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Development and application of a parallelized version of the advanced modeling system for transport, emissions, reactions and deposition of atmospheric matter (AMSTERDAM): 1. Model performance evaluation and impacts of plume–in–grid treatment

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ABSTRACT

The Community Multiscale Air Quality Model (CMAQ) is a comprehensive three–dimensional "one–atmosphere" air quality model that is now routinely used to address urban, regional–scale and continental–scale multi–pollutant issues such as ozone, particulate matter, and air toxics. Several updates have been made to CMAQ by the scientific community to enhance its capabilities and to provide alternative science treatments of some of the relevant governing processes. The Advanced Modeling System for Transport, Emissions, Reactions and Deposition of Atmospheric Matter (AMSTERDAM) is one such adaptation of CMAQ that adds an Advanced Plume–in–grid Treatment (APT) for resolving sub–grid scale processes associated with emissions from elevated point sources. It also incorporates a state–of–the–science alternative treatment for aerosol processes based on the Model of Aerosol Dynamics, Reaction, Ionization and Dissolution (MADRID). AMSTERDAM is configured to provide flexibility to the model user in selecting options for the new science modules. This paper describes the parallelization of AMSTERDAM to make it a practical tool for plume–in–grid (PinG) treatment of a large number of point sources, and presents results from its application to the central and eastern United States for summer and winter periods in 2002. Over 150 coal–fired power plants in the domain with high emissions of sulfur dioxide (SO_2) and nitrogen oxides (NO_x) were selected for PinG treatment in the CMAQ–MADRID–APT configuration of AMSTERDAM used for this application. Although both model configurations (grid–only and PinG) give similar model performance results (an aggregate measure of model skill), the results show significant differences between the two versions in the specific nature of the predicted spatial distribution of ozone and $PM_{2.5}$ concentrations. These differences can be important in determining source contributions to ambient concentrations. A companion paper examines the differences in the predicted contributions of hypothetical source regions from the two configurations of the model.

Keywords:
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1. Introduction

The U.S. Environmental Protection Agency (EPA) Community Multiscale Air Quality (CMAQ) model (Byun and Schere, 2006), is a one–atmosphere three–dimensional grid model that is being used to predict the impacts of emission controls on the atmospheric concentrations and depositions of multiple pollutants such as ozone (O_3), fine particulate matter ($PM_{2.5}$) and air toxics. Because it is a community model, several enhancements to the model have been made by the air quality modeling community to provide alternative science treatments of some of the governing processes or to include treatments that are not supported in the base CMAQ.

The Advanced Modeling System for Transport, Emissions, Reactions and Deposition of Atmospheric Matter (AMSTERDAM) is a version of CMAQ that incorporates an alternative treatment of aerosol processes and also adds a plume–in–grid treatment to simulate the subgrid–scale features associated with pollutant emissions from point sources. Grid models, such as CMAQ, necessarily average emissions within the volume of the grid cell where they are released. This averaging process may be appro–priate 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 approximately 40 km in continental 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 (PinG) 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; Godowitch, 2004; Karamchandani et al., 2006a; Vijayaraghavan et al., 2008; Karamchandani et al.,

2009). In this approach, the errors associated with the grid–averaging of stack emissions are addressed by using a subgrid–scale representation of stack plumes that is imbedded in the 3D grid system of the air quality model.

While PinG modeling provides a more accurate and detailed representation of point source emissions than a traditional grid model, it increases the computational time required for model simulations, particularly when a large number of point sources are treated explicitly with the embedded plume model. This additional computational overhead can make it impractical to use PinG modeling for large modeling domains and long simulation periods. In this paper, we describe the development of a parallelized version of AMSTERDAM to overcome these limitations, and present model performance results with and without PinG treatment. We also compare the spatial patterns of predicted ozone and $PM_{2.5}$ concentrations from the two model configura–tions to illustrate the differences between the two approaches. In a companion paper (Karamchandani et al., 2010), we present results from hypothetical emission control scenarios to illustrate the effect of PinG modeling on predicted impacts of emissions reductions on ozone and $PM_{2.5}$ concentrations and sulfur and nitrogen deposi–tion.

2. AMSTERDAM

AMSTERDAM is actually a suite of models, based on CMAQ, with user–selectable configurations for chemistry, aerosols and plume–in–grid (PinG) treatment. In addition to the standard CMAQ configurations, the new configurations offered by AMSTERDAM include CMAQ–AERO3–APT, CMAQ–MADRID, and CMAQ–MADRID–APT. MADRID, which refers to the Model of Aerosol Dynamics, Reaction, Ionization and Dissolution, is an advanced alternative aerosol treatment developed by Zhang et al. (2004). MADRID is available with both the Carbon Bond IV and SAPRC–99 gas–phase chemistry options. PinG treatment is provided with the Advanced Plume Treatment (APT) option (Karamchandani et al., 2002; Karamchandani et al., 2006a). This option is available with both the AERO3 aerosol module of CMAQ and the MADRID aerosol treatment. AMSTERDAM also includes options for the treatment of mercury (Hg) species based on Seigneur et al. (2004; 2006).

The embedded reactive plume model for the APT option is adapted from the Second–Order Closure Integrated puff model with Chemistry (SCICHEM) (Karamchandani et al., 2000). SCICHEM simulates plume transport and dispersion using a second–order closure approach to solve the turbulent diffusion equations. The plume is represented by a myriad of three–dimensional puffs that are advected and dispersed according to the local micrometeo–rological 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. The model 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 computa–tional 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.

The APT option for PinG treatment was initially developed and applied for ozone (Karamchandani et al., 2002; Vijayaraghavan et al., 2006) and subsequently extended to particulate matter (Karamchandani et al., 2006a) and mercury (Karamchandani et al., 2006b; Karamchandani et al., 2006c, Vijayaraghavan et al., 2008).

Because of the computational overhead associated with the PinG treatment (about 20 to 30% for 50 sources), early model applications were limited to small domains and/or short–term simulations, with no more than 50 point sources treated explicitly with the embedded plume model. However, these constraints limited the utility of the model and it became apparent that it would be necessary to reduce the turn–around time for PinG applications. In the next section, we describe our approach to achieve this speed–up, based on parallelization of the PinG code. This approach relies on the widespread availability of multi–processor workstations and workstation clusters that are commonly used today for air quality model simulations. A parallel PinG code allows efficient utilization of the available compute cycles in these modern computer systems.

3. Parallelization of AMSTERDAM

The traditional approach to parallelizing a grid model such as CMAQ is to perform domain decomposition by subdividing the horizontal domain into a number of roughly equal subdomains, with each subdomain assigned to a separate processor. Each processor then performs the transport/chemistry/removal calcula–tions on the grid cells within the subdomain. However, inter–processor communication is required for I/O purposes and horizontal transport calculations. In CMAQ, this inter–processor communication is accomplished by using the parallel input output (PARIO) management library based on the Message Passing Interface (MPI) library (http://www.mcs.anl.gov/research/projects/mpi/), a standard for message passing in parallel comput–ing. CMAQ uses the Argonne National Laboratory open–source implementation of MPI, referred to as MPICH (http://www.mcs.anl.gov/research/projects/mpich2/), because of its widespread usage and availability.

While the domain decomposition paradigm is appropriate for the grid model, the plume component (SCICHEM) in the PinG model requires a different approach because the puffs are not distributed uniformly among the subdomains. For example, one could expect a higher density of puffs in subdomains with many point sources than in other subdomains. Thus, using a domain decomposition approach for SCICHEM would result in inefficient utilization of processors. Furthermore, there could be potential issues with puffs crossing subdomain boundaries during a simulation time step.

Hence, we selected “puff decomposition” as the approach for parallelizing the plume component of the model. The total puffs at any given time step are divided uniformly among the available processors. However, the strongly interactive nature of the puff calculation, including splitting, merging, and overlap calculations posed an additional challenge in the parallelization of the plume–in–grid code. Because these puff interactions could occur between puffs distributed among different processors, there would be a significant communication overhead associated with performing the interaction calculations on independent processors.

To overcome this issue, we focused our parallelization effort on the chemistry component of the plume model. This component that includes gas–phase chemistry, aerosol calculations, and aqueous–phase chemistry, requires more computing resources

than any of the other components of the model. Thus, the largest benefit could be achieved by conducting these chemistry calculations in parallel.

Based on these considerations, the overall parallelization approach of the embedded plume model consists of the following steps:

(1) The grid model subdomain concentrations are collected to construct a full domain 3D concentration field as background concentrations for SCICHEM, which is called at each CMAQ transport time step.

(2) The overall puff stepping control is maintained on the root processor, which performs the calculations for puff emissions, transport, dispersion and interactions and assembles the complete meteorology and ambient chemistry fields from the Eulerian subdomains.

(3) The root processor distributes the total puffs among itself and the slave processors to perform the chemistry calcu–lations.

(4) At the end of the chemistry calculations, the root pro–cessor collects the puff information from the individual processors.

(5) At the end of the SCICHEM simulation time step, the root processor performs a puff dumping calculation if necessary (i.e., transfers the puff information back to the grid model) to adjust the full domain gridded concentration field.

(6) The full domain 3D concentration field is distributed among the various subdomains for the host grid model parallel computations for the next transport time step.

As in the case of the host grid model, all the inter–processor communication required for distributing and collecting the puff data and subdomain 3D concentration fields is accomplished using MPI methods. The data exchange between the host grid model and the plume model consists of transfer of the full 3D concentration field from the host model to the plume model at the initiation of a SCICHEM time step, and the transfer of the adjusted 3D concen–tration field at the end of the time step from SCICHEM to the host grid model. In a parallel application, this data exchange requires the storage of an extra copy of the 3D concentration field, in–creasing the memory requirements of the model by nearly a factor of 2. All other data required by the plume model (i.e., meteorology, emissions) are directly read from model input files using the I/O API and netCDF libraries.

Figure 1 shows the hybrid decomposition scheme used for the parallelization of AMSTERDAM. The scheme is illustrated for a workstation with four computing or processing elements (PEs). The left panel shows the traditional domain decomposition method used in the grid model component, in which the four processors are used for 4 subdomains of the modeling domain. The right panel shows the puff decomposition method used for the reactive plume component, in which the puffs in the entire domain are divided among the 4 PEs for the chemistry calculations.

In the following section, which describes the model appli–cation with 158 point sources treated explicitly with the plume model, we present the computational speed–up obtained from parallelizing the PinG code.

4. Model Application

The parallelized version of CMAQ–MADRID–APT was applied to a modeling domain that covers the central and eastern United States (see Figure 2). The grid is based on a Lambert Conformal map projection, with the origin located at 97° west longitude and 40° north latitude and reference latitudes at 33° and 45° north latitude. The horizontal grid system consists of 243 x 246 grid cells, with a resolution of 12 km. The vertical grid is pressure–based and extends from the surface to about the tropopause (100 mb or ~15 km) and is discretized using 19 layers of variable thickness.

Based on an analysis of their emissions of SO_2 and NO_X, 158 large coal–fired power plants (CFPPs) in the United States were chosen for PinG treatment. The locations of these point sources are also shown in Figure 2. The SO_2 and NO_X emissions from these sources represent over 75% and 68% of the respective total CFPP emissions in the entire modeling domain.

Baseline simulations with the CMAQ–MADRID and CMAQ–MADRID–APT configurations of AMSTERDAM were conducted for a summer period (August 1 to August 31, 2002) and a winter period (January 15 to February 15, 2002). The CMAQ transport options for the AMSTERDAM simulations included the Yamartino horizontal advection scheme and the Asymmetric Convective Model (ACM2) for the vertical mixing. For the gas–phase chemistry, the EBI solver was used with the MADRID version of the Carbon Bond IV (CB–IV) mechanism.

The boundary conditions for the 12–km resolution modeling domain shown in Figure 2 were obtained from a coarse grid (36–km resolution) simulation over the continental United States, using the CMAQ–MADRID configuration of AMSTERDAM. The boundary conditions for the 36–km resolution coarse domain were obtained from a simulation with the global Goddard Earth Observing System with Chemistry (GEOS–CHEM) model (Bey et al., 2001), provided by Harvard University.

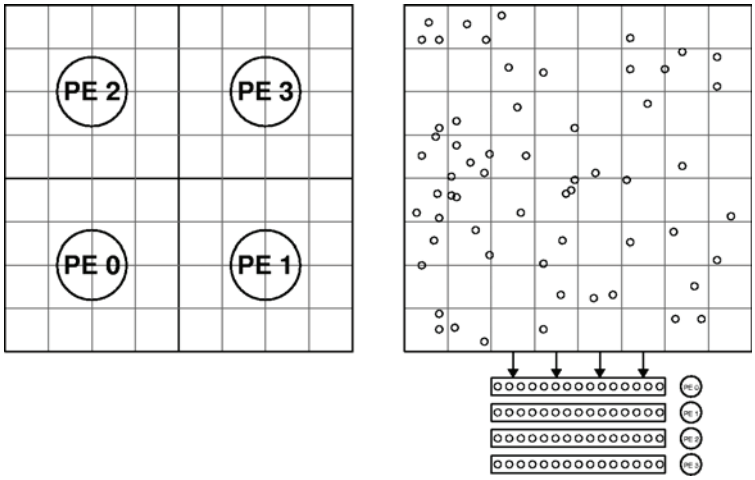

Figure 1. *Hybrid parallelization approach for the grid and embedded plume model components of AMSTERDAM.*

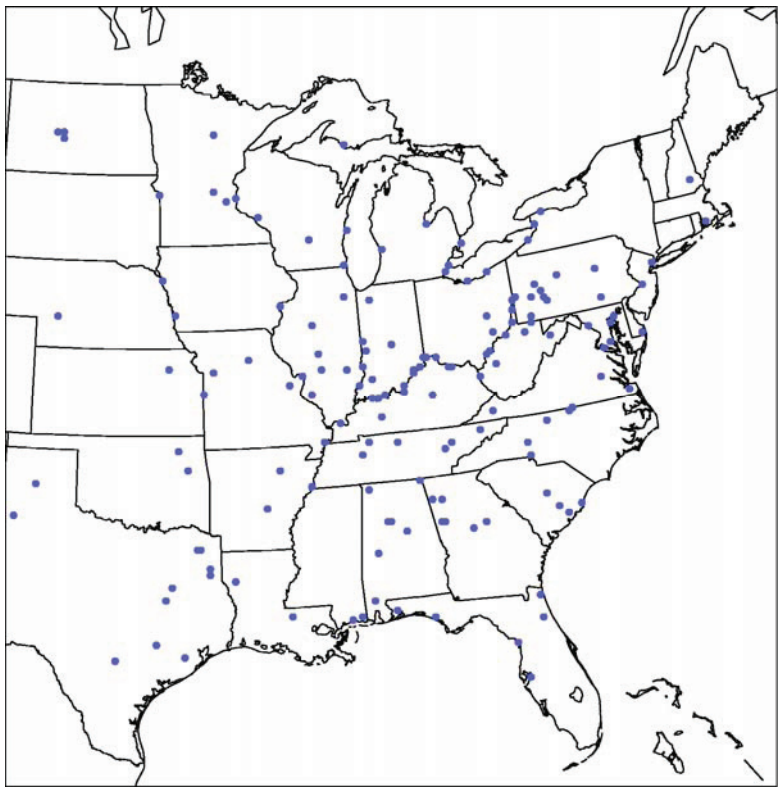

Figure 2. *Modeling domain and locations of point sources treated explicitly with SCICHEM.*

The meteorological inputs for the simulations were provided by ENVIRON International Corporation and were based on the outputs of the Fifth–Generation NCAR/Penn State Mesoscale Model (MM5) (Grell et al., 1994). Emissions data were also pro–vided by ENVIRON and were based on 2002 Continuous Emissions Monitoring Systems (CEMS) data for CFPPs in the U.S. and on 2002 data from the Regional Planning Organizations (RPOs) for other sources. These were processed to develop emissions for the AMSTERDAM simulations.

AMSTERDAM run–times for the MADRID–APT application with 158 point sources treated explicitly were about 40 to 50% higher than the corresponding run–times for the grid model only configu–ration. On a machine with 8 processors, the speed–up from the parallelization of the plume–in–grid configuration was about a factor of 4 to 5. This was comparable to the speed–up for the grid–only configuration of the model, indicating that the degree of parallelization of the plume–in–grid version was similar to that of the grid version. From Amdahl's law, this suggests that nearly 90% of the code is parallelized for both model configurations.

In the following section, we present results from the model performance evaluation for the summer and winter periods and the two configurations of AMSTERDAM.

5. Model Performance Evaluation

Data from the AIRS/AQS, IMPROVE, and SEARCH monitoring networks were used for the model performance evaluation of ambient ozone and $PM_{2.5}$ concentrations. The Aerometric Information Retrieval System (AIRS)/Air Quality System (AQS) (http://www.epa.gov/ttn/airs/airsaqs/), contains ambient air pollution data collected by EPA, state, local, and tribal air pollution control agencies from thousands of monitoring stations across the U.S. The Interagency Monitoring of Protected Visual Environments (IMPROVE) monitoring network (http://vista.cira. colostate.edu/improve/) consists of aerosol, light scatter, light extinction and scene samplers in a number of National Parks and Wilderness areas. In contrast to these two data sources, which have a nation–wide focus and provide routine measurements, the SouthEastern Aerosol Research and Characterization study (SEARCH) network is a highly instrumented regional network in the states of Alabama, Florida, Georgia and Mississippi (Hansen et al., 2003). In 2002, there were eight stations in the network, arranged in urban–rural pairs in each of the four states. SEARCH provides integrated filter–based measurements as well as year–round continuous measurements (1–60 min averages) of $PM_{2.5}$ and gas components.

Table 1 shows the model performance statistics for hourly ozone concentrations for the CMAQ–MADRID and CMAQ–MADRID–APT configurations of AMSTERDAM for the summer and winter periods using measurements from AIRS/AQS and SEARCH. Although EPA has revoked the one–hour ozone standard, the calculation of model performance statistics for one–hour ozone concentrations is still recommended (EPA, 2007). The statistical measures shown in Table 1 are defined by EPA (2007). A cut–off value of 20 ppb for the observed hourly ozone concentrations was used to remove the influence of very low observed concentrations on the performance statistics. EPA modeling guidance recom–mends using a cut–off value of 60 ppb (EPA, 2007); however, this cut–off would eliminate most of the observations for the winter period for our performance evaluation. Other studies have investigated the use of 20 ppb and 40 ppb cut–off values in addition to the 60 ppb recommended value (e.g., Hogrefe et al., 2001; Baker, 2005; Tong and Mauzerall, 2006). We selected the 20 ppb threshold to reduce the number of data points discarded for the performance evaluation.

As shown in Table 1, both configurations of the model (i.e., with and without PinG treatment) result in very similar perfor-mance statistics. Summer ozone concentrations tend to be slightly under–predicted and winter concentrations are slightly over–predicted. The normalized bias is less than 2% for the summer period and less than 10% for the winter period, within the EPA guidance value of ± 5 to ± 15% for hourly ozone concentrations (EPA, 1991; Russell and Dennis, 2000). Similarly, the normalized errors for both the summer and winter periods are less than the EPA guidance value of ±30 to ±35%. The modeled and observed

Table 1. *Model performance statistics for hourly ozone concentrations using AIRS/AQS and SEARCH measurements*

	Summer 2002		Winter 2002	
AMSTERDAM Configuration	MADRID	MADRID-APT	MADRID	MADRID-APT
Mean Observed Value (ppb)	45.8		31.5	
Mean Modeled Value (ppb)	42.9	43. 2	33.2	33.5
Gross Bias (ppb)	-2.9	-2.6	1.7	2.0
Mean Normalized Bias (%)	0.8	1.4	7.5	8.5
Normalized Mean Bias (%)	-6.9	-6.2	5.4	6.4
Fractional Bias (%)	-3.4	-2.8	2.0	2.9
Gross Error (ppb)	9.9	9.8	7.7	7.7
Mean Normalized Error (%)	22.8	22.7	26.0	26.2
Normalized Mean Error (%)	23.1	22.7	24.4	24.5
Fractional Error (%)	22.5	22.3	25.6	25.6
Normalized RMSE (%)	27.7	27.5	32.4	32.6
Coefficient of Determination (r^2)	0.55	0.56	0.23	0.23

ozone concentrations are well correlated for the summer period, when the model explains more than 50% of the variance in observed concentrations. The correlation for the winter period is lower.

Figure 3 shows scatter–plots comparing observed daily maximum 8–hour average ozone concentrations with the simulated maximum 8–hour average concentrations for the summer period. The left panel shows the results for the MADRID configuration while the right panel shows the results for the MADRID–APT configuration of AMSTERDAM. The results are similar for the two model configurations – for both configurations, we note a high degree of correlation between the modeled and observed maximum 8–hour ozone concentrations, consistent with the correlation noted earlier in Table 1 for the hourly concentrations. However, we also note some over–prediction biases at the low end of the measured concentrations and under–prediction biases at the high end. This is also illustrated in the residual scatter plots (i.e., errors of prediction vs. measured values) for the daily maximum 8–hour average ozone concentrations, shown in Figure 4. Note that residual plots traditionally show the predicted values on the x–axis; however, we have used measured concentrations in Figure 4 to determine the accuracy of model predictions over the range of observed values.

The corresponding scatter–plots and residual scatter plots for the winter period are shown in Figures 5 and 6, respectively. The results for the two model configurations are again similar. As expected, peak winter ozone concentrations are lower than summer values, because of the lower photochemical production in winter. We see from Figure 6 that most of the over–prediction bias in modeled ozone results for the winter period is associated with 8–hour maximum ozone concentrations less than 40 ppb, and more than 50% of the observed values are lower than this threshold. The bias is considerably lower at the higher concentrations. Figure 5 shows that there is considerably more scatter in the winter period results than the summer results, again associated with the difficulties in predicting concentrations lower than 40 ppb. The lower correlation for the 8–hour maximum ozone concentrations for the winter period is consistent with the lower coefficient of determination noted in Table 1 for the hourly ozone concentrations. Cai et al. (2008) have evaluated an air quality forecast modeling system for summer and winter seasons and have noted that the performance for the winter season was lower than for the summer season. They point out that ozone concentrations in the winter season are dominated by background concentrations and titration by NO_X with little photochemical production.

EPA modeling guidance also recommends the calculation of the average peak prediction bias and error for an operational evaluation of model performance for ozone (EPA, 2007). We calculated these measures for the daily maximum 8–hour average concentrations (the basis of the primary and secondary ozone standards). For the summer period, the average peak prediction bias for both configurations of AMSTERDAM is –8% and the average peak prediction error is 17%. For the winter period, the average peak prediction bias is 9% for the MADRID configuration and 10% for the MADRID–APT configuration; the average prediction error for both configurations is 21%.

Figure S1 (see the Supporting Material, SM) shows the average station peak estimation accuracy for daily maximum 8–hour average ozone concentrations for each day of the summer period for the MADRID–APT configuration of AMSTERDAM (the results for the MADRID configuration are similar and are not shown here). This statistic is the average over all stations of the spatially paired peak estimation accuracy, i.e., the discrepancy between the magnitude of the peak daily 8–hour average measurement at a monitoring station and the peak daily estimated value at the same monitor. As shown in Figure S1a, the average station peak estimation accuracy is within ±20% for all but 4 days of the summer period. For the winter period, shown in Figure S1b, the average station peak 8–hour average estimation accuracy is within ±20% for only about 60% of the days in the period, again confirming that the model performs better for the summer period than the winter period.

The performance statistics for 24–hour average $PM_{2.5}$ concentrations for the summer and winter periods are shown in Table 2. These statistics are based on measured concentrations from the AIRS/AQS, IMPROVE, and SEARCH networks. The performance statistics for the individual $PM_{2.5}$ components (sulfate, nitrate and others) are provided in the SM (Tables S1 through S5). As in the case of the hourly ozone concentrations, the $PM_{2.5}$ performance statistics for the two model configurations are almost identical. The PinG configuration (MADRID–APT) tends to predict slightly lower total $PM_{2.5}$ concentrations than the grid–only configuration (MADRID). The modeled and observed concentrations are well correlated, especially for the summer period. Although the mean normalized bias is significantly higher for the summer period than for the winter period, both the normalized mean bias and fractional bias are small for the two periods. Because no cut–off or threshold was used for the observed $PM_{2.5}$ concentrations in calculating the model performance statistics, the mean normalized bias is the least reliable indicator of model performance among the three measures. The performance statistics shown in Table 2 indicate that both model configurations do a good job of estimating total $PM_{2.5}$ concentrations. However, the

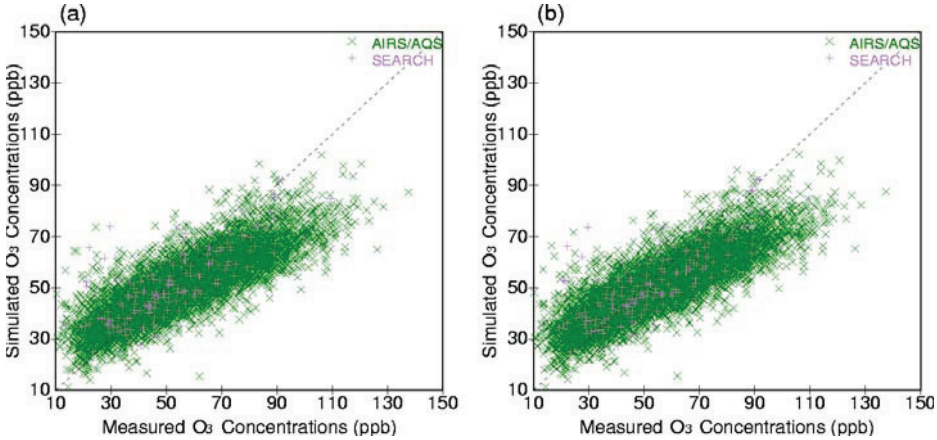

Figure 3. *Comparison of maximum 8–hour average simulated and observed ozone concentrations for the summer period for* ***(a)*** *the grid–only or CMAQ–MADRID configuration and* ***(b)*** *the PinG or CMAQ–MADRID–APT configuration of AMSTERDAM.*

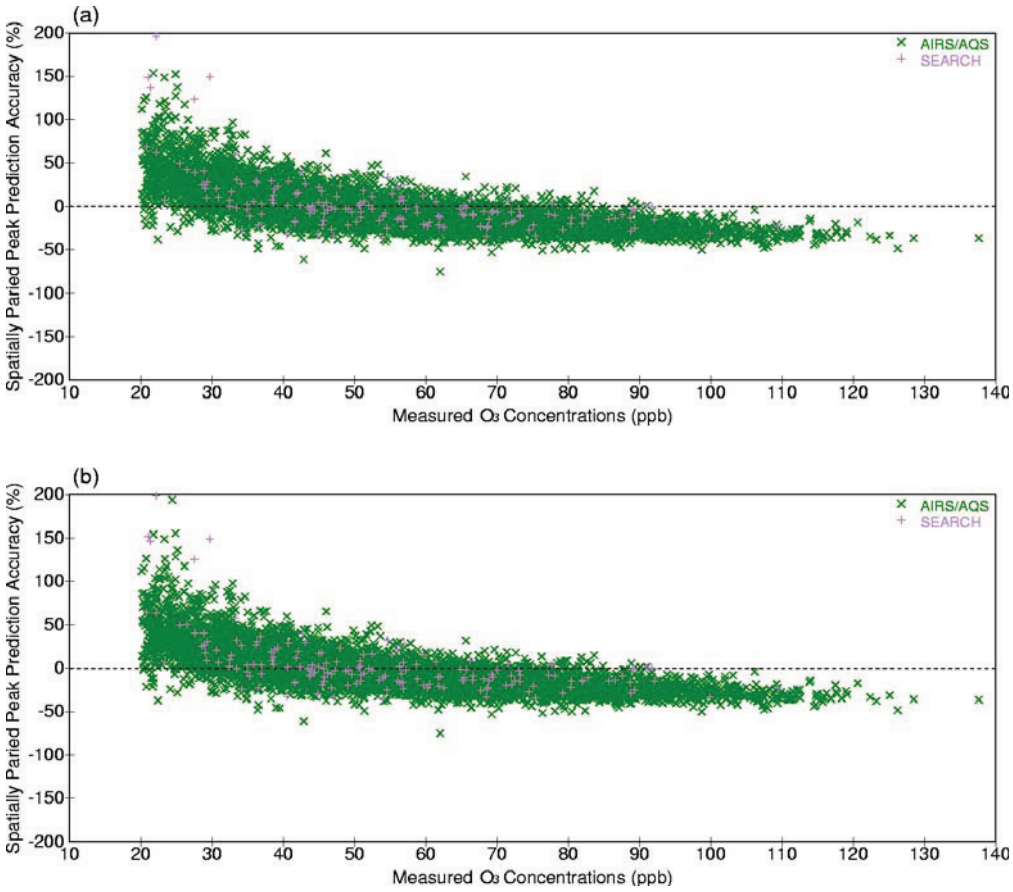

Figure 4. *Spatially paired peak prediction accuracy for 8–hour average ozone concentrations for the summer period for* ***(a)*** *the grid–only configuration and* ***(b)*** *the PinG configuration of AMSTERDAM.*

performance for individual $PM_{2.5}$ components is more variable, with better performance for sulfate (see the SM, Table S1) than for organic matter and black (elemental) carbon (Tables S4 and S5) or nitrate and ammonium (Tables S2 and S3).

Figure 7 shows scatter plots of the observed and estimated 24–hour average total $PM_{2.5}$ concentrations for the summer period and the two model configurations. The observed and modeled values are generally in good agreement, and are well correlated (note high r^2 value in Table 2). The agreement for the winter period is not as good and there is considerably more scatter between the observed and modeled values, as shown in Figure 8. Appel et al. (2008) have also noted better model performance for $PM_{2.5}$ in spring and summer as compared to fall and winter. Note that sulfate constitutes a large fraction of the total $PM_{2.5}$ mass in summer and is well–predicted, as shown in Table S1 (see the SM). On the other hand, nitrate, which is a much larger component of $PM_{2.5}$ mass in winter than in summer, is poorly predicted (see the SM, Table S2).

6. Plume–in–Grid Impacts: Beyond Model Performance

The model evaluation, discussed above, shows that the grid–only and PinG configurations of AMSTERDAM display very similar statistical performance characteristics at the locations of the monitors. The similarities in model performance, as reported in

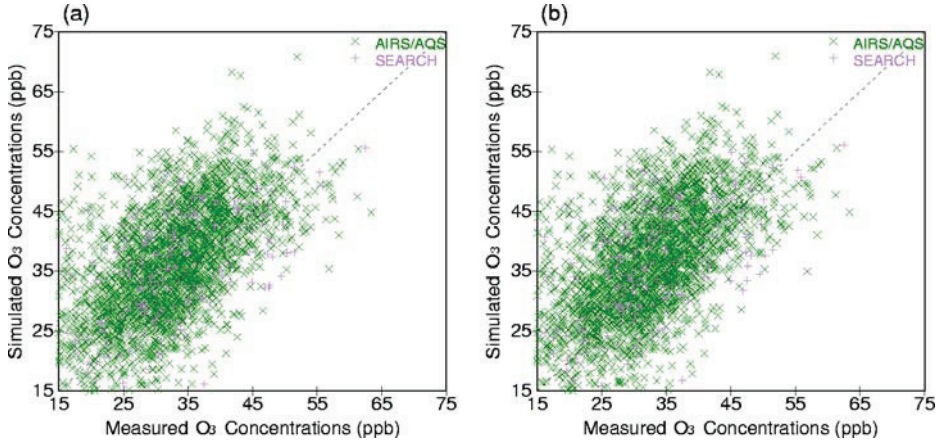

Figure 5. *Comparison of maximum 8–hour average simulated and observed ozone concentrations for the winter period for* ***(a)*** *the grid–only configuration and* ***(b)*** *the PinG configuration of AMSTERDAM.*

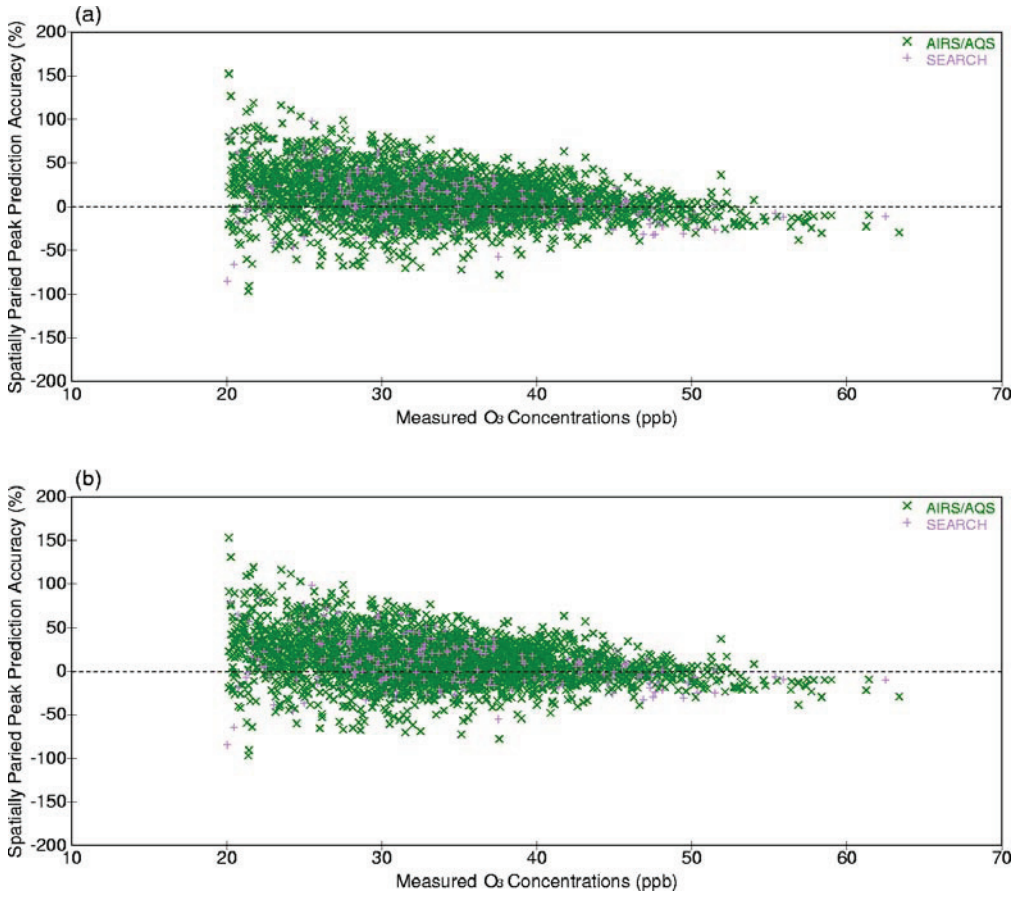

Figure 6. *Spatially paired peak prediction accuracy for 8–hour average ozone concentrations for the winter period for* ***(a)*** *the grid–only configuration and* ***(b)*** *the PinG configuration of AMSTERDAM.*

past comparisons of the grid model and PinG model (Karamchandani et al., 2002; Karamchandani et al., 2006a; Vijayaraghavan et al., 2006), are associated with the majority of the monitoring sites being unaffected or only slightly affected by the plumes from the point sources selected for explicit treatment by the plume model.

Model evaluation is an aggregate measure of model skill. Although having similar performance statistics, the two configu–rations can yield significantly different results on the specific nature of predicted ozone and $PM_{2.5}$. These differences can influ–ence the estimation of the contributions of major point sources to ozone and $PM_{2.5}$ concentrations.

In a companion paper (Karamchandani et al., 2010), we discuss the effects of hypothetical emission scenarios using the two configurations of AMSTERDAM (i.e., grid–only and PinG), as well as the contributions of the 158 CFPPs selected for PinG treatment to ozone and $PM_{2.5}$ concentrations. Here, we present results showing the differences between the results from the two configurations for the base simulation. These differences arise because of differences in the treatment of transport and chemistry of elevated point source emissions in the two configurations of the model. As discussed in Karamchandani et al. (2000), the chemistry of a coal–fired power plant plume is significantly different from the

Table 2. *Model performance statistics for daily $PM_{2.5}$ concentrations using AIRS/AQS, IMPROVE and SEARCH measurements*

	Summer 2002		Winter 2002	
AMSTERDAM Configuration	MADRID	MADRID-APT	MADRID	MADRID-APT
Mean Observed Value (μg m^{-3})	14.1		10.5	
Mean Modeled Value (μg m^{-3})	14.0	13. 8	10.9	10.9
Gross Bias (μg m^{-3})	-0.1	-0.3	0.35	0.32
Mean Normalized Bias (%)	27.2	26.4	14.7	14.2
Normalized Mean Bias (%)	-0.9	-1.9	3.3	3.0
Fractional Bias (%)	2.4	1.4	-0.5	-0.9
Gross Error (μg m^{-3})	4.4	4.5	4.27	4.28
Mean Normalized Error (%)	55.5	55.7	45.1	44.9
Normalized Mean Error (%)	31.7	32.2	40.5	40.6
Fractional Error (%)	38.1	38.3	39.3	39.4
Normalized RMSE (%)	42.5	42.7	48.0	48.2
Coefficient of Determination (r^2)	0.62	0.61	0.35	0.35

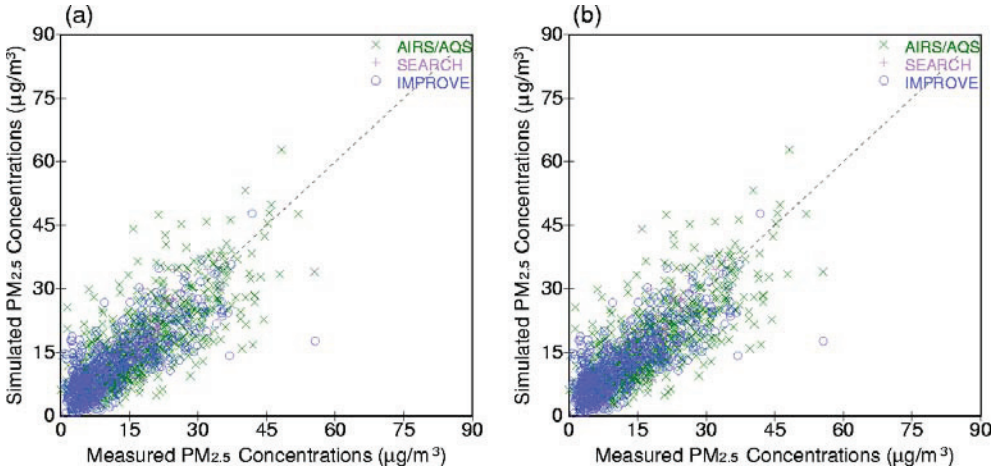

Figure 7. *Comparison of monthly average simulated and observed $PM_{2.5}$ concentrations for the summer period for* ***(a)*** *the grid–only configuration and* ***(b)*** *the PinG or configuration of AMSTERDAM.*

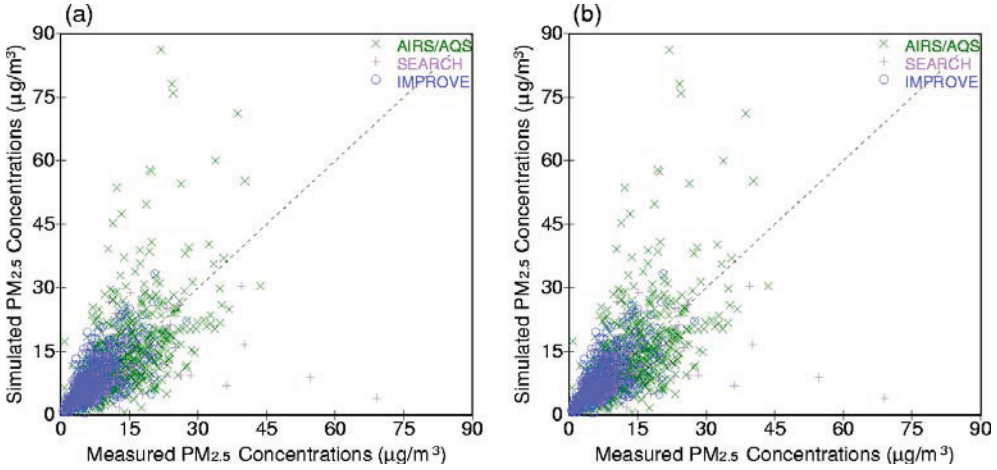

Figure 8. *Comparison of monthly average simulated and observed $PM_{2.5}$ concentrations for the winter period for* ***(a)*** *the grid-only configuration and* ***(b)*** *the PinG configuration of AMSTERDAM.*

background chemistry and the plume traverses several stages of differing chemistry before its chemical characteristics approach that of the background atmosphere.

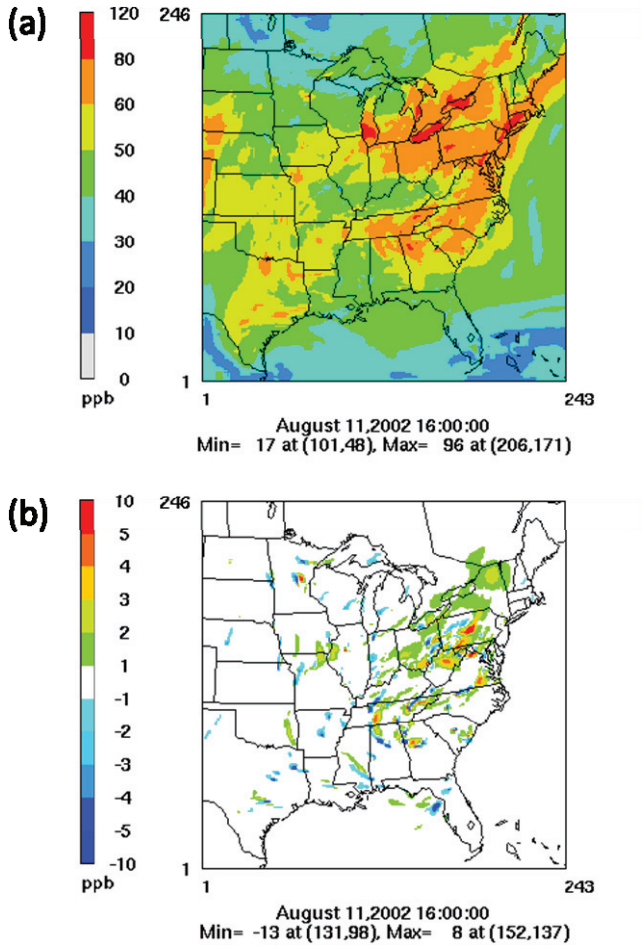

Figure 9. *Spatial patterns of* ***(a)*** *8–hour average surface ozone concentrations on August 11, 2002, predicted with the grid–only configuration of AMSTERDAM and* ***(b)*** *differences in 8–hour average surface concentrations between the PinG and grid–only configurations of AMSTERDAM.*

Figure 9a shows the spatial pattern of 8–hour average surface ozone concentrations, on August 11, 2002, estimated with the PinG configuration of AMSTERDAM. This day was selected for analysis because some of the highest 8–hour average concentrations in August 2002 were predicted on this day. As seen in Figure 9b, which shows the differences in 8–hour average ozone concentrations between the PinG and grid configurations, using the PinG approach results in decreases of up to 13 ppb and increases of about 8 ppb in predicted ozone concentrations. As discussed in Karamchandani et al. (2002) and Vijayaraghavan et al. (2006), some of the increases in surface ozone due to PinG treatment at the locations of elevated point sources can be attributed to reduced near–source titration of background surface ozone by the NO_X emissions from those sources. In the grid model, the NO_X emissions are instantaneously brought to the surface resulting in artificially enhanced titration of ozone, while the PinG version allows the emissions to be transported for a longer distance downwind before they affect surface ozone concentrations. Other ozone increases in the PinG approach can be attributed to delayed production of ozone in the plume as it is transported downwind to NO_x–limited environments. The decreases in ozone using PinG are associated with lower production of ozone in the early stages of plume transport.

Figure 10a shows the distribution of the predicted monthly average surface $PM_{2.5}$ concentrations across the modeling domain for August 2002, using the PinG configuration of AMSTERDAM. The high $PM_{2.5}$ concentrations (over 1 000 μg m^{-3}) along the northern border of the modeling domain are associated with wild fires in the region during August 2002. The effect of using the PinG approach on $PM_{2.5}$ predictions is illustrated in Figure 10b, which shows regions of both decreases and increases in $PM_{2.5}$ concentrations. The decreases are generally larger in magnitude than the increases; the maximum decrease is larger than 12 μg m^{-3}, while the maximum increase is less than 1 μg m^{-3}. The differences between the grid–only and the PinG approaches can again be explained by the differences in their treatment of the transport and chemistry of SO_2 and NO_X emissions from large CFPPs. The NO_X in the plumes from these elevated sources inhibits the oxidation of the primary species to the secondary products (sulfate and nitrate) in the early stages of plume dispersion, and the emissions are transported aloft for larger distances than surface emissions (Karamchandani et al., 2000; Karamchandani et al., 2006a). These phenomena are treated in the PinG model but cannot be captured by the grid model, which is limited by its framework to mix emissions instantaneously within one or more grid cell volumes.

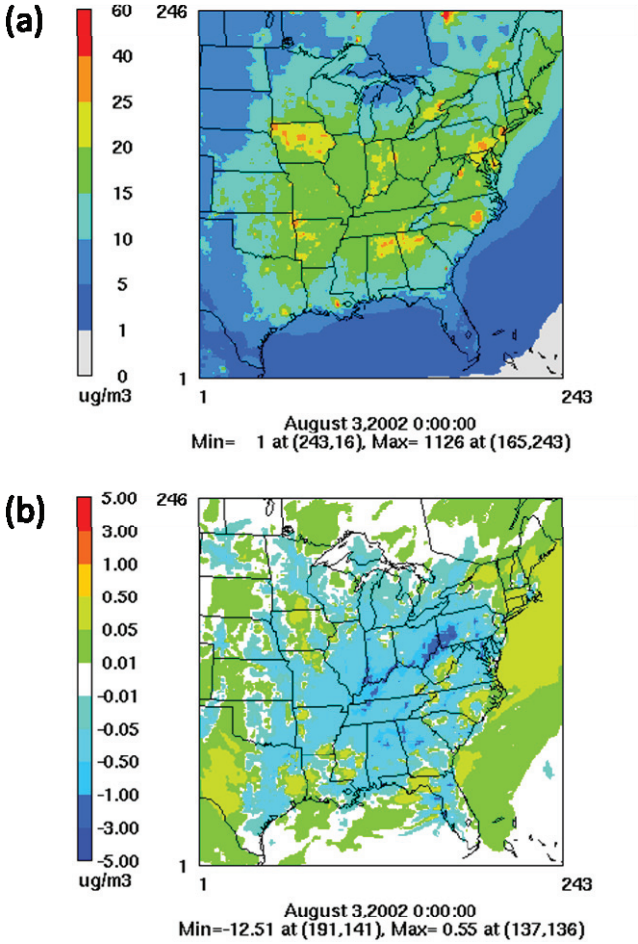

Figure 10. *Spatial patterns of* ***(a)*** *monthly average surface $PM_{2.5}$ concentrations in August 2002, predicted with the grid–only configuration of AMSTERDAM and* ***(b)*** *differences in monthly average surface $PM_{2.5}$ concentrations between the PinG and grid–only configurations of AMSTERDAM.*

7. Summary and Conclusions

We have presented the development of a parallel version of an advanced air quality modeling system, based on the framework of the U.S. EPA community model, CMAQ, but with alternative treatments of particulate matter formation and an advanced treatment of sub–grid scale plumes from large elevated point sources. The development and application of the single–processor

version of this modeling system has been described previously. However, computational constraints limited the application of this version to small–domain/short–term simulations, and allowed the explicit treatment with the embedded plume model of only a small fraction of large point sources of interest. The development of the parallel version increases the utility of the model and allows us to conduct long–term simulations over a regional domain, as well as to explicitly simulate over 150 coal–fired power plants (CFPPs), representing approximately 70% of the CFPP SO_2 and NO_X emis–sions in the modeling domain.

We describe the application of the model for summer and winter months in 2002 and present results from the evaluation of the model using available ambient measurements of ozone and $PM_{2.5}$. The model was applied in two configurations – the grid–only version, with all sources treated with the grid model, and the plume–in–grid (PinG) version, with 158 CFPPs explicitly treated with the plume model. For both the summer and winter periods, the model performance statistics for both 1–hour ozone concen–trations and maximum daily 8–hour ozone concentrations are within suggested guidelines (EPA, 1991; Russell and Dennis, 2000). The performance for the summer period is better than the winter period. Ozone concentrations are much lower in the winter than in summer due to reduced photochemical production and this influ–ences the model performance in winter. Total $PM_{2.5}$ concentrations and $PM_{2.5}$ sulfate concentrations are well–predicted by the model. However, $PM_{2.5}$ nitrate concentrations are poorly predicted. The observed and simulated total $PM_{2.5}$ concentrations are highly correlated for the summer period, but the agreement is lower for the winter period. The lower agreement in winter is possibly associated with the difficulties in predicting concentrations of $PM_{2.5}$ nitrate, which is a large component of the winter aerosol.

Both model configurations (grid–only and PinG) give very similar model performance results. This similarity has been noted in previous PinG modeling studies and is associated with the majority of the monitoring sites being only slightly affected or unaffected by the plumes from the point sources selected for explicit treatment by the plume model. However, it should be noted that when measurements are available at monitoring locations located downwind of major point sources, then the plume–in–grid simulation can capture plume events more successfully than the grid model simulation. This was shown in the study by Karamchandani et al. (2006a), who evaluated grid model and PinG model performance for plume events, using data from the Southeastern Aerosol Research and Characterization study (SEARCH) network. Thus, the similarities in aggregate model per–formance conceal the smaller–scale differences between the grid and PinG models.

More importantly, previous studies and this study show significant differences between the PinG and grid–only versions in their predicted spatial distribution of ozone and $PM_{2.5}$ concen–trations. These differences can be important in determining source contributions to ambient concentrations. A companion paper (Karamchandani et al., 2010) examines the differences in the predicted impacts from the two configurations of the model for hypothetical emission scenarios, and shows that the two models predict significantly different source contributions, particularly to summertime $PM_{2.5}$ concentrations.

Although PinG modeling increases the computational requirements for an air quality model simulation, we believe that the additional overhead (about 20 to 30% for 50 sources and about 40 to 50% for over 150 sources) is acceptable in exchange for correctly treating elevated point source plumes at the subgrid–scale to overcome an inherent and well–recognized limitation of grid–only models. With the advances in computing capabilities over the last few years and expected advances in the future, conducting a seasonal or annual PinG simulation with over 100 sources for a large modeling domain is no longer a research–only exercise, as demonstrated in this study.

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Supporting Material Available

Model performance statistics for daily $PM_{2.5}$ sulfate concentrations using AIRS/AQS, IMPROVE and SEARCH measurements (Table S1), Model performance statistics for daily $PM_{2.5}$ nitrate concentrations using AIRS/AQS, IMPROVE and SEARCH measurements (Table S2), Model performance statistics for daily $PM_{2.5}$ ammonium concentrations using AIRS/AQS, IMPROVE and SEARCH measurements (Table S3), Model perform-ance statistics for daily $PM_{2.5}$ organic matter concentrations using AIRS/AQS, IMPROVE and SEARCH measurements (Table S4), Model performance statistics for daily $PM_{2.5}$ black carbon concentrations using AIRS/AQS, IMPROVE and SEARCH measurements (Table S5), Average station peak prediction accuracy for daily maximum 8–hour average ozone concentrations with the PinG model for (a) the summer period and (b) the winter period in 2002 (Figure S1). This information is available free of charge via the Internet at http://www.atmospolres.com.

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