EVALUATION OF THE TRAJECTORY-GRID AND THE BOTT SCHEMES FOR SOLVING AEROSOL CONDENSATION AND EVAPORATION EQUATIONS

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1. INTRODUCTION

Atmospheric aerosols have adverse effects on human health and visibility and play an important role in climate changes. Understanding of their chemical composition, ambient concentrations, as well as dynamic processes is essential for the development of effective control strategies to reduce their adverse impacts. Numerical models provide a fundamental tool to investigate the formation and fate of atmospheric aerosols. Accurately simulating atmospheric aerosols requires model treatments of all major processes including emission, nucleation, coagulation, condensation, gas/particle mass transfer, transport, deposition, and cloud-processing of aerosols.

Large uncertainties exist in aerosol treatments in air quality models (AQMs). The use of different approaches to represent the particle size distribution is one of the factors that contribute to the discrepancies of the model predictions. Zhang et al. (1999, 2004) summarized several approaches used in 3-D AQMs to simulate the condensation/evaporation processes. The modal approach is used in several models such as the Regional Particulate Matter Model (Binkowski and Shankar, 1995) and the Models-3/Community Multiscale Air Quality (CMAQ) modeling system (Binkowski and Roselle, 2003). The sectional approach is used in a number of models including the Urban Airshed Model with Aerosols (UAM-AERO)(Lurmann et al., 1997), the San Joaquin Valley Air Quality Study (SJVAQS) and the Atmospheric Utility Signatures Predictions and Experiments Study (AUSPEX) Regional Modeling Adaptation Project (SARMAP) Air Quality Model with Aerosols (SAQM-AERO)(Dabdub et al., 1998), the California /Carnegie-Mellon Institute of Technology (CIT) (Meng et al., 1998), the Urban Airshed Model with Aerosol Inorganics Model (UAM-AIM) (Sun and Wexler, 1998), and the Gas, Aerosol, Transport, Radiation, general circulation, mesoscale, and ocean model (GATOR-GCMOM) and its predecessors (Jacobson, 1997, 2001, 2002, 2004; Jacobson et al., 2004). One problem that is associated with the use of the traditional sectional approach is its numerical diffusion.

Using the time splitting method, the condensation/evaporation equation can be divided into two parts: $\frac{dp_i}{dt} = H_i p$ and $\frac{\partial p_i}{\partial t} = -\frac{1}{3}\frac{\partial H p_i}{\partial u}$. The second part is an advection-like form. Solving an advection equation accurately and efficiently is a challenge in atmospheric modeling because of the numerical diffusion problems arisen in the solutions. Various methods have been developed in the past to minimize the numerical diffusion associated with these solutions. Among them, the Bott scheme (Bott, 1989) and the Trajectory-Grid (T-G) scheme (Chock et al., 2000) are the two advection schemes that can be adapted to solve the condensation/evaporation equations for multicomponent aerosols.

In this study, the performance of the T-G scheme as a condensation scheme is first evaluated against those of several other condensation approaches used in three-dimensional AQMs (e.g., CIT, GATOR, Models-3/CMAQ, and UAM-AERO) in a simple system that only considers the condensation of sulfuric acid. The T-G scheme is then implemented in a box model version of CMAQ-Model of Aerosol Dynamics, Reaction, Ionization and Dissolution (MADRID) that combines the Carbon-Bond Mechanism Version IV gas phase mechanism with the MADRID aerosol module. The T-G and Bott schemes are being further evaluated in a system of multicomponent aerosols in which both condensation and evaporation occur. The strengths and limitations of each scheme in solving condensation/evaporation equations for multicomponent aerosols are being identified and analyzed.

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2. DESCRIPTION OF THE TRAJECTORY-GRID (T-G) APPROACH FOR CONDENSATIONAL GROWTH

Almost all the variables simulated in AQMs are temporally and spatially continuous. In the AQMs that use the sectional approach, however, we have to discretize the particle size distribution in space and time to approximate the continuous distribution. This discretization creates inevitable numerical problems. The trajectory-grid (T-G) approach contains a Lagrangian advection scheme coupled with a fully Eulerian diffusion scheme to reduce the numerical problems associated with the condensation equation in air quality modeling. The T-G scheme assigns the spatial locations of points on a given concentration profile to a set of concentration pulses; then tracks the pulse positions as they move downwind undergoing diffusion and reactions (Chock et al., 1996).

Similar to the semi-Lagrangian methods, the T-G scheme requires interpolation to obtain the fixed grid concentration distribution as a model-predicted distribution. Errors will be introduced during the interpolation process into the fixed grid concentration. In semi-Lagrangian methods the error may be propagated since the fixed grid concentration obtained from interpolation is used to update the concentration distribution profile during the next time step. The T-G scheme is a fully Lagrangian scheme and has advantage over the semi-Lagrangian methods in the following case: If pulses are also used for all the other aerosol processes in the host AQMs (e.g., nucleation, emission, and deposition), the error of interpolation of the T-G scheme will not be propagated through the feedback mechanism since the concentration pulses (instead of the fixed grid concentration) will always be used to update the concentration distribution profile.

In a host aerosol module or an Eulerian AQM, however, if the fixed grid concentration is used as a global variable in the other aerosol processes, pulses need to be determined based on the fixed grid concentration profile to provide initial conditions for the T-G scheme and the information of the pulses need to be mapped back to the fixed grid concentration profile (that will be used as inputs by the other processes) at the end of the simulation with the T-G scheme. Under such cases, T-G scheme becomes a semi-Lagrangian scheme and generally does not conserve mass, which is similar to other semi-Lagrangian schemes (Chock et al., 1996). Mass adjustment is necessary to overcome the mass inconservation problem.

3. SIMULATION RESULTS

3.1 Analyses of Results from a Single Component System Test

The performance of T-G scheme is first tested in a simple case, in which sulfuric acid is the only condensing species. The two test cases with sulfuric acid condensation rates of 5.5 and 11 μm^3 cm^{-3} under the hazy conditions in Zhang et al. (1999) are used here. These two cases are the most stringent tests for the simulation of condensational growth among the test cases used in Zhang et al. (1999). The performance of T-G scheme is compared with that of the other schemes tested in Zhang et al. (1999) under the same test conditions.

Figure 1 shows the initial and resultant volume concentration distributions after a 12-hr simulation period under the hazy conditions. A total of 12 size sections are used for the particle diameter range of 0.001 -10 µm for all the schemes. Simulating the hazy conditions is difficult because the high condensation rate and the particle size dependence of the growth law lead to a narrow but significant nuclei mode centered around 0.1 µm (Zhang et al., 1999). The "exact" result is obtained using the full-moving approach of the Analytical Predictor of Condensation (APC) of Jacobson (1999) with 500 size sections (Zhang et al., 1999). The "exact'' solution shows a peak of the nuclei mode of about 14 μm^3 cm^{-3}. A time step of 1 second is used for the T-G scheme. Two pulses are used in each section (a total of 24 pulses in the entire size range are considered here).

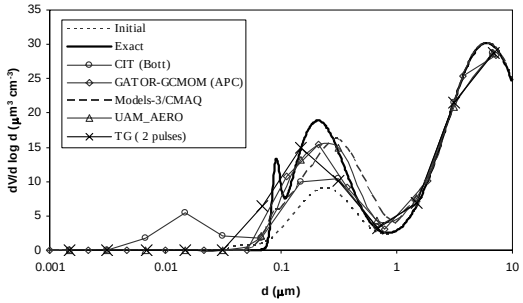

Figure 1. Simulations of condensation for hazy conditions with a condensation rate of 5.5 μm^3 cm^{-3} per 12-h.

The Bott scheme of CIT predicts a spurious peak centered at 0.0147 µm below 0.1 µm (i.e., an upstream numerical diffusion); consequently it

underpredicts the concentration in the accumulation mode. The T-G scheme with a total of 12 bins and 2 pulses per bin is also diffusive upstream, overpredicting the concentration for the bin centered at 0.07 μm. In contrast, other schemes do not have the upstream numerical diffusion problem. They predict a peak value closer to the "exact" peak value in terms of both magnitude (although the peaks are still somewhat lower than the "exact" peak) and size coordinate. The positions of the peak volume concentration predicted by GATOR-GCMOM and UAM_AERO are more consistent with the "exact" solution, whereas the peak predicted by the T-G scheme occurs at a smaller diameter and that predicted by Models-3/CMAQ occurs at a larger diameter. The Bott scheme gives the worst performance among all algorithms considered under this test condition. Since only 12 sections are used here, the resolution is not sufficiently fine for them to replicate the "continuous" distribution of the nuclei and accumulation modes in the "exact" solution.

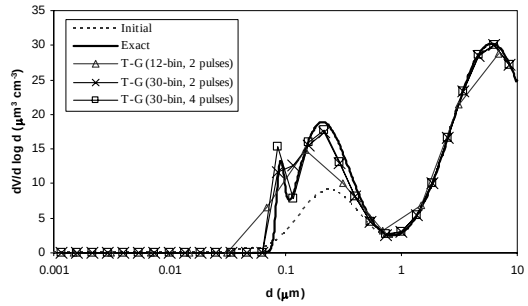

Figure 2. Simulations of condensation for hazy conditions with the T-G scheme with different total number of bins and total number of pulses in each bin.

Since the number of the sections and the pulses in each section used in the T-G scheme can affect the performance of the T-G scheme, several sensitivity simulations are conducted using the T-G scheme with different total number of sections and pulses in each section. Figure 2 shows that given more sections (i.e., 30, instead of 12), the predicted shape of the distribution is no longer diffusive upstream; and it is much closer to the "exact" solution in terms of both peak volume concentration and its position in the size coordinate. Given the same number of size bins (i.e., 30), the T-G scheme with 4 pulses per bin performs better than that with 2 pulses per bin. With 4 pulses in each section, the T-G scheme is capable of predicting two distinct nuclei and accumulation modes. The use of 4 pulses (instead of 2 pulses) in each size bin does not significantly increase the total cpu time (67.3 s vs. 64.43 s).

The system with a high condensation rate is more difficult to simulate than that with a lower condensation rate. Additional simulation with a higher condensation rate (i.e., 11 μm^3 cm^{-3} per 12-h) is conducted to further test the T-G scheme. Figure 3 shows the simulation results. As shown, the T-G scheme can also simulate the higher condensation rate case very well, especially when sufficient number of sections and more pulses in each section are used. With 30 bins and with 4 and 8 pulses in each bin, the T-G scheme predicts concentration distribution profile with two distinct nuclei and accumulation modes.

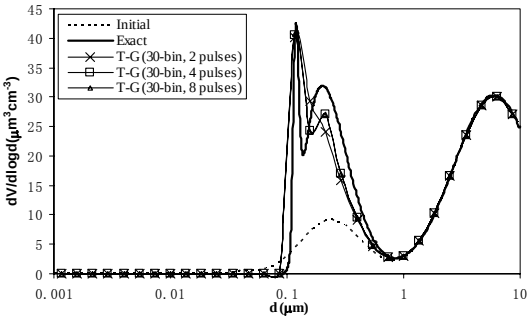

Figure 3. Simulations of condensation for hazy conditions with the T-G scheme with a condensation rate of 11 μm^3 cm^{-3} per 12-h.

3.2 Analyses of Results from A Multicomponent System Test Using MADRID

The T-G and Bott schemes are implemented in a kinetic gas/particle mass transfer approach in the box model that combines a gas phase mechanism with a modified Euler backward iteration (mebi) numerical solver and the MADRID1 aerosol module. Different from the condensation scheme, many other aerosol processes (e.g., nucleation, emission, deposition, and transport) require a fixed grid instead of the moving pulses. The results need to be interpolated to a fixed grid concentration distribution at the end of each time step. After the fixed grid concentration distribution is updated during the next time step, new pulses need to be assigned according to the updated fixed grid concentration distribution. This is the largest difference in the application of the T-G scheme between condensation only test case and multicomponent test case in which other aerosol processes are also simulated. In the multicomponent test system, the error of the interpolation may prorogate

through the time integrations. The T-G scheme thus becomes a semi-Lagrangian approach after its incorporation into a host aerosol module or 3-D AQM.

The initial conditions for the gas/particle mass transfer test case 2 in Zhang et al. (1999) are used here. This case represents conditions typical of the Los Angeles Basin, where it has been demonstrated that different treatments for gas/particle transfer (i.e., equilibrium and kinetic approaches) will make a difference on the particulate concentrations (Zhang et al., 1999). An

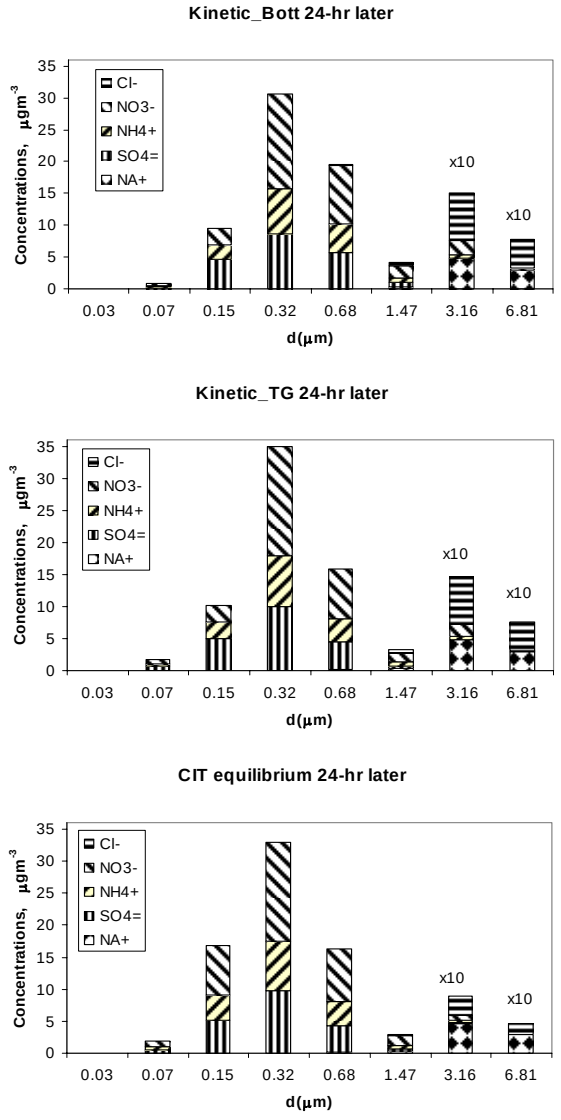

Figure 4. Size-resolved chemical composition predicted by kinetic and equilibrium approaches. The T-G and the Bott schemes are used in the kinetic approach.

initial gas phase mixing ratios of 15 ppb is assumed for both HNO_3 and NH_3. An initial concentration of NaCl is assumed to be 3 μg m^{-3}. To demonstrate different performance of the equilibrium and kinetic gas/particle mass transfer approaches, the size-resolved aerosol composition predicted by the CIT equilibrium approach along with those from the two kinetic approaches (one with the T-G scheme, the other one with the Bott scheme) are shown in Figure 4. The advantages of equilibrium approaches are their speed and simplicity. But in some cases (e.g., coarse particles, low temperatures, and low accommodation coefficients for condensing species) equilibrium cannot be established, then kinetic gas/particle mass transfer approach is required to obtain an accurate prediction (Zhang et al., 1999; Koo et al., 2003).

Figure 4 shows the predicted size-resolved chemical composition by the kinetic gas/particle transfer approach with the T-G and Bott schemes, and the CIT equilibrium approach. 0.5 second is used as the time step for solving condensation equations for the T-G and Bott schemes. Although the T-G scheme loses its advantage as a fully Lagrangian scheme in the box model that simulates all other processes, its performance does not deteriorate significantly. It predicts the size-resolved chemical composition similar to that predicted by the kinetic approach with the Bott scheme. Compared with the results with the equilibrium approach, the kinetic approach with the two condensation schemes predict more chloride and nitrate in the coarse mode. The kinetic approach predicts a different concentration distribution from the equilibrium approach around the peak. CIT equilibrium approach predicts around 6 μg m^{-3} more mass in the 3rd section (centered at 0.15 μm) than the two kinetic approaches.

4. SUMMARY

In simulating the condensation process alone, the T-G scheme uses a fully Lagrangian approach to solve the advection-like term in the condensation/evaporation equation. The results show that the T-G scheme is numerically more accurate than the Bott scheme used in CIT under the conditions tested here. The performance of the T-G scheme can be improved with a finer size resolution and more number of pulses per size bin. The T-G scheme can perform well in a system with a high condensation rate.

After incorporated into a host aerosol module or a 3-D AQM, T-G scheme becomes a semi-Lagrangian scheme since the other processes simulated in the module or AQM use the fixed grid concentration rather than the pulse concentration.

The test simulation in this study in a multicomponent system shows that the kinetic gas/particle approach with the T-G scheme predicts similar size-resolved chemical composition as the kinetic approach with the Bott scheme.

Additional test simulations are being conducted for both condensational schemes for other representative cases in MADRID. Both schemes will be further evaluated in 3-D CMAQ-MADRID or the NOAA's Weather Research and Forecasting/Chemistry prediction system with MADRID (WRF/Chem-MADRID).

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