Cumulus parameterization	Kain-Fritsch
Dry deposition for trace gases	Wesely
Chemistry option	CBM-Z
Aerosol option	MOSAIC
Photolysis option	FAST-J

4. RESULTS

Since our model development activities are not yet complete, the results presented here are preliminary. An example of observed and simulated meteorological quantities at one station in Houston is shown in Fig. 1.

Mostly clear skies, increasing temperatures, decreasing humidity, and regular diurnal wind direction variations were observed during the five-day period. As a sea-breeze circulation developed during the afternoon, the near-surface winds shifted from westerly to easterly. The winds then rotated clockwise at night so that they gradually became southwesterly by sunrise. While the model reproduced much of the observed diurnal and multi-day meteorological variations, the evening wind speeds were too high and the afternoon temperatures were as much as 5°C too low. The predictions have also been compared with radiosonde and radar wind profile data (not shown). Although relatively large forecast errors were produced at times, the overall performance was satisfactory so that the predicted meteorology could be used to simulate the transport and mixing of trace gases and aerosols.

An example of the surface ozone and particulate matter three and a half days into the simulated period at 18 UTC 31 August is shown in Fig. 2. Off-shore flow during the morning transported ozone precursors and particulates to the southeast over the Gulf of Mexico. The winds became light and variable so that pollutants remained in the vicinity of Houston during the afternoon. The predicted distribution of ozone and particulates was consistent with the observations at most locations. Predicted peak ozone concentrations in central Houston were about 10 ppb lower than the observations between 90 and 110 ppb. The highest concentrations of ozone precursors and particulates were produced over Galveston Bay at this time; NO titration reduced ozone to less than 50 ppb.

The predicted aerosol composition was also compared with measurements at two sites as shown in Fig. 3. While the predicted aerosol composition was often similar to measurements, at times the predicted concentrations of NO₃ and NH₄ were too high and organic carbon was too low. Other measurements indicated that a large fraction of the organic matter was composed of secondary organic compounds. Since the MOASIC aerosol mechanism does not yet include secondary organic aerosol, it is not surprising that afternoon total organic mass was underestimated. The predicted aerosols significantly affected radiative quantities. For example, peak NO₂ photolysis rates (that affect ozone photochemical production) were about 0.0075 s⁻¹, while predicted JNO₂ from FAST-J with and without aerosols was 0.008 and 0.0095 s⁻¹, respectively.

Many of the differences between the observed and simulated trace gases and particulates are likely the result of the coarse 16-km grid spacing. For example, a grid spacing of 1 km was needed in the simulations of Fast and Jiang (2004) to resolve the local peak ozone concentrations observed by a research aircraft around Galveston Bay. Therefore, WRF-chem will be run at a higher resolution to better resolve the urban-scale pollutant variations.

Our WRF-chem simulation took 41 h on a SGI Altix machine using 25 processors; a meteorology-only run required 2.5 h. Additional benchmark tests will be performed to assess the speed-up of WRF-chem with its

167 additional prognostic species and how the speed-up differs from WRF.

5. SUMMARY

We have implemented trace-gas and aerosol mechanisms from the PEGASUS model into WRF-chem. Preliminary simulations indicate that the model is producing forecasts of trace gases and aerosols that are consistent with measurements made in the vicinity of Houston. After our model development activities are complete, our objective is to use WRF-chem as a framework to evaluate and improve aerosol parameterizations to better understand urban to regional-scale variations in direct and indirect forcing.

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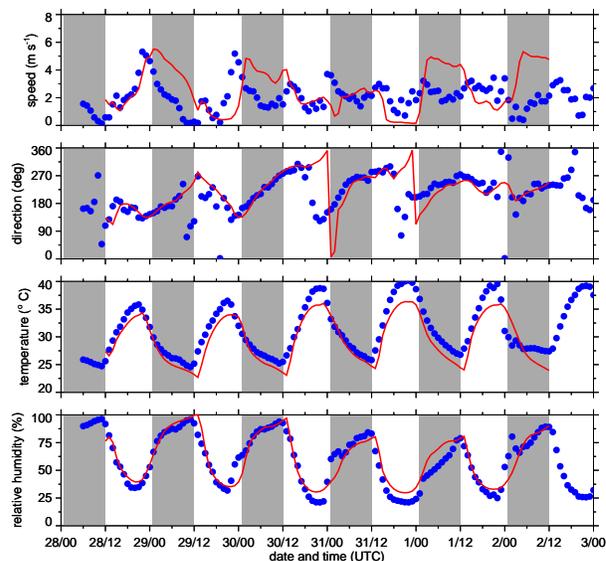


Fig. 1. Observed (blue dots) and predicted (red line) wind speed, direction, temperature, and humidity. Gray shading denotes nighttime periods.

6. 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, 2981-2999.
- Barnard, J.C., E.G. Chapman, J.D. Fast, J.R. Schmelzer, J.R. Schlusser, and R.E. Shetter, 2004: An evaluation of the FAST-J photolysis model for predicting nitrogen dioxide photolysis rates under clear and cloudy sky conditions. *Atmos Environ.*, 38, 3393-3403.
- Binkowski, F. S., and U. Shankar, 1995: The Regional Particulate Matter Model, 1, Model description and preliminary results. *J. Geophys. Res.*, 100, 26191-26209.
- Fast, J. D., R.A. Zaveri, X. Bian, E.G. Chapman, and R.C. Easter, 2002: The effect of regional-scale transport on oxidants in the vicinity of Philadelphia during the 1999 NE-OPS field campaign. *J. Geophys. Res.*, 107, 10.1029/2001JD000980.

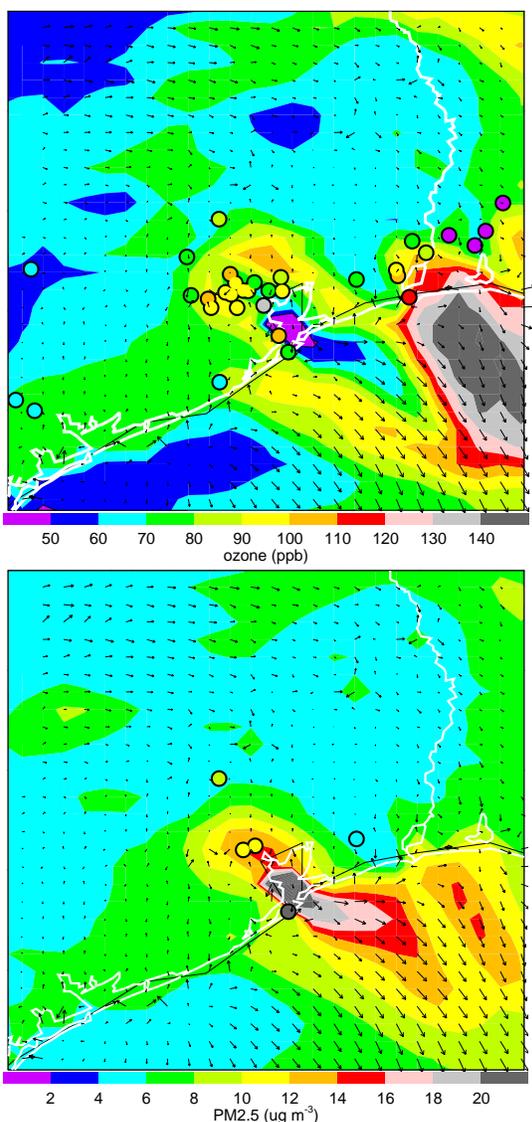


Fig. 2. Observed (dots) and predicted (contours) ozone and PM_{2.5} at 18 UTC 31 August over a portion of the model domain.

Ghan, S.J., L.R. Leung, R.C. Easter, and H. Abdul-Razzak, 1997: Prediction of cloud droplet number in a general circulation model. *J. Geophys. Res.*, 102, 21,777-21,794.

Grell, G.A., S.E. Penkham, R. Schmitz, and S.A. McKeen, 2004: Fully coupled "online" chemistry with the WRF model. *Sixth Conference on Atmospheric Chemistry: Air Quality in Megacities*. Amer. Meteor. Soc., Seattle, WA, paper 12.6.

Jacobson M.Z., 1997: Development and application of a new air pollution modeling system – II. Aerosol phase simulations. *Atmos. Environ.*, 31, 587-608.

Jiang, G., and J.D. Fast, 2004: Modeling the effects of VOC and NO_x emission sources on ozone formation in Houston during the TexAQS 2000 field campaign. In press, *Atmos. Environ.*

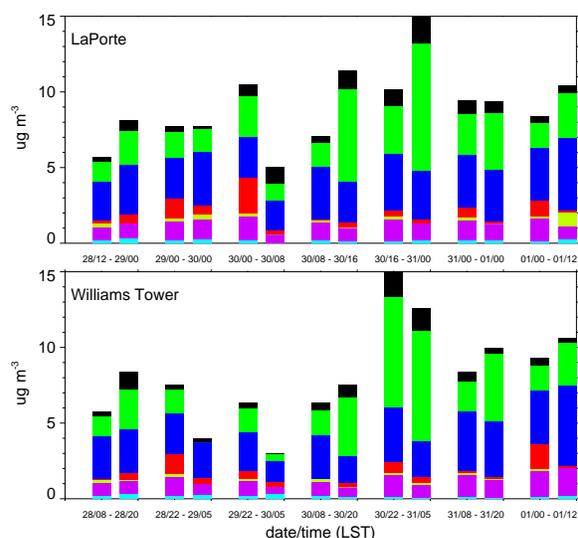


Fig. 3. Simulated (left bar) aerosol composition averaged over seven sampling periods corresponding to measurements (right bar) at two locations in Houston. Colors denote sodium (light blue), ammonium (purple), chloride (light green), nitrate (red), sulfate (blue), organic carbon (green), and elemental carbon (black).

Schell, B., I.J. Ackermann, H. Hass, F.S. Binkowski, and A. Ebel, 2001: Modeling the formation of secondary organic aerosol within a comprehensive air quality modeling system. *J. Geophys. Res.*, 106, 28275-28293.

Penkham, S.E., G.A. Grell, S.A. McKeen, and R. Schmitz, 2004: Comparisons between observations made during NEAQS and forecasts from MM5 and WF chemistry models. *Sixth Conference on Atmospheric Chemistry: Air Quality in Megacities*. Amer. Meteor. Soc., Seattle, WA, paper J2.15.

Tzivion, S., G. Feingold, and Z. Levin, 1989: The evolution of raindrop spectra. Part II: Collisional collection/breakup and evaporation in a rainshaft. *J. Atmos. Sci.*, 46, 3312-3327.

Wild, O., X. Zhu, M.J. Prather, 2000: Fast-J: Accurate simulation of in- and below cloud photolysis in tropospheric chemical models. *J. Atmos. Chem.*, 37, 245-282.

Zaveri, R.A., and L.K. Peters, 1999: A new lumped structure photochemical mechanism for large-scale applications. *J. Geophys. Res.*, 104, 30,387-30,415.

Zaveri, R.A., R.C. Easter, J.D. Fast, and L.K. Peters, 2004a: Model for simulating aerosol interactions and chemistry. To be submitted to *J. Geophys. Res.*

Zaveri, R.A., R.C. Easter, and L.K. Peters, 2004b: A computationally efficient multi-component equilibrium solver for aerosols (MESA). Submitted to *J. Geophys. Res.*

Zaveri, R.A., R.C. Easter, A.S. Wexler, 2004c: A computational efficient model for multi-component activity coefficients in aqueous solutions: Application to atmospheric inorganic aerosols. In press, *J. Geophys. Res.*