Efficient Splitting Characteristic Method for Solving Multi-component Aerosol Spatial Transports in Atmospheric Environment

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***Abstract*—In this work, we develop a splitting characteristic method for solving multi-component atmospheric aerosol spatial dynamics, which can efficiently evaluate aerosol spatial transports by using large time step sizes. The method can compute the multi-component aerosol distributions in high-dimensional domains with large ranges of concentrations and for different aerosol types. Numerical tests show the computational efficiency of the proposed method. An actual simulation focusing on the sulfate pollution is taken in the domain with a varying wind field. We also simulate multi-component aerosol transports in a large area in the southeast of America. The developed algorithm can be applied for the large scale predictions of multi-component aerosols in multi-regions and multi-levels in atmospheric environment.**

Keywords–Atmospheric aerosol transport; Multi-component; Splitting; Characteristic method; Efficiency; High accuracy.

I. INTRODUCTION

Global climate change and warming in atmosphere have been widely recognized. As one of most important constituents, aerosols have a direct radiative forcing by scattering and absorbing solar and infrared radiation in atmosphere, while they have an indirect radiative forcing associated with the changes in cloud properties by decreasing the precipitation efficiency of warm clouds. In these processes, the physical states including the gas and aerosol phases (i.e., gas, liquid and solid) and the composition of aerosols including the aerosol-associated water mass, and the multi-components of aerosols are of great significance. Numerical modeling has been playing a key role in the study of aerosol processes and aerosol concentration distributions in the atmospheric environment prediction and the air quality control.

Aerosol transport model in atmosphere is a complex multi-component system that involves several physical and chemical processes, such as emission, transport, dispersion, aerosol dynamics and aerosol chemistry processes. The studied area usually covers a large region. Odman and Russell [12] studied the URM model and the UAM-AIM model in the aerosol simulation in the southern California. Grell et al. [5] developed the WRF/Chem model and simulated the aerosol distributions in the eastern United States and contiguous areas. Further applications have been done to some areas in the Europe [11]. However, in these computations, very small time steps have to be used in order to ensure the numerical stability of the numerical schemes, which brings a huge cost of computation and some limitation of applications. Therefore, it has been an important task to develop efficient numerical algorithm for multi-component aerosol transports.

In this paper, we present our new development of the efficient splitting method for solving aerosol transports in atmosphere. We consider general spatial aerosol transport problems in atmosphere and develop the splitting characteristic method for modeling multi-component aerosol transports. For the spatial transport systems, we propose the characteristic finite difference method to solve the transport process by combining with the operator splitting technique to deal with processes of emission, aerosol dynamics and chemical process. The methods of characteristics to treat convection terms were studied for high dimensional convection-diffusion problems in porous media [1][2][4][9]. In this study, we take the important advantages of both the characteristic method and the operator splitting technique to solve the spatial aerosol transport dynamical system, which can be solved in parallel computation. The developed method can efficiently compute the multi-component aerosol transport dynamics in high-dimensional domains with a large range of aerosol concentrations and for different types of aerosols. Numerical tests show the computational efficiency of the proposed method for both the spatially homogeneous aerosol dynamic problems and the spatial transport aerosol dynamic problems. An actual simulation focusing on the sulfate pollution is then taken in the domain with a varying wind field. Finally, a simulation of multi-component aerosol transports in a large area in the southeast of America is taken, which shows clearly that city areas usually have high $PM_{2.5}$ concentration and marine aerosols have affection in marine and coast areas. The developed algorithm can be applied for the large scale predictions of multi-component aerosols in multi-regions and multi-levels in environment.

The paper is arranged as follows. The mathematical model of multi-component aerosol dynamics is presented in Section II. The splitting numerical method is proposed for the multi-component aerosol transport equations in Section III. Numerical simulations are given in Section IV. Some conclusions are addressed in Section V.

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II. Multi-component Aerosol Dynamic Models

The multi-component aerosol spatial transport models are:

$$\begin{aligned}\frac{\partial c_l}{\partial t} &= -\mathbf{U}\cdot\nabla c_l + \nabla\cdot(K\nabla c_l) + \mathcal{L}_{Aerosol}(\vec{c}) \\ &\quad + E_{l_{Emission}}(\vec{x},v,t), \quad l=1,2,\cdots,s, \qquad (1)\end{aligned}$$

$$\begin{aligned}c_l(\vec{x},v,t) &= c_l^{IN}(\vec{x},v,t),\ \vec{x}\in\Gamma_{IN}, \qquad (2)\\ K\nabla c_l\cdot\vec{\nu} &= 0,\ \vec{x}\in\Gamma_{OUT}, \qquad (3)\\ K\nabla c_l\cdot\vec{\nu} &= 0,\ \vec{x}\in\Gamma_{TOP}, \qquad (4)\\ K\nabla c_l\cdot\vec{\nu} &= E_{l,g},\ \vec{x}\in\Gamma_{GR}, \qquad (5)\\ c_l(\vec{x},v,0) &= c_l^0(\vec{x},v), \qquad (6)\end{aligned}$$

where $c_l(\vec{x},v,t)$ is the mass concentration of aerosol species l at position $\vec{x}=(x,y,z)$ in space at time t, and at particle volume v; $\vec{c}=(c_1,c_2,\cdots,c_s)$; c_l^0 and c_l^{IN} are initial and inflow boundary values. Ω is a three dimensional rectangle computational domain, and the boundary of the domain is partitioned into $\partial\Omega=\Gamma_{IN}\cup\Gamma_{OUT}\cup\Gamma_{GR}\cup\Gamma_{TOP}$, where Γ_{IN} denotes the inflow lateral boundary, Γ_{OUT} denotes the outflow lateral boundary, Γ_{GR} is the ground level portion of the boundary, and Γ_{TOP} is the top level portion of the boundary. $\vec{\nu}$ is the unit outward normal to the boundary $\partial\Omega$. The volume interval is $[V_{\min},V_{\max}]$. At $v=V_{\min}$, the concentrations of aerosols are zero. $E_{l,g}$ is the rate of ground level emission of the specie.

III. The Splitting Numerical Scheme

Let Δt be the splitting time step size, and $t^n=n\Delta t$. In a time interval $(t^n,t^{n+1}]$, the splitting numerical scheme for the multi-component aerosol spatial transport problems is proposed as:

Step 1. From c_l^n, the value at $t=t^n$, solve the aerosol emission process over $(t^n,t^{n+1}]$

$$\begin{aligned}\frac{\partial c_l}{\partial t} &= E_{l_{Emission}}(\vec{x},v,t), \qquad (7)\\ c_l(\vec{x},v,t^n) &= c_l^n(\vec{x},v)\ \vec{x}\in\Omega. \qquad (8)\end{aligned}$$

The emission rates of multi-component aerosol particles are different due to differences in the emission characteristics of emitted particles from a variety of natural and anthropogenic sources. We can get the solution $c_{l,emission}^{n+1}$ of problem (7)(8) at $t=t^{n+1}$.

Step 2. Solve the multi-component aerosol dynamic process in $(t^n,t^{n+1}]$ with $c_{l,emission}^{n+1}$ being the initial value at $t=t^n$.

$$\begin{aligned}\frac{\partial c_l}{\partial t} &= \mathcal{L}_{Aerosol}(c_1,c_2,\cdots,c_s;\vec{x},v,t) \qquad (9)\\ c_l(\vec{x},v,t^n) &= c_{l,emission}^{n+1}(\vec{x},v),\ l=1,2,\cdots,s. \qquad (10)\end{aligned}$$

The aerosol process includes the aerosol dynamic processes (condensation, coagulation), and the aerosol chemical process. Numerical methods, such as modal methods, sectional methods, and wavelet methods ([10]), can be used to solve the aerosol dynamic process. The aerosol chemical process is described by aerosol thermodynamic equilibrium equations, which can be solved ISORROPIA II ([7]) and the moving-cut HDMR method ([3] [8]). Denote the solution by $c_{l,aero}^{n+1}$.

Step 3. From $c_{l,aero}^{n+1}$ as the value at $t=t^n$, the spatial transport process is solved in $(t^n,t^{n+1}]$.

$$\begin{aligned}\frac{\partial c_l}{\partial t} &= -\mathbf{U}\cdot\nabla c_l + \nabla\cdot(K\nabla c_l), \qquad (11)\\ c_l(\vec{x},v) &= c_l^{IN}(\vec{x},v,t^n),\vec{x}\in\Gamma_{IN}, \qquad (12)\\ K\frac{\partial c_l(\vec{x},v)}{\partial\nu} &= 0,\vec{x}\in\Gamma_{OUT}, \qquad (13)\\ K\frac{\partial c_l(\vec{x},v)}{\partial\nu} &= 0,\vec{x}\in\Gamma_{TOP}, \qquad (14)\\ K\frac{\partial c_l(\vec{x},v)}{\partial\nu} &= E_{l,g},\ \vec{x}\in\Gamma_{GR}, \qquad (15)\\ c_l(\vec{x},v,t^n) &= c_{l,aero}^{n+1}(\vec{x},v). \qquad (16)\end{aligned}$$

We propose a characteristic method to solve systems (11)-(16) in this step. Finally, the solution c_l^{n+1} is obtained at $t=t^{n+1}$.

IV. Numerical simulation

We now give numerical simulations of the spatially homogeneous aerosol dynamics and the multi-component atmospheric aerosol spatial transport dynamics.

Example 1.

We consider the spatially homogeneous three-component aerosol dynamics of aerosol water, black carbon and sulfate components. The initial values are tri-modal log-normal distributions. The spatially homogeneous three-component aerosol dynamic equations are

$$\begin{aligned}\frac{\partial c_i(m,t)}{\partial t} &= \mathcal{L}_{Aerosol}(\vec{c}) := \eta_i c(m,t) - \frac{\partial(m\eta c_i)}{\partial m}\\ &\quad + \int_{M_{\min}}^{m-M_{\min}} \beta(m,m-m')c_i(m,t)\frac{c(m-m',t)}{m-m'}dm'\\ &\quad - c_i(m,t)\int_{M_{\min}}^{M_{\max}}\beta(m,m')\frac{c(m',t)}{m'}dm', \qquad (17)\\ c_i(M_{min},t) &= 0,\quad t\in[0,T], \qquad (18)\\ c_i(m,0) &= c_i^0(m),\quad m\in\Omega,\quad i=1,2,3. \qquad (19)\end{aligned}$$

where $t>0$ is the time, and $T>0$ is the time period; the finite mass interval $I=[M_{\min},M_{\max}]$ where $M_{\min}>0$ is the minimal mass and $M_{\max}>0$ is a finite maximal mass. $\beta(m,m')$ is the coagulation kernel function. $\eta_i(m,t)$ is the growth rate of species i, $\eta(m,t)=\sum_{i=1}^3\eta_i(m,t)$. $c_i(m,t)$ is the mass concentration of aerosol species i and $c(m,t)=\sum_{i=1}^3 c_i(m,t)$. Eq. (17) forms a system of nonlinear integral-differential equations on time and particle mass m.

Let

$$\begin{aligned}&F_i\big(m,t,c_i(m,t),c(m,t)\big) \qquad (20)\\ &= \int_{M_{\min}}^{m-M_{\min}}\beta(m',m-m')c_i(m',t)\frac{c(m-m',t)}{m-m'}dm'\\ &\quad - c_i(m,t)\int_{M_{\min}}^{M_{\max}}\beta(m,m')\frac{c(m',t)}{m'}dm'.\end{aligned}$$

We propose the time second-order characteristic method for the three-component aerosol dynamic equations (17) as

$$\frac{c_{i,h}^{k+1}(m)-c_{i,h}^{k}(\bar{m})}{\Delta t}+\eta\frac{c_{i,h}^{k+1}(m)+c_{i,h}^{k}(\bar{m})}{2}$$
$$-\eta_i\frac{c_{h}^{k+1}(m)+c_{h}^{k}(\bar{m})}{2}=\frac{3}{2}F_i\big(\bar{m},c_{i,h}(\bar{m},t^k),c_h(\bar{m},t^k)\big)$$
$$-\frac{1}{2}F_i\big(\bar{\bar{m}},c_{i,h}(\bar{\bar{m}},t^{k-1}),c_h(\bar{\bar{m}},t^{k-1})\big),$$
$$i=1,2,3, \tag{21}$$

where $\bar{m}^k$ and $\bar{\bar{m}}^{k-1}$ are the intersection points of the characteristic curve at time level $t=t^k$ and $t=t^{k-1}$ respectively.

TABLE I. COMPARISON OF ERRORS BY OUR METHOD AND S-C-FEM FOR THE THREE-COMPONENT AEROSOL CONDENSATION PROBLEM WITH A TRI-MODAL INITIAL DISTRIBUTION.

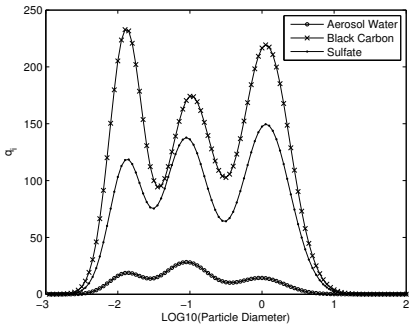
	Δt (hour)	$\frac{T}{8}$	$\frac{T}{16}$	$\frac{T}{32}$	$\frac{T}{48}$
Our method	E_∞	5.1358e-3	1.3000e-3	3.3209e-4	1.4579e-4
	Ratio	-	1.9821	1.9688	2.0304
	E_2	3.2027e-3	7.9209e-4	1.9431e-4	8.4280e-5
	Ratio	-	2.0155	2.0273	2.0601
S-C-FEM	E_∞	3.6335e-1	1.7538e-1	7.8061e-2	4.5141e-2
	Ratio	-	1.0509	1.1678	1.3508
	E_2	2.2616e-1	1.0915e-1	4.8613e-2	2.8123e-2
	Ratio	-	1.0510	1.1669	1.3498

We solve the general problem on the $\Omega = [5.236 \times 10^{-22}, 5.236 \times 10^{-7}]$g and time interval $[0,T]=[0,5]$ hours. Table I presents the comparison of the errors and ratios in time step of the predicted results by our method and the standard characteristics FEM scheme (S-C-FEM), for the three-component problems with a same three-modal distribution for each species. It is obvious that our method is of second-order accuracy in time step but the standard characteristic finite element method (S-C-FEM) is only of first-order accuracy in time step.

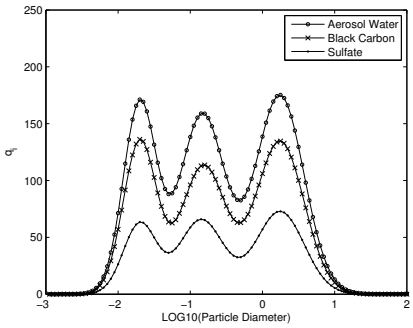
The predicted mass concentration distributions of the of aerosol water, black carbon and sulfate components are shown in Fig. 1 for problem with different initial species distributions. We can see that the peaks of the distributions of aerosol water, black carbon and sulfate components change a lot during the simulation. For example, at time $T=0$, the mass distribution of aerosol water has the highest peak value of 28.307 in the accumulation mode, while at time $T=5$, the highest peak value is 175.28 located in the coarse mode. Due to the highest condensation rate among the three species, the concentration of aerosol water is the smallest at time $T=0$ hour, but becomes the largest at time $T=5$ hours. For the distribution of the total mass, we can see that the peak values keep almost unchange.

Example 2.

We then do a simulation to the spatial multicomponent aerosol transport by the developed method, where the WRF ([13]) is used to provide the information of wind components, temperature, pressure, water vapor, clouds, and rainfalls, etc. The studied domain is a 2400 km×1800 km area centered at 79.25°W longitude and 43.40°N latitude with the horizontal grid dimension 40 (west-east)×30 (south-north) with the spacing of 60 km, the vertical interval of the domain consist of 27 layers up to approximately 20.1 km. The layers of the aerosol transport model are aligned with the layers in



(top)



(bottom)

Figure 1. Initial distributions of the mass concentrations of aerosol water, black carbon and sulfate components (top) and the numerical solutions at time $T=5$ hours (bottom) for the multi-component condensation problem. The condensation rates of the aerosol water (aw), black carbon (bc) and sulfate (sul) are $\alpha_{aw}=1.6\times10^{-8}$ hour $^{-1}$, $\alpha_{bc}=7\times10^{-9}$ hour $^{-1}$, $\alpha_{sul}=3\times10^{-9}$ hour $^{-1}$, $M_{\min}=5.236\times10^{-22}$g, $M_{\max}=5.236\times10^{-7}$g.

the meteorological WRF model, with a vertical mesh interval from 60 m near the surface to 1.5 km at the domain top. The emission inventory used in the simulation is the EPA U.S. National Emissions Inventory (NEI-05) [14]. The simulation period presented here consists of the last 72 hours of a 108-hour simulation.

Fig. 2 presents the hourly predicted concentrations of total $PM_{2.5}$ total mass and the sum of $PM_{2.5}$ sulfate, ammonium, nitrate, sodium and chloride concentrations in New York, the rural and marine area, which show that these five species are the mainly constituents of $PM_{2.5}$ total mass, and the sum of their concentrations determines the variation trends of total concentration of $PM_{2.5}$ total mass.

The variation of the nitrate concentration and temperature of NY are presented in Fig. 3. Thus, it is obvious that low temperatures are more favorable to the generation of the nitrate particulate phase. This result agrees well with existing theories and experiment.

Example 3.

Finally, we simulate multi-component aerosol spatial transports in a large regional area in the southeast of the America. The studied domain is a 40×40 mesh grid, which centered at 82.5°W longitude and 36°N latitude and the spacing is 20km. We take a 228 hours simulation and analyze numerically the last 120 hours simulation results, which will limit the influence of the default initial condition of chemical species

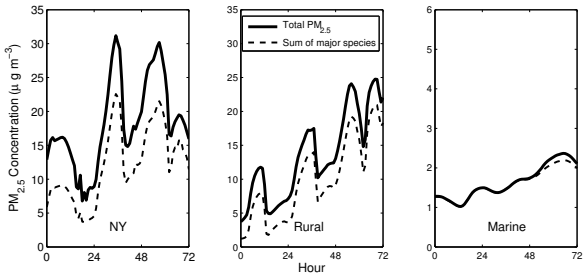


Figure 2. Predicted concentrations of total $PM_{2.5}$ mass concentration and the sum of major species, $PM_{2.5}$ sulfate, ammonium, nitrate, sodium and chloride concentrations in New York, the rural and marine area.

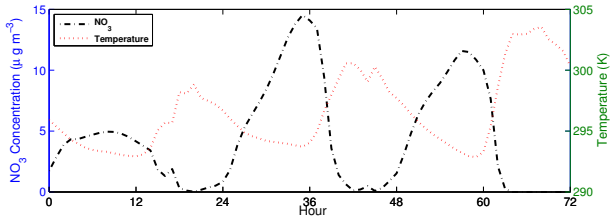


Figure 3. Comparisons of hourly predicted concentrations of $PM_{2.5}$ nitrate and temperature in New York.

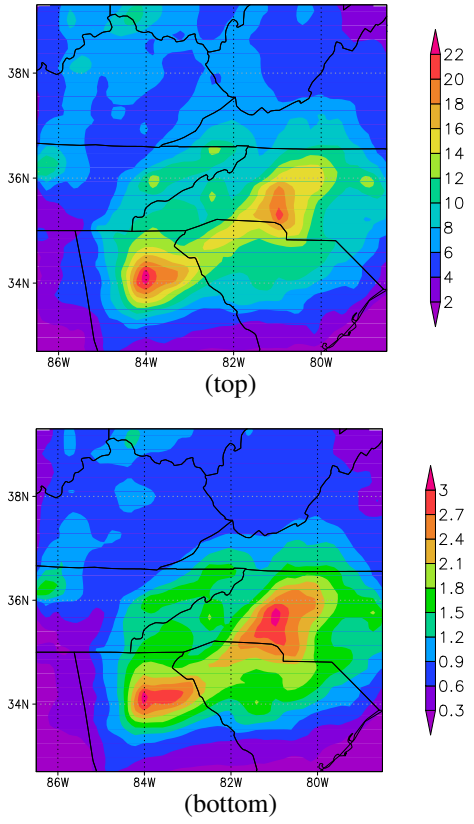


Figure 4. The predicted ground-level concentration distributions of $PM_{2.5}$ total mass (top) and ammonium (bottom) (μg m^{-3}).

concentration. For transport process, the time step size is used as 1800s for the characteristic finite difference method, and the time step of the aerosol process step is used as 120s in the system.

Fig. 4 and Fig. 5 show the averaged ground-level concentrations of $PM_{2.5}$ total mass, nitrate, ammonium, chloride (μg m^{-3}). We can see that, nitrate accounts for a large portion of the $PM_{2.5}$ total mass in the predicted domain, and $PM_{2.5}$ nitrate, ammonium generally have high concentrations near cities. For instance, the area near Atlanta (84.39°W, 33.67°N) has the highest predicted concentrations of $PM_{2.5}$ nitrate and ammonium, and total mass with over 7 μg m^{-3}, 3 μg m^{-3}, and 22 μg m^{-3}, respectively. Marine aerosol of chloride only exists in the marine and coast area,which is 0.03 to 0.14 μg m^{-3} for chloride from the coast to the open sea.

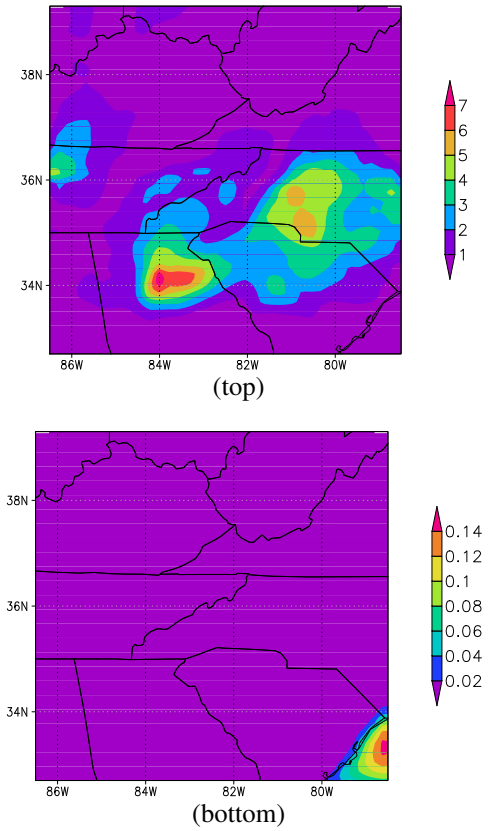


Figure 5. The predicted ground-level concentration distributions of nitrate (top) and chloride (bottom) (μg m^{-3}).

V. Conclusion

In this paper, we developed a characteristic method, combining with the splitting technique, for multi-component aerosol spatial transports which involve several physical and chemical processes of advection, dispersion, emission, deposition and aerosol dynamic processes. The developed algorithm is robust, efficient, and highly accurate for the spatially homogeneous aerosol dynamics and the spatial multi-component aerosol transports, in which large time steps can be used in simulation. A simulation of multi-component aerosol transports over an area in the northeast America was further done by the algorithm. The 72-hour simulation results at the New York, a rural area and a marine area show that the $PM_{2.5}$ has the highest concentration among the three areas, while the marine area has the lowest. The major species of aerosols in

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the New York and rural areas are sulfate, nitrate and ammonia, while chloride and sodium make the most portion of the marine aerosol. The results also exhibited that the nitrate concentration varies inversely with the temperature. The comparisons of simulation results with and without dry deposition provided the fact that dry deposition makes great contribution to the aerosol decrease, and has more influence to larger particles. Finally, a real life multi-component aerosol transport simulation was carried out over a large area in the southeast of America, which showed that, city areas usually have high concentration of $PM_{2.5}$, and marine aerosols only have affection of marine and coast areas. The developed algorithm can be applied to efficiently simulate multi-component atmospheric aerosol spatial transports in large domains.

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