

# Monte Carlo solution of wet removal of aerosols by precipitation

Haibo Zhao\*, Chuguang Zheng

*State Key Laboratory of Coal Combustion, Huazhong University of Science and Technology, Wuhan, 430074 Hubei, PR China*

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## Abstract

The time evolution of aerosol size distribution (ASD) during precipitation describes quantitatively aerosols wet scavenging process. Scavenging coefficient, which takes account of the three most important wet removal mechanisms: Brownian diffusion, interception and inertial impaction, is used to parameterize wet scavenging process. A new multi-Monte Carlo method (MMC) is promoted to solve general dynamic equation for wet removal of aerosols. Two special cases in which analytical solutions exist are adopted to validate computation precision of MMC method. Furthermore, the influence of precipitation type on aerosols wet scavenging process is investigated by numerical simulation of the method. The results show that for lognormal raindrop size distribution and lognormal ASD (1) the increase of rainfall intensity (from light precipitation to moderate precipitation and then to heavy precipitation) can help scavenge aerosols with any size; (2) any precipitation type scavenges large aerosols ( $>2\ \mu\text{m}$ ) more effectively than small aerosols ( $<0.01\ \mu\text{m}$ ) and intermediate aerosols ( $>0.01\ \mu\text{m}$  and  $<2\ \mu\text{m}$ ) (in that order); (3) the three precipitation types have a weak effect of wet scavenging on intermediate aerosols.

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*Keywords:* Scavenging coefficient; Collision efficiency; Brownian diffusion; Interception; Inertial impaction

## 1. Introduction

The wet removal of aerosol suspended in the atmosphere by precipitation plays an outstanding role on air quality. Aerosols wet scavenging by precipitation is usually named as below-cloud scavenging. The falling raindrops collide with aerosols and collect them. The three most important wet removal mechanisms are as follows: Brownian diffusion, interception and inertial impaction. The

aerosol wet removal mechanisms are very complicated because wet scavenging process is influenced by some kinds of external factors (for example, aerosol size distribution (ASD), raindrop size distribution (RSD), water content, rainfall intensity, even environmental temperature, physical and chemical properties of aerosol and raindrop) and internal factors (for example, aerosol–raindrop collision domain). The introduction of scavenging coefficient is a simple, intuitionistic method and is widely used to parameterize wet scavenging process (Mircea and Stefan, 1998; Mircea et al., 2000; Chate and Pranesha, 2004; Loosmore and Cederwall, 2004). Scavenging coefficient depends

\*Corresponding author. Tel.: +86 27 8754 4779;  
fax: +86 27 8754 5526.

E-mail address: [klinmannzhb@163.com](mailto:klinmannzhb@163.com) (H. Zhao).

Nomenclature		Greek letters	
$C_c$	the Cunningham slip correction factor	$\alpha$	multiplicative constant
$d_p$	the diameter of aerosol, $\mu\text{m}$	$\lambda$	the mean free path length of the gas, m
$d_{pg}$	the geometric mean diameter of aerosol, $\mu\text{m}$	$\mu_w, \mu_a$	the viscosity of raindrop and air, respectively, $\text{kg m}^{-1} \text{s}^{-1}$
$D_d$	the diameter of raindrop, mm	$\rho_w, \rho_a, \rho_p$	the density of raindrop, air and aerosol, respectively, $\text{kg m}^{-3}$
$D_{dg}$	the geometric mean diameter of raindrop, mm	$\sigma_{pg}$	the geometric standard deviation based on the aerosol diameter
$D_{diff}$	the diffusion coefficient of aerosol	$\sigma_{dg}$	the geometric standard deviation based on the raindrop diameter
$E(d_p, D_d)$	collision efficiency	$\tau_p$	the relaxation time of aerosol, s
$J$	rainfall intensity, $\text{mm h}^{-1}$	$\tau_{sca}$	the characteristic time scale of wet removal, s
$K(d_p, D_d)$	collision kernel, $\text{m}^3 \text{s}^{-1}$	$A(d_p)$	the scavenging coefficient, $\text{s}^{-1}$
$k_b$	Boltzmann constant, $1.38054 \times 10^{-23} \text{JK}^{-1}$	<i>Subscripts</i>	
kwt	Transform-weight of fictitious particle	0	refers to initial condition
$n_d(D_d, t)$	raindrop size distribution function, $\text{mm}^{-1} \text{m}^{-3}$	d,w	refers to raindrops
$N_d$	total raindrop number concentration, $\text{m}^{-3}$	p	refers to aerosols
$n_p(d_p, t)$	aerosol size distribution function, $\mu\text{m}^{-1} \text{m}^{-3}$	a	refers to air
$N_p$	total aerosol number concentration, $\text{m}^{-3}$	f	refers to fictitious particles
$Pe$	the Peclet number	sca	refers to scavenging process
$Re$	the Reynolds number of a raindrop based on its radius	$i, j, k, m$	refer to indices of fictitious aerosols or fictitious raindrops
$S^*$	a dimensionless parameter	min	refers to minimum value
$Sc$	the Schmidt number of aerosol	max	refers to maximum value
$St$	the Stokes number of aerosol		
$T$	the absolute temperature of the air, K		
$U$	the falling velocity, $\text{m s}^{-1}$		
$v_{pg}$	the geometric mean volume of aerosol, $\mu\text{m}^3$		
$w_1$	water content, $\text{g m}^{-3}$		

mathematically on collision kernel, collision efficiency and RSD. Mircea and Stefan (1998) summarized the derived power-style relationships for the scavenging coefficients and rainfall intensity for different precipitation types, i.e.,  $A = aEJ^b$ , where  $A$  is the scavenging coefficient with dimension  $\text{h}^{-1}$  in the formula; collision efficiency,  $E$ , is assumed as an unknown constant in the formula;  $J$  is rainfall intensity with dimension  $\text{mm h}^{-1}$ ;  $a$  and  $b$  are two empirical coefficients which depends on local real ASD and precipitation type. In fact the study of Jung et al. (2003) shows that constant collision efficiency leads a significant error in estimating ASD by precipitation. Mircea et al. (2000) also derived the linear relationships between the polydisperse scavenging coefficients and rainfall intensity for different aerosol types as following,  $A = a + bJ$ ,

where two empirical coefficients,  $a$  and  $b$ , are also determined by different aerosol and precipitation types. Whereas, those semi-empirical relationships cannot used to obtain the detailed information about time evolution of ASD because those relationships are only associated with the rainfall intensity and then there is no any information about ASD.

Only the microphysical parameterization of scavenging coefficient is insufficient to describe the process of aerosols wet scavenging by precipitation. In fact, the information about the time evolution of ASD is of fundamental interest in atmospheric physics and chemistry, because many important properties of aerosols (such as light scattering, electrostatic charging, toxicity, radioactivity, hygroscopy, etc.) depend on their size distribution. On the

other side, properties of aerosols are also concerned with its wet scavenging. For example, the hygroscopic degree of aerosols, which is an important external factor of aerosols wet scavenging, is associated with their size distribution. Therefore, the information about the time evolution of ASD when raindrop falling can help understand in details the process of aerosols wet scavenging. ASD with time is described by size function,  $n_p(d_p, t)$ , so that  $n_p(d_p, t)dd_p$  is the number concentration of aerosol particles whose size range is between  $d_p$  and  $d_p + dd_p$  per volume unit at time  $t$ . The dimension of  $n_p(d_p, t)$  is  $\mu\text{m}^{-1}\text{m}^{-3}$ . The familiar general dynamic equation (GDE) describes mathematically the time-dependent size function by wet removal as following:

$$\frac{\partial n_p(d_p, t)}{\partial t} = -A(d_p)n_p(d_p, t) \quad (1)$$

in which  $A(d_p)$ , the scavenging coefficient or the wet deposition kernel, expresses the removal rate of the aerosol number concentration,  $\text{s}^{-1}$ .

The detailed information of the time-dependent ASD can be obtained quantitatively by the solutions of GDE for wet removal, which is a typical partially integro-differential equation. Exponential-style analytical solution of Eq. (1) could be obtained (Jung et al., 2002), if ASD and RSD is monodisperse or is represented by special functions such as exponential or lognormal or gamma function, meanwhile scavenging coefficient is expressed as the simple function of aerosol diameter  $d_p$  (for example,  $A(d_p) = ad_p^c + bd_p^d + \dots$ , which is limited in some special cases). However, as far as complicated polydisperse ASD and RSD or complicated model of scavenging coefficient are considered, not only analytical solutions do not exist, but also normal numerical methods, for example, finite volume method and finite difference method, take hardly account of Eq. (1) or generate amounts of numerical errors.

Jung et al. (2003) utilized the moment method to take account of Eq. (1) for the entire ambient aerosol size range. Moment method is computationally efficient and is used to investigate the time evolution of ASD. However, moment method for the simulation of aerosols wet scavenging by precipitation has those following limitations or disadvantages: (a) ASD and RSD must be represented by some special function, says, lognormal, Gamma function, etc.; (b) collision efficiency must

be expressed as a function of the power of aerosol size  $d_p$ ; (c) moment method assumes ASD is “self-preserving” during the evolution of time; (d) those moment equations are very complicated and are difficult to program even using the familiar Runge–Kutta fourth-order method. The above disadvantages constrain mostly its application.

The difficulty for the description of aerosols wet scavenging lies in that the process of aerosols wet scavenging is related to not only ASD but also RSD. Usually RSD could not be expressed as special function style, so the numerical definite integral over RSD (seeing Eq. (2)) will cost amounts of CPU time or generate some numerical errors. Factually, both ASD and RSD are nature-dispersed, and discrete nature of Monte Carlo (MC) method adopts itself naturally to discrete ASD and RSD and those involved discrete wet removal event. In a MC simulation, one uses a finite sample of the population and follows its evolution under the action of wet removal event that are implemented with probabilities proportional to the corresponding rates. Zhao et al. (2005a–c) have promoted a new stochastic approach to describe particle coagulation, simultaneous coagulation and condensation/evaporation, simultaneous coagulation and breakage, successively. The stochastic algorithm has characteristics of time-driven MC method (Liffman, 1992), constant-volume MC method (Garcia et al., 1987) and constant-number MC method (Lin et al., 2002). The stochastic algorithm is named as multi-Monte Carlo (MMC) method. The paper will extend the MMC method to solve GDE for wet removal. Firstly, the process of aerosols wet scavenging by precipitation is introduced in Section 2; then in Section 3, the stochastic algorithm for wet removal is described in details; and then two special cases in which analytical solutions exist are used to validate the algorithm in Section 4. In Section 5, using the stochastic algorithm, some numerical simulations are taken to investigate the process of aerosols wet scavenging when different precipitation types.

## 2. Theoretical description of aerosols wet scavenging by precipitation

The following major assumptions are made for the process of aerosols wet scavenging:

- (A) Precipitation process is considered to be steady-state.

- (B) Since the size of raindrop is far greater than that of aerosol, it's considered that the size of raindrop does not change although it collects amounts of aerosols. Moreover, the paper doesn't consider raindrop collision, coagulation, condensation/evaporation, nucleation, breakup, etc. So RSD is maintained steadily during precipitation. Raindrop particle is a regular sphere.
- (C) Other dynamic events of aerosol, for example, collision, coagulation, condensation/evaporation, nucleation, breakup, etc., are not considered; aerosol particle is a regular sphere.

Scavenging coefficient  $A(d_p)$  encapsulates any possible wet removal mechanisms.  $A(d_p)$ , which describes physically  $d_p$ -aerosol diameter collides with raindrops and then is collected, is given by

$$A(d_p) = \int_{D_{d,\min}}^{D_{d,\max}} K(d_p, D_d) E(d_p, D_d) dD_d, \quad (2)$$

where  $K(d_p, D_d)$  is collision kernel or collection kernel describing the possibility of collision between  $d_p$ -aerosol diameter and  $D_d$ -raindrop diameter when the trajectory of aerosol intercrosses geometrically that of raindrop,  $E(d_p, D_d)$  is the collision efficiency or collection efficiency, which represents the fraction of aerosols in the raindrop sweep volume that are actually captured.  $K(d_p, D_d)$  is defined as

$$K(d_p, D_d) = \pi D_d^2 |U(D_d) - U(d_p)| n_d(D_d) / 4 \quad (3)$$

in which  $U(D_d)$  and  $U(d_p)$  are the falling velocity of  $D_d$ -raindrop diameter (mm) and  $d_p$ -aerosol diameter ( $\mu\text{m}$ ), respectively;  $n_d(D_d)$  is raindrop size function,  $\text{mm}^{-1} \text{m}^{-3}$ . In order to be convenient for the microphysical parameterizations of scavenging coefficient (Mircea and Stefan, 1998) and moment method (Jung et al., 2002, 2003), it's considered the falling velocity of  $d_p$ -aerosol diameter,  $U(d_p)$ , approaches to zero for relatively quiet atmospheric environment, so usually  $K(d_p, D_d) \approx \pi D_d^2 U(D_d) n_d(D_d) / 4$ . Factually, if the difference between the size of raindrop and that of aerosol is little, the difference between of the falling velocity of raindrop and that of aerosol should not be neglected when calculating collection kernel. In the paper  $U(d_p)$  is calculated according to Seinfeld and Pandis (1998) as

$$U(d_p) = \rho_p d_p^2 g C_c / (18 \mu_a), \quad (4)$$

where

$$C_c = 1 + 2.493 \frac{\lambda}{d_p} + 0.84 \frac{\lambda}{d_p} \exp\left(-0.435 \frac{d_p}{\lambda}\right)$$

in which  $\rho_p$  is the material density of aerosol;  $g$  is the gravitational acceleration;  $C_c$  is the Cunningham slip correction factor;  $\lambda$  is the mean free path length of the gas molecules;  $\mu_a$  is the viscosity of the air.

The smallest size of raindrop is about 100  $\mu\text{m}$  and the largest size is about 6 mm for precipitation process in nature fields (Mason, 1971). If the size of raindrop is < 100  $\mu\text{m}$ , it will be vaporized when it falls from the clouds. If the size of raindrop below clouds is greater than 6 mm, it will break up undergoing simultaneous gravity force and interfacial tension. With regard to the falling velocity of raindrop, it is approximately fitted as (Pruppacher and Klett, 1978)

$$\begin{aligned} U(D_d) &= 30.75 D_d^2 \times 10^6 \quad \text{if } D_d < 100 \mu\text{m}, \\ U(D_d) &= 38 D_d \times 10^3 \quad \text{if } 100 \mu\text{m} < D_d < 1000 \mu\text{m}, \\ U(D_d) &= 133.046 D_d^{0.5} \quad \text{if } D_d > 1000 \mu\text{m}, \end{aligned} \quad (5)$$

where  $D_d$  is with dimension mm and the maximum value of  $U(D_d)$  is about  $9.17 \text{ m s}^{-1}$ .

The model of collision efficiency takes account of the contribution of those important wet removal mechanisms such as Brownian diffusion, interception and inertial impaction. Slinn (1983) parameterized collision efficiency originating from Navier–Stokes equation and using the dimensionless analysis and coupling with the experimental data:

$$\begin{aligned} E(d_p, D_d) &= \left\{ \frac{4}{ReSc} [1 + 0.4 Re^{1/2} Sc^{1/3} + 0.16 Re^{1/2} Sc^{1/2}] \right\}_{\text{Brownian diffusion}} \\ &+ \left\{ 4 \frac{d_p}{D_d} \left[ \frac{\mu_a}{\mu_w} + (1 + 2 Re^{1/2}) \frac{d_p}{D_d} \right] \right\}_{\text{interception}} \\ &+ \left\{ \left( \frac{\rho_w}{\rho_p} \right)^{1/2} \left( \frac{St - S^*}{St - S^* + 2/3} \right)^{3/2} \right\}_{\text{inertial impaction}}, \end{aligned} \quad (6)$$

where  $Re$  is the Reynolds number of a raindrop based on its radius;  $Sc$  is the Schmidt number of aerosol;  $\rho_w$  is the density of the raindrop;  $St$  is the Stokes number of aerosol;  $S^*$  is a dimensionless

parameter. Those parameters are calculated as

$$\begin{aligned} Re &= D_d U(D_d) \rho_a / (2\mu_a), \quad Sc = \mu_a / (\rho_a D_{diff}), \\ St &= 2\tau_p U(D_d) / D_d, \\ S^* &= \{1.2 + [\ln(1 + Re)]/12\} / [1 + \ln(1 + Re)], \\ \tau_p &= \rho_p d_p^2 C_c / (18\mu_a), \quad D_{diff} = k_b T C_c / (3\pi\mu_a d_p), \end{aligned} \quad (7)$$

where  $\rho_a$  is the density of the air;  $D_{diff}$  is the diffusion coefficient of aerosol;  $k_b$  is the Boltzmann constant ( $= 1.38054 \times 10^{-23} \text{ J K}^{-1}$ );  $T$  is the absolute temperature of the air;  $\mu_w$  is the viscosity of the raindrop;  $\tau_p$  is the relaxation time of aerosol. When  $St \leq S^*$ , it is considered that the “inertial impaction” part of collision efficiency,  $\{E(d_p, D_d)\}_{inertial \text{ impaction}}$  is equal to zero. It’s noticeable that  $0 \leq E(d_p, D_d) \leq 1$ . In the paper, for any numerical calculation the following conditions are adopted:  $T = 296.15 \text{ K}$ ,  $\rho_a = 1.193 \text{ kg m}^{-3}$ ,  $\mu_a = 1.83245 \times 10^{-5} \text{ kg m}^{-1} \text{ s}^{-1}$ ,  $\rho_w = 997.45 \text{ kg m}^{-3}$ ,  $\mu_w = 9.591 \times 10^{-4} \text{ kg m}^{-1} \text{ s}^{-1}$ ,  $\rho_p = 2270 \text{ kg m}^{-3}$  (the typical material density of fly ash from coal-fired power plant),  $\lambda = 6.73 \times 10^{-8} \text{ m}$ .

### 3. Description of multi-Monte Carlo method

#### 3.1. Weighted fictitious aerosol and weighted fictitious raindrop

The object of any MC method for GDE is the dispersed particles. A reasonably sized system contains approximately  $10^{10}$  or more particles, for example, the number concentration of the particulate matter (PM) in pulverized-coal boiler exceeds  $10^{15} \text{ m}^{-3}$  (Linak and Wendt, 1993). However, MC code can only examine  $10^3$ – $10^7$  particles at a time on fast PCs because of the limitation of CPU speed and memory capacity. Generally speaking, simulation particle in MC methods has a number weight, where some simulation particles, whose number is far less than that of real particles, represent those real particles. The most prevalent measure evaluating the number weight of simulation particle is the introduction of the concept of “subsystem” (Liffman, 1992). One assumes the whole system is fully stirred and spatially isotropic, and the subsystem is an indicator of the whole system. “Subsystem” contains  $10^3$ – $10^7$  simulation particles, and satisfies the constraint of periodic boundary conditions, that is, as some particles move out from one boundary of the subsystem, some identical particles move in from the symmetrical boundary of the subsystem; by those hypotheses one simulation

particle in the “subsystem” represents some real particles in the whole system, and the behavior of the subsystem duplicates the system as a whole. The introduction of “subsystem” can hardly take account of space dispersion of size function, boundary conditions, or particle and medium dynamics owing to the periodic boundary and the hypothesis of spatial isotropy.

MC method can be classified into two general classes according to whether or not the total number of simulation particles and simulation domain are changed during MC simulation. In constant-volume MC (Garcia et al., 1987), the total number of simulation particles continues to decrease as aerosols are scavenged by precipitation, which fatigues statistical accuracy of MC method. As far as constant-number MC (Lin et al., 2002) is considered, in the cases of wet removal event which results in net depletion of real aerosols, the domain of “subsystem” is expanded in order to maintain the constant number of simulation particles, which constrains greatly the application in engineering.

In the MMC method, both computational domain and the total number of simulation particles are conserved during simulation, that is, MMC method exhibits not only the characteristics of constant-volume MC but also those of constant-number MC. MMC method introduces the concept of “weighted fictitious aerosol”. It is believed that those real aerosol particles which have same or similar size have same properties and hence the same behaviors. One or several weighted fictitious aerosols represent those real aerosols, and fictitious aerosols are the indicator of those real aerosols. So the time evolution of fictitious aerosols duplicates that of real aerosols. The generation of fictitious aerosols follows those steps:

- (A) The aerosol population is divided into  $C_p$  sections, and the total number of real aerosols is  $N_p$ . The initial total of fictitious aerosols is set as  $N_{pf}$ , and the average transform-weight is set as  $kw t_p = N_p / N_{pf}$  ( $kw t_p$  is not required to be an integral value).
- (B) Aerosol section looping:
  - I. As for aerosol section  $i$  in a given size range, the representative size is  $d_{pi}$ , and the total number of real aerosols falling in section  $i$  is  $N_{pi}$ . The number of fictitious aerosols representing the aerosol section  $i$  is calculated as follows:  $N_{pfi} = \text{integer}[N_{pi}/kw t_p]$ . If  $N_{pfi} = 0$ , let  $N_{pfi} = 1$  in order to not

omit any information about real aerosol population.

- II. Some fictitious aerosols, the number of which is  $N_{pfi}$ , are generated and indexed. As for one fictitious aerosol whose index is  $j$ , its transform-weight is updated:  $kwt_{pj} = N_{pi}/N_{pfi}$ , and its size is still  $d_{pi}$ .
- (C) The total number of fictitious aerosols is renewed as  $N_{pf} = \sum_i N_{pfi}$ .

The detailed description and a factual example are shown in Zhao et al. (2005b). In fact, those fictitious aerosols of the same section have same size and same value of “transform-weight”, however different size and different value of “transform-weight” for different section. The size of fictitious aerosol is just equal to the representative size of the section, and the value of transform-weight “ $kwt_p$ ” of one fictitious aerosol is equal to the number concentration of those real aerosols represented by the fictitious aerosol. Generally speaking, MMC method still maintains high computation precision even though the value of “ $kwt_p$ ” reaches to the magnitude of  $O(10^3)$ – $O(10^4)$ .

During the evolution of the system, the transform-weight “ $kwt_p$ ” and size of those related fictitious aerosols are changed, instead the computational domain and the number of fictitious particles are maintained, which will be described in “Section 3.5.”

The calculation of scavenging coefficient needs the definite integral over RSD. This operation of definite integral will cost plenty of time or generate some numerical errors, or even be unable to be gone along. In order to avoid the operation of definite integral, the concept of “weighted fictitious raindrop”, which is similar to “weighted fictitious aerosol”, is introduced in the paper. The generation of fictitious raindrops is fully similar to that of fictitious aerosols, which will not be repeated. Now the transform-weight of fictitious raindrop  $i$  is  $kwt_{di}$  and size  $D_{di}$ . If the total number of fictitious raindrops is  $N_{df}$ , the calculation of scavenging coefficient is as follows:

$$\begin{aligned}
 A(d_p) &= \int_{D_{d,\min}}^{D_{d,\max}} \int \frac{\pi D_d^2}{4} |U(D_d) - U(d_p)| E(d_p, D_d) n_d(D_d) dD_d \\
 &= \sum_{i=1}^{N_{df}} \left[ \frac{\pi D_{di}^2}{4} |U(D_{di}) - U(d_p)| E(d_p, D_{di}) kwt_{di} \right]. \quad (8)
 \end{aligned}$$

### 3.2. The scheme of multi-Monte Carlo method

MMC method for wet removal is based on time-driven technique (Liffman, 1992). The time-driven technique tracks every simulation particle and considers every possible event within a special adjustable time step. The technique assumes dynamic events are decoupled within a sufficiently small time step. Time step must be less than or equal to the minimum time within which every possible event takes place once at most for every simulation particle. Time-driven MC need divide explicitly time window into intervals, and it's possible for one simulation particle that no any event is examined within one interval. The scheme of MMC method for wet removal is as following:

- (A) Fictitious aerosols are generated to represent the real aerosol population, and the number of fictitious aerosols is far less than that of real aerosols; in the same way, fictitious raindrops are generated to represent those real raindrop population (seen in Section 3.1).
- (B) Time-step looping:
- I. The minimum time scale of wet removal event is calculated, and then time step  $\Delta t$  is set (seen in Section 3.3).
  - II. Fictitious aerosol looping:
    - i For every fictitious aerosol, the judgment of the occurrence of wet removal event is taken using the random number (seen in Section 3.4).
  - III. The consequence of every wet removal event is treated, where computational domain and the total number of fictitious aerosols are maintained (seen in Section 3.5).
- (C) The results of numerical simulation are counted and output consisted of when the appointed time point is reached.

### 3.3. Time step

The scavenging coefficient of fictitious aerosol  $i$ ,  $A_i$ , denotes the probability of a wet removal event in unit time. So the time scale of wet removal event for fictitious aerosol  $i$  follows  $t_{sca,i} = 1/A_i$ . Time step of time-driven MC technique (Liffman, 1992) should be less than or equal to the minimum time scale in which the number of wet removal event must be less than or equal to one for every fictitious aerosol. So

time step should be less than the minimum time scale of wet removal event, i.e.,  $\Delta t \leq \min(t_{\text{sca},i}) = 1/\max(A_i)$ . In order to increase the number of MC loop, time step is usually defined as  $\Delta t = \alpha/\max(A_i)$ , where the multiplicative constant,  $\alpha$ , has the value of 0.01 or less.

Along with the occurrence of wet removal event, ASD must have been changed, which makes the minimum time scale of wet removal event change. So time step must be adjusted real-timely, not just as a fixed value.

The computational cost of the calculation of time step will be  $O(N_{\text{pr}} \times N_{\text{dt}})$  if all of the calculations of scavenging coefficient of every fictitious aerosol use Eq. (8). In fact, scavenging coefficient for an aerosol size depends on RSD. Fortunately, the process of precipitation and RSD are assumed to be steady state. And then, the characteristic of wet removal is that the range of ASD is only contracted. So scavenging coefficient of every fictitious aerosol is calculated and stored in the first time step. After that, the value of scavenging coefficient for a fictitious aerosol size is just the stored value of scavenging coefficient for the corresponding aerosol size. By those means the computational cost of every calculation of time step will decrease to  $O(N_{\text{pr}})$ .

#### 3.4. The judgment of the occurrence of wet removal event

Its deemed that the process of dynamic evolution due to wet depositions in dispersed system is a Markov process (Garcia et al., 1987). As far as a standard procedure for a Markov process is considered, the probability of fictitious particle  $i$  scavenging by precipitation within  $\Delta t$ ,  $Pr_{\text{sca},i}(\Delta t)$ , is represented by an exponential function (Liffman, 1992):  $Pr_{\text{sca},i}(\Delta t) = 1 - \exp(-A_i \Delta t) \approx A_i \Delta t$ . A random number  $R_1$  from a uniform distribution in the interval [0,1] is generated. As for a standard MC simulation, fictitious aerosol  $i$  will be scavenged if the relation  $R_1 \leq A_i \Delta t$  is satisfied. Here the transform-weight  $kw_{t_{pi}}$  and the size  $d_{pi}$  of the current tracked fictitious aerosol  $i$  are stored in order to treat the consequence of the wet removal event after fictitious aerosol looping. Because it's considered in time-driven MC technique that any events do not change immediately the properties and the behaviors of the tracked fictitious aerosol and the related fictitious aerosol within current time step, the consequential treatment of aerosol wet removal

event should be delayed until the end of current time step.

#### 3.5. The consequential treatment of wet removal event

If the fictitious particle  $i$  be scavenged, it will not be tracked next time step and its vacancy should be filled. The consequential treatment of aerosol wet removal event should maintain both the computational domain and the total number of fictitious aerosols. Here one random fictitious aerosol  $j$  is selected by a stochastic process, and then those real aerosols which are represented by fictitious aerosol  $j$  are halved, and every half of those real aerosols is represented by one new fictitious aerosol, respectively. The two new fictitious aerosols are indexed respectively by  $k$  and  $m$ . The following treatments are adopted:  $kw_{t_{pj}} = 2kw_{t_{pk}} = 2kw_{t_{pm}}$ , and  $d_{pj} = d_{pk} = d_{pm}$ , where  $kw_{t_{pj}}$ ,  $kw_{t_{pk}}$  and  $kw_{t_{pm}}$  are the transform-weight of fictitious particle  $j$ ,  $k$  and  $m$ , respectively;  $d_{pj}$ ,  $d_{pk}$  and  $d_{pm}$  is the size of fictitious aerosol  $j$ ,  $k$  and  $m$ , respectively. And then fictitious aerosol  $i$  and  $j$  are replaced by fictitious aerosol  $k$  and  $m$ , respectively. Factually, those measures mean that the leaving vacancy of the removed fictitious particle  $i$  is filled with the half of the randomly selected fictitious particle  $j$ .

#### 4. Validation of multi-Monte Carlo method

In the paper, ASD is described by a set of lognormal distributions:

$$n_p(d_p, t) = \frac{N_p}{\sqrt{2\pi} \ln \sigma_{pg}} \exp \left[ -\frac{\ln^2(d_p/d_{pg})}{2 \ln^2 \sigma_{pg}} \right] \frac{1}{d_p} \quad (9)$$

in which  $N_p$  is the total aerosol number concentration at  $t$  ( $\text{m}^{-3}$ );  $d_{pg}$  is the geometric mean diameter at  $t$  ( $\mu\text{m}$ );  $\sigma_{pg}$  is the geometric standard deviation based on the aerosol diameter at  $t$ .

For the purpose of convenient integral over RSD when the acquisition of accurate analytical solutions, RSD is also considered to be a lognormal function (Jung et al., 2002):

$$n_d(D_d) = \frac{N_d}{\sqrt{2\pi} \ln \sigma_{dg}} \exp \left[ -\frac{\ln^2(D_d/D_{dg})}{2 \ln^2 \sigma_{dg}} \right] \frac{1}{D_d} \quad (10)$$

in which  $N_d$  is the total raindrop number concentration at  $t$  ( $\text{m}^{-3}$ );  $D_{dg}$  is the geometric mean diameter at  $t$  (mm);  $\sigma_{dg}$  is the geometric standard deviation based on the raindrop diameter at  $t$ .

Jung and Lee (1998) simplified the collision efficiency for small size aerosol mainly dominated by Brownian diffusion mechanism:

$$E_{JD}(d_p, D_d) = 2 \left( \frac{\sqrt{3}\pi}{4Pe} \right)^{2/3} \left[ \frac{(1-\alpha)(3\sigma+4)}{J+\sigma K} \right]^{1/3} \quad (11)$$

in which  $\alpha$  is the volume fraction or packing density of raindrop;  $\sigma$  is the viscosity ratio of raindrop and air,  $\sigma = \mu_w/\mu_a$ ;  $Pe$  is the Peclet number,  $Pe = D_d U(D_d)/D_{diff}$ ;  $J = 1 - 6\alpha^{1/3}/5 + \alpha^2/5$ ;  $K = 1 - 9\alpha^{1/3}/5 + \alpha + \alpha^2/5$ .

As for aerosol population with range  $d_p < 0.05 \mu\text{m}$  and  $0.05 < d_p < 1.0 \mu\text{m}$ , Jung et al. (2002) obtained analytical solutions of the time evolution of size distribution of scavenging aerosols by precipitation, respectively. Those analytical solutions are used to validate the computation precision of MMC method for wet removal. The computational conditions are listed in Table 1, where the characteristic time scale of wet removal is defined as  $\tau_{sca} = 1/\Lambda(d_{pg,0})$ ;  $N_{pf}$  and  $N_{df}$  are the initial number of fictitious aerosols and that of fictitious raindrops; the parameters of RSD are as follows:  $N_d = 10^5 \text{m}^{-3}$ ,  $D_{dg} = 0.1 \text{mm}$ ,  $\sigma_{dg} = 1.2$ .

Figs. 1 and 2 show the comparison of MMC solutions and analytical solution for Cases 1 and 2, respectively, where Fig. 1(a) and Fig. 2(a) are the curves of ASD at 5 special time-point, respectively; Fig. 1(b) and Fig. 2(b) are the time evolution of the total number concentration of real aerosols, geometric mean volume ( $v_{pg} = \pi d_{pg}^3/6$ ) and geometric standard deviation, respectively. Even though the total number concentration of real aerosols decreases sharply due to wet removal at the end of time evolution ( $N_p = 81080.9 \text{m}^{-3}$  at  $t = 10^4 \text{s}$  for Case 1;  $N_p = 2565.9 \text{m}^{-3}$  at  $t = 10^7$  for Case 2), the results of MMC method agree with the corresponding analytical solutions very well, except a little bias for geometric standard deviation.

Computation costs for Cases 1 and 2 are about 25 and 78 s in PCs, respectively. The low computation

cost of MMC method will encourage its application in engineering.

## 5. Results and discussions

### 5.1. Numerical simulation of MMC method

Water content ( $w_1$ ) and rainfall intensity ( $J$ ) are two parameters describing the classification of precipitation. They are related to RSD:

$$w_1 = \int_{D_{d,min}}^{D_{d,max}} \frac{\pi}{6} \rho_w D_d^3 n_d(D_d) dD_d,$$

$$J(\text{mm h}^{-1}) = 3.6 \int_{D_{d,min}}^{D_{d,max}} \frac{\pi}{6} \rho_w D_d^3 U \times (D_d) n_d(D_d) dD_d. \quad (12)$$

RSD in the Mediterranean area can be approximated fitted by a unimodal log-normal function according to long-time measurements in Israel (Feingold and Levin, 1986) and Spain (Cerro et al., 1997). The three parameters of RSD,  $N_d$ ,  $d_{dg}$  and  $\sigma_{dg}$ , are expressed as a function of rainfall intensity. In the paper, we choose the parameterizations of Feingold and Levin (1986) to describe RSD, and it's considered the geometric standard deviation is constant. The parameters in the typical light, moderate and heavy precipitation are listed in Table 2.

Scavenging coefficient is directly associated with collision efficiency, RSD and the falling velocity of raindrop and aerosol. Fig. 3 shows the relationship between scavenging coefficient and aerosol size for typical light, moderate and heavy precipitation, respectively. For every precipitation type, the curve of scavenging coefficient as a function of aerosol diameter has obvious similarity with the curve of collision efficiency as a function of aerosol diameter, which implies collision efficiency plays an important role on scavenging coefficient and cannot be considered to be a constant value.

As shown in Fig. 3, the scavenging coefficient of aerosol for different precipitation type is different,

Table 1  
Computational condition

Case	$N_{p,0}, \text{m}^{-3}$	$d_{pg,0}, \mu\text{m}$	$\sigma_{pg,0}$	$\tau_{sca}, \text{s}$	$N_{pf}$	$N_{df}$	Time evolution, s
Case 1	$10^6$	0.001	1.5	3221.80	3000	3000	$10^4$
Case 2	$10^6$	0.1	1.5	1213571.72	3000	3000	$10^7$



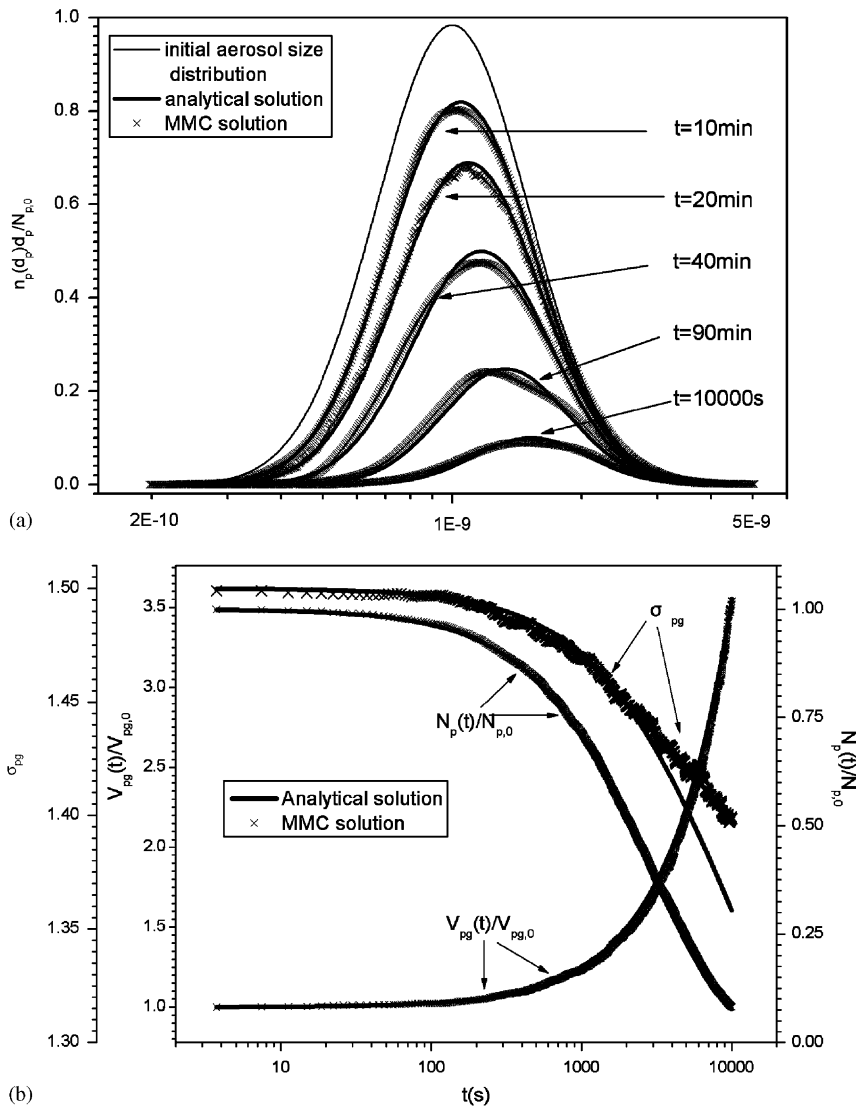


Fig. 1. The comparison between MMC solutions and analytical solutions in Case 1.

which indicates precipitation type will affect the process of aerosols wet scavenging. Figs. 4–6 investigate the effect of aerosols wet scavenging for different precipitation type, where ASD is expressed by lognormal function, and the initial total of real aerosols per unit volume is  $N_{p,0} = 10^6 \text{ m}^{-3}$ , geometric standard deviation  $\sigma_{pg,0} = 1.3$ , geometric mean diameter  $d_{pg,0}$  for small, intermediate and large particle size distribution are 0.01, 0.5 and  $5 \mu\text{m}$ , respectively. RSD is represented by log-normal function, and rainfall intensity is 1, 10 and  $100 \text{ mm h}^{-1}$  for light, moderate and heavy precipitation, respectively.

Aerosols with  $d_{pg,0} = 0.01 \mu\text{m}$  are mainly collected by raindrop due to Brownian diffusion mechanism. The smaller size, the higher collision efficiency is. As shown in Fig. 4(b), the geometric mean volume ( $v_{pg}$ ) continues to increase; however, the total number of real aerosols ( $N_p$ ) continues to decrease. Furthermore, the bigger rainfall intensity, the faster velocity of the ascending of  $v_{pg}$  or the descending of  $N_p$  is. The numerical results shown in Fig. 4(a) indicate that short heavy precipitation (for example, 2 h) scavenges as many aerosols as long moderate precipitation (7.5 h) and longer light precipitation (30 h), which accords with the model

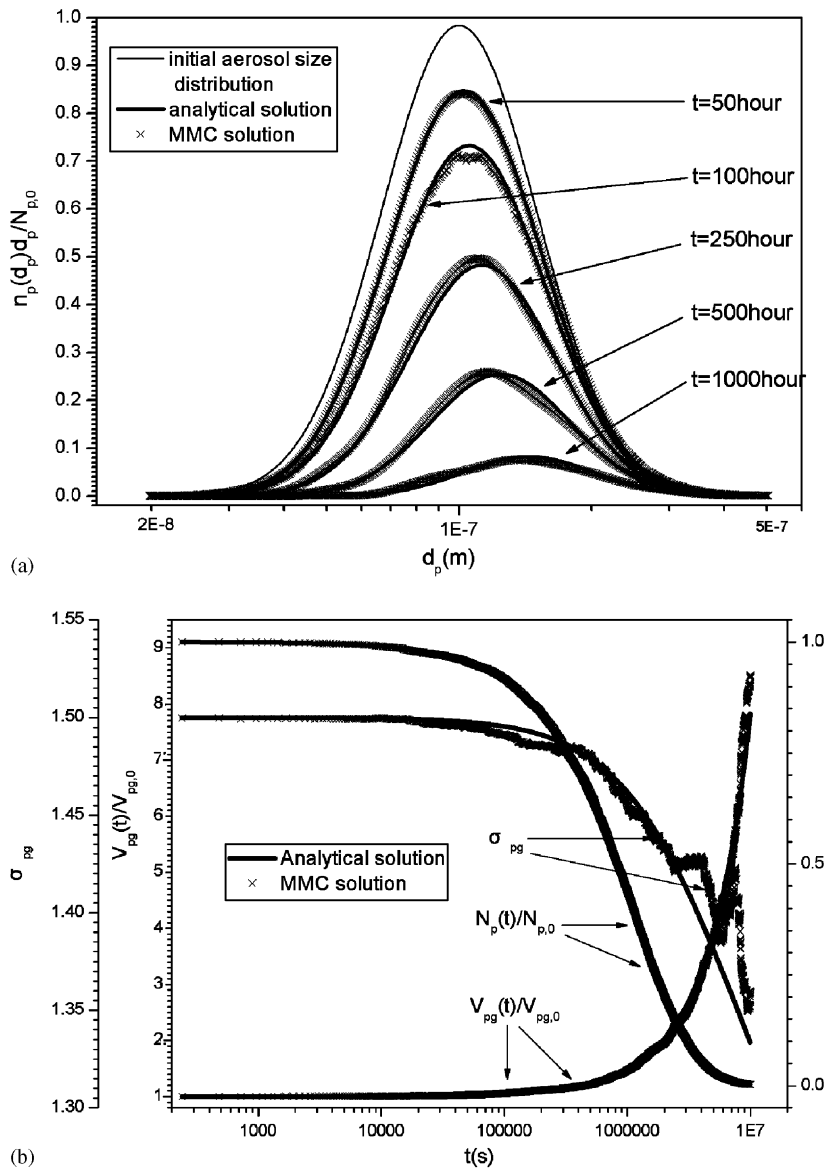


Fig. 2. The comparison between MMC solutions and analytical solutions in Case 2.

Table 2  
Classification of precipitation

Precipitation type	$J, \text{mm h}^{-1}$	$N_d, \text{m}^{-3}$	$D_{dg}, \text{mm}$	$\sigma_{dg}$
Light precipitation	1	172	0.72	2.0
Moderate precipitation	10	285.45	1.22	2.0
Heavy precipitation	100	473.73	2.08	2.0

results of Garcia Nieto et al. (1994). The location of peak of ASD continues to close up right side (the side of larger aerosols). With respect to aerosols with  $d_{pg,0} = 5 \mu\text{m}$ , they are scavenged due to mainly inertial impaction with raindrops. The larger size of aerosol, the higher collision efficiency is. Here the geometric mean volume continues to decrease as well as the total number of real aerosols. The curves of ASD continue to decline to left side (the side of smaller aerosols), which shows those large aerosols are scavenged more easily and effectively.

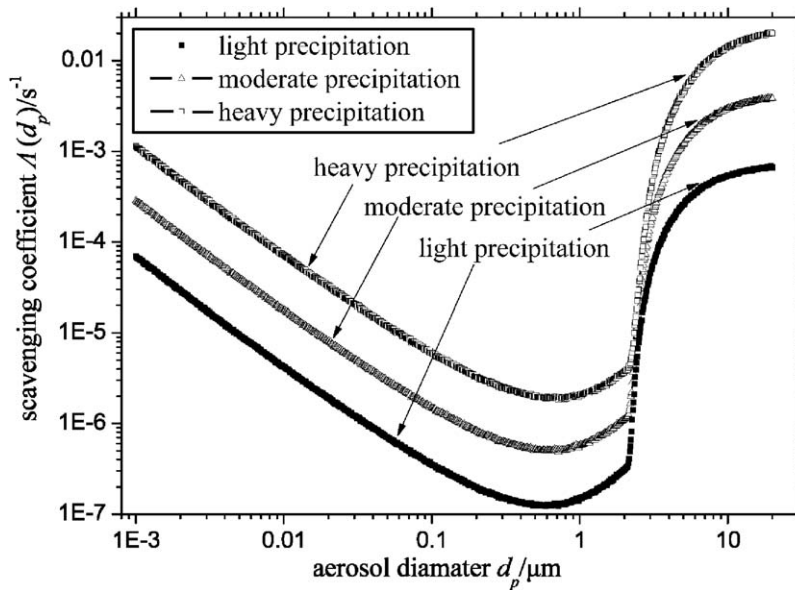


Fig. 3. Scavenging coefficient as a function of aerosol size.

Obviously, heavy precipitation exhibits a better effect of aerosols wet scavenging than moderate and light precipitation (in that order). Aerosols with  $d_{pg,0} = 0.5 \mu\text{m}$  (situated in “Greenfield gap” range (Greenfield, 1957)) are hardly removed by precipitation process because the two important wet removal mechanisms, Brownian diffusion and inertial impaction, have the minimum effect on those aerosols and the contribution of interception mechanism on collision efficiency is also weak. The numerical results of Fig. 5 show the geometric mean volume is nearly constant however the aerosol number concentration continues to decrease with time evolution. Similar with small aerosols ( $d_{pg,0} = 0.01 \mu\text{m}$ ) and large aerosols ( $d_{pg,0} = 5 \mu\text{m}$ ), those intermediate aerosols ( $d_{pg,0} = 0.5 \mu\text{m}$ ) are scavenged more effectively by heavy precipitation than moderate and light precipitation (in that order). It’s noticeable that geometric standard deviation for any aerosol size and for any precipitation type is decreasing on the average, which indicates the curve of ASD continues to incline to be flat and the size of aerosols continues to incline to uniform and monodisperse. Synthetically, the wet removal of small aerosols is dominated mainly by Brownian diffusion mechanism, and inertial impaction is the most important mechanism for large aerosols; however, both Brownian diffusion and inertial impaction have the minimum influence on the wet removal of intermediate aerosols; interception mechanism plays

a relatively weak role on aerosols with any size. The numerical results which are shown in Figs. 4–6 accord with qualitatively the results of Garcia Nieto et al. (1994).

The same precipitation type has different effect of aerosols wet scavenging on those aerosols with different size. In order to reach to the same effect of aerosols wet scavenging, for example, to scavenge 40% aerosols, light precipitation spends about 35 min when scavenging large aerosols ( $d_{pg,0} = 5 \mu\text{m}$ ); however, 33 h when scavenging small aerosols ( $d_{pg,0} = 0.01 \mu\text{m}$ ), and even about 1094 h when scavenging intermediate aerosols ( $d_{pg,0} = 0.5 \mu\text{m}$ ). Similarly, moderate or heavy precipitation scavenges large aerosols more effectively than small aerosols and intermediate aerosols (in that order).

On the whole, the increase of rainfall intensity can help scavenge not only small aerosols and large aerosols but also intermediate aerosols. The three precipitation types have a better effect of aerosols wet scavenging for large aerosols than small aerosols and intermediate aerosols (in that order). The three precipitation types can hardly scavenge intermediate aerosols; comparatively, the effect of heavy precipitation on intermediate aerosols is better than the other precipitation types. Those conclusions accord with our conventional viewpoint, which can be explained by the scavenging coefficient of aerosol. Since scavenging

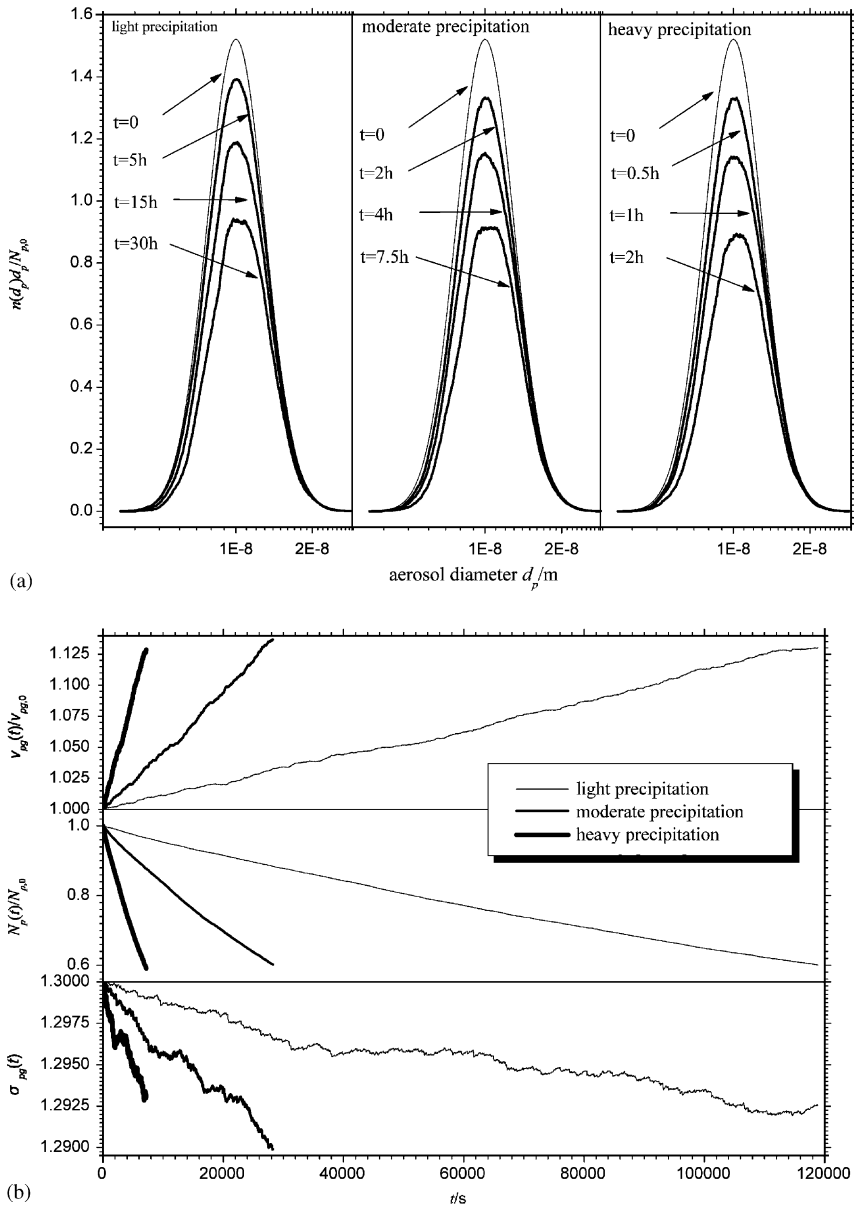


Fig. 4. The wet scavenging of small aerosols by three precipitation types.

coefficient of aerosol represents the removal rate of aerosols by raindrop, the information about scavenging coefficient as a function of aerosol size when different precipitation processes, which is shown in Fig. 3, can help research the effect of wet scavenging of aerosols with different size when different precipitation processes from the other view. The increasing mass of raindrops results in the increasing of volume and surface area of raindrops, which will increase the collision efficiency of small aerosols due to Brownian diffusion mechanism

and then the scavenging coefficient of small aerosols. However, the collision efficiency due to interception mechanism, which depends mathematically on the ratio of aerosol size and raindrop size ( $d_p/D_d$ ), will decrease with the increasing size of raindrops. With respect to the collision efficiency due to inertial impaction mechanisms which is approximate to  $(1-0.9St^{-0.5})$  according to Jung et al. (2003), it correlates positively with Stokes number of aerosol ( $St$ ). However,  $St$  may correlate positively or negatively with raindrop size  $D_d$ ,

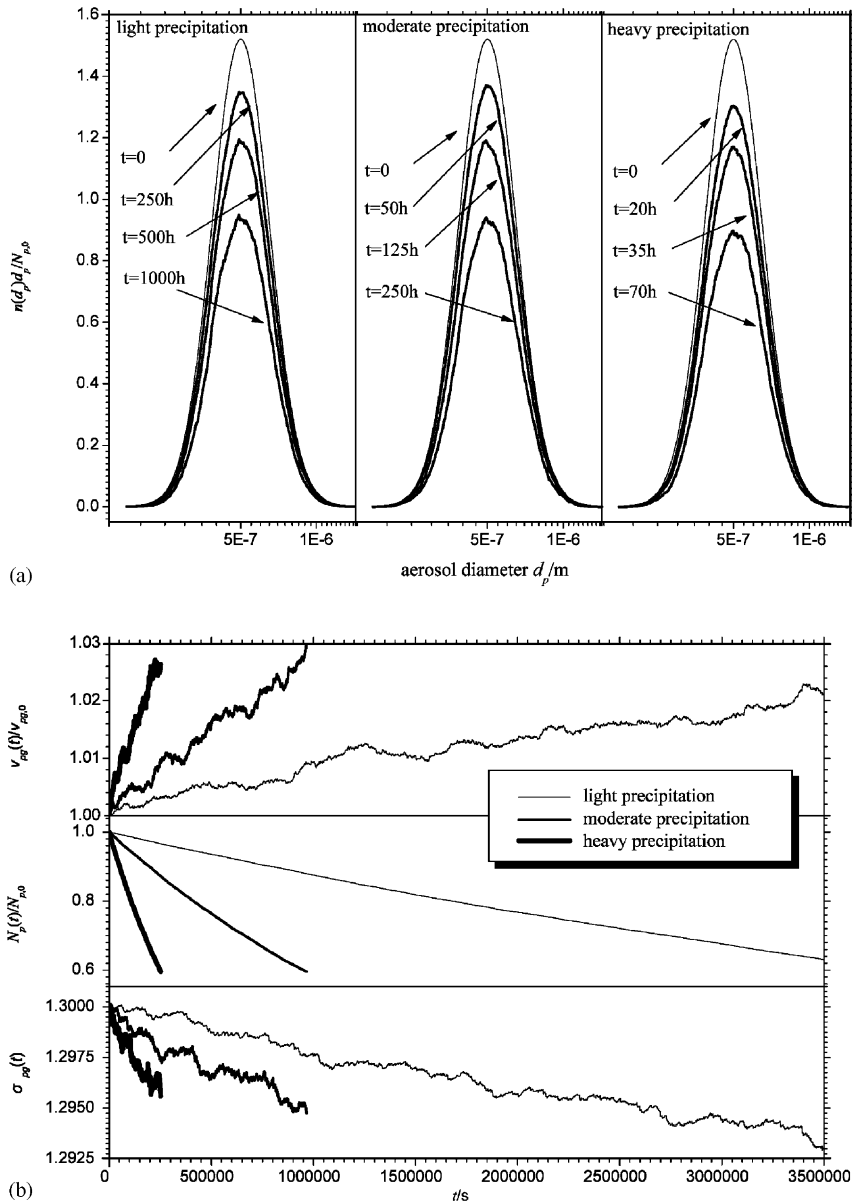


Fig. 5. The wet scavenging of intermediate aerosols by three precipitation types.

depending on the “three-section” representations of the falling velocity of the raindrop within different size range of raindrops. So the collision efficiency due to inertial impaction mechanism may increase or decrease with the increasing size of raindrops according to the model of collision efficiency promoted by Slinn (1983). Factually, the scavenging coefficient of aerosol is directly associated with not only collision efficiency but also RSD. Therefore, the increase of rainfall intensity (water content) and then the increase of mass and size of raindrops will

increase the scavenging coefficient of aerosol with any size on the common contribution of the increasing number concentration and size of raindrops, although there exist some complicated relation between the collision efficiency and rainfall intensity. The increasing scavenging coefficient for any aerosol size will benefit aerosols wet scavenging. Furthermore, there are more obvious increase of scavenging coefficient for large aerosols and small aerosols than intermediate aerosols with the increase of rainfall intensity. So precipitation

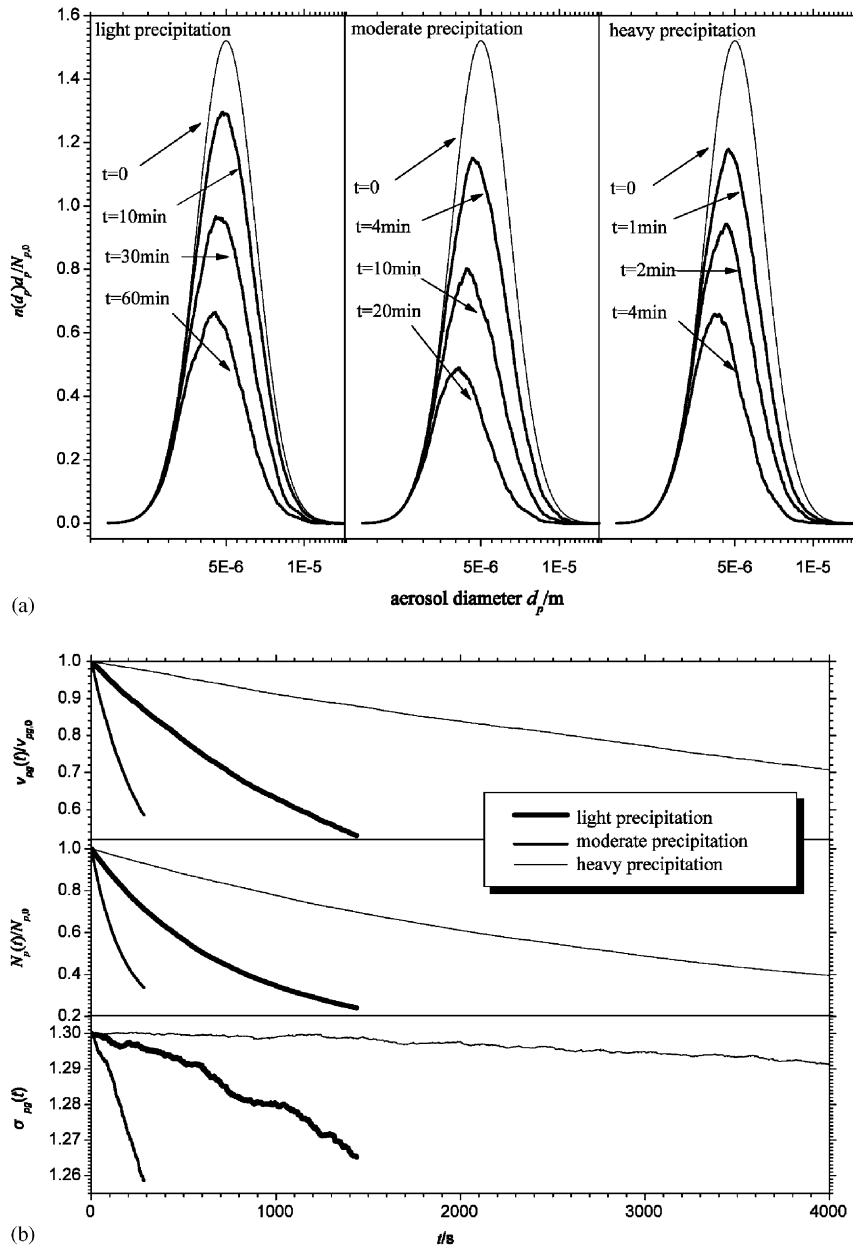


Fig. 6. The wet scavenging of large aerosols by three precipitation types.

scavenges more effectively large aerosols and small aerosols along with the increase of rainfall intensity. On the other hand, the scavenging coefficient of large aerosols is greater than that of small aerosols and intermediate aerosols (in that order), which is the reason why precipitation scavenges more effectively large aerosols than small aerosols and intermediate aerosols (in that order).

It's noticeable that the time evolutions of ASD of small and intermediate aerosols are basically "self-preserving". However, the "self-preserving" ASD is not rigid for aerosols with range 2–5  $\mu\text{m}$  because scavenging coefficient increases sharply within the size range of aerosols. Moment method (Jung et al., 2003), which is based on the "self-preserving" ASD, will generate some numerical errors.

### 5.2. The analysis of numerical errors of MMC method for wet removal

MMC method is based on time-driven technique, which assumes dynamic events in dispersed systems are decoupled each other within a sufficiently small time step  $\Delta t$ . In fact, within time-step  $\Delta t$ , those dynamic events may associate with each other and are NOT uncoupled absolutely each other, so any “time-driven” MC method shows the so-called “uncoupling error”. The introduction of multiplicative constant  $\alpha$  is just for the purpose of the reduction of the “uncoupling error”. The smaller multiplicative constant, the fewer deposition events take place within one time-step and then the “uncoupling error” decreases, however, on expense of computation time. The other source of numerical errors originates from the consequential treatment of dynamic event. The disturbing of transform-weight of fictitious aerosols due to the forcible maintenance of constant volume and constant number contributes to the so-called “constant volume and number error”. The third numerical error is random error. MMC method for wet removal adopts many random number and random process, for example, when judging the occurrence of wet removal event. Those random procedures exhibit some inevitable error, which is called as random error. Lastly, the statistical error is inevitably demonstrated by all MC methods.

### 5.3. Future works

Aerosols wet scavenging by precipitation is a very complicated process. The microphysical model adopted by the paper does not take account of fractal aerosol, condensation/evaporation, nucleation or crystallization (Ackerman et al., 1995), coagulation (Zhang et al., 2004) of raindrops and aerosols, etc. Furthermore, the different chemical nature of aerosols (Chate and Kamra, 1997; Chate et al., 2003) affects wet removal process, for example, the size and density of hygroscopic aerosols and raindrops relates closely to their height, humidity and chemical composition of air. The description of those complicated factors needs the foundation of micro-chemical model.

The paper only considers the contribution of the three wet removal mechanisms: Brownian diffusion, interception and inertial impaction. In fact, electrostatic collection is also an important wet removal

mechanism (Tinsley et al., 2001; Andronache, 2004). Furthermore, how to improve the collision efficiency of “Greenfield gap” aerosols is an important issue.

The aerosols wet scavenging by precipitation may illumine the removal of PM from pulverized-coal fired power plants in electrostatic precipitator.

## 6. Conclusions

A new stochastic approach, MMC method, is firstly promoted to describe quantitatively the time evolution of ASD when raindrop falling. MMC method, which is based on “time-driven” technique, introduces “fictitious aerosols” and “fictitious raindrops” to represent those real aerosols population and real raindrop population, respectively. MMC method maintains synchronously the total number of fictitious aerosols and computational domain. The good agreement between MMC solution and analytical solution for diffusion dominant range shows its stable and high computation precision. The precision of MMC method is restrained together by “uncoupling error”, “constant volume and number error”, random error and statistical error. When describing the process of aerosols wet scavenging by precipitation, MMC method does not need any assumption on collision efficiency, ASD and RSD, which contrasts nearly with moment method. MMC method can describe any case of aerosols wet scavenging so long as the model of scavenging coefficient can be obtained mathematically.

MMC method is used to investigate the effects of the different precipitation type on wet scavenging of aerosols with different size. Numerical results show, short heavy precipitation scavenges as many aerosols with any size as long moderate precipitation and even longer light precipitation; large aerosols are scavenged more effectively by any precipitation type than small aerosols and intermediate aerosols (in that order).

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## References

- Ackerman, A.S., Hobbs, P.V., Toon, O.B., 1995. A model for particle microphysics, turbulent mixing, and radiative transfer in the stratocumulus-topped marine boundary layer and comparisons with measurements. *Journal of Atmospheric Sciences* 52, 1204–1236.
- Andronache, C., 2004. Diffusion and electric charge contributions to below-cloud wet removal of atmospheric ultra-fine aerosol particles. *Journal of Aerosol Science* 35, 1467–1482.
- Cerro, C., Codina, B., Bech, J., Lorente, J., 1997. Modelling raindrop size distribution and Z(R) relations in the Western Mediterranean Area. *Journal of Applied Meteorology* 36, 1470–1479.
- Chate, D.M., Kamra, A.K., 1997. Collection efficiencies of large water drops collecting aerosol particles of various densities. *Atmospheric Environment* 31, 1631–1635.
- Chate, D.M., Pranesha, T.S., 2004. Field studies of scavenging of aerosols by rain events. *Journal of Aerosol Science* 35, 695–706.
- Chate, D.M., Rao, P.S.P., Naik, M.S., Momin, G.A., Safai, P.D., Ali, K., 2003. Scavenging of aerosols and their chemical species by rain. *Atmospheric Environment* 37, 2477–2484.
- Feingold, G., Levin, Z., 1986. The lognormal fit to raindrop spectra from frontal convective clouds in Israel. *Journal of Climate and Applied Meteorology* 25, 1346–1364.
- Garcia, A.L., van den Broek, C., Aertseens, M., Serneels, R., 1987. A Monte Carlo simulation of coagulation. *Physica* 143A, 535–546.
- Garcia Nieto, P.J., Arganza Garcia, B., Fernandez Diaz, J.M., Rodriguez Brana, M.A., 1994. Parametric study of selective removal of atmospheric aerosol by below-cloud scavenging. *Atmospheric Environment* 28, 2335–2342.
- Greenfield, S., 1957. Rain scavenging of radioactive particulate matter from the atmosphere. *Journal of Meteorology* 14, 115–125.
- Jung, C.H., Lee, K.W., 1998. Filtration of fine particles by multiple liquid droplet and gas bubble systems. *Aerosol Science and Technology* 29, 389–401.
- Jung, C.H., Kim, Y.P., Lee, K.W., 2002. Analytic solution for polydispersed aerosol dynamics by a wet removal process. *Journal of Aerosol Science* 33, 753–767.
- Jung, C.H., Kim, Y.P., Lee, K.W., 2003. A moment model for simulating raindrop scavenging of aerosols. *Journal of Aerosol Science* 34, 1217–1233.
- Liffman, K., 1992. A direct simulation Monte-Carlo method for cluster coagulation. *Journal of Computational Physics* 100, 116–127.
- Lin, Y., Lee, K., Matsoukas, T., 2002. Solution of the population balance equation using constant-number Monte Carlo. *Chemical Engineering Science* 57, 2241–2252.
- Linak, W.P., Wendt, J.O.L., 1993. Toxic metal emission from incineration: mechanisms and control. *Progress in Energy and Combustion Science* 19, 145–185.
- Loosmore, G.A., Cederwall, R.T., 2004. Precipitation scavenging of atmospheric aerosols for emergency response applications: testing an updated model with new real-time data. *Atmospheric Environment* 38, 993–1003.
- Mason, B.J., 1971. *The Physics of Clouds*. Oxford University Press, London, UK.
- Mircea, M., Stefan, S., 1998. A theoretical study of the microphysical parameterization of the scavenging coefficient as a function of precipitation type and rate. *Atmospheric Environment* 32, 2931–2938.
- Mircea, M., Stefan, S., Fuzzi, S., 2000. Precipitation scavenging coefficient: influence of measured aerosol and raindrop size distributions. *Atmospheric Environment* 34, 5169–5174.
- Pruppacher, H.R., Klett, J.D., 1978. *Microphysics of Clouds and Precipitation*. Reidel, Boston, MA 714pp.
- Seinfeld, J.H., Pandis, S.N., 1998. *Atmospheric Chemistry and Physics*. Wiley, New York 1326pp.
- Slinn, W.G.N., 1983. Precipitation scavenging. In: Raderson, D. (Ed.), *Atmospheric Sciences and Power Production*. Division of Biomedical Environmental Research, US Department of Energy, Washington, DC (Chapter 11).
- Tinsley, B.A., Rohrbaugh, R.P., Hei, M., 2001. Electroscavenging in clouds with broad droplet size distributions and weak electrification. *Atmospheric Research* 59–60, 115–135.
- Zhang, L.M., Michelangeli, D.V., Taylor, P.A., 2004. Numerical studies of aerosol scavenging by low-level, warm stratiform clouds and precipitation. *Atmospheric Environment* 38, 4653–4665.
- Zhao, H.B., Zheng, C.G., Xu, M.H., 2005a. Multi-Monte Carlo method for general dynamic equation considering particle coagulation. *Applied Mathematics and Mechanics* 26, 953–962.
- Zhao, H.B., Zheng, C.G., Xu, M.H., 2005b. Multi-Monte Carlo method for coagulation and condensation/evaporation in dispersed systems. *Journal of Colloid and Interface Science* 286, 195–208.
- Zhao, H.B., Zheng, C.G., Xu, M.H., 2005c. Multi-Monte Carlo approach for general dynamic equation considering simultaneous particle coagulation and breakage. *Powder Technology* 154, 164–178.