

Development and initial application of a sub-grid scale plume treatment in a state-of-the-art online Multi-scale Air Quality and Weather Prediction Model

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H I G H L I G H T S

- ▶ First implementation of plume-in-grid (PinG) treatment in online-coupled WRF/Chem.
- ▶ Improved model capability in simulating sub-grid scale plumes with PinG.
- ▶ Large differences in predicted spatial patterns of pollutants with and without PinG.
- ▶ Important implications to simulated aerosol direct and indirect feedbacks.

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Traditional Eulerian air quality models are unable to accurately simulate sub-grid scale processes, such as the near-source transport and chemistry of point source plumes, because they assume instantaneous mixing of the emitted pollutants within the grid cell containing the release, and neglect the turbulent segregation effects that limit the near-source mixing of emitted pollutants with the background atmosphere (e.g., Kramm and Meixner, 2000). Observations by Dlugi et al. (2010) show that the segregation of chemically reactive species can slow effective second-order reaction rates by as much as 15%, due to inhomogeneous mixing of the reactants. This limitation of traditional grid models applies to both “off-line” models, in which externally derived meteorology is used to drive the chemistry model, and newer “on-line” models, such as the Weather Research and Forecasting model with Chemistry (WRF/Chem), that simulate the emissions, transport, mixing, and chemical transformation of trace gases and aerosols simultaneously with the meteorology. While a number of approaches have been used in the past to address this limitation, the approach that has been most effectively used in operational models is the plume-in-grid (PinG) approach, in which a reactive plume model is embedded within the grid model to resolve sub-grid scale plumes. This paper describes the implementation of such a PinG treatment in WRF/Chem, based on a similar extension to the U.S. EPA Community Multi-scale Air Quality (CMAQ) model. The treatment, referred to as Advanced Plume Treatment, has been tested in CMAQ over more than a decade and has been used successfully in both episodic and long-term applications for assessing point source contributions to ozone and particulate matter. This paper presents the application of the PinG version of WRF/Chem for a three-day episode in July 2001, including a model performance evaluation and comparison of model results with and without PinG treatment. The results from the model application show that overall model performance is only slightly affected when the PinG treatment is used, although there are some generally small improvements, with the PinG treatment showing a 5% lower bias in predicting ozone concentrations, and 3% lower bias in sulfate predictions. However, the predicted spatial patterns of ozone and PM_{2.5} concentrations from the two simulations show both large decreases of up to 40 ppb ozone and 14 μg m⁻³ PM_{2.5}, and increases of up to 80 ppb ozone and 33 μg m⁻³ PM_{2.5} as a result of using the PinG treatment. These differences are attributed to both direct effects of the PinG treatment (i.e., differences in dispersion, transport and chemistry of point source emissions) and indirect effects (i.e., impacts of air quality changes on meteorology).

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1. Introduction

Meteorological and chemical processes in the atmosphere are closely coupled and an accurate simulation of the climate-chemistry-aerosol-cloud-radiation feedbacks requires the use of fully coupled meteorology, climate, and chemistry models (e.g., Zhang, 2008). A number of models with such an online coupling are now available. The Weather Forecasting and Research with Chemistry (WRF/Chem) model (Grell et al., 2005; Peckham et al., 2009, 2011) is an example of such a fully-coupled meteorology and chemistry model. WRF/Chem is a community model for the regional scale that is being adapted to simulate the range of scales from global through urban (Zhang et al., 2009a,b; Peckham et al., 2011) to understand the interactions between climate and air quality over scales ranging from hundreds of kilometers to tens of kilometers using multiple nested grids. However, even the finest grid resolution of 12 km or 4 km is insufficient to resolve sub-grid scale effects, such as the near-source transport and chemistry of emissions from elevated point sources, because the initial dimensions of stack plumes from these sources are of the order of tens of meters, and the dimensions of these plumes typically approach the model grid resolution only after they have traveled several grid cells downwind of the source locations. Because a significant fraction of emissions in the U.S. (particularly the eastern U.S.) can be attributed to elevated point sources (Chapman et al., 2009; Tran and Mölders, 2012), this limitation of all traditional grid models, including WRF/Chem, can lead to errors in simulating the effects of these sources on air quality.

Several modeling studies (e.g., Karamchandani et al., 2002, 2006, 2010a, 2010b; Vijayaraghavan et al., 2009) have demonstrated how the inability of traditional grid models to resolve sub-grid scale plumes can lead to errors in determining (1) the contribution of elevated point sources to ambient concentrations and deposition fluxes, and (2) the model response to emissions changes from elevated point sources or from other sources that may affect the chemistry of the elevated point source emissions. A number of approaches can be employed to address this limitation, such as plume-in-grid (PinG) modeling (e.g., Karamchandani et al., 2002), the use of interactive static nested grids (e.g., Odman and Russell, 1991; Kumar et al., 1994; Kumar and Russell, 1996a) and adaptive grid techniques (e.g., Tomlin et al., 1997; Srivastava et al., 2001; Garcia-Menendez et al., 2009). However, the PinG approach, has been used most often and is most practical from an operational point of view (e.g., Seigneur et al., 1983; Gillani, 1986; Sillman et al., 1990; Morris et al., 1991; Kumar and Russell, 1996b; Myers et al., 1996; Gillani and Godowitch, 1999; Karamchandani et al., 2002, 2006, 2010a,b; Vijayaraghavan et al., 2006, 2009). In this approach, a reactive puff model is embedded within the host grid model to track the near-source transport and chemistry of sub-grid scale plumes. In this paper, we present the development of a PinG version of WRF/Chem using this approach.

2. Plume-in-grid modeling

In the PinG modeling approach, the errors associated with the grid-averaging of stack emissions are reduced by imbedding a puff or plume model within the grid model. The embedded model tracks the sub-grid scale process occurring in the point source plume until the fine scale variability becomes unimportant (referred to as the “puff dumping” or “hand-over” point), at which point the grid model takes over the calculations for these mature puffs, while the embedded puff model continues tracking other active puffs and puffs arising from new releases of the emissions from the point sources.

PinG modeling has a long history, particularly for ozone modeling, and is now being used for multi-pollutant modeling as well. For example, the first sub-grid scale treatment of plumes in 3-D air quality models, referred to as the Plume-Airshed Reactive-Interacting System (PARIS), was developed nearly 3 decades ago by Seigneur et al. (1983), who embedded a simple puff model into the Urban Airshed Model (UAM) (Reynolds et al., 1973), and used it to calculate ozone concentrations in urban areas such as Los Angeles, California, and St. Louis, Missouri. Morris et al. (1991) described a PinG treatment in the variable grid version of UAM (UAM-V). Other early treatments include those of Sillman et al. (1990), who implemented PinG treatment in a two-dimensional (single layer) Eulerian grid model, and Myers et al. (1996), who implemented PinG treatment in the SARMAP Air Quality Model (SAQM) of Chang et al. (1997). Kumar and Russell (1996b) incorporated a sub-grid scale treatment of plumes in the Urban and Regional Multiscale (URM). Initial versions of CMAQ also included a PinG treatment (Gillani and Godowitch, 1999; Godowitch, 2004), based on a reactive plume model developed by Gillani (1986).

These early PinG models treated plumes at a sub-grid scale, thereby eliminating some of the errors associated with the grid model. However, they could not represent the complex dispersion processes associated with the plume mixing into the background air because the plume dimensions were represented by simple geometric functions (columns, grids, ellipses, or Gaussian distributions). Physical phenomena such as the effect of wind shear on plume dispersion, the effect of plume overlaps (e.g., under conditions of flow reversal or merging of adjacent plumes), and the effect of atmospheric turbulence on chemical kinetics were not represented in these models.

In the last decade, new plume models, such as the Second-Order Closure Integrated Puff with Chemistry model (SCICHEM) (Karamchandani et al., 2000), have been developed that address the above deficiencies and also include a detailed treatment of in-plume atmospheric chemistry. SCICHEM has been embedded (Karamchandani et al., 2002, 2006) into the U.S. EPA Community Multi-scale Air Quality (CMAQ) modeling system (Byun and Schere, 2006) to develop a PinG model, referred to as CMAQ with an Advanced Plume Treatment (CMAQ–APT). The latest available version of CMAQ–APT includes SCICHEM embedded in CMAQ version 4.6 (Karamchandani et al., 2010a, 2010b).

SCICHEM has been well tested in both stand-alone (e.g., Karamchandani et al., 2000, 2011) and plume-in-grid modes (e.g., Karamchandani et al., 2002), and the PinG version in CMAQ–APT has been applied successfully for a long-term application with over 100 point sources (e.g., Karamchandani et al., 2010a, 2010b). It is thus selected for the PinG implementation in WRF/Chem in this study. The following section provides a brief summary of SCICHEM and its features.

3. SCICHEM

SCICHEM is a state-of-the science puff model that represents puff expansion using second-order closure with an advanced chemistry module that represents detailed chemical interactions between plume pollutants and the surrounding atmosphere (Karamchandani et al., 2000).

The puff dispersion component of the model is referred to as Second-Order Closure Integrated Puff (SCIPUFF) (Sykes et al., 1988). Plumes are represented by a collection of three-dimensional puffs that are advected and dispersed according to the local characteristics (wind speed and direction, turbulence) of the atmosphere. Puff transport and dispersion are simulated using a second-order closure approach to solve the turbulent diffusion

equations. 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 effects of buoyancy on plume rise and initial dispersion are simulated by solving the conservation equations for mass, heat, and momentum.

The plume 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, based on the size of the puff relative to the grid cell size. 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. Note that, in the plume-in-grid implementation of SCICHEM, splitting is based only on the vertical spread of the puff, while the horizontal spread is used to determine if the puff should be handed over (dumped) to the grid model.

SCIPUFF has undergone continuous development and improvement since its inception more than two decades ago (Sykes et al., 1993; Sykes and Henn, 1995). Improvements include: (i) adaptive time stepping, allowing small time steps for puffs near the source, without additional computation for larger puffs that evolve on longer time scales; (ii) generalized description of shear distortion within the Gaussian framework; (iii) generalized puff splitting/merging algorithms to describe inhomogeneous meteorology; and (iv) incorporation of mesoscale turbulence description to simulate long-range transport.

The chemistry module in SCICHEM includes a gas-phase photochemical mechanism, aerosol chemistry (including sulfate/nitrate/ammonium and secondary organic aerosols), and aqueous-phase chemistry (Karamchandani et al., 2006). Chemical species concentrations in the puffs are treated as perturbations from the background concentrations. The gas-phase chemical reactions within the puffs are simulated using a general framework that allows any chemical kinetic mechanism to be treated, with relatively straightforward modifications to the code to handle the rate constant calculations for new reaction types in alternative chemical mechanisms, and to the SCICHEM control file which contains the list of reactions, reaction types and rate constants. However, the aerosol and aqueous-phase chemistry module codes are more hard-wired and require substantial modifications for alternative gas-phase chemistry mechanisms and alternative aerosol and aqueous-phase chemistry treatments.

SCICHEM has been successfully evaluated using helicopter measurements of day-time power plant plumes, including plume widths and plume chemistry (Karamchandani et al., 2000). It was recently applied and evaluated using day-time aircraft measurements of a power plant plume in northeastern Texas (Vijayaraghavan et al., 2010). More recently, the model has been successfully evaluated for night-time power plant plumes in northern Texas (Karamchandani et al., 2011), using data collected by the NOAA P-3 helicopter during the second Texas Air Quality Study in 2006 (TexAQ5 II or TEXAQ5 2006).

In the following section, we describe the implementation of SCICHEM inside WRF/Chem, including updates to the SCICHEM gas-phase chemistry and aerosol chemistry modules for consistency with the corresponding treatments in WRF/Chem.

4. The implementation of SCICHEM into WRF/Chem

The host model, WRF/Chem, used for the implementation of SCICHEM in this work is an updated version of WRF/Chem with the Model of Aerosol Dynamics, Reaction, Ionization and Dissolution (MADRID) (referred to as WRF/Chem-MADRID) of Zhang et al. (2010, 2012). WRF/Chem-MADRID is developed based on WRF/Chem 3.0. It provides the option to use any of three gas-phase mechanisms that are coupled with the MADRID aerosol module. For this application, the 2005 Carbon Bond Mechanism (CB05) of Yarwood et al. (2005) was used. The major update for the version of WRF/Chem-MADRID used in this work is the coupling of the CB05 gas-phase mechanism with two additional aerosol modules available in WRF/Chem 3.0, i.e., the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) (Zaveri et al., 2008) and the Modal Aerosol Dynamics Model for Europe/the Secondary Organic Aerosol Model (MADE/SORGAM; Ackermann et al., 1998; Schell et al., 2001). The WRF/Chem aqueous-phase chemistry module is based on the Carnegie Mellon University (CMU) aqueous-phase mechanism of Fahey and Pandis (2001). For the plume-in-grid application, the MADE/SORGAM aerosol option was selected.

The version of SCICHEM in CMAQ-APT (Karamchandani et al., 2010a) was used as the starting point for the PinG implementation in the updated WRF/Chem. This version includes chemistry treatments that are different from those in WRF/Chem. The gas-phase chemistry and aerosol chemistry treatments are based on the Carbon Bond IV mechanism (Gery et al., 1989) and the MADRID aerosol treatment (Zhang et al., 2004), respectively, and the aqueous-phase chemistry treatment is similar to that used in CMAQ 4.6, which is based on the Walcek and Taylor (1986) treatment in the Regional Acid Deposition Model (RADM; Chang et al., 1987). Therefore, the chemistry modules in SCICHEM were updated to be consistent with those in WRF/Chem. As the first step of the PinG code development and testing, only the CB05 gas-phase chemistry and the MADE/SORGAM aerosol modules were implemented in SCICHEM for the PinG application described in this paper. Thus, this version only allows the examination of the impact of PinG on the formation of O₃ and PM_{2.5} under cloud-free conditions. Future versions of the plume model will incorporate an aqueous-phase chemistry module compatible with WRF/Chem when resources are available.

In addition to the above differences in the science modules of WRF/Chem and CMAQ, there are significant differences in their frameworks. Thus, the implementation required substantial modifications to the reactive plume model code as well as the interface codes controlling the flow of information between SCICHEM and the host model. Modifications were also required to WRF/Chem and auxiliary files (such as the WRF namelist and registry files) to accommodate the new PinG option. These modifications are discussed in more detail below.

Because of the significantly different architectures of CMAQ and WRF/Chem, the plume model interfaces were completely rewritten to be compatible with WRF/Chem. WRF employs a layered software architecture, with a driver layer, mediation layer, model layer, and external libraries for time-keeping and input/output (I/O) (Michalakes et al., 2005). The science modules (e.g., physics, chemistry, aerosols, SCICHEM) that perform the actual model computations reside in the low-level “model layer” of WRF and are invoked by subroutine calls in the mediation layer. Only primitive or intrinsic Fortran data types and simple arrays are passed through the argument list for the model layer subroutine calls. However, the grid properties (e.g., meteorological state variables, pollutant concentrations, etc.) in WRF and WRF/Chem are not intrinsic data types but are actually components of a derived data type, “domain”. Thus, before these state variables can be passed to the model layer

subroutines, they need to be extracted or “de-referenced” from the derived data type corresponding to the model grid. This dereferencing is performed in the mediation layer.

Following this WRF convention, the main PinG interface between WRF/Chem and SCICHEM resides in the mediation layer. All WRF grid description and meteorology state variables are de-referenced in this interface and passed as simple arrays to the SCICHEM driver. This approach is different from that used in CMAQ–APT, in which these variables were read from Input/Output Applications Programming Interface (I/O API) data files. This difference arises primarily because meteorology and chemistry in CMAQ–APT are uncoupled, unlike WRF/Chem, which is a fully coupled model. Similarly, the WRF/Chem grid chemistry variables are de-referenced in the PinG interface and passed as a simple array to the SCICHEM driver to provide the 3-D background concentrations as both inputs to the plume model, and as outputs that are updated whenever mature puffs are dumped to the grid. Other WRF/Chem variables that are de-referenced in the interface are the photolysis rates and dry deposition velocities of chemical species. Note that the latter are not state variables in the base version of WRF/Chem and were added to the WRF Registry (see below) so that the code to declare, allocate and initialize these variables could be generated automatically at model build time. In addition, new code was written to extract and pass these variables from the dry deposition velocity module to the SCICHEM model layer via the interface.

In addition to the main PinG interface, a number of auxiliary interface and driver routines in the SCICHEM code were updated to be compatible with the WRF/Chem architecture and science codes,

such as the routines that set up the SCICHEM model grid, the SCICHEM arrays, the chemical species lists, the emissions routines, and the chemistry module drivers.

Other changes to the WRF/Chem framework include changes to the Registry and namelist files. The Registry is a text file that specifies model state arrays, I/O, coupling, inter-process communication, physics and chemistry packages, and model configuration options (NCAR, 2011). This file is used at model build time to auto-generate several thousand lines of code, e.g., declaration, allocation, and initialization of state data, generation of dummy argument lists and their declarations, and generation of actual argument lists used when passing state data between subroutines at the interface between layers in the WRF software architecture. When new science code requires access to WRF variables that are not currently saved as state variables, the Registry needs to be modified by the code developer. Similarly, changes need to be made to the Registry to specify new model configuration options, such as the PinG option implemented in this study. The namelist file is used at model run time to select simulation options, such as the gas-phase chemistry module, or the PinG option introduced in this study. Corresponding changes to the WRF/Chem code are required to read these selections from the namelist file and use the selected options for the simulation.

In addition to the model code development, codes were developed for the pre-processing and post-processing required for PinG applications. The emissions pre-processor creates the point source emissions file for SCICHEM by reading stack parameters and stack emissions for the point sources selected for PinG from data downloaded from the Clean Air Markets Division (CAMD) web site.

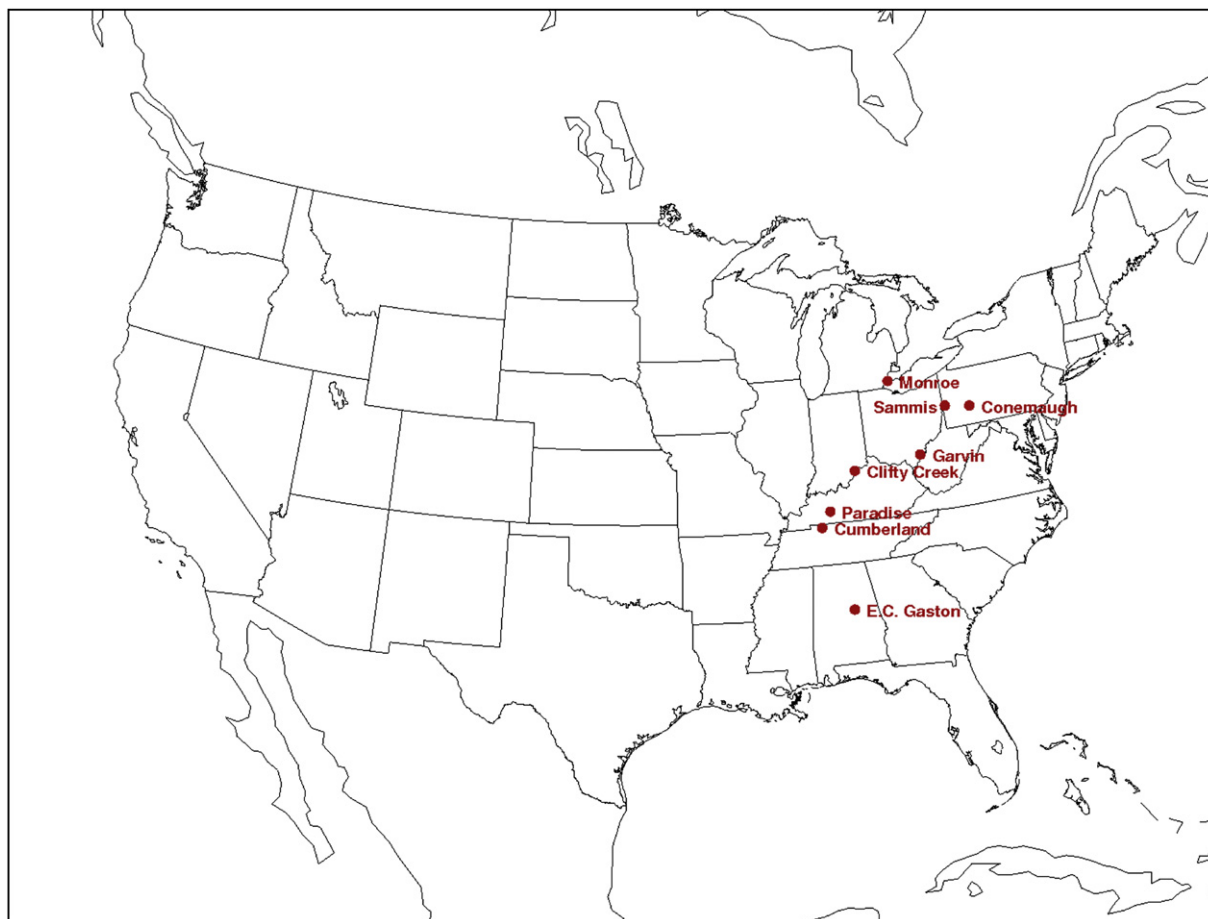


Fig. 1. CONUS modeling domain for the WRF/Chem simulations with and without PinG, and point sources selected for PinG treatment.

These emissions are then subtracted from the WRF/Chem gridded emissions file. The post-processor reads the outputs of the PinG simulation, i.e., the gridded WRF/Chem netCDF chemistry variables, and the corresponding variables in the SCICHEM puff file for active puffs (i.e., puffs not dumped to the host grid), to create a new gridded netCDF file that contains the combined (merged) concentrations.

5. Model application and results

WRF/Chem with the new PinG treatment was applied for a three-day period in July 2001 (July 8–10) during which high ozone concentrations were observed. The model was applied for the CONUS domain with a 36 km horizontal grid resolution and 34 logarithmic structure layers from the surface to a fixed pressure of ~100 mb (or ~16 km above ground level). The meteorological initial conditions (ICONS) and boundary conditions (BCONS) were generated using the National Centers for Environmental Prediction (NCEP) Final Analysis (FNL) reanalysis data. The EPA's National Emissions Inventories (NEI) 2001 (also referred to as NEI 1999 version 3) was used to generate a gridded anthropogenic emission inventory for gaseous and PM species. Chemical ICONs and BCONS for both gas and aerosol species were generated using the simulation results from a global chemistry model (GEOS-Chem).

Fig. 1 shows the modeling domain as well as the locations of the power plants in the eastern United States that were explicitly simulated with the plume model. In this initial PinG application of WRF/Chem, we selected 8 sources for PinG treatment, although the model currently allows for up to 200 PinG sources. Karamchandani et al. (2010a) have conducted a PinG application (using CMAQ as the host model) in which over 150 power plants were treated as PinG sources. The 8 power plants shown in Fig. 1 were selected on the basis of their SO₂ and NO_x emissions as well as their geographical distributions in the domain. Stack parameters and SO₂ and NO_x emissions for these sources were obtained from the CAMD database for the simulation days. For the PinG application, the emissions from these sources were treated in the embedded plume model using a separate point source emissions file. To avoid double-counting these sources, their emissions in the grid cells containing them were removed from the base WRF/Chem gridded emissions files using the emissions pre-processor described in the previous section.

In addition to the PinG application, a base simulation was performed with the standard WRF/Chem–MADE/SORGAM configuration without the PinG option. The purpose of this simulation was to provide a basis for comparison with the PinG application from both a model performance perspective as well as the spatial distributions of ozone and PM_{2.5} sulfate and nitrate concentrations.

Initial conditions for the three-day period from July 8 to 10, 2001 were obtained by conducting an initialization (“spin-up”) simulation with the base WRF/Chem configuration for seven days (July 1 to 7, 2001). The concentrations at the end of July 7 were then used as initial conditions for both the base and PinG simulations of the three-day period.

5.1. Model performance evaluation

An operational model performance evaluation was performed in which WRF/Chem estimates of ozone and fine particulate matter (PM_{2.5}) sulfate and nitrate concentrations, as well as total PM_{2.5} concentrations, for the July 8–10, 2001 period were compared with observed values. The comparisons were performed for both the base and PinG configurations of WRF/Chem. Ozone observations for the evaluation were obtained from the Air Quality System (AQS) monitoring network. Sulfate, nitrate, and total PM_{2.5} observations

Table 1

Model performance statistics for 1-h and 8-h average ozone concentrations with a 40 ppb cut-off on observed ozone concentrations.

Metric	1-h average O ₃		8-h average O ₃	
	Base	PinG	Base	PinG
Mean Observed Value (ppb)	55.4	55.4	59.9	59.9
Mean Predicted Value (ppb)	49.2	52.2	53.8	57.3
Ratio of Means	0.89	0.94	0.90	0.96
Gross Bias (ppb)	−6.2	−3.2	−6.15	−2.6
Normalized Bias (%)	−9.2	−4.1	−8.8	−3.0
Fractional Bias (%)	−13.8	−8.2	−12.0	−5.7
Gross Error (ppb)	12.9	12.0	12.0	11.1
Normalized Error (%)	22.9	21.6	19.7	18.5
Fractional Error (%)	25.6	22.6	21.7	18.8
Normalized RMSE (%)	26.7	25.6	21.5	20.3
Normalized Mean Bias (%)	−11.1	−5.8	−10.3	−4.3
Normalized Mean Error (%)	23.2	21.6	20.1	18.5
Correlation Coefficient (<i>r</i>)	0.32	0.37	0.23	0.28
Coefficient of Determination (<i>r</i> ²)	0.10	0.13	0.05	0.08

from the Interagency Monitoring of Protected Visual Environments (IMPROVE) and Speciation Trends Network (STN) monitoring networks (representing rural and urban locations, respectively) were used to evaluate model performance for fine particulate matter.

Table 1 shows the model performance statistics for 1-h and 8-h average ozone concentrations for both the base and PinG

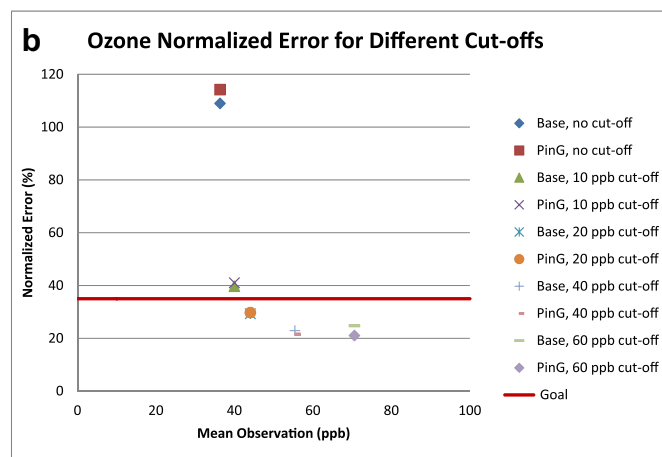
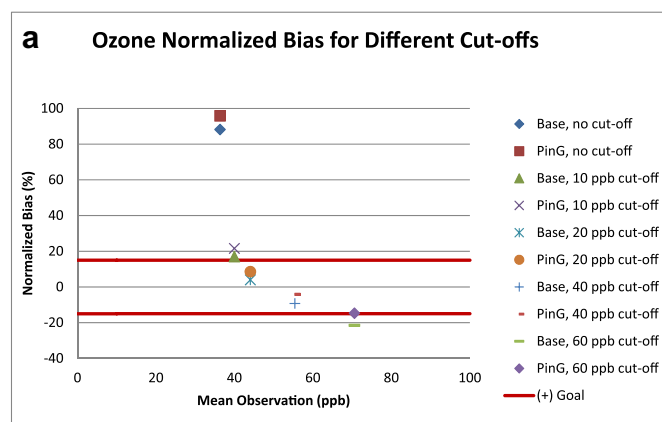


Fig. 2. (a) Normalized bias and (b) normalized error as a function of mean 1-h observed ozone concentrations using different cut-offs for observed ozone. The solid lines show the goals for model performance.

configurations of the model. For the comparison of 8-h average ozone concentrations, the maximum observed 8-h average concentration at each monitoring location is compared with the maximum 8-h average model prediction in the grid cell containing the monitor. 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). EPA modeling guidance also recommends using a cut-off value of 60 ppb for the observed hourly ozone concentrations to remove the influence of very low observed concentrations on the performance statistics. However, this cut-off results in a large number of data points being discarded in the 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). Table 1 shows the model performance statistics using a 40 ppb cut-off on observed ozone concentrations to keep the number of data points discarded reasonably low. As shown in Table 1, the overall model performance statistics for 1-h and 8-h ozone concentrations are comparable for both model configurations, but the PinG simulation shows a 5% lower bias and 1% lower error than the base simulation.

We also calculated model performance statistics for one-hour ozone concentrations using no cut-off, as well as cut-offs of 10 ppb, 20 ppb, 40 ppb and 60 ppb. The results are shown graphically in the normalized bias and error plots shown in Fig. 2a and b, which are similar to the bugle plots of Boylan and Russell (2006) that are often used to assess model performance for PM_{2.5} and visibility. The solid lines in Fig. 2a and b show the goals and criteria for ozone model performance ($\pm 15\%$ goals for normalized bias and 35% criteria for normalized error). We see from Fig. 2a that both the base and PinG configurations of WRF/Chem overestimate the low ozone concentrations (cut-offs less than 20 ppb) and underestimate the high ozone concentrations (cut-offs greater than 20 ppb). For the lower cut-offs, the performance for the PinG configuration is slightly worse than the base configuration performance. However, the PinG configuration shows a lower underestimation bias than the base configuration at the high ozone concentrations. Similarly, Fig. 2b shows that the PinG configuration results in lower normalized errors than the base configuration for the high ozone concentrations.

Table 2
Model performance statistics for 24-h average sulfate, nitrate, and total PM_{2.5} concentrations at IMPROVE sites.

Metric	Sulfate		Nitrate		Total PM _{2.5}	
	Base	PinG	Base	PinG	Base	PinG
Mean Observed Value ($\mu\text{g m}^{-3}$)	2.56	2.56	0.25	0.25	9.69	9.69
Mean Predicted Value ($\mu\text{g m}^{-3}$)	0.90	0.93	0.10	0.11	4.69	4.23
Ratio of Means	0.35	0.36	0.41	0.45	0.48	0.44
Gross Bias ($\mu\text{g m}^{-3}$)	-1.67	-1.64	-0.15	-0.14	-5.00	-5.47
Normalized Bias (%)	-44.1	-42.3	-13.5	-29.5	-47.9	-49.5
Fractional Bias (%)	-67.9	-66.0	-139.1	-146.0	-66.2	-72.8
Gross Error ($\mu\text{g m}^{-3}$)	1.70	1.68	0.24	0.25	5.00	5.47
Normalized Error (%)	51.8	50.9	143.6	132.6	47.9	49.5
Fractional Error (%)	74.0	72.5	162.4	166.3	66.2	72.3
Normalized RMSE (%)	51.8	50.9	143.6	132.6	47.9	49.5
Normalized Mean Bias (%)	-65.1	-64.0	-59.1	-55.0	-51.6	-56.4
Normalized Mean Error (%)	66.3	65.5	96.6	98.1	51.6	56.4
Correlation Coefficient (<i>r</i>)	0.77	0.74	0.22	0.31	0.83	0.66
Coefficient of Determination (<i>r</i> ²)	0.60	0.55	0.05	0.10	0.68	0.44

Table 3
Model performance statistics for 24-h average sulfate, nitrate, and total PM_{2.5} concentrations at STN sites.

Metric	Sulfate		Nitrate		Total PM _{2.5}	
	Base	PinG	Base	PinG	Base	PinG
Mean Observed Value ($\mu\text{g m}^{-3}$)	4.48	4.48	0.75	0.75	14.51	14.51
Mean Predicted Value ($\mu\text{g m}^{-3}$)	1.34	1.41	0.25	0.51	7.25	7.40
Ratio of Means	0.30	0.32	0.33	0.67	0.50	0.51
Gross Bias ($\mu\text{g m}^{-3}$)	-3.14	-3.07	-0.50	-0.24	-7.27	-7.11
Normalized Bias (%)	-63.9	-60.5	-59.7	-45.1	-44.0	-43.9
Fractional Bias (%)	-97.5	-94.8	-121.6	-139.4	-64.7	-68.3
Gross Error ($\mu\text{g m}^{-3}$)	3.15	3.16	0.62	0.90	8.02	8.65
Normalized Error (%)	64.3	64.8	78.8	115.1	50.8	55.1
Fractional Error (%)	97.9	98.1	134.8	153.4	69.0	75.2
Normalized RMSE (%)	63.8	63.6	84.0	122.6	51.6	56.4
Normalized Mean Bias (%)	-70.1	-68.5	-66.9	-32.6	-50.1	-49.0
Normalized Mean Error (%)	70.3	70.5	82.5	120.3	55.3	59.6
Correlation Coefficient (<i>r</i>)	0.82	0.63	0.04	0.15	0.41	0.35
Coefficient of Determination (<i>r</i> ²)	0.68	0.40	<0.01	0.02	0.17	0.12

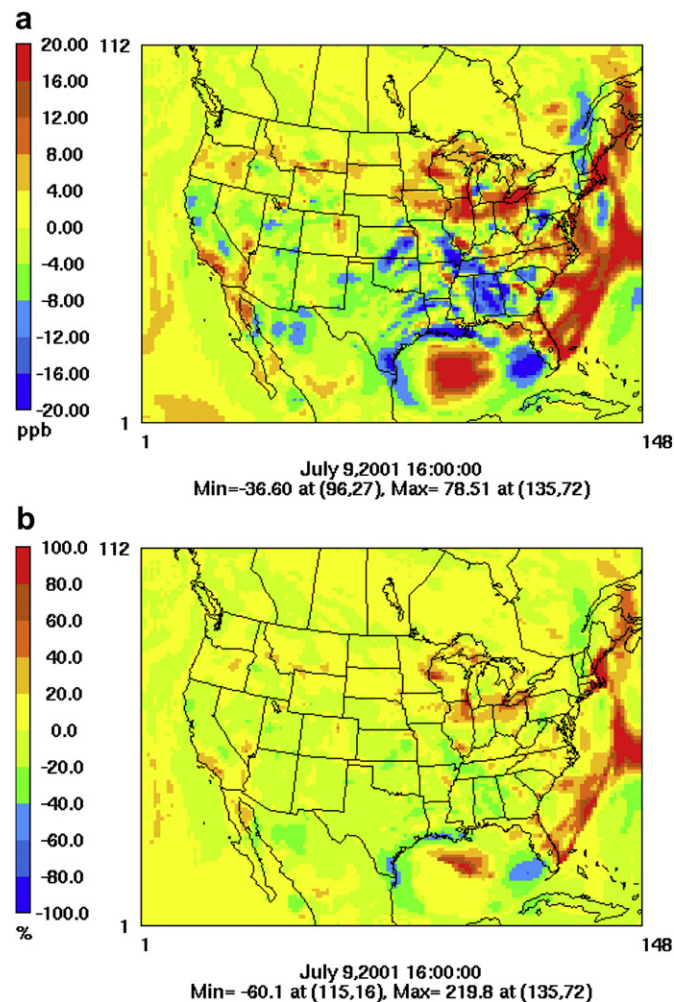


Fig. 3. (a) Absolute and (b) relative differences between 8-h ozone concentrations (ppb) on July 9, 2001 predicted by the PinG and base configurations of WRF/Chem.

Both model configurations show large under-predictions of 24-h average sulfate, nitrate, and total PM_{2.5} concentrations, as shown in Table 2 for the IMPROVE monitoring network and Table 3 for the STN network. The mean sulfate and nitrate concentrations are underpredicted by factors of 2–3, while the mean PM_{2.5} concentrations are underpredicted by about a factor of 2. About 40% of the predicted PM_{2.5} concentrations are within a factor of 2 of the observed values, and about 80% are within a factor of 3.

Some of the large under-predictions in sulfate and PM_{2.5} concentrations can be attributed to the fact that the WRF/Chem simulations were conducted without aqueous-phase chemistry for this initial application (see Section 4). Using the same emissions and chemical inputs, Zhu and Zhang (2011) conducted a WRF/Chem simulation with the CB05 gas-phase mechanism, the MADE-SORGAM aerosol module, and the CMU aqueous-phase chemistry for July 1–31, 2001 and reported NMBs of sulfate and PM_{2.5} of –3.4% and –23.1% at the STN sites and 20.4% and –27.1% at the IMPROVE sites, respectively.

Other reasons for the PM_{2.5} under-predictions include some limitations of the MADE-SORGAM aerosol module in simulating PM_{2.5}. For example, the MADE-SORGAM aerosol module gives very low secondary organic aerosol (SOA) concentrations since it neglects several SOA precursors such as isoprene and sesquiterpenes (Zhu and Zhang, 2011). Zhang et al. (2012) conducted WRF/Chem

simulations with the same CB05 and aqueous-phase chemistry but with a more advanced aerosol module (MADRID) using the same emissions and chemical inputs as those used in Zhu and Zhang (2011) and this work. They reported NMBs of sulfate and PM_{2.5} of –20.2% and –2.2% at STN sites and –5.9% and 5.5% at IMPROVE sites, respectively. Furthermore, uncertainties in emissions could also contribute to the under-predictions of PM_{2.5} concentrations, as pointed out by Zhang et al. (2012), who cited underestimations of emissions of primary PM_{2.5} at urban sites as possible reasons for underestimating PM_{2.5} concentrations.

Although the overall performances of both the base and PinG configurations of WRF/Chem are similar, the PinG configuration shows 2% lower bias at the IMPROVE sites and 3% lower bias at the STN sites for sulfate concentrations. For nitrate concentrations, the results are mixed, with the PinG configuration showing a higher bias (about 16%) at the IMPROVE sites and lower bias (about 15%) at the STN sites, as compared to the base configuration. Note that there are only 14 data points available for the IMPROVE sites and over 1000 data points for the STN sites.

The similarity in model performance with and without PinG treatment has been noted in previous PinG modeling studies with CMAQ (e.g., Karamchandani et al., 2002, 2006, 2010a) and is not unexpected given that monitoring stations may only sometimes be impacted by plumes from upwind point sources. However, the previous PinG modeling studies have also shown that predicted

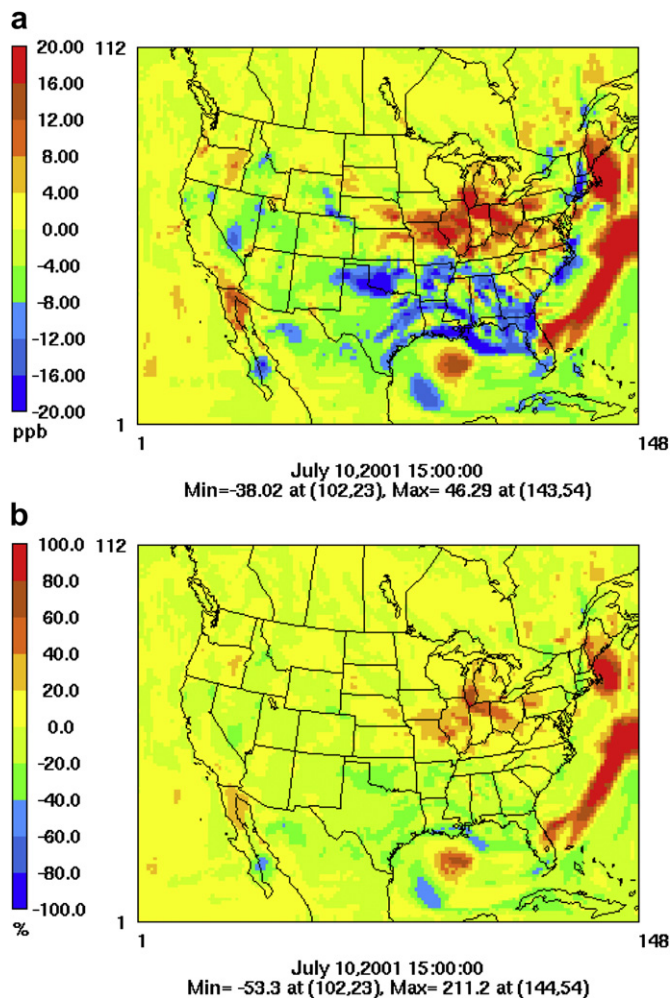


Fig. 4. (a) Absolute and (b) relative differences between 8-h ozone concentrations on July 10, 2001 predicted by the PinG and base configurations of WRF/Chem.

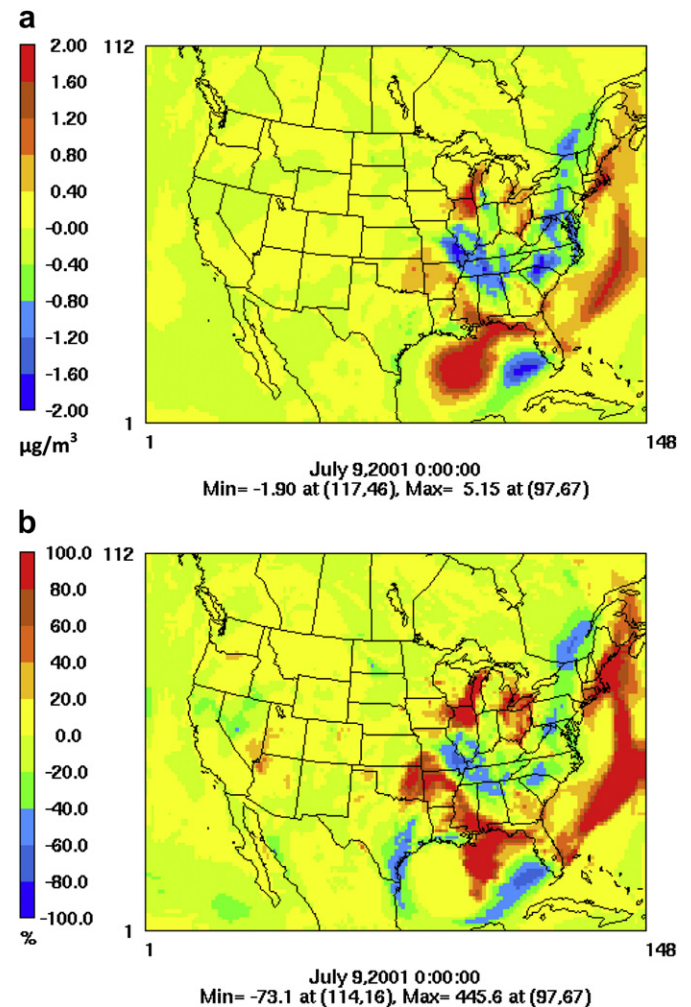


Fig. 5. (a) Absolute and (b) relative differences between 24-h sulfate concentrations on July 9, 2001 predicted by the PinG and base configurations of WRF/Chem.

spatial patterns of pollutant concentrations using the PinG approach can be significantly different from those predicted using the traditional gridded approach. In the following section, we look at these spatial differences.

5.2. PinG impacts on spatial distributions of pollutant concentrations

Figs. 3a and b show the absolute and relative differences, respectively, between the predicted 8-h average (from 1600 UTC to 2400 UTC) surface ozone concentrations from the PinG and base configurations for July 9, 2001. The results show both large increases and decreases in ozone concentrations in the eastern U.S. as a result of using the PinG approach. As discussed in Karamchandani et al. (2002), these differences can be attributed to differences in the plume transport, mixing, and chemistry in the two model configurations. In the base simulation without PinG, the plume NO_x is rapidly mixed artificially horizontally and vertically. This artificial mixing in the base simulation results in not only excessive titration of surface ozone near and upwind of the source, but also quicker downwind photochemical production of ozone depending on the VOC/NO_x ratios downwind of the source. In contrast, in the PinG simulation, the NO_x in the plume remains aloft for larger distances, resulting in less titration of surface ozone

near the source, and also higher photochemical production of ozone further downwind as the NO_x in the plume is transported over a larger distance than in the base simulation. In addition, the point source plumes in the base and PinG simulations may be transported in different directions, depending on the near-source vertical distribution of plume material in the two simulations and the vertical variation of wind speed and direction. Thus, in general, it is found that photochemical ozone production near the sources is suppressed in the PinG simulation, but the production increases as the plumes travel further downwind. However, in some regions, such as the Ohio River Valley region in the upper Midwest, the PinG simulation predicts about 20–40% higher 8-h average ozone concentrations than the base simulation near the sources (see Fig. 2b). This pattern has also been noted in a previous PinG modeling study for ozone with CMAQ (Karamchandani et al., 2002), and is likely due to the excessive titration of surface ozone in the base simulation as compared to the PinG simulation. The region has a high density of power plants and is thus not NO_x-limited and local NO_x emissions quench the background ozone (e.g., Godowitch et al., 2008), particularly in the base simulation, where the point source NO_x emissions are well-mixed to the surface instantaneously. These findings are consistent with the theoretical concepts of Kramm and Meixner (2000) and the observational findings of Dlugi et al. (2010). We also note higher ozone concentrations in the PinG simulation than in the base simulation west of this region,

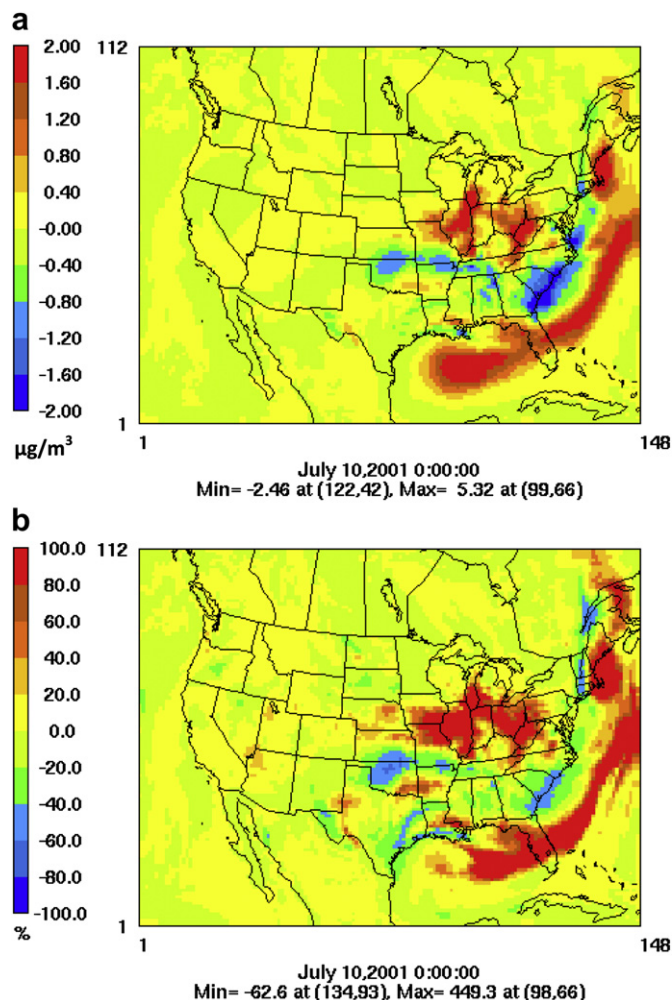


Fig. 6. (a) Absolute and (b) relative differences between 24-h sulfate concentrations on July 10, 2001 predicted by the PinG and base configurations of WRF/Chem.

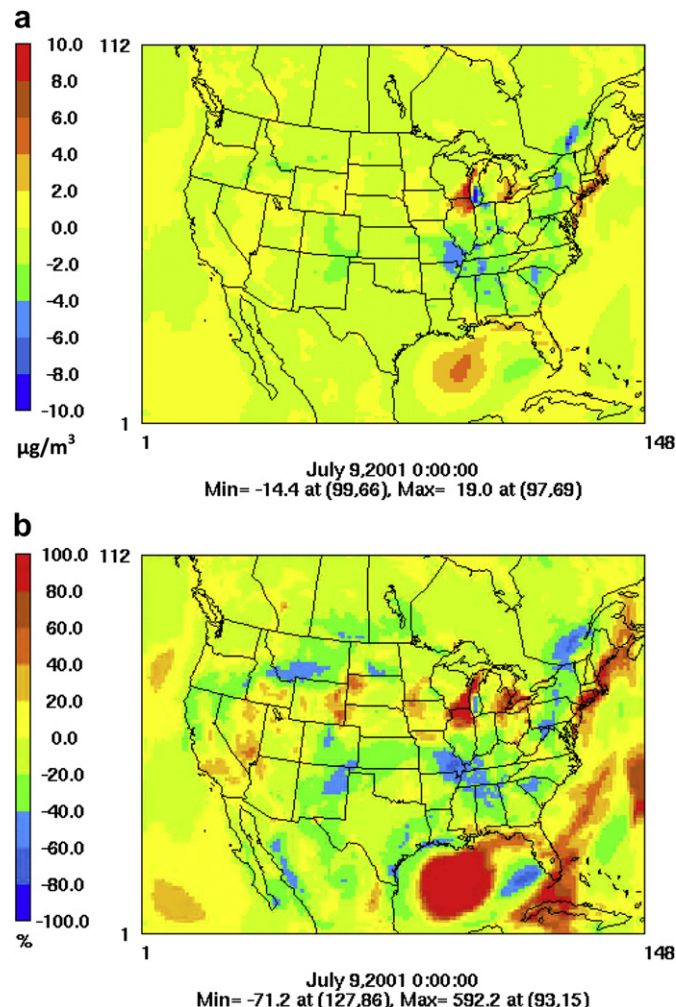


Fig. 7. (a) Absolute and (b) relative differences between 24-h total PM_{2.5} concentrations on July 9, 2001 predicted by the PinG and base configurations of WRF/Chem.

suggesting that significantly more plume material is being transported to the west in the PinG simulation.

The absolute and relative differences for 8-h average ozone concentrations on July 10, shown in Fig. 4, are qualitatively similar to the results for July 9. For both days, we also note some differences in the predicted ozone concentrations from the base and PinG simulations in the western portion of the modeling domain, which is somewhat surprising, given that the PinG sources are in the eastern portion of the domain. It is possible that the feedback between the meteorology and chemistry in the coupled WRF/Chem system may be affected by perturbations to the system resulting from the PinG approach. Additional sensitivity studies, including simulations in the uncoupled mode, may provide further insight.

The absolute and relative differences in the spatial distribution of predicted 24-h average sulfate concentrations are shown in Figs. 5 and 6, for July 9 and 10, respectively. As in the case of 8-h average surface ozone concentrations, regions of both decreases and increases in the sulfate concentrations are found. In particular, on July 9, we note generally large decreases in sulfate concentrations near most of the PinG sources, and large increases further downwind, due to the plume material being transported to longer downwind distances in the PinG simulation than in the base simulation, and also due to slower initial oxidation of SO₂ to sulfate in the concentrated NO_x plume, consistent with previous PinG results with CMAQ (Karamchandani et al., 2006). However, as in the case of the ozone results, discussed previously, the exception to this

pattern is the upper Midwest region, where we see higher sulfate concentrations to the west of the point sources in the PinG simulation than in the base simulation, indicating more transport of plume material to the west in the PinG simulation. In contrast to the ozone results, we see that the sulfate differences between the two simulations are primarily confined to the eastern U.S., with some small differences (less than 0.4 μg m⁻³) in the western part of the domain on both days.

Fig. 7 shows the absolute and relative differences in the spatial distribution of predicted 24-h average total PM_{2.5} concentrations on July 9. The patterns of PM_{2.5} decreases and increases are qualitatively similar to those for sulfate. We see lower PM_{2.5} concentrations in the PinG simulation near the sources in the southern part of the domain with higher concentrations further downwind. In the upper Midwest, we see higher PM_{2.5} concentrations in the PinG simulation both near and to the west of the point sources. Fig. 8 shows a similar pattern in the total PM_{2.5} results for July 10.

6. Summary and conclusions

We have incorporated a plume-in-grid capability into a fully-coupled meteorology and chemistry model, WRF/Chem–MADE/SORGAM. Combined with the global-through-urban simulation capability incorporated by Zhang et al. (2009a,b), the PinG enhancement to WRF/Chem will enable the simulation of air pollutants on a wide range of scales to understand the interactions between global climate and local air quality. Our initial application with the model shows that there are substantial differences between the spatial patterns of species concentrations predicted the base and PinG configurations of WRF/Chem, but the model performances for both configurations using routine air quality observations are essentially the same. The results indicate that additional studies will be useful in understanding the effects of the PinG treatment for a range of conditions. These studies include sensitivity studies in which the model is applied in the uncoupled mode (i.e., no feedback between meteorology and air quality), as well as model applications for other modeling periods and a larger number of point sources treated with PinG.

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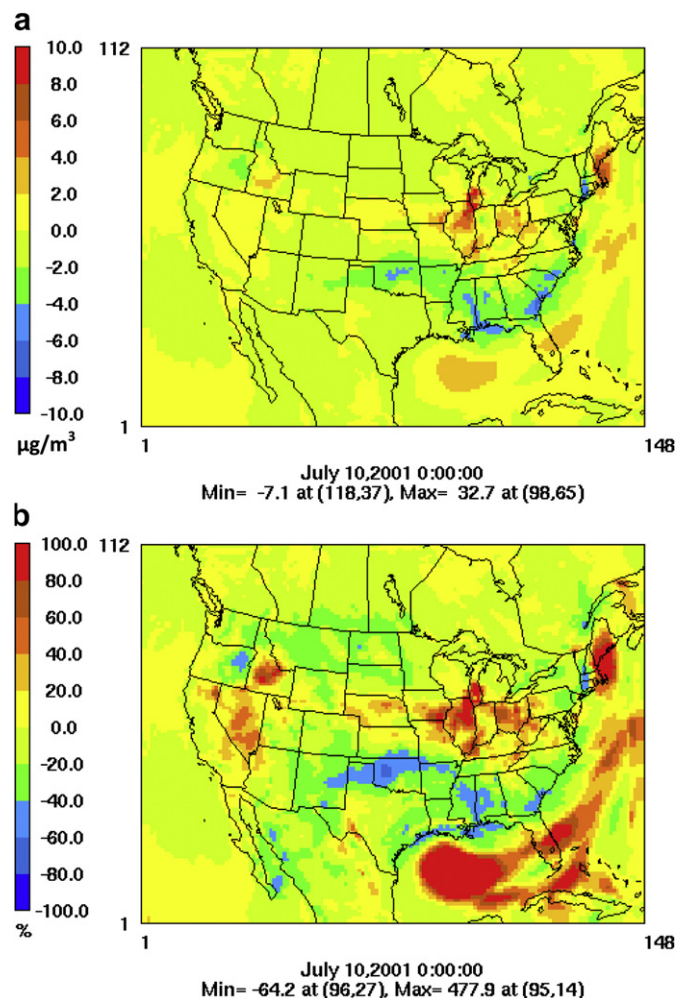


Fig. 8. (a) Absolute and (b) relative differences between 24-h total PM_{2.5} concentrations on July 10, 2001 predicted by the PinG and base configurations of WRF/Chem.

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