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Plume-in-grid modeling for particulate matter

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Abstract

Three-dimensional grid models are now being commonly used to simulate particulate matter (PM) concentrations, especially fine PM ($PM_{2.5}$), which includes a significant fraction of secondary species formed in the atmosphere. These models usually do not address the subgrid-scale effects associated with emissions from large elevated point sources. This can lead to errors in the calculation of $PM_{2.5}$ concentrations downwind of these sources because of unrealistic representations of the dispersion and transformation processes that govern $PM_{2.5}$ formation. Here, we describe the development, application and evaluation of a plume-in-grid (PiG) model for PM. The model, based on an existing PiG model for ozone, is extended to include in the plume component state-of-the-science treatments of aerosol chemistry and dynamics as well as aqueous chemistry that are consistent with the treatments used in the host grid model. Application of this model to several SO_2 and NO_x emitting power plants in the southeastern United States shows that the PiG treatment leads to significant differences in sulfate and total inorganic nitrate concentrations. Comparisons of model results, with and without PiG treatment, against measurements characterizing specific plume events, show that the PiG treatment captures the plume events more often and generally better than the standard grid-based approach.

Keywords: $PM_{2.5}$; Subgrid-scale; Plume chemistry; CMAQ; SCICHEM; SEARCH

1. Introduction

Three-dimensional (3-D) grid models are frequently used to predict the impacts of emission controls on concentrations of pollutants such as ozone and fine particulate matter ($PM_{2.5}$). However, such models are limited in their ability to correctly represent the near-source dispersion, transport, and chemistry of emissions from elevated sources. These limitations are due to the inability of grid models, with typical horizontal resolutions of a few kilometers to tens of kilometers, to resolve stack plumes with initial dimensions of tens of meters. A common approach to address these limitations is to use Plume-in-Grid (PiG) modeling, in which a subgrid-scale representation of stack plumes is embedded in the 3-D model. This approach has traditionally been limited to ozone modeling studies (Seigneur et al., 1983; Kumar and Russell, 1996; Karamchandani et al., 2002; Vijayaraghavan et al., 2006). A detailed discussion of the errors associated with ignoring the subgrid-scale features of point source plumes on the

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formation of gaseous pollutants has been provided by Karamchandani et al. (2002). The errors associated with the gridded representation of point source plumes are just as important for $PM_{2.5}$ as they are for ozone, since the chemistry leading to formation of $PM_{2.5}$ constituents, such as sulfate and nitrate, is significantly slower in the plume of a large NO_x point source than it is in the background air (Karamchandani et al., 1998; Karamchandani and Seigneur, 1999). This is particularly true in the initial stages of plume dispersion, when the plume dimensions are small relative to the grid resolution. Thus, using a purely grid-based approach can potentially lead to an incorrect estimation of the impacts of large SO_x and NO_x point sources on ambient $PM_{2.5}$ concentrations.

Karamchandani et al. (2002) previously developed a PiG model for ozone based on the US EPA Community Multiscale Air Quality model (CMAQ). This model is referred to as CMAQ with Advanced Plume Treatment (CMAQ-APT), and includes a state-of-the-science reactive puff model, SCICHEM, to resolve stack plumes within CMAQ. Here, we present the improvement of SCICHEM and CMAQ-APT to include a state-of-the-science treatment for PM. This treatment, referred to as the Model of Aerosol Dynamics, Reaction, Ionization, and Dissolution (MADRID), was incorporated in CMAQ by Zhang et al. (2004), and is extended in this work to SCICHEM and CMAQ-APT. The resulting PiG model, CMAQ-MADRID-APT, is applied to the southeastern United States for 2 months in 2002 with stacks from 14 power plants selected for PiG treatment. For comparison purposes, the grid model without PiG treatment, CMAQ-MADRID, is applied for the same domain and time periods.

2. The PiG model

CMAQ-MADRID-APT consists of a reactive plume model, SCICHEM (Karamchandani et al., 2000), embedded into the "host" 3-D grid-based model, CMAQ-MADRID (Zhang et al., 2004). Brief descriptions of CMAQ-MADRID and SCICHEM are provided in the following sub-sections.

SCICHEM was embedded into the host grid model following the established protocols for incorporating new science modules into CMAQ (www.cmascenter.org/help/model_docs/cmaq/madrid/CMAS-CodingGuidelines.pdf). The general approach for the coupling of the plume model with the host model is described in detail by Karamchandani et al. (2002).

2.1. The host grid model, CMAQ-MADRID

The host model is based on the October 2004 release (Version 4.4) of the US EPA's CMAQ model and includes a condensed version of the MADRID $PM_{2.5}$ treatment of Zhang et al. (2004). CMAQ was developed by EPA to address multiscale multi-pollutant air pollution problems (Byun and Schere, 2006). CMAQ treats the emissions, transport, dispersion, chemical transformations, gas-particle conversion and removal processes that govern the behavior of chemical pollutants in the atmosphere. Emissions include those from area sources (e.g., industrial, residential, agricultural, mobile and biogenic emissions) and point sources (e.g., power plants, smelters, and refineries). The plume rise of point source emissions is treated in a pre-processor to CMAQ. Transport processes include advection, large-scale convection and, in the presence of cumulus clouds, subgrid-scale convection. Dispersion includes both horizontal and vertical dispersion. Chemical transformations modeled include reactions in the gas phase and reactions in the aqueous phase (i.e., in cloud droplets). The formation of secondary aerosols and the gas-particle partitioning of semi-volatile chemical species are simulated. Dry deposition is simulated for gases and particles. Wet deposition is simulated for precipitating clouds that are resolved by the grid system as well as for clouds that are treated at the subgrid-scale.

In CMAQ-MADRID, the EPA treatment of aerosols is replaced by a condensed version of the MADRID treatment of Zhang et al. (2004). The primary differences between the EPA and MADRID treatments are in their representation of the particle size distribution and the formation of secondary organic aerosols (SOA). The former uses a modal representation of the size distribution, while the latter uses a sectional representation. Here, MADRID was applied with two size sections. The condensed version of MADRID used here includes 8 explicit SOA precursors (Pun et al., 2004) instead of the 14 precursors used in the original MADRID formulation. This condensed version was found to produce slightly higher SOA concentrations than the original formulation, but the differences between the two versions were negligible compared to current uncertainties in our knowledge