PLUME-IN-GRID MODELING FOR PM AND MERCURY

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1. INTRODUCTION

One-atmosphere three-dimensional (3-D) grid models are now being widely used to predict the impacts of emission controls on the concentrations and deposition of pollutants such as ozone (O_3), fine particulate matter ($PM_{2.5}$) and mercury (Hg). Such a grid-based approach necessarily averages emissions within the volume of the grid cell where they are released. This averaging process may be appropriate for sources that are more or less uniformly distributed at the spatial resolution of the grid system. However, it may lead to significant errors for sources that have a spatial dimension much smaller than that of the grid system. For example, stack emissions lead to plumes that initially have a dimension of tens of meters, whereas the horizontal resolution in grid-based air quality models is typically several kilometers in urban applications and up to about 100 km in regional applications. This artificial dilution of stack emissions leads to (1) lower concentrations of plume material, (2) unrealistic concentrations upwind of the stack, (3) incorrect chemical reaction rates due to the misrepresentation of the plume chemical concentrations and turbulent diffusion, and (4) incorrect representation of the transport of the emitted chemicals. The errors associated with the grid-averaging of stack emissions can be eliminated by using a subgrid-scale representation of stack plumes that is imbedded in the 3-D grid system of the air quality model, i.e., a plume-in-grid (PiG) approach.

Such PiG models have been developed for O_3 (e.g., Karamchandani et al., 2002) and PM (Karamchandani et al., 2006a). We describe here the extension of this PiG modeling approach to the transport, chemistry and deposition of Hg. The new model uses the U.S. EPA Multiscale Air Quality model (CMAQ) as the grid-based host model, the Model of Aerosol Dynamics, Reaction, Ionization and Dissolution (MADRID) for PM processes and an Advanced Plume Treatment (APT) to resolve stack plumes at the sub-grid scale. The model, CMAQ-MADRID-APT-Hg, is applied to the southeastern United States for the 2002 calendar year, and a performance evaluation is conducted using available databases.

2. DESCRIPTION OF CMAQ-MADRID-APT-Hg

CMAQ (Byun and Schere, 2006) is the grid-based host model. The processes that govern PM concentrations are simulated with MADRID (Zhang et al., 2004). MADRID uses a sectional representation of the particle size distribution and simulates the formation of secondary inorganic and organic aerosols. Two PM sizes (fine and coarse) were used in this application.

Mercury processes were incorporated into CMAQ using the chemical mechanism and deposition algorithms of Seigneur et al. (2004). The base formulation of CMAQ-MADRID-Hg and its application to Hg deposition in the contiguous United States for an entire year have been described by Vijayaraghavan et al. (2006).

The plume-in-grid model consists of a reactive plume model, the second-order closure integrated puff model with chemistry (SCICHEM), imbedded into CMAQ. SCICHEM uses a second-order closure approach to solve the turbulent diffusion equations (Sykes et al., 1993; 1995). The plume is represented by a myriad of three-dimensional puffs that are advected and dispersed according to the local micrometeorological characteristics. SCICHEM can simulate the effect of wind shear as well as plume overlaps. The effects of buoyancy on plume rise and initial dispersion are simulated by solving the conservation equations for mass, heat, and momentum.

The formulation of nonlinear chemical kinetics within the puff framework is described by Karamchandani et al. (2000). Chemical species

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concentrations in the puffs are treated as perturbations from the background concentrations. The Carbon-Bond Mechanism (CBM-IV) is used in both SCICHEM and the host grid model. The formulation of PM processes in SCICHEM follows that of the host model and has been described by Karamchandani et al. (2006a). Hg processes also follow the formulation used in the host model (Vijayaraghavan et al., 2006). An empirical reaction is used to represent the potential reduction of Hg^{II} to Hg^0 in coal-fired power plant plumes (Lohman et al., 2006); the kinetics that is taken to be proportional to the SO_2 gas-phase concentration affects Hg^{II} concentrations in plumes but has little effect in the background (Seigneur et al., 2006).

SCICHEM was imbedded into the host grid model (CMAQ-MADRID, based on CMAQ version 4.5.1) according to the established protocols for incorporating new science modules into CMAQ. The transfer of puff material to the 3-D grid system occurs when the horizontal puff size becomes commensurate with the horizontal grid size of the host model. For fine grid resolutions (e.g., on the order of a few km), another criterion based on the chemical maturity of the plume can be added as discussed by Karamchandani et al. (2002).

3. APPLICATION OF CMAQ-MADRID-APT-Hg

The PiG model described above was applied to a domain covering the southeastern U.S. with a 12-km horizontal resolution for the entire year 2002. The vertical grid structure consists of 19 layers from the surface to the tropopause with finer resolution near the surface (e.g., the surface layer is about 35 m deep). The meteorological fields for the air quality modeling simulations were obtained from a prognostic simulation conducted with the non-hydrostatic meteorological model, MM5. Forty coal-fired power plants in the 12 km resolution domain with the highest emissions of SO_2, NO_x and Hg were selected for explicit PiG treatment. Figure 1 shows the modeling domain and the locations of the 40 power plants used in the APT simulation.

Two simulations with CMAQ-MADRID-APT-Hg were conducted for the model performance evaluation. One simulation includes an empirical reaction between Hg^{II} and SO_2 to represent the potential reduction of Hg^{II} to Hg^0 in coal-fired power plant plumes, while the other simulation does not include this reaction. As shown in a previous study (Karamchandani et al., 2006b), the incorporation of this reaction provides significantly better agreement with Hg concentration measurements downwind of power plants as compared to simulations without this reaction. For the study described here, the results from the two simulations are compared with data from national and regional ambient and deposition monitoring networks, as well as with measurements of ambient Hg concentrations downwind of several coal-fired power plants during plume events. This comparative model performance evaluation will determine which approach provides better agreement with observed Hg concentrations and wet depositions.

4. CONCLUSION

CMAQ-MADRID-APT-Hg is a new model that provides a comprehensive treatment of O_3 formation, PM processes and Hg deposition with an advanced PiG treatment. This model has been thoroughly evaluated against available measurements and represents the state-of-the-science for “one-atmosphere” simulations. CMAQ-MADRID-APT-Hg is available from www.cmascenter.org.

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6. REFERENCES

Byun, D., and K. L. Schere, 2006: Review of governing equations, computational algorithms, and other components of the Models-3 Community Multiscale Air Quality (CMAQ) modeling system, *Appl. Mech. Revs.*, **59**, 51–77.

Karamchandani, P., A. Koo, and C. Seigneur, 1998: A reduced gas-phase kinetic mechanism for atmospheric plume chemistry, *Environ. Sci. Technol.*, **32**, 1709–1720.

Karamchandani, P., and C. Seigneur, 1999: Simulation of sulfate and nitrate chemistry in power plant plumes, *J. Air Waste Manage. Assoc.*, **49**, PM-175-181.

Karamchandani, P., L. Santos, I. Sykes, Y. Zhang, C. Tonne, and C. Seigneur, 2000: Development and evaluation of a state-of-the-science reactive plume model, *Environ. Sci. Technol.*, **34**, 870–880.

Karamchandani, P., C. Seigneur, K. Vijayaraghavan, and S.-Y. Wu, 2002: Development and application of a state-of-the-science plume-in-grid model, *J. Geophys. Res.*, **107**, 4403-4415.

Karamchandani, P., K. Vijayaraghavan, S.-Y. Chen, C. Seigneur and E. Edgerton, 2006a: Plume-in-grid modeling for particulate matter, *Atmos. Environ.*, doi:10.1016/j.atmosenv.2006.06.033.

Karamchandani, P., K. Vijayaraghavan and C. Seigneur, 2006b: Detailed treatment of power plant plumes in the regional modeling of atmospheric mercury, *8th International Conference on Mercury as a Global Pollutant*, Madison, WI, August 6-11.

Lohman, K., C. Seigneur, E. Edgerton, and J. Jansen, 2006: Modeling mercury in power plant plumes, *Environ. Sci. Technol.*, **40**, 3848-3854.

Seigneur, C., K. Vijayaraghavan, K. Lohman, P. Karamchandani and C. Scott, 2004: Global source attribution for mercury deposition in the United States, *Environ. Sci. Technol.*, **38**, 555-569.

Seigneur, C., K. Vijayaraghavan and K. Lohman, 2006: Atmospheric mercury chemistry: sensitivity of global model simulations to chemical reactions, *J. Geophys. Res.*, in press.

Sykes, R. I., S. F. Parker, D. S. Henn, and W. S. Lewellen, 1993: Numerical simulation of ANATEX tracer data using a turbulence closure model for long-range dispersion, *J. Appl. Meteor.*, **32**, 929–947.

Sykes, R. I., and D.S. Henn, 1995: Representation of velocity gradient effects in a Gaussian puff model, *J. Appl. Meteor.*, **34**, 2715–2723.

Vijayaraghavan, K., C. Seigneur, P. Karamchandani and S.-Y. Chen, 2006: Development and application of a multi-pollutant model for atmospheric mercury deposition, *J. Appl. Meteor. Climatol.*, in press.

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, *J. Geophys. Res.*, **109**, D01202, doi:10.1029/2003JD003501.

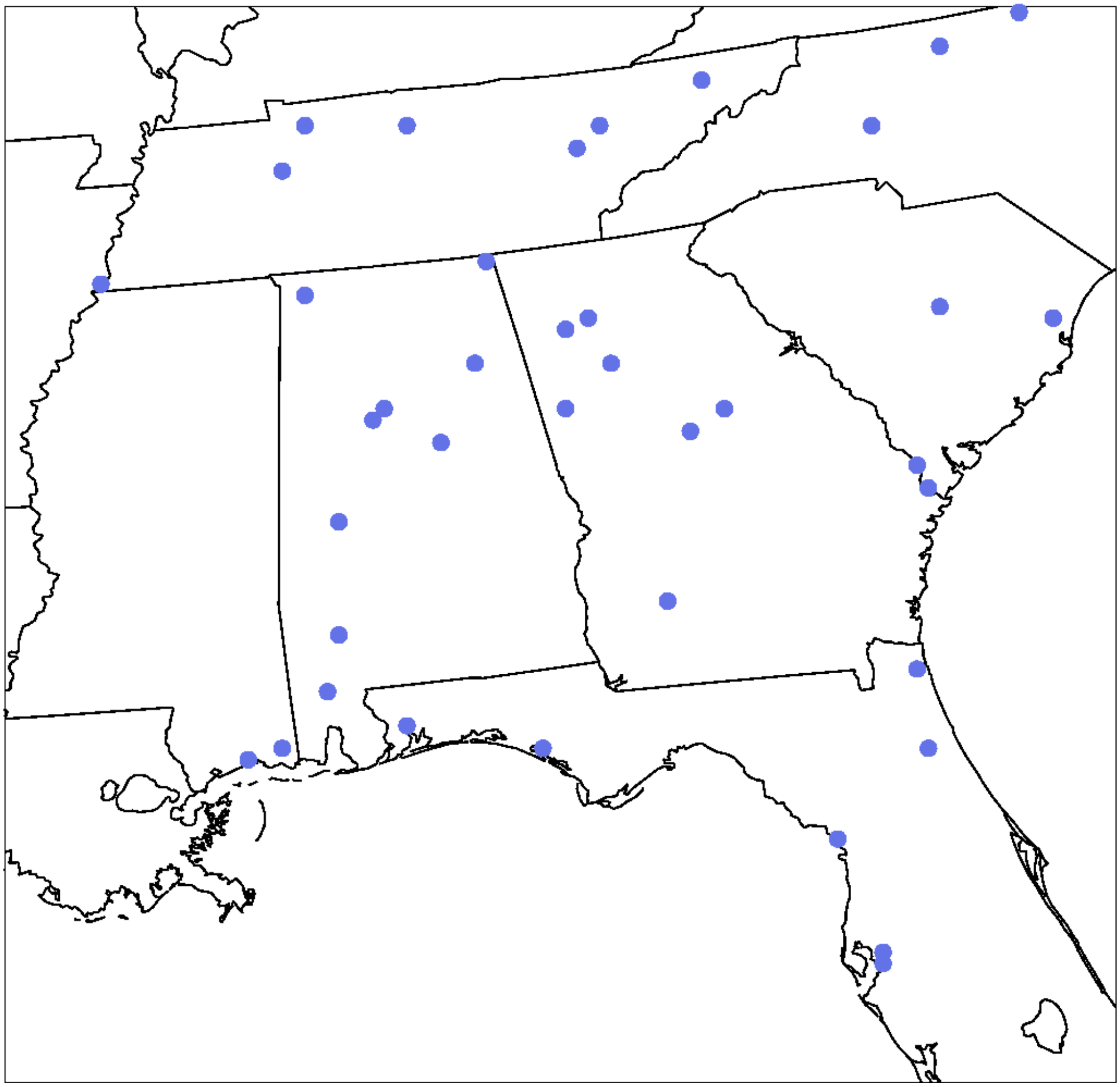

Figure 1. Modeling domain for CMAQ-MADRID-APT-Hg simulations and locations of 40 power plants selected for explicit PiG treatment.