

DEVELOPMENT AND EVALUATION OF A STATE-OF-THE-SCIENCE PLUME-IN-GRID MODEL, CMAQ-APT

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1. INTRODUCTION

Three-dimensional (3-D) modeling of air quality is typically based on a gridded representation of the atmosphere where atmospheric variables such as chemical concentrations are assumed to be uniform within each grid cell. 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 grid resolution in Eulerian 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.

We describe here the development and application of a new state-of-the-science plume-in-

grid (PiG) air quality model that addresses the physical phenomena mentioned above explicitly, thereby providing a more realistic representation of the behavior of reactive plumes in the atmosphere.

2. DESCRIPTION OF CMAQ-APT

The plume-in-grid model consists of a reactive plume model, SCICHEM, imbedded into a three-dimensional grid-based model, the U.S. EPA Models-3 Community Multiscale Air Quality modeling system (CMAQ). The PiG model is referred to as CMAQ-APT, where APT stands for "Advanced Plume Treatment". Below, we provide brief descriptions of the embedded plume model and the host grid model.

The reactive plume component of CMAQ-APT is the Second-order Closure Integrated puff model (SCIPUFF) with CHEMistry (SCICHEM). Plume transport and dispersion are simulated with SCIPUFF, a model that uses a second-order closure approach to solve the turbulent diffusion equations (Sykes et al., 1988, 1993; Sykes and Henn, 1995). The plume is represented by a myriad of three-dimensional puffs that are advected and dispersed according to the local micrometeorological characteristics. SCIPUFF can simulate the effect of wind shear as well as plume overlaps. Also, 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

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Karamchandani et al. (2000). Chemical species concentrations in the puffs are treated as perturbations from the background concentrations. The chemical reactions within the puffs are simulated using a general framework that allows any chemical kinetic mechanism to be treated. Here, the Carbon-Bond Mechanism (CBM-IV) was used in both SCICHEM and the host grid model for consistency. SCICHEM allows the option of explicitly simulating the effect of turbulence on chemical kinetics for selected reactions. This effect is more pronounced near the stack (Karamchandani et al., 2000) and requires additional computational time for its simulation. More details on the SCICHEM model formulation and its evaluation with plume data from the 1995 Southern Oxidants Study (SOS) in Nashville/Middle Tennessee are presented elsewhere (Karamchandani et al., 2000).

SCICHEM was imbedded into the host grid model following the established protocols for incorporating new science modules into Models-3/CMAQ (Byun and Ching, 1999). The transfer of puff material to the 3-D grid system (referred to as "puff dumping") occurs when the puff is determined to be chemically mature with respect to the host model. Optionally, a physical criterion for dumping can be used. The physical criterion is based on the horizontal size of the puff relative to the horizontal grid cell size. The chemical criterion for maturity is based on Karamchandani et al. (1998), who defined three stages in the evolution of the chemistry of NO_x plumes, and Gillani and Godowitch (1999), who used the plume concentration ratio of O_3/O_x , where $\text{O}_x = \text{O}_3 + \text{NO}_2$, as a surrogate for the chemical age of the plume.

3. APPLICATION OF CMAQ-APT

The PiG model described above was applied to a domain covering the northeastern U.S. with two nested grids for a five-day episode from 11 to 15 July 1995. The outer (coarse) grid had a horizontal resolution of 12 km while the inner (fine) grid had a 4 km horizontal resolution. The vertical grid structure consisted of 13 layers from the surface to the tropopause with finer resolution near the surface (e.g., the surface layer is 18 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, using four-dimensional data assimilation (FDDA) (Seaman and Michelson, 2000). The thirty largest NO_x point sources in the 12 km resolution domain were explicitly simulated with PiG treatment. The

effect of the PiG treatment is analyzed by comparing the CMAQ-APT and base CMAQ simulations. Figures 1 and 2 show such comparisons over the 12 km resolution domain on July 13 for ozone (O_3) and nitric acid (HNO_3), respectively.

The simulation results show that the use of CMAQ-APT has a significant effect on the spatial patterns of O_3 and HNO_3 surface concentrations downwind of the sources considered for PiG treatment to distances of 100 to 200 km. Surface O_3 concentrations from the APT simulation show both decrements and increments with respect to the Base simulation. The maximum decrement is about 80 ppb, and the maximum increment is about 77 ppb.

The decrements between the APT and Base simulations are generally associated with lower production of O_3 in the APT simulation than in the Base simulation immediately downwind of large NO_x point sources. Conversely, the large increments in surface O_3 concentrations between the APT and Base simulations are primarily due to the artificially high titration of background surface O_3 in the Base simulation near and upwind of the point sources, particularly in VOC-limited environments. In the APT simulation, the plumes from these sources are explicitly treated, and the plume NO remains aloft near the sources and is not available for titrating surface O_3 concentrations near the source.

An exception to the above generalization occurs in West Virginia, downwind of the Ohio River Valley point sources. Large increments in surface O_3 concentrations between the APT and Base simulations are noted, particularly on July 14, that are primarily associated with higher O_3 production further downwind in the APT simulation than in the Base simulation. In the Base simulation, the NO_x in the plume undergoes rapid conversion to HNO_3 and O_3 within 25 to 50 km of the sources, leading to a depletion of NO_x , and a consequent reduction in O_3 production, at larger downwind distances. In the APT simulation, the conversion of NO_x to HNO_3 occurs at a slower rate, so that more NO_x is available for O_3 production at larger downwind distances (about 150 to 200 km from the sources in southeastern Ohio). Note that the combined NO_x emissions from the cluster of sources in southeastern Ohio and West Virginia are almost three times higher than that from the single largest source in the modeling domain (the Cumberland power plant in northwestern Tennessee).

The smaller increments (typically less than 10 ppb) between the APT and Base simulations are

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associated with both higher titration of existing O₃ in the Base simulation and delayed production of O₃ further downwind in the APT simulation, as the plume NO_x is transported and exposed to a NO_x-limited environment.

The generally lower production of O₃ downwind of major NO_x point sources in the APT simulation is more or less compensated by the unrealistically large titration of existing surface O₃ by the NO_x emissions in the Base simulation without PiG treatment. Thus, over the entire domain and episode, the difference between the total O₃ mass in the two simulations is negligible (0.2%).

For surface HNO₃ concentrations, the differences between the APT and Base simulations range from a maximum decrement of 24 ppb to a maximum increment of 9 ppb. The APT simulation generally gives lower HNO₃ concentrations than the Base simulation over a large portion of the modeling domain because NO_x is converted to HNO₃ more slowly in a power plant plume than in the ambient background (Karamchandani et al., 1998; Karamchandani and Seigneur, 1999). Thus, over the entire domain and episode, the PiG treatment leads to a decrease in HNO₃ mass of 3%.

4. CONCLUSION

These results suggest that it is desirable to use a PiG treatment for major NO_x point sources, since both O₃ and HNO₃ production downwind of these sources are generally overpredicted, if a PiG treatment is not used.

CMAQ-APT has been subjected to a comprehensive beta-testing by third-party organizations. Minor modifications were made as a result of this beta-testing. CMAQ-APT has been shown to be numerically robust under a variety of simulation conditions and currently represents the state-of-the-science for the simulation of large point sources in 3-D models.

5. ACKNOWLEDGMENTS

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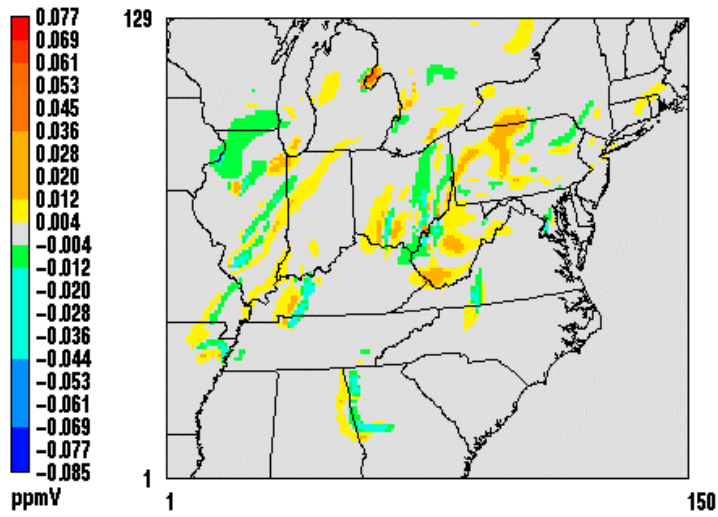


Fig. 1 Difference in O₃ concentrations between the CMAQ-APT and CMAQ base simulations at 3 p.m. EDT, 13 July 1995.

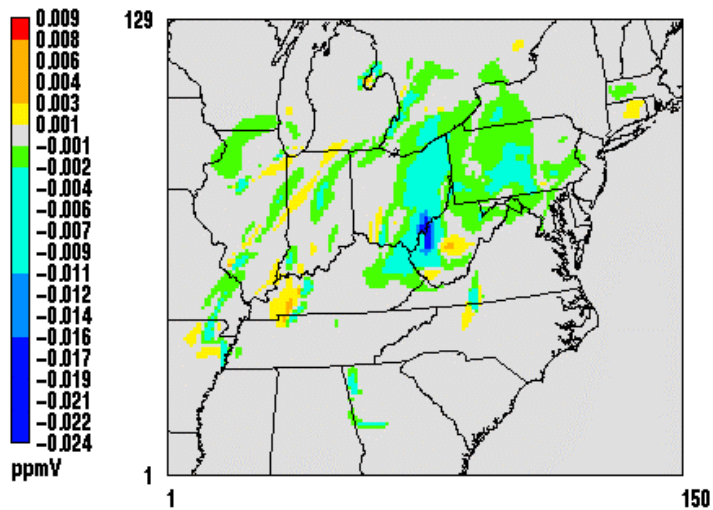


Fig. 2 Difference in HNO₃ concentrations between the CMAQ-APT and CMAQ base simulations at 3 p.m. EDT, 13 July 1995.