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A New Weighted Fraction Monte Carlo Method for Particle Coagulation

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Abstract

Purpose – The purpose of this study is to investigate the aerosol dynamics of the particle coagulation process using a newly developed weighted fraction Monte Carlo (WFMC) method.

Design/methodology/approach – The weighted numerical particles are adopted in a similar manner to the multi-Monte Carlo (MMC) method, with the addition of a new fraction function, α. Probabilistic removal is also introduced to maintain a constant number scheme.

Findings– Three typical cases with constant kernel, free-molecular coagulation kernel and different initial distributions for particle coagulation are simulated and validated. The results show an excellent agreement between the Monte Carlo method and the corresponding analytical solutions or sectional method results. Further numerical results show that the critical stochastic error in the newly proposed WFMC method is significantly reduced when compared with the traditional MMC method for higher-order moments with only a slight increase in computational cost. The particle size distribution (PSD) is also found to extend for the larger size regime with the WFMC method, which is traditionally insufficient in the classical direct simulation Monte Carlo (DSMC) and MMC methods. The effects of different fraction functions on the weight function are also investigated.

Originality/value– Stochastic error is inevitable in Monte Carlo simulations of aerosol dynamics. To minimize this critical stochastic error, many algorithms, such as MMC method, have been proposed. However, the weight of the numerical particles is not adjustable. This newly developed algorithm with an adjustable weight of the numerical particles can provide improved stochastic error reduction.

Keywords: General dynamic equation; Multi-Monte Carlo method; Fraction function; Weighted fraction Monte-Carlo method; Particle coagulation

Paper type Research paper

1. Introduction

Aerosol dynamics covers a wide variety of scientific fields (Friendlander, 2000,

Ramkrishna, 2000, Knopf *et al.*, 2018) such as atmospheric physics (aerosol particles), combustion science (polycyclic aromatic hydrocarbons and soot), and chemical engineering (gelation and crystallization). Aerosol dynamic processes include nucleation, surface growth (condensation), and coagulation (Zhang *et al.*, 1999). The coagulation process plays an important role in the time evolution of the particle size distribution (PSD) (Meng *et al.*, 1998). The evolution of the PSD is always described using the population balance equation (PBE) (Ramkrishna, 2000). The PBE is a convection-diffusion equation with source terms including nucleation, condensation, and coagulation.

The PBE is a partial integral-differential equation; analytical solutions of the PBE are available for only a few ideal cases (Von Smoluchowski, 1916). In general, approximate solutions can be obtained using various numerical methods such as the sectional method (SM) (Gelbard *et al.*, 1980, Prakash *et al.*, 2003), method of moments (MOM) (Frenklach, 2002, Yu *et al.*, 2008, Yu *et al.*, 2016, Chan *et al.*, 2018), and Monte Carlo (MC) method (Zhao *et al.*, 2009, Zhou and He, 2014, Zhou *et al.*, 2014, Liu and Chan, 2017b, Liu and Chan, 2017a, Liu and Chan, 2018a). However, there are two disadvantages to MC methods, i.e., stochastic error and a high computational cost (Xu *et al.*, 2015). Because only a finite number of numerical particles can be used, uncertainty in PSD function is inevitable. This stochasticity can be determined by repeating a sufficient number of numerical simulations with different random seeds (e.g., system time) to restrict the stochastic error (e.g., variance, standard deviation) (Zhou *et al.*, 2020). However, the tails of the particle size spectrum always contain only a small number of numerical particles, which are poorly represented in the numerical simulation. This problem becomes especially severe when the PSD is in a logarithmic form (Zhao et al., 2009).

In recent years, some modified numerical algorithms have been developed to improve the numerical efficiency and accuracy of stochastic methods. The constant-number method (Smith and Matsoukas, 1998, Lee and Matsoukas, 2000, Lin *et al.*, 2002) continuously changes the numerical simulation volume, while the number of numerical particles is maintained constant. The stepwise constant-volume method (Kruis *et al.*, 2000, Maisels *et al.*, 2004) doubles the numerical simulation volume while the number of numerical particles become halves, to reset it to the initial values. Although these numerical algorithms can constrain the total number of numerical particles within appropriate ranges, the numerical particles at the tails of the particle size spectrum are always poorly represented, resulting in stochastic error. To avoid this problem, the mass flow algorithm (MFA) (Eibeck and Wagner, 2001) was proposed, in which numerical particles with different weights were adopted for coagulation dynamics. The MFA was shown to provide significant improvement in numerical efficiency and variance reduction. More generally, the weighted flow algorithm (WFA) proposed by DeVille et al. (2011) can work with arbitrary weighting functions, especially in the power law functions of particle size. The new stochastically and differentially weighted operator splitting Monte Carlo (i.e., SWOSMC and DWOSMC) methods were first developed by the research group (Liu and Chan, 2017b, Liu and Chan, 2018b, Liu and Chan, 2019, Liu *et al.*, 2019). Another stochastic error reduction technique is the multi-Monte Carlo (MMC) method proposed by the research group (Zhao *et al.*, 2005, Zhao et al., 2009), which leads the concept of 'fictitious particles' where the number of fictitious particles and the simulation volume are both maintained constant. The MMC

method shows high numerical efficiency and low stochastic error, which makes this method applicable for many problems in population balance modeling. Kotalczyk and Kruis (2017) introduced 'stochastic resolution' to construct a constant-number coagulation scheme, which was very similar to the MMC method but with an asymmetric coagulation kernel function.

Although the MMC method can maintain the number of numerical particles, the variation in the weight of numerical particles in the size interval is not adjustable. However, the number of large numerical particles is still insufficient for some situations. To simulate large particles more accurately, the weighted fraction Monte Carlo (WFMC) method is newly proposed and developed in the present study. The novelty of the WFMC method is the adjustable weight function of the numerical particles, which can ensure that the number of numerical particles in different particle size intervals is in the appropriate range by introducing a fraction function, α. Furthermore, the simulation volume and number of numerical particles are maintained constant in the WFMC method by implementing probabilistic removal. The remainder of this paper is organized as follows. The WFMC method is first proposed and described in detail. The numerical efficiency and precision of the WFMC method are then verified through the numerical simulations with different coagulation kernel functions and initial distributions. A comparison of stochastic errors for different algorithms is also performed. Furthermore, the effects of different fraction functions on the weight function and computational efficiency of this algorithm are analyzed. Finally, some major conclusions are drawn.

2. Description of the Weighted Fraction Monte Carlo Method

2.1. Smoluchowski Equation and the Monte Carlo Method

The Smoluchowski equation describes the time evolution of the PSD during the coagulation process (Von Smoluchowski, 1916):

$$\frac{\partial n(v,t)}{\partial t}=\frac{1}{2}\int_0^v \beta(u,v-u)n(u,t)n(v-u,t)du-\int_0^\infty \beta(v,u)n(v,t)n(u,t)du \tag{1}$$

where $n(v, t)$ is the particle number density of volume v at time t, and $\beta(u, v)$ is the coagulation kernel function which describes the coagulation rate for two particles with volumes u and v.

Gillespie (1975) developed a stochastic algorithm to solve Equation (1), which is the corner-stone of many subsequent developments. The basics of the algorithm are sketched as follows.

A coagulation event will occur at a random time, τ, which satisfies a Poisson distribution:

$$P(\tau \geq \tau_0)=\exp(-C_0\tau_0) \tag{2}$$

where

$$C_0=\sum_{i=1}^{N-1}\sum_{j=i+1}^{N} C_{ij} \tag{3}$$

$$C_{ij} = \frac{\beta(v_i, v_j)}{V} \tag{4}$$

where i and j are the indices of the numerical particles, N; v_i and v_j are the volumes of the corresponding particles, respectively; τ_0 is the reference coagulation time; C_{ij} depicts the coagulation rate in the simulation volume, V, between particles i and j; and C_0 represents the total coagulation rate between any two particles, which determines how quickly a coagulation event can occur. The coagulation pair is selected according to the probability as follows:

$$P(i, j) = \frac{C_{ij}}{C_0} \tag{5}$$

Then, both particles i and j are removed and a particle of volume $v_i + v_j$ is added. The details of the actual implementation can be found in the original work (Gillespie, 1975).

2.2. Derivation of the Coagulation Rate

In the classical direct simulation Monte Carlo (DSMC) method, numerical particles are assigned the same weight. The number of numerical particles in different particle size intervals is proportional to the particle number density. As a result, there is always an insufficient number of numerical particles at the tails of the PSD. This introduces stochastic error and a narrower PSD in stochastic simulations, which has constrained the application of the classical DSMC method (Zhao et al., 2009). These limitations can be overcome by introducing different particle weights. This approach considers that each numerical particle is related to a certain number of physical particles, and the action (e.g., add/remove/change) is implemented on the numerical particle with certain probabilities instead of directly implemented on a real physical particle.

For the coagulation process of numerical particles with different weights, the new coagulation rates between numerical particles need to be determined. Specifically, the coagulation rate depends on how the coagulation event between a coagulation pair is implemented, and then the jump Markov process is constructed based on this new coagulation rate.

A numerical particle, i, represents a number of physical particles, w_i, with size v_i; thus, i represents a group of physical particles with number concentration, w_i/V. This analysis is the same for numerical particle j. From the definition of the collision kernel function (Friendlander, 2000), the number of real coagulation events occurring among ith-group particles and jth-group particles per unit time and volume is given as:

$$\Phi_{ij} = \beta_{ij} \times \frac{w_i}{V} \times \frac{w_j}{V} \tag{6}$$

where β_{ij} is the collision kernel function for particles i and j, and Φ_{ij} is the coagulation rate for the group of physical particles, i and j.

However, if the probabilistic coagulation rule from Zhao et al. (Zhao et al., 2009) is applied, the coagulation event is related to two numerical particles, but not every real particle from the numerical particles is considered to participate in the coagulation event. For example,

the mean number of real coagulation events per real particle from i or j are considered in the MMC method as:

$$\Omega = \min(w_i, w_j) \tag{7}$$

If fewer coagulation events between real particles are considered, only a fraction, α_{ij}, of coagulation events takes place among numerical particles i and j, and the mean number of real coagulation events is then considered as:

$$\Omega' = \alpha_{ij} \min(w_i, w_j), \qquad \alpha_{ij} \le 1 \tag{8}$$

It should be noted that the fraction function, α_{ij}, cannot have a value greater than 1 because the maximum number of coagulation pairs between i and j is equal to $\min(w_i, w_j)$. In addition, the fraction function is not restricted to be constant; it can be a function of any specific properties (e.g., particle weight or particle volume) of the numerical particles, i and j, considered for coagulation.

Irrespective of the coagulation rule adopted, the coagulation rate of a physical particle from the same numerical particle in the simulation volume, V, should always be the same and is expressed as follows:

$$\Omega C_{ij} = \Omega' C'_{ij} = V \Phi_{ij} \tag{9}$$

$$\begin{aligned} C'_{ij} &= \frac{1}{\Omega'} \Phi_{ij} = \frac{1}{\alpha_{ij}} \frac{\max(w_i, w_j) \beta_{ij}}{V} \\ &= \frac{\beta'_{ij}}{V} \end{aligned} \tag{10}$$

where C'_{ij} is the new coagulation rate of numerical particle pair i and j with numerical simulation volume V; $\beta'_{ij} = 1/\alpha_{ij} \max(w_i, w_j) \beta_{ij}$ is the normalized coagulation kernel, in which β'_{ij} is related not only to the size of the particles but also could be related to their other specific properties. Then, the new jump Markov model for the WFMC method is constructed based on β'_{ij}.

The waiting time between two successive coagulation events of the numerical particles obeys an exponential distribution as follows:

$$P(\tau) = C_0' \exp(-C_0' \tau) \tag{11}$$

where

$$C_0' = \sum_{i=1}^{N} \sum_{j<i} C_{ij}' \tag{12}$$

For the time-driven Monte Carlo scheme, the occurrence probability, $P'_{\text{coag}}(\Delta t)$, of a coagulation event for numerical particles within Δt and V is an exponentially distributed random variable (Garcia *et al.*, 1987):

$$P'_{\text{coag}}(\Delta t) = 1 - \exp(-\Delta t C'_0) \tag{13}$$

Coagulation particle pairs can be randomly selected by the cumulative probability method (Liffman, 1992) or the acceptance-rejection method (Lin *et al.*, 2002). However, the acceptance-rejection method adopts a simpler and more straightforward criterion for selecting the particle pairs compared with the cumulative probability method (Wei, 2013). Hence, the acceptance-rejection method is used in the present study. A possible particle pair, *i*, *j*, is first selected randomly. Then, if the following condition is satisfied, numerical particles *i* and *j* are accepted as a coagulation particle pair:

$$r \le \beta'_{ij} / \max_{\forall k, \forall m}(\beta'_{km}) \tag{14}$$

where *r* is a random number, *r*~ *U* [0,1].

2.3. Treatment of a Coagulation Event

Once the coagulation particle pair, *i* and *j*, is selected, numerical particle *i* will coagulate with its coagulation particle partner, *j*. As a result, some number of physical particles from numerical particle *i* will also coagulate with those from numerical particle *j*. However, if numerical particles *i* and *j* have different weights (e.g., $w_i > w_j$), that means there will not be enough physical particles in numerical particle *j* to form particle pairs with those from numerical particle *i*. Therefore, after coagulation events between particle pairs with different weights, the physical particles can be separated into "coagulated" physical particles and "non-coagulated" physical particles.

For a coagulated numerical particle, a new weight, $w''_{\text{coag}} = \alpha_{ij} \min(w_i, w_j)$, is obtained with a new volume equal to the summed volume of the two coagulation particle partners, while the other two numerical particles are assigned a new weight equal to the number of non-coagulated real particles. The weights for these two numerical particles are $w''_i = w_i - \alpha_{ij} \min(w_i, w_j)$ and $w''_j = w_j - \alpha_{ij} \min(w_i, w_j)$. If a coagulation particle pair has equal weight and all physical particles will coagulate, then these physical particles can be separated into two groups. With an equal weight of two numerical particles, the operations for a coagulation event are similar to those of Kotalczyk and Kruis (2017). Then, the consequences of a coagulation event can be denoted as follows:

$$\text{if } w_i \ne w_j, \begin{cases} w''_i = w_i - \alpha_{ij} \min(w_i, w_j); \quad v''_i = v_i \\ w''_j = w_j - \alpha_{ij} \min(w_i, w_j); \quad v''_j = v_j \\ w''_{\text{coag}} = \alpha_{ij} \min(w_i, w_j); \quad v''_{\text{coag}} = v_i + v_j \end{cases} \tag{15}$$

$$\text{if } w_i = w_j, \begin{cases} w''_i = w_i / 2; \quad v''_i = v_i + v_j \\ w''_j = w_j / 2; \quad v''_j = v_i + v_j \end{cases} \tag{16}$$

where w'' and v'' represent the new weight and volume after the coagulation event, respectively. In Equations (15) and (16), mass is conserved during coagulation. It should be noted that the number of numerical particles remains constant when $\alpha_{ij} \equiv 1$; under this condition, the WFMC method coincides with the MMC method (Zhao et al., 2009). Although the WFMC method can assimilate certain traditional differential weight method schemes under some specific conditions, this also demonstrates that the WFMC method is a new scheme without such specific conditions. Compared with the traditional MMC method, the weight distribution in WFMC is adjustable. For other weighted particle schemes, e.g., the mass-flow algorithm (MFA) (Babovsky, 1999, Eibeck and Wagner, 2001) and weighted flow algorithm (WFA) (DeVille et al., 2011), the weight of numerical particles is not explicitly given in the WFMC method, which coincides with the MMC method.

2.4. Probabilistic Removal

For two coagulated numerical particles with different weights when $\alpha_{ij} < 1$, additional new numerical particles need to be created as a result of the coagulation event, which will lead to non-constant-number schemes (Kotalczyk and Kruis, 2017). The statistical accuracy and numerical efficiency vary as the total number of numerical particles changes.

To maintain the number of numerical particles, a new coagulation rule for the coagulation event must be constructed. A common method is the removal of one selected numerical particle partner after the coagulation event, followed by adjusting weight of the other particle partner is to ensure that the particle properties are statistically unchanged (e.g., the particle number/mass). Such a method is always adopted in Monte-Carlo simulations. For the constant-number method (Lee and Matsoukas, 2000), the additional numerical particles are randomly removed after fragmentation events to force the number of numerical particles in the simulation to remain constant. For the algorithms of stochastic weighted particle method schemes (Eibeck and Wagner, 2001, Patterson *et al.*, 2011, DeVille *et al.*, 2011), to solve the Smoluchowski coagulation equation for weighted particles, particle creation and destruction processes are also introduced with probabilities of p_{birth} and p_{death}, respectively. Therefore, a new probabilistic removal scheme is proposed and developed in the WFMC based on previous studies. The probabilistic removal scheme can not only maintain a constant number of numerical particles, but also guarantees that the statistics of the particle properties remain on average intact before and after the probabilistic removal. The probability of random removal and the weight of particles should be first determined. The schematic implementation of this probabilistic removal is shown in Figure 1.

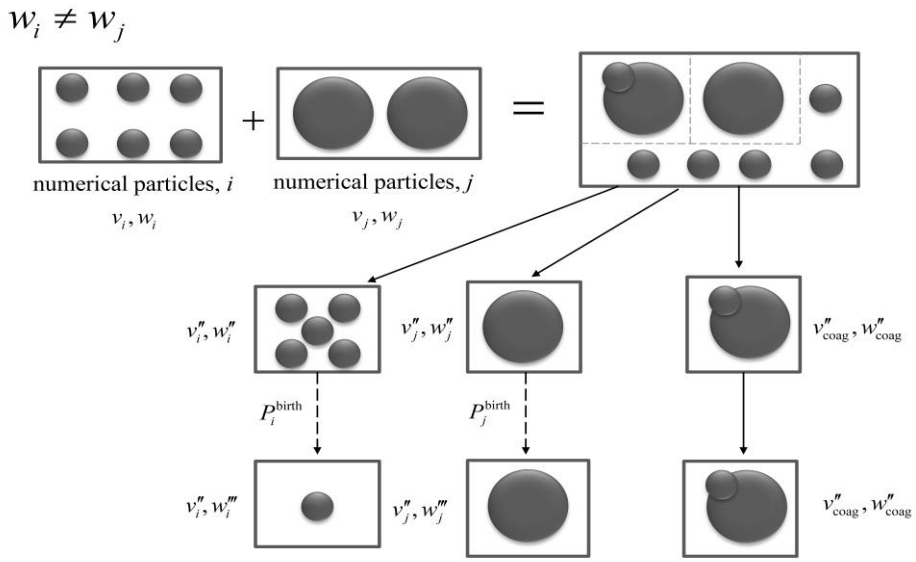


Figure 1 Treatment of a coagulation event with the probabilistic removal.

When numerical particle *i* coagulates with its coagulation pair, *j*, this means that some real particles from numerical particle *i* coagulate with those from numerical particle *j*. After a coagulation event, these particles can be divided into three groups: "coagulated" real particles and "non-coagulated" real particles from the *i*th and *j*th numerical particles. These are then subjected to probabilistic removal to ensure that the number of numerical particles remains constant. The "coagulated" particles are always added and maintain their weight in the next stage. One of the "non-coagulated" particles is removed, and the other numerical particle survives based on the probabilities of P_i^{birth} and P_j^{birth}. For example, with a probability of P_i^{birth}, the *i*th particle remains and the *j*th particle is removed; otherwise, the *j*th particle remains and the *i*th particle is removed. w''' means numerical weight after probabilistic removal, which should be adjusted simultaneously to ensure the conservation of particle properties.

To ensure that only one numerical particle will be removed, the probability of survival for numerical particles *i* and *j*, P_i^{birth} and P_j^{birth}, should satisfy the following:

$$P_i^{\text{birth}} + P_j^{\text{birth}} = 1 \tag{17}$$

To satisfy the conservation of some particle properties (e.g., particle number/mass) during the random removal, the new weight after the random removal particles, w_i''' and w_j''', should satisfy the following:

$$\begin{cases} w_i'''\chi_i = w_i''\chi_i + w_j''\chi_j \\ w_j'''\chi_j = w_i''\chi_i + w_j''\chi_j \end{cases} \tag{18}$$

where χ_i and χ_j represent the expected conserved scalar of particles *i* and *j*, respectively. For $\chi_i \equiv 1$, the number of represented real particles does not change during the random removal;

this is called the conserved number removal scheme in the present numerical model development. For $\chi_i \equiv v_i$, where v_i is the volume of numerical particle *i*, the total mass/volume of the represented real particles does not change during the random removal; this is called the conserved volume removal scheme in the present numerical model development.

Combining Equations (15) and (18), yields the following:

$$\begin{cases} w_i''' = w_i - \alpha_{ij}\min(w_i, w_j) + [w_j - \alpha_{ij}\min(w_i, w_j)]\dfrac{\chi_j}{\chi_i} \\ w_j''' = w_j - \alpha_{ij}\min(w_i, w_j) + [w_i - \alpha_{ij}\min(w_i, w_j)]\dfrac{\chi_i}{\chi_j} \end{cases} \tag{19}$$

In order to maintain the arbitrary particle properties, Γ_i, they are statistically unchanged before and after the random removal. The expectation of these particle properties after random removal must be equal to those before random removal; therefore, P_i^{birth} and P_j^{birth} should also satisfy the following:

$$\Gamma_i w_i''' P_i^{\text{birth}} + \Gamma_j w_j''' P_j^{\text{birth}} = \Gamma_i w_i'' + \Gamma_j w_j'' \tag{20}$$

By combining Equations (15), (17), (19), and (20), the probabilities of coagulation particles, *i* and *j* remaining, P_i^{birth} and P_j^{birth}, can then be obtained as follows:

$$\begin{cases} P_i^{\text{birth}} = \dfrac{w_i - \alpha_{ij}\min(w_i, w_j)}{w_i - \alpha_{ij}\min(w_i, w_j) + [w_j - \alpha_{ij}\min(w_i, w_j)]\dfrac{\chi_j}{\chi_i}} \\ P_j^{\text{birth}} = \dfrac{w_j - \alpha_{ij}\min(w_i, w_j)}{w_j - \alpha_{ij}\min(w_i, w_j) + [w_i - \alpha_{ij}\min(w_i, w_j)]\dfrac{\chi_i}{\chi_j}} \end{cases} \tag{21}$$

The above method is called probabilistic removal. After implementing probabilistic removal, the constant number scheme remained without loss of its inherent computational accuracy.

The values of the total coagulation rate, C_0', for all numerical particles must be re-evaluated after a coagulation event because the size and weight of the coagulation particle pair change during the coagulation event. In the present study, the smart bookkeeping method (Kruis et al., 2000) is used to evaluate the value of C_0' in order to avoid a large number of recalculations of the coagulation rates for particles not participating in coagulation events. The details of the newly proposed WFMC method are summarized in Numerical Algorithm I.

Numerical Algorithm I: Weighted fraction Monte-Carlo (WFMC) method for particle coagulation

Step 1. Initialize time $t = 0$. Set the initial particle size matrix, $X_1, X_2, \ldots, X_N$ of N particles.

Step 2. Calculate the fraction function, α_{ij}, and coalescence kernels, C_{ij}', for $N(N-1)/2$ as unique particle pairs.

Step 3. Specify the stopping time, t_{stop}, and the time step, Δt.

Step 4. Randomly choose r from a uniform particle distribution in [0, 1]. If $r \le P'_{coag}(\Delta t) = 1 - \exp(-\Delta t C_0')$, then go to Step 5; else, go to Step 9.

Step 5. Generate a random numerical particle pair, i, j, and random number, r_1, from a uniform distribution in [0, 1] using the Monte Carlo method.

Step 6. If $r_1 \le \beta'_{ij} / \max_{\forall k, \forall m}(\beta'_{km})$, go to Step 7; else, return to Step 5.

Step 7. Generate a random number, r_2, from a uniform distribution in [0, 1]. If $r_2 \le P_i^{\text{birth}}$, particle X_j is removed and the weight of w_i''' is assigned to the particle X_i; else, the particle X_i is removed and the weight of w_j''' is assigned to particle X_j.

Step 8. Add a new particle with size $X_i + X_j$ and weight $\alpha_{ij} \min(w_i, w_j)$.

Step 9. Advance the time, t, by time step, Δt.

Step 10. If $t > t_{stop}$, the calculation is terminated; else, go to Step 11.

Step 11. Update the coagulation rate, C_{ij}', for new particle pairs.

Step 12. Return to Step 4.

2.5. Choice of Fraction Functions

The general algorithm for the WFMC method with an arbitrary fraction function, α, is described in Numerical Algorithm I. However, from the expression of probabilistic removal, if there is a large size discrepancy between particle pairs i and j, fluctuations in the total number of real particles (for the conserved volume removal scheme) or total mass of real particles (for the conserved number removal scheme) will be introduced, resulting in additional stochastic error. To minimize this type of stochastic error, an appropriate fraction function must be derived. For the probabilistic removal of numerical particle pairs with large size discrepancies, the fraction function should be close to or equal to 1; thus, the stochasticity of the probabilistic removal can be ignored. This can be formulated as follows:

$$\lim_{\max(v_i, v_j)/\min(v_i, v_j) \to \infty} \alpha_{ij} = 1 \tag{22}$$

where v_i is the volume of numerical particle i. For numerical simulations in Section 3, the fraction functions are focused on three forms in Equations (23) - (25).

$$\alpha_{ij} = \begin{cases} C, & \text{if} \quad \max(v_i, v_j) / \min(v_i, v_j) \le p_{\text{critical}} \\ 1, & \text{if} \quad \max(v_i, v_j) / \min(v_i, v_j) > p_{\text{critical}} \end{cases} \tag{23}$$

where $C < 1$ is the fraction constant and $p_{\text{critical}} > 1$ is the critical ratio. The form of the function in Equation (23) is called the stepwise constant fraction function. The other two forms are also introduced as follows:

$$\alpha_{ij} = \frac{1}{1 + \min(v_i, v_j) / \max(v_i, v_j)} \tag{24}$$

$$\alpha_{ij} = 1 - 2^{-\max(v_i, v_j)/\min(v_i, v_j)} \tag{25}$$

The hyperbolic fraction function (HFF) is given in Equation (24), and the exponential fraction function (EFF) is given in Equation (25).

2.6. Connection to the Mass-Flow Algorithm

The mass-flow algorithm (MFA) was proposed by Babovsky (1999) and Eibeck and Wagner (2001). In the MFA, a numerical particle of size v_i represents a number, $1/v_i$, of real particles of the same size, where the weight function $w(v_i)$ is $1/v_i$. The MFA algorithm is given as follows:

The coagulation pair, i and j, is chosen with the coagulation rate:

$$\beta_{ij}^{\text{MFA}} = \beta_{ij}\left(\frac{1}{v_i} + \frac{1}{v_j}\right) \tag{26}$$

The algorithm always adds particle $v_i + v_j$ to the population, and then one of v_i or v_j is removed. With probability $v_i/(v_i + v_j)$, particle v_i is removed; otherwise, particle v_j is removed.

To compare the MFA and WFMC algorithms, the HFF is chosen, with the conserved volume removal scheme and an initial weight of $w(v_i)=1/v_i$. In Equations (19) and (21), and the coagulation kernel is then expressed as follows:

$$\beta_{ij}^{\text{WFMC}} = \beta_{ij}\left(\frac{1}{v_i} + \frac{1}{v_j}\right) \tag{27}$$

$$\begin{cases} w_i^{\text{WFMC}} = \dfrac{1}{v_i}; \quad v_i^{\text{WFMC}} = v_i \\ w_j^{\text{WFMC}} = \dfrac{1}{v_j}; \quad v_j^{\text{WFMC}} = v_j \\ w_{\text{coag}}^{\text{WFMC}} = \dfrac{1}{v_i + v_j}; \quad v_{\text{coag}}^{\text{WFMC}} = v_i + v_j \end{cases} \tag{28}$$

$$\begin{cases} P_i^{\text{birth}} = \dfrac{v_j}{v_i + v_j} \\ P_j^{\text{birth}} = \dfrac{v_i}{v_i + v_j} \end{cases} \tag{29}$$

where Equations (27) to (29) in the WFMC method exactly follow the coagulation rule in the MFA method. Therefore, the WFMC method can assimilate the MFA method only if all of the above conditions are satisfied. It should be noted that the initial particle weight is not forced to satisfy $w(v_i) = 1/v_i$ in the present numerical simulation case. Hence, the HFF method used in the present study is a newly proposed scheme compared with the MFA method.

3. Numerical Simulations

Different orders of moments and PSDs are considered as references for evaluating the numerical simulation results. The kth order moment is defined as:

$$M_k = \int_0^\infty v^k n(v) dv \tag{30}$$

Low-order moments typically have some physical meaning, e.g., M_0 is the total particle number density and M_1 is the total particle mass/volume faction.

3.1. Case 1: Constant Coagulation Kernel and Initial Monodispersed Population

3.1.1. Numerical Validation

A case with a constant coagulation kernel and initial monodispersed population is considered in which the analytical solution is well known (Friendlander, 2000). Numerical results of the DSMC (Gillespie, 1975), MMC (Zhao et al., 2009) and newly developed WFMC methods for different fraction functions, α are compared with the analytical solutions. For the initial conditions of this numerical case, the initial particle size distribution is monodispersed as follows:

$$n(u,0) = \begin{cases} 10^6, & \text{if} \quad u = 1 \\ 0, & \text{if} \quad u > 1 \end{cases} \tag{31}$$

The constant coagulation kernel is expressed as:

$$\beta(i,j) = 10^{-5}, \qquad i, j = 1,2,3,..., \tag{32}$$

where the exact solutions of the PSD and kth moment are available in Friedlander (2000).

Figure 2 shows the four orders of moments (M_0, M_1, M_2, M_3) simulated by different Monte Carlo method schemes and a comparison to the analytical solutions. In the present study, 2000 numerical particles are used in all of the numerical simulations with 400 repetitions because this provided highly accurate numerical results and stable variances, respectively. Other

numerical results are simulated using the newly proposed WFMC method for different fraction functions, α (i.e., stepwise constant fraction functions, $C = 0.5$, 0.7, and 0.9 with $p_{\text{critical}} = 2.0$, HFF and EFF). In the present study, two different conservation schemes are defined and studied in Section 2.4. The conserved number removal scheme, CN is used, otherwise the conserved volume removal scheme, CV is chosen.

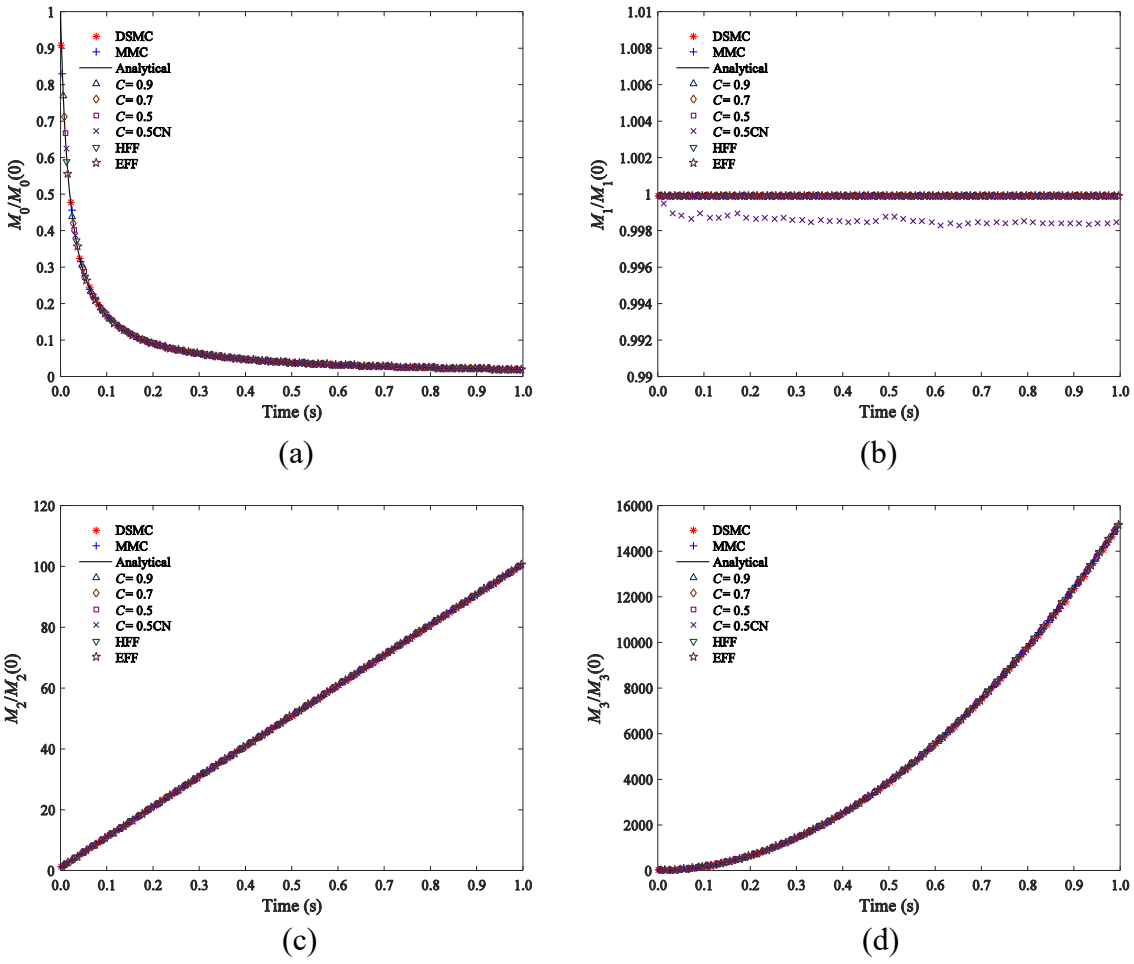


Figure 2 Comparison of four orders of moments obtained with different Monte Carlo methods (DSMC, MMC and WFMC) and the analytical solutions.

Figure 2 shows an excellent agreement between the analytical solutions and numerical results for the four orders of moments obtained with different Monte Carlo methods (DSMC, MMC and the newly proposed WFMC methods) with stepwise constant fraction functions, C, and hyperbolic and exponential fraction functions (HFF and EFF). However, it should be noted that the conserved number removal scheme (i.e., 0.5CN) may result in an extremely small deviation within a relative error of 0.2% in the total particle volume (M_1), as shown in Figure 2(b).

3.1.2. Stochastic Error of the Moments

The relative standard deviation (RSD) is used to quantify the stochastic error as follows:

$$\mathrm{RSD} = \frac{\sqrt{\dfrac{1}{P-1}\sum_{i=1}^{P}(M_{ki} - \bar{M}_k)^2}}{\bar{M}_k} \tag{33}$$

In Figure 3(a), the RSDs of M_0 obtained with the DSMC, MMC and newly developed WFMC method with a stepwise constant fraction function exhibit similar trends and lower RSDs compared with the WFMC method with the HFF and EFF. However, the RSDs of M_1 are maintained at zero for all of the MC method schemes except for the WFMC method with the conserved number removal scheme (0.5CN), as shown in Figure 3(b). Generally speaking, all of the MC method schemes can keep the perfect mass balance expect for the newly developed WFMC method with the conserved volume removal scheme.

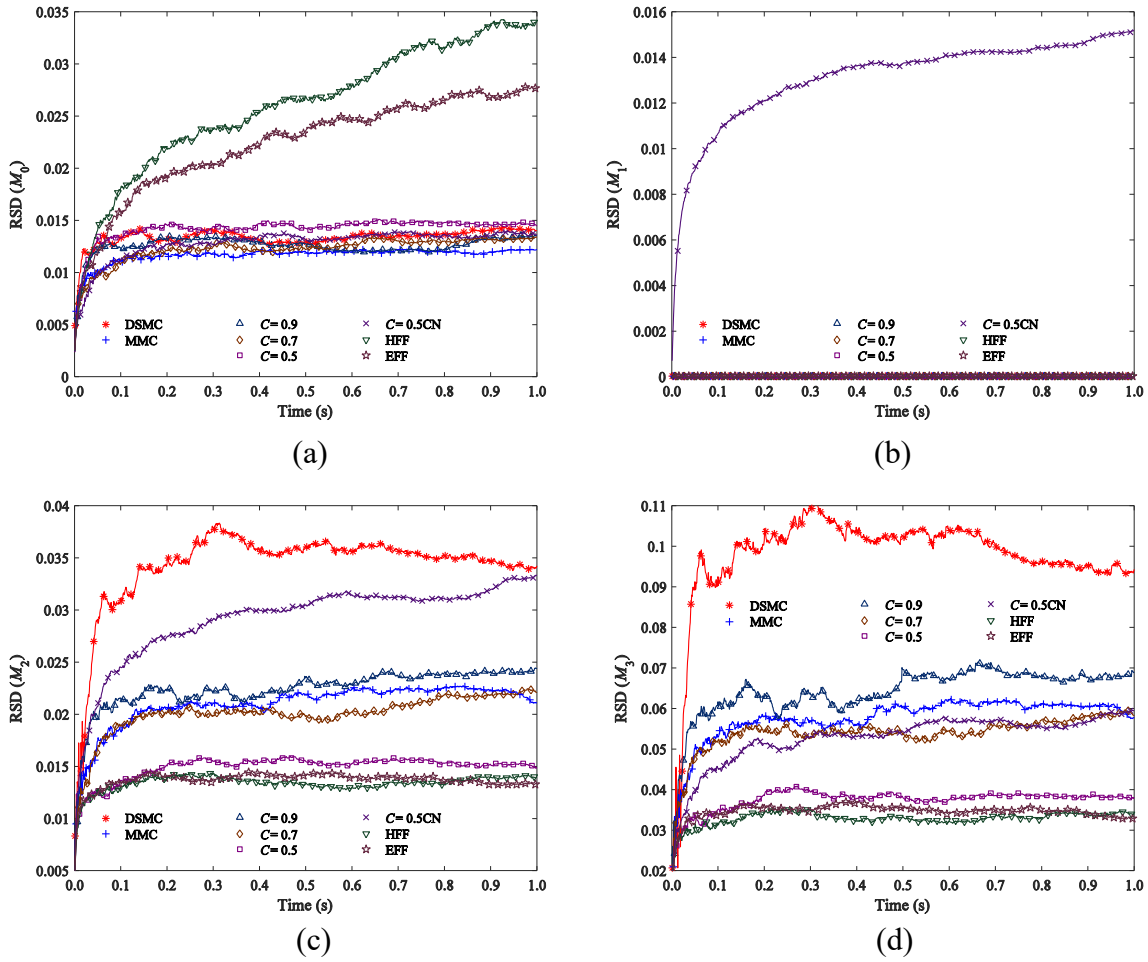
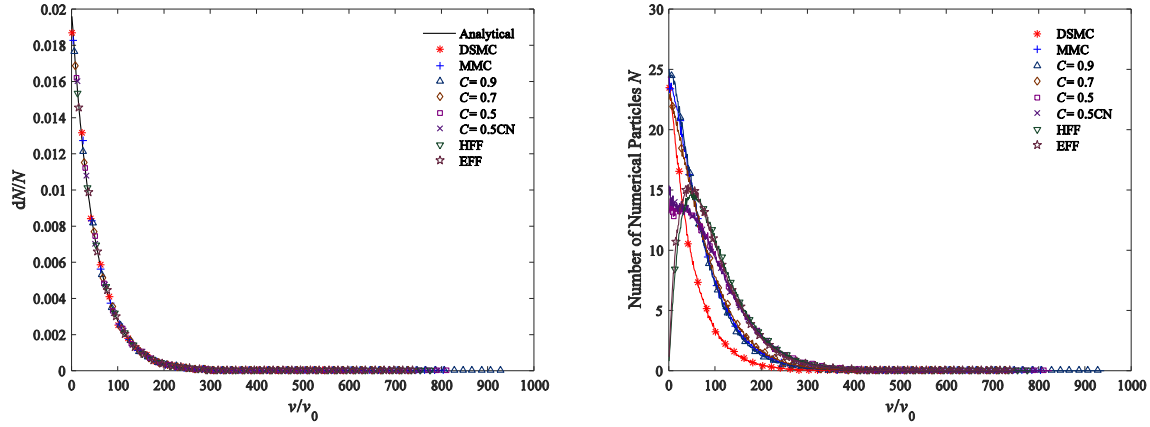


Figure 3 Comparison of RSDs for four orders of moments obtained with different MC method schemes.

For the RSDs of the M_2 and M_3 moments in Figures 3(c) and (d), the DSMC method has the highest RSD. The WFMC method shows a great capability for reduction of the stochastic error, especially with the HFF, EFF and stepwise constant fraction function with C = 0.5. In

addition, lower RSDs for higher-order moments (i.e., M_2 and M_3) are observed for the stepwise constant fraction function with a decrease in the fraction constant, C.

Figures 4(a) shows the PSDs at the end of the simulation time, t = 1 s, where the PSD is fully developed. An excellent agreement between the numerical results and analytical solutions is obtained. The results of different MC method schemes are found to have almost the same trends and values.



(a) (b)

Figure 4 Comparison of PSDs from the analytical solutions and the number of numerical particles in each particle size interval for different MC method schemes at t = 1.0 s.

Figure 4(b) shows the number of numerical particles used in the particle size interval for different MC method schemes at t = 1.0 s. The DSMC method has the narrowest numerical PSD; therefore, only a few (or no) numerical particles are used to represent the large real particles, which results in large fluctuations in the higher-order moments (i.e., M_2 and M_3). The numerical PSD of the MMC method is slightly wider than that of the DSMC method, and more large numerical particles are used to represent the real large particles. On the other hand, the fraction functions are introduced to 'twist' this numerical PSD in the WFMC method. It can be observed that although there are fewer numerical particles used to represent the small real particles, a greater number of large numerical particles is taken place. These results become clearer with a decrease in the stepwise constant fraction function, C, from 0.9 to 0.5.

Figure 5 shows the average particle weight in each size interval, $\bar{w}$, for different MC method schemes at t = 1.0 s. The average particle weight is defined as:

$$\bar{w}(v) = \frac{\sum_{i=1}^{N} w(v_i)\delta(v_i, v)}{\sum_{i=1}^{N} \delta(v_i, v)} \tag{34}$$

where δ is the Dirac delta function. The DSMC method has a constant average particle weight for different particle sizes because all of the numerical particles in the DSMC method have an equal particle weight. The average particle weight decreases slightly with increasing particle size in the MMC method. Different stepwise constant fraction functions in the WFMC method

can lead to different weight functions, which implies that fraction functions are adopted to adjust the weight functions; this is one of the most important characteristics of the newly proposed WFMC method. For the stepwise constant fraction function in the WFMC method, a similar trend is found to that obtained with the MMC method, while the WFMC method has a higher weight for small particles and lower weight for large particles. Another interesting finding is that the weight functions of the WFMC method with both the HFF and EFF are nearly linear in logarithmic coordinates but have different slopes. This linearity indicates that these types of weight functions are power-law weight functions that are similar to the weight function in the WFA (DeVille et al., 2011).

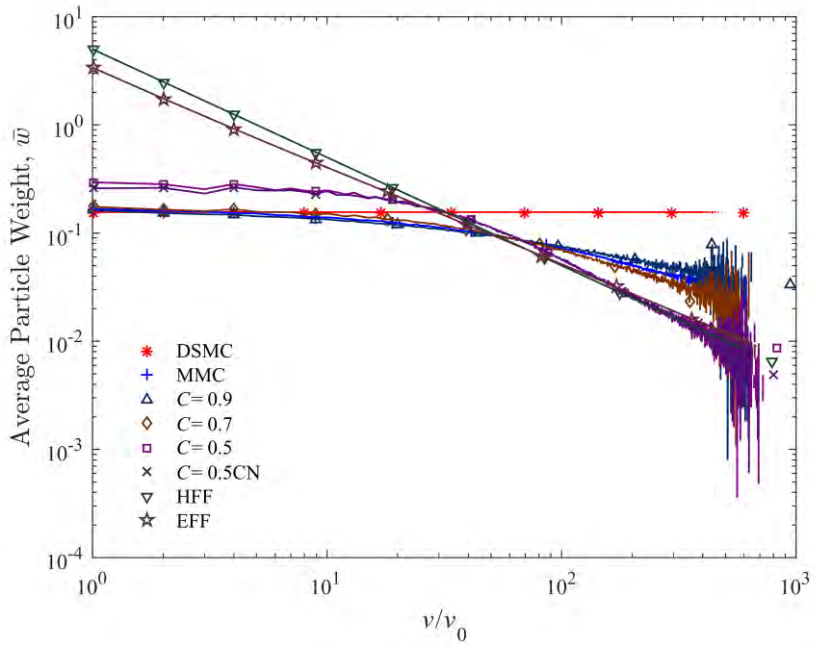


Figure 5 Average particle weight distributions for different MC schemes.

3.2. Case 2: Free Molecular Coagulation Kernel and Initial Monodispersed Population

Free molecular coagulation kernel has been widely used in aerosol science and technology for particles in the free molecular regime. However, there is no analytical solution for this coagulation kernel; thus, the numerical simulation results of the sectional method (SM) are used as a reference (Prakash et al., 2003). In the present study, different Monte Carlo method schemes are compared with the SM results. To simplify, only the HFF in the WFMC method is used. The initial particle size distribution is monodispersed with a typical initial total number density of 10^{17} #/m^3 and initial particle size of 1.2407 nm.

The coagulation kernel of particles i and j in the free molecular regime is given by the following (Friendlander, 2000):

$$\beta_{ij}=\left(\frac{3}{4\pi}\right)^{\frac{1}{6}}\left(\frac{6k_bT}{\rho_p}\right)^{\frac{1}{2}}\left(\frac{1}{v_i}+\frac{1}{v_j}\right)^{\frac{1}{2}}\left(v_i^{\frac{1}{3}}+v_j^{\frac{1}{3}}\right)^2 \tag{35}$$

where T is the local temperature, which is set to be 300K in the present study; v_i is the volume of the ith particle; and ρ_p is the particle density, which is defined as 1800 kg/m^3. In the present study, all of the numerical simulations used 2000 numerical particles, and the number of simulation repetitions is 400.

Figure 6 shows an excellent agreement for four orders of moments in the free molecular coagulation kernel case between the different MC method schemes and SM results. All of the MC method schemes show a perfect mass balance.

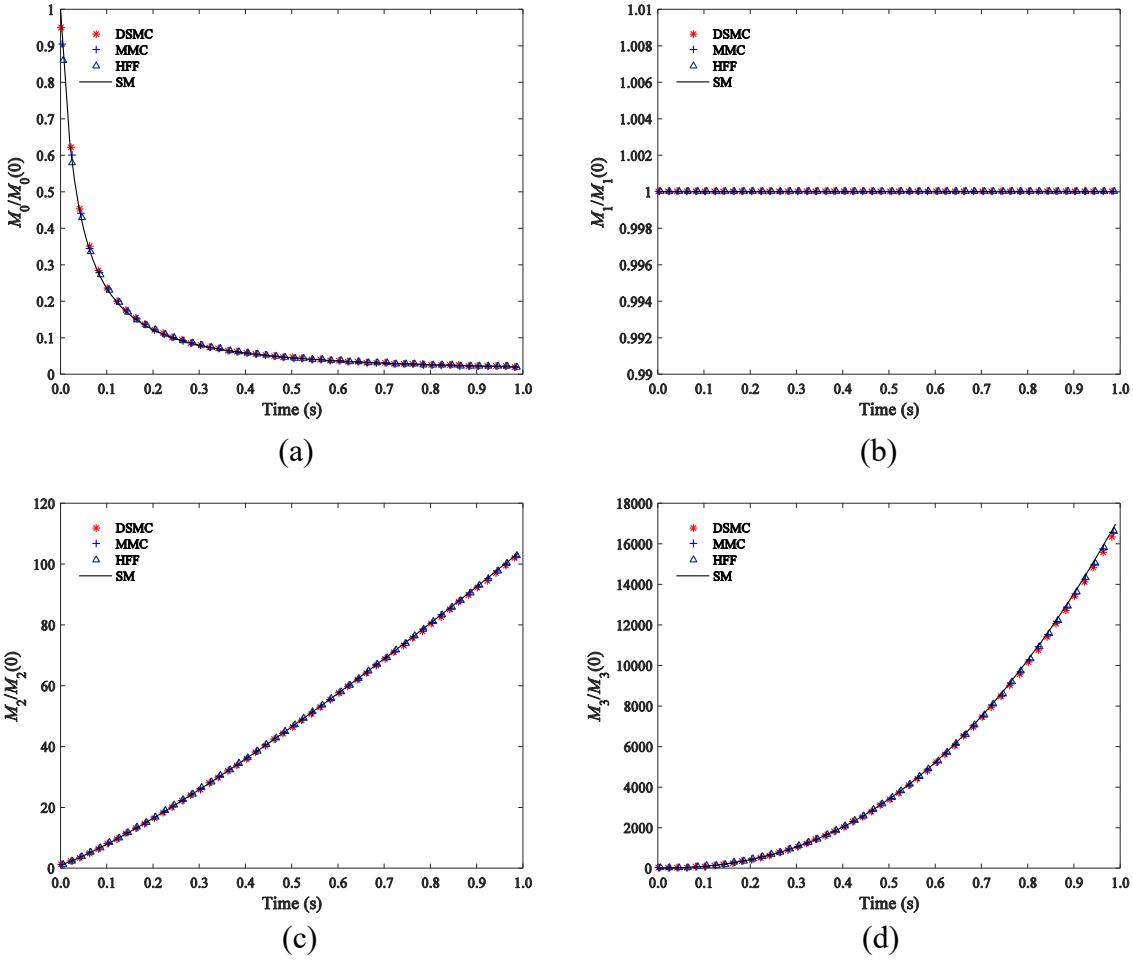


Figure 6 Comparison of four orders of moments with the free molecular coagulation kernel obtained with different MC method schemes and the sectional method.

In Figure 7, there are similar RSD trends for each moment, M_i, for a free molecular coagulation kernel obtained with different MC method schemes. Moreover, it is worth noting that the proposed WFMC method with HFF has a higher RSD for M_0, but it has lower RSDs for higher-order moments (i.e., M_2, M_3) compared with the DSMC and MMC methods. The proposed WFMC method shows a significant reduction in stochastic error for higher-order moments.

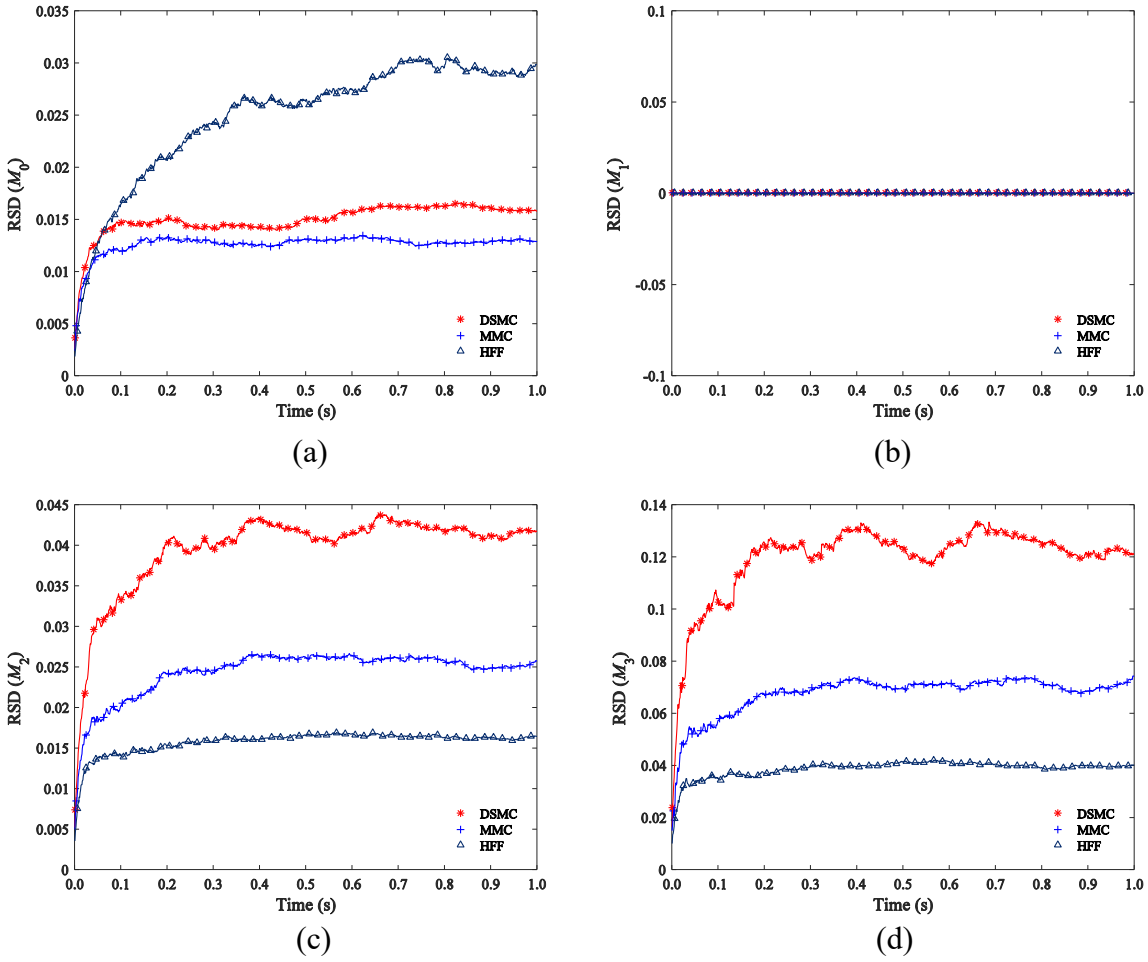


Figure 7 Comparison of RSDs for four orders of moments with a free molecular coagulation kernel under different MC method schemes.

3.3. Case 3: Free Molecular Coagulation Kernel and Initial Exponential Distributed Particle Population

In Case 3, a free molecular coagulation kernel is adopted and the initial size distribution is represented by an exponential function as follows (Zhao *et al*., 2005):

$$n(v,0) = [N_0 \exp(-v / v_{g0})] / v_{g0} \tag{36}$$

where v_{g0} is the initial mean volume, which is defined as $v_{g0}=1\times10^{-22}\mathrm{m}^3$ (Liu and Chan, 2018b) in the present study. The numerical results of the DSMC, MMC, and WFMC methods are shown in Figure 8.

Figure 8 shows an excellent agreement in the four orders of moments with the free molecular coagulation kernel and initial exponential distribution between the different MC method schemes and SM results.

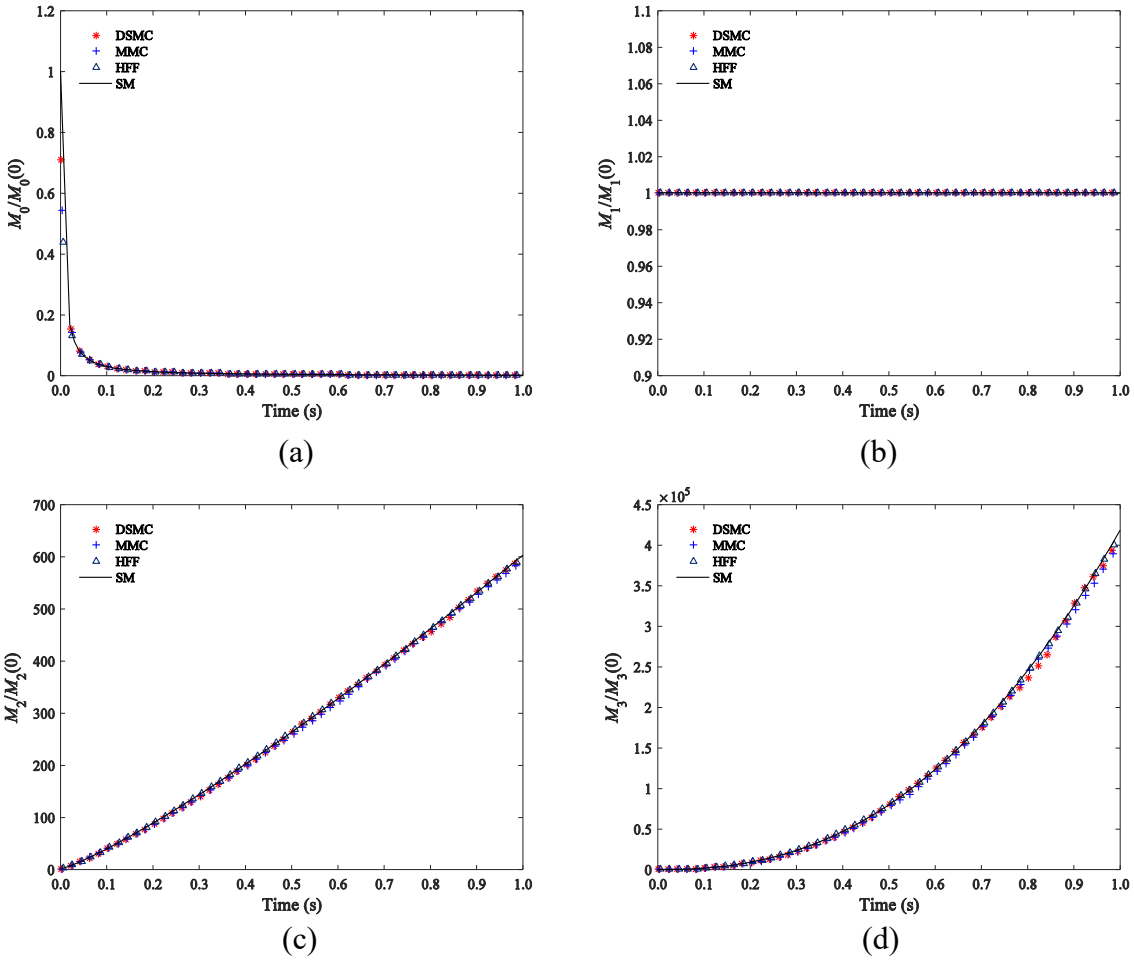


Figure 8 Comparison of four orders of moments with a free molecular coagulation kernel and initial exponential distribution.

Figure 9 shows the PSDs at different simulation times: t = 0.1 s, t = 0.5 s, and t = 1.0 s (t = 0 s is the initial exponential distribution). An excellent agreement between the MC methods and SM results is obtained. The results of different MC method schemes are also found to have excellent agreement. As the simulation time advances, the PSDs shift to a larger-size regime due to the occurrence of coagulation events. Figure 9 also shows that the widest PSD can be found in the WFMC results compared with the DSMC and MMC results, especially for the larger-size regime, which will have a higher contribution to higher-order moments. Therefore, this suggests that the WFMC method can predict the larger particle size distribution more accurately as well as the higher-order moments.

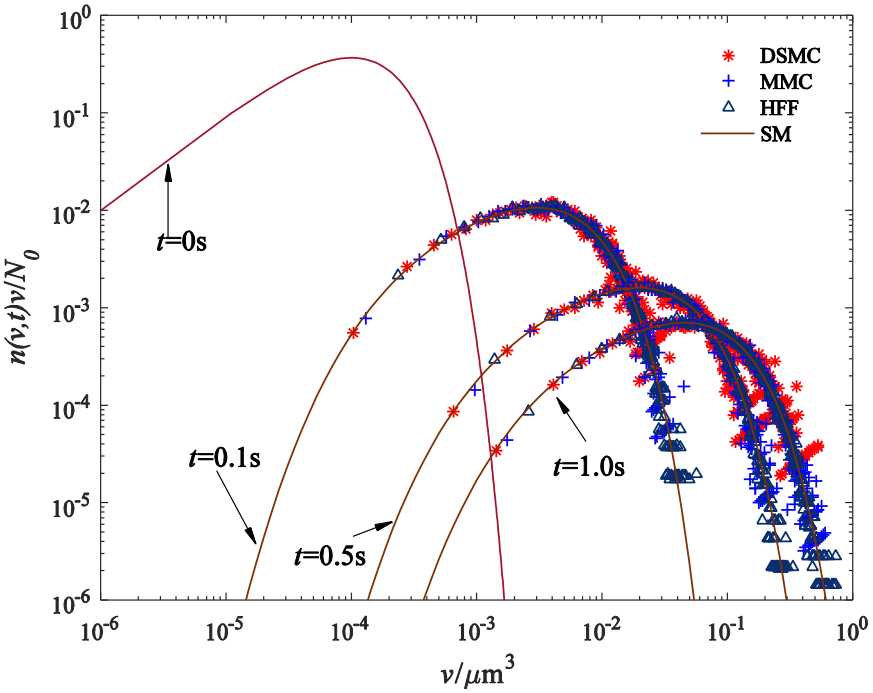


Figure 9 Particle size distributions obtained with different MC method schemes and the sectional method.

3.4. Computational Efficiency Analysis

To evaluate the computational efficiency of the WFMC method, the normalized computational time, τ, is defined as follows:

$$\tau = t / t_{\text{ref}} \tag{37}$$

where t_{ref} is the reference time, and t is the amount of computational time required for the corresponding DSMC, MMC, and WFMC methods. In the present study, the reference value is the computational time for the constant coagulation case (Case 1) obtained with the DSMC method. The normalized computational times, τ, for the studied cases are listed in Table 1.

Table 1 Normalized computational times, τ, obtained for different cases using the DSMC, MMC and WFMC methods.

Parameter	DSMC	MMC	*C*= 0.5CN	*C*= 0.5	*C*= 0.7	*C*= 0.9	EFF	HFF
Case 1	1.0	1.4	1.5	1.8	1.5	1.5	2.8	1.6
Case 2	4.3	7.4	-	-	-	-	-	8.1
Case 3	4.1	7.8	-	-	-	-	-	8.3

Table 1 indicates that the DSMC method generates the lowest values of τ compared with the other MC methods. This is because although the same initial number of numerical particles is used for these MC methods, the number of numerical particles will continuously decrease as time advances owing to the coagulation events, resulting in lower computational cost. However, the computational accuracy will also decrease as the number of total numerical particles decreases.

In Table 1, the numerical simulation time of the WFMC method is slightly higher than that of the MMC method, which is attributed to two factors. On one hand although both the WFMC and MMC methods can maintain a constant number of numerical particles, the WFMC method must deal with a more complex coagulation kernel compared with the MMC method (i.e., the kernel function is $\max(w_i, w_j)\beta_{ij}/\alpha_{ij}$ in the WFMC method and $\max(w_i, w_j)\beta_{ij}$ in the MMC method). Hence, a longer CPU time is required to calculate the coagulation kernel in the WFMC method. On the other hand, the coagulation kernel function in the WFMC method is always larger than that in the MMC method (for example, $\alpha_{ij} \le 1$). The larger total coagulation rate, C_0, in the WFMC method leads to a shorter coagulation waiting time for event-driven schemes, as shown in Equation 11, or more coagulation events for time-driven schemes, as shown in Equation 13, thus resulting in higher computational time. This can also explain the varying calculation times for different types of fraction functions in the WFMC method.

4. Conclusions

A new weighted fraction Monte Carlo (WFMC) method is well developed and validated in the present study. In the WFMC method, a new coagulation particle rule is derived that allows changes in the mean number of real coagulation events between two numerical particles. Then, to maintain a constant number of numerical particles, the probabilistic removal method is also proposed. The WFMC method can reduce stochastic error by adjusting the weight function to 'twist' the numerical particle size distribution (PSD) and change the number of numerical particles that represent real particles in each size interval. The method for particle coagulation is validated through three classical cases. An excellent agreement between the numerical simulation results of the WFMC and analytical solutions/sectional method results is obtained. The WFMC method shows a significant reduction in stochastic error for higher-order moments with a slightly higher computational cost because there are more numerical particles used to represent real physical particles in larger-size regime**.** In addition, the proposed WFMC method also shows the significant advantage of tracking the PSD over the larger size regime, which is traditionally insufficient in the classical DSMC and MMC methods.

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