MULTI-POLLUTANT PLUME-IN-GRID MODELING

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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 atmospheric concentrations and deposition of pollutants such as ozone (O_3), fine particulate matter ($PM_{2.5}$), mercury (Hg) and other air toxics. 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.

Plume-in-Grid (PiG) modeling has been demonstrated to be an effective approach to resolve sub-grid scale effects associated with discrete sources (e.g., Seigneur et al., 1983; Sillman et al., 1990; Kumar and Russell, 1996; Gillani and Godowitch, 1999; Karamchandani et al., 2002; 2006a; Godowitch, 2004). In this approach, 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.

This paper describes the development and evolution of one such PiG model that was originally developed and applied for ozone (Karamchandani et al., 2002; Vijayaraghavan et al., 2006) and then extended to particulate matter (PM) (Karamchandani et al., 2006a), mercury (Karamchandani et al., 2006b; 2006c), and more recently, to investigate the sub-grid scale impacts of air toxics emissions from roadways (Karamchandani et al., 2008a). We present results from the application of the model for a number of case studies to illustrate the differences in results from traditional grid modeling versus PiG modeling, and the implication of these differences.

2. PLUME-IN-GRID MODEL

The PiG model presented here consists of a reactive plume model that is embedded within a three-dimensional grid model. The 3-D grid model, also referred to as the "host" model, is based on the U.S. EPA Community Multiscale Air Quality (CMAQ) model (Byun and Schere, 2006). The embedded reactive plume model is adapted from the Second-Order Closure Integrated puff model with CHEMistry (SCICHEM) (Karamchandani et al., 2000). The resulting PiG model is referred to as CMAQ with Advanced Plume Treatment or CMAQ-APT.

In SCICHEM, the reactive plume component of CMAQ-APT, plume transport and dispersion are simulated using a second-order closure approach to solve the turbulent diffusion equations (Sykes et al., 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. Each puff has a Gaussian representation of the concentrations of emitted inert species. The overall plume, however, can have any spatial distribution of these concentrations, since it consists of a multitude of puffs that are independently affected by the transport and dispersion characteristics of the atmosphere. SCIPUFF can simulate the effect of wind shear since individual puffs will evolve according to their respective locations in an inhomogeneous velocity field. As puffs grow larger, they may encompass a volume that cannot be considered homogenous in terms of the meteorological variables. A puff splitting algorithm accounts for such conditions by dividing puffs that have become too large into a number of smaller puffs. Conversely, puffs may overlap significantly, thereby leading to an excessive computational burden. A puff-merging

algorithm allows individual puffs that are affected by the same (or very similar) micro-scale meteorology to combine into a single puff. 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 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. The puff chemical mechanism is the same as the host grid model mechanism for consistency. For example, typical applications of CMAQ-APT for ozone (Karamchandani et al., 2002) and fine particulate matter (Karamchandani et al., 2006a) have used the Carbon-Bond Mechanism (CBM-IV) in both SCICHEM and CMAQ. Another feature of SCICHEM is that it 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).

The formulation of PM processes in SCICHEM follows that of the host model and has been described by Karamchandani et al. (2006a). Two versions of CMAQ-APT are available based on the treatment of aerosols in the host and plume models. The first version uses the AERO3 module of CMAQ for the treatment of aerosols and is referred to as CMAQ-AERO3-APT. The second version uses the Model of Aerosol Dynamics, Reaction, Ionization and Dissolution (MADRID) (Zhang et al., 2004) for aerosol treatment and is referred to as CMAQ-MADRID-APT. The primary differences between the two aerosol modules are in their representation of particle size distribution (sectional in MADRID and modal in AERO3) and in their treatment of the formation of secondary organic aerosols (SOA). Both aerosol treatments use the ISOROPPIA module of Nenes et al. (1999) for the thermodynamics of inorganic species.

The consistency between the host model and reactive plume model is extended to their treatment of heterogeneous chemistry on aerosol particles and cloud droplets as well as aqueous-phase chemistry in clouds and removal by dry and wet deposition.

The treatment for mercury in CMAQ-AERO3-APT and CMAQ-MADRID-APT (Karamchandani et al., 2006b) is based on the chemical mechanism and deposition algorithms of Seigneur et al. (2004). Additionally, 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 of this reaction is taken to be proportional to the SO_2 gas-phase concentration and significantly affects Hg^{II} concentrations in plumes but has little effect in the background (Seigneur et al., 2006). Another recent improvement to the treatment of mercury processes in the host and reactive plume models is the addition of modules to simulate the adsorption of reactive gaseous mercury (Hg^{II}) to atmospheric particulate matter (Vijayaraghavan et al., 2008).

The coupling of SCICHEM and CMAQ follows the established protocols for incorporating new science modules into CMAQ. SCICHEM is invoked in CMAQ-APT by a single subroutine call similar to the invocation of any other physical or chemical process module in the host model. Like the other process modules in CMAQ, all relevant information related to the emissions and the dynamic state of the atmosphere required by SCICHEM are accessed directly from the input files via the Input/Output Applications Programming Interface (I/O API) (Coats et al., 1993); only the three-dimensional concentration fields are shared directly between the host model and SCICHEM. On input to SCICHEM, these concentrations serve as the background (ambient) concentrations for SCICHEM calculations. On output from SCICHEM, these concentrations are updated whenever plume-to-grid transfer occurs and are returned to the host model. The transfer of puff material to the 3-D grid system (referred to as "puff dumping") is triggered when the puff size is commensurate with the grid cell size. As an option, a chemical dumping criterion may be selected for situations where the physical criterion may result in premature transfer of the plume material to the grid (Karamchandani et al., 2002). However, for most applications, the physical dumping criterion is sufficient.

The computational overhead associated with PiG modeling can make it impractical to use for long-term simulations with a large number of point sources treated explicitly with the plume component of the model. To overcome this limitation, we have recently parallelized the PiG

code (Karamchandani et al., 2008b) to exploit the widespread availability of multi-processor workstations and workstation clusters that are commonly used today for air quality model simulations. A fully parallel PiG code allows efficient utilization of all the available compute cycles in these modern computer systems.

3. MODEL APPLICATIONS

Applications for Ozone

The first version of the PiG model described above was applied to examine the effects of the PiG formulation on model predictions of ozone concentrations in the vicinity of and downwind of large power plants in the northeastern U.S (Karamchandani et al., 2002). This early version of the model did not include a treatment for aerosols, aqueous-phase chemistry or mercury. The model was applied to a domain covering the northeastern U.S. for a five-day episode from 11 to 15 July 1995. The grid had a horizontal resolution of 12 km and 30 large point sources were selected for PiG treatment. The model was evaluated with available observations.

The simulation results showed that the use of the PiG model had a significant effect on the spatial patterns of ozone (O_3) and nitric acid (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 PiG simulation showed both decrements and increments with respect to the base simulation (a simulation with all sources treated with the grid model). The maximum decrement was about 80 ppb, while the maximum increment was about 77 ppb.

Most of the surface O_3 decrements were associated with lower production of O_3 immediately downwind of the point sources in the PiG simulation relative to the base simulation. Furthermore, downwind O_3 production was generally delayed in the PiG simulation as compared to the base simulation. The large increments in surface O_3 concentrations in the PiG simulation relative to the base simulation were primarily associated with the higher titration of background surface O_3 in the base simulation near and upwind of the point sources, particularly in VOC-limited environments. When the PiG treatment was used, the plume NO remained aloft for longer distances and was not immediately available for titrating surface O_3 concentrations near the source. The smaller increments (typically less than 10 ppb) in the PiG simulation relative to the base simulations were associated with both higher titration of existing surface O_3 in the Base simulation and delayed production of O_3 further downwind in the PiG simulation, as the plume NO_x was transported and exposed to a NO_x-limited environment.

For surface HNO_3 concentrations, the differences between the PiG and base simulations range from a maximum decrement of 24 ppb to a maximum increment of 9 ppb. The PiG simulation generally predicted lower surface HNO_3 concentrations than the base simulation over a large portion of the modeling domain. Over the entire domain and episode, the PiG treatment resulted in a decrease in HNO_3 mass of about 3%.

Vijayaraghavan et al. (2006) applied the PiG model to simulate O_3 and HNO_3 formation during a four-day July/August 2000 episode in central California. The top ten NO_x emitting plants in the Central California Ozone Study (CCOS) domain were selected for explicit plume treatment. Their results were qualitatively similar to those of Karamchandani et al. (2002) for the northeastern U.S. However, the magnitudes of the differences between the PiG and base simulations were smaller in the central California application than in the northeastern U.S. simulation. This can be explained by the relatively smaller magnitude of NO_x emissions from large point sources in California as compared to the eastern United States.

Application for Fine Particulate Matter ($PM_{2.5}$)

The PiG model for ozone was extended by Karamchandani et al. (2006a) to include in the plume component state-of-the-science treatments of aerosol chemistry and dynamics as well as aqueous chemistry that were consistent with the treatments used in the host grid model. The model was applied to the southeastern United States with a 12 km horizontal grid resolution for the months of January and July 2002. Emissions from fourteen coal-fired power plants (CFPPs) in Alabama, Georgia, Florida and Mississippi were explicitly simulated with the PiG treatment.

The model evaluation included both comparisons with routine measurements of $PM_{2.5}$ components from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network, the Clean Air Status and Trends Network (CASTNet), the Southeastern Aerosol Research and Characterization study (SEARCH) network. In addition, the SEARCH network measurements also included more detailed observations corresponding to plume events from specific

upwind power plants. This special database provided an opportunity to conduct a refined evaluation of the PiG treatment. The comparisons of measured peak plume increments of SO_2 concentrations for plume events at the SEARCH monitoring sites during January and July 2002 with the corresponding peaks from the base simulation (i.e., all sources treated with the grid model) and PiG simulations showed that, for a large majority (nearly 90%) of the cases, the PiG simulation provided a better representation of the plume events than the base simulation.

The results showed that the use of the PiG model had a significant effect on the spatial patterns of particulate sulfate and total inorganic nitrate (gas-phase nitric acid + particulate phase nitrate) concentrations. The effects were largest in the four states containing the 14 power plants selected for PiG treatment. However, differences in sulfate and total inorganic nitrate concentrations were also predicted in some of the surrounding states. Using a PiG treatment resulted in a reduction in the contributions of the 14 CFPPs to sulfate and total inorganic nitrate concentrations over most of the regions impacted by these sources. In July 2002, using the PiG treatment reduced the calculated contributions of the power plants to sulfate concentrations by 5% to 40% in most of Alabama, north-western Georgia, south-eastern Mississippi and north-western Florida and by up to 5% in eastern Tennessee and most of the Carolinas. The maximum CFPP contributions in the PiG simulation to monthly-average sulfate and total inorganic nitrate concentrations in July 2002 were about 41% and 45% lower respectively than the corresponding values in the base simulation.

These differences between the base and PiG simulation results are due to the more correct treatment of elevated point source emissions and the near-field transport and chemistry of these emissions in the PiG simulation. The purely gridded approach used in the base simulation cannot resolve these subgrid-scale phenomena and overestimates the oxidation rates of SO_2 and NO_x emissions from the power plants. Over the entire modeling domain, about 77% of the SO_2 emissions from the power plants were oxidized to sulfate in July 2002 in the base simulation, as compared to 69% in the PiG simulation, a difference of about 10%.

This application was one of the first studies to use a PiG treatment for $PM_{2.5}$ and the results from this study showed that it is important to use a PiG treatment for emissions from large elevated point sources to accurately capture the subgrid-scale features of the transport and chemistry of the plumes from these sources, so that the impacts of these sources on downwind $PM_{2.5}$ sulfate and nitrate concentrations can be correctly simulated.

Applications for Mercury (Hg)

Karamchandani et al. (2006b) subsequently extended the PiG model for ozone and $PM_{2.5}$ to include a treatment for mercury. The model 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 consisted 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.

Two simulations with the PiG model were conducted for the model performance evaluation. One simulation included 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., 2006c), the incorporation of this reaction provides significantly better agreement with Hg concentration measurements downwind of power plants as compared to simulations without this reaction. The results from the two simulations were 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.

Vijayaraghavan et al. (2008) applied the PiG model for 2001 over a domain covering the U.S. at 36 km horizontal resolution. The top thirty Hg emitting power plants in the U.S. were selected for explicit plume treatment. The version of the PiG model used in this application included a treatment of the gas-phase adsorption of reactive gaseous mercury (RGM) on atmospheric particulate matter. The PiG treatment resulted in improved performance for Hg wet deposition over a purely Eulerian grid-based model, partial correction of over-predictions of wet deposition downwind of coal-fired power plants in the northeastern U.S., and in decreases of approximately 10% in simulated dry and wet deposition over large parts

of the eastern U.S. with larger decreases near the plants selected for PiG treatment.

Application for Nitrogen Deposition

The PiG model was also used to estimate the decrease in atmospheric nitrogen deposition in Escambia Bay and its watershed in Florida and southern Alabama due to planned emissions controls of NO_x and SO_2 at a nearby coal-fired power plant (Vijayaraghavan et al., 2007). The model was applied for 2002 to a domain covering the southeastern USA and centered on Alabama (AL) and Georgia (GA), the ALGA domain, with a horizontal resolution of 12 km. The use of PiG treatment resulted in less simulated dry deposition of atmospheric nitrogen than a purely gridded model, approximately half as much in the immediate vicinity of the plant and by 10% over the Escambia Bay watershed. This difference is due to the correct treatment in the PiG model of the depletion of oxidants by NO in the early stages of plume development in NO_x-rich plumes.

Application for Sub-Grid Scale Modeling of Air Toxics Concentrations Near Roadways

The PiG model was recently adapted by Karamchandani et al. (2008a) to develop a prototype model that could be used to simulate near-roadway concentrations due to mobile emissions from roadways. The motivation for the study was the increasing concern about population exposure to hazardous air pollutants (HAPs), especially in the vicinity of the sources of these pollutants. For example, measurements of toxic air pollution levels near a busy freeway in Los Angeles showed that exposures near the freeway were up to 10 times greater than those at background locations and dropped to background levels within 300 m downwind of the freeway (Zhu et al., 2002).

The model was applied to a busy interstate highway in New York City. The model was able to successfully capture the observed spatial variability in exposure levels from near the source to several hundreds of meters from the source.

4. SUMMARY

We have described the evolution of a multi-pollutant plume-in-grid model from its initial development for ozone applications to a model that can be used to calculate air quality impacts for a suite of pollutants including fine particulate matter, mercury, and other air toxics. Newer developments include the parallelization of the model code to improve its computational efficiency and the application (currently ongoing) of the parallelized code to conduct annual base simulations and emission scenario simulations with over 150 point sources explicitly treated with the plume component of the model.

Applications of the PiG model show that the model can predict significantly different air quality impacts of emissions from large point sources than the traditional gridded approach. These differences are due to the more realistic treatment of the transport and chemistry of the plumes from these sources in the PiG model.

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