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Research Article

Study on the Interaction between Modes of a Nanoparticle-Laden Aerosol System

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Nanoparticle-laden two-phase flow systems, especially atmospheric aerosols, are usually found with several modes for particle size distribution (PSD). For the first time, a mathematical method is proposed to study the interaction of nanoparticle dynamics between modes by establishing two joint population balance equations (PBEs). The PBEs are solved using the sectional method, which divides the PSD into discrete bins. The nanoparticle-laden system involves Brownian coagulation, ventilation, and injection. The interaction between modes within a size distribution is studied quantitatively with and without injection and ventilation. The study shows that particles with smaller size are easier to be removed by background particles, but the lag time to be removed is affected by not only the total number concentration of small particles but also their sizes. Background particles play an important role in determining the evolution of small particle system, whose presence makes the secondary model absent for the small particles.

1. Introduction

Nanoparticle-laden two-phase flows exist in lots of natural and chemical engineering processes, such as atmospheric aerosols [1], gels in colloid [2], nanoparticle synthesis [3–5], and combustion reactors [6, 7]. In these processes, the evolution of system, especially the particle size distribution (PSD), is determined by more than one dynamics [3]. Regardless of a closed or an open nanoparticle-laden system, multiple dynamics lead to more than one mode for the PSD [8, 9]. Each of these modes mainly comes or is determined by one dynamical process; the interaction among these modes dominates the evolution of the whole system. Therefore, the study on the interaction of dynamics between modes rather than focusing on the whole system becomes necessary.

Since first proposed by von Smoluchowski in 1917 [4], the Smoluchowski mean-field theory has become the main theory to investigate the evolution of nanoparticle size distribution. In this theory, the key is to establish the population balance equation (PBE) according to particular dynamics [5]. Traditionally, PBE is established for the whole size distribution rather than a special part of the whole size distribution. Thus, the traditional PBE cannot meet the requirement of distinguishing modes within a system. Recently, Yu and Chan proposed a method to establish two joint PBEs, each of PBEs accounting for one mode [6]. In this method, the system is artificially divided into two subsystems; the transfer of mass between modes is reserved. Using this way, they studied the evolution of a bimodal size-distributed system and claimed that this method has capability to study the interaction of modes. Unfortunately, in their study, the PBEs are solved using the Taylor-series expansion method of moments (TEMOM). All methods of moments (MOM) have a common shortcoming; that is, the size-resolved information of PSD cannot be resolved or cannot be grasped. Thus, in Yu and Chan's study, some important information about the interaction between the modes might be lost. Similar studies can be also found in [7, 10].

In our surroundings, atmospheric aerosol is one of the most common nanoparticle-laden two-phase systems. Lots of works have been carried out for the study of nanoparticle evolution of PSD determined by various dynamical processes, including both experiments and numerical simulations [11–13]. Depending on the number of dynamical processes involved, atmospheric aerosols usually exhibit two or more modes. For atmospheric aerosols, three key processes, namely, Brownian coagulation, ventilation, and injection, are usually involved. In mathematics, these three processes can be easily represented by existing models. Thus, the atmospheric aerosol, especially in an open homogeneous isotropic chamber, is usually selected by scientists as an ideal system for the comparative study of PSD [8, 13]. By selecting an aerosol inside a homogeneous chamber as investigated objective, Liu et al. studied the evolution of nanoparticle size distribution due to continuous injection using the sectional method (SM) [8]. However, in their study, the interaction of dynamics between modes is not involved.

PBE is a nonlinear integral-differential equation, which has no analytical solutions as real coagulation kernels are selected [9]. Until now, three main methods have been proposed and used for the solution of PBE, namely, the SM, MOM [11, 14–18], and Monte Carlo (MC) method. These methods have both advantages and disadvantages regarding accuracy and efficiency and are used in PBE studies according to specific requirements [12]. Among all the numerical methods, the SM is regarded as the most suitable method for studying PSD without losing size-distributed information but consumes much computational costs. In this work, we applied the SM to the solution of PBE involving two modes mainly because in some aerosol systems there are usually two or more modes, and thus, the interaction between modes inevitably exists. We separate the aerosol system into two subsystems each representing one mode; therefore, we can investigate the interaction between modes. We used the SM to solve population balance equation, and thus, the size-resolved particle distribution can be exactly traced with time. In fact, the scheme separating the size distribution into two parts has been used by Yu and Chan in the implementation of the Taylor-series expansion method of moments [6], which has been verified in their work.

The remainder of this paper is organized as follows. The mathematical forms of two joint PBEs as well as their numerical solutions using the SM are analyzed in Section 2. The specifications of the initial conditions for comparison are outlined in Section 3. The results and discussion are presented in Section 4.

2. Theory and Model

In this work, only two modes are considered in a size distribution for simplification, in which the nanoparticle-laden system is artificially separated into two subsystems. Similar to Liu et al.'s work [8], in which one accounts for small particles (SPs) originating from source or for small particle mode and another for background particles (BPs). For each subsystem, it needs to establish its corresponding particle general dynamic equation based on the Smoluchowski mean-field theory. In order to make the mathematical equations approach to real physical phenomenon as much as possible, the transfer of particle between two systems is considered, which is similar to the work [6]. Because the particle size distribution of BP system is little influenced by the existence of small particles and especially there is very large difference between background and source particles in their sizes, a novel disposition is proposed here in which particles belonging to SP system are considered to become those belonging to BP system once these small particles are attached to background particles. Therefore, in theory, the mass of SP system always decreases with time while the total mass of the whole system is conserved if there is no ventilation and no source.

By including coagulation, injection, and ventilation, the particle general dynamic equation for SP system is represented as follows:

$$\frac{\partial n(v,t)}{\partial t} = \underbrace{\frac{1}{2}\int_0^v \beta(v-v',v')n(v-v',t)n(v',t)dv' - n(v,t)\int_0^\infty \beta(v,v')n(v',t)dv'}_{\text{Coagualtion within SP mode}}$$
$$\underbrace{-n(v,t)\int_0^\infty \gamma(v,v')p(v',t)dv'}_{\text{SP mode attached to BP mode}} \underbrace{-\frac{F_{\text{out}}}{V}n(v,t)}_{\text{Ventilation}} \underbrace{+\frac{f(v,t)F_{\text{in}}}{V}}_{\text{Injection}}, \quad (1)$$

where $n(v, t)dv$ is the particle number of SP system whose volume is between v and $v + dv$ at time t and $\beta(v, v')$is the collision kernel for two particles of volumes v and v'. On the right hand of (1), the first two terms account for particle number increase and decrease due to interparticle collision and join together within the SP system, while the third term serves as particle loss due to collision between SP system and BP system. The last two terms account for particle loss and increase due to ventilation and source production, respectively. $p(v', t)\mathrm{d}v'$ is the particle number between v' and $v' + \mathrm{d}v'$ for BP system. $f(v, t)$ is the source number concentration intensity with volume v and time t.

Similarly, the particle general dynamic equation for BP system is as follows:

$$\frac{\partial p(v,t)}{\partial t} = \underbrace{\frac{1}{2}\int_0^v \beta(v-v',v')p(v-v',t)p(v',t)dv' - p(v,t)\int_0^\infty \beta(v,v')p(v',t)dv'}_{\text{Coagulation within BP mode}}$$
$$+\underbrace{\int_0^v \gamma(v-v',v')n(v-v',t)p(v',t)dv' - p(v,t)\int_0^\infty \gamma(v,v')n(v',t)dv'}_{\text{Interaction between SP and BP mode}} \quad (2)$$
$$\underbrace{-\frac{F_{\text{out}}}{V}p(v,t)}_{\text{Ventilation}}.$$

Equations (1) and (2) comprise a governing equation describing nanoparticle evolution due to coagulation, ventilation, and injection for the first time. By introducing sectional method, the above two equations can be written as

$$\frac{dN_k}{dt} = \omega_k - N_k\sum_{i=1}\gamma_{ik}P_i - \frac{F_{\text{out}}}{V}N_k + F_{\text{in}}\frac{F_k}{V},$$
$$\frac{dP_k}{dt} = \epsilon_k + \sum_{\substack{i=1\\j=1}}\eta_{ijk}\gamma_{ij}N_iP_j - P_k\sum_{i=1}\gamma_{ik}N_i - \frac{F_{\text{out}}}{V}P_k, \quad (3)$$

with

$$\omega_k = \begin{cases} -N_1\sum_{i=1}\beta_{i1}N_i, & k=1,\\ \frac{1}{2}\sum_{\substack{i=1\\j=1}}\chi_{ijk}\beta_{ij}N_iN_j - N_k\sum_{i=1}\beta_{ik}N_i, & k>1,\end{cases}$$

$$\epsilon_k = \begin{cases} -P_1\sum_{i=1}\beta_{i1}N_i, & k=1,\\ \frac{1}{2}\sum_{\substack{i=1\\j=1}}\chi_{ijk}\beta_{ij}P_iP_j - P_k\sum_{i=1}\beta_{ik}P_i, & k>1,\end{cases} \quad (4)$$

$$\chi_{ijk} = \begin{cases} \frac{v_{k+1}-(v_i+v_j)}{v_{k+1}-v_k}, & \text{if} \quad v_k \le v_i+v_j \le v_{k+1},\\ \frac{(v_i+v_j)-v_{k-1}}{v_k-v_{k-1}}, & \text{if} \quad v_{k-1} \le v_i+v_j \le v_k,\\ 0, & \text{else.}\end{cases}$$

In order to simplify the program code, the above two systems use the same division of particle size. So, the coagulation rate $\gamma_{ij} = \beta_{ij}$, and also the collision coefficient $\chi_{ijk} = \eta_{ijk}$.

In this study, the particle loss due to deposition was described by Lai and Nazaroff's model [13], and the coagulation kernel was from Fuchs model [19].

3. Computations

The numerical computations were all performed on an Intel (R) Pentium 4 CPU 3.00 GHz computer with memory 4 GB. The 4-order Runge–Kutta method with fixed time step was used to solve the set of ordinary differential equations, and the error function in the SM model was computed using the incomplete gamma function method. In all the computations, the time step for the SM model was set to be 27 s. All the programs were written by the C Programming language and were performed on Microsoft Visual C++ 6.0 compiler. In all the simulations, the temperature and pressure of the surrounding air were assumed to be 300 K and 1.013×105 Pa, respectively. At the condition, the viscosity and the mean free path of gas molecules were 1.85×10^{-5} Pa·s and 68.41 nm, respectively. The morphology of the nanoparticle was assumed to be spherical or fractal-like and its bulk density is 1,000 kg/m^3.

4. Analysis and Results

4.1. Evolution of NSD of a Bimodal Aerosol. In theory, large particles have an ability to scavenge SP with large efficiency [19]. In this section, the interaction between BP and SP modes is investigated as only coagulation is considered. The characteristics of particles at BP and SP modes are shown in Table 1 where the BP mode is fixed, but the SP mode is changed in its GMD.

In Figure 1, two aerosols are considered: one is composed by the BP mode and SP mode (1), while the second is composed by the BP mode and SP mode (2). In the evolution of NSD, it shows that the BP mode changes negligibly, but SP modes change largely, and especially, the particle size in the SP mode moves in the large size direction. Due to the scavenge effect of the BP mode on the SP mode, particles with smaller size (SP mode (1)) in the SP mode are found to be easier to be removed. It needs to be noted here larger difference of two modes in SP mode (1) and in SP mode (2). From the comparison of two different aerosols, it is found that the larger the difference of two modes in their size, the easier to be scavenged for particles at the SP mode. This finding is further validated in Figure 2 where three aerosols with different sources (1–3) and with the same BP mode are investigated. In Figure 2, it shows that the total particle number concentration decreases

Table 1: Characteristics of both BP and SP modes.

Mode	TPN (#/m^3)	GMD (nm)	GSD	Df	F_{out} (lpm)	F_{in} (lpm)	Deposition (friction velocity, cm/s)
BP	4.34×10^{10}	500	1.32	3.0	0.0	0.0	0.0
SP (1)	4.34×10^{12}	100	1.32	3.0	0.0	0.0	0.0
SP (2)	4.34×10^{12}	50	1.32	3.0	0.0	0.0	0.0
SP (3)	4.34×10^{12}	24	1.32	3.0	0.0	0.0	0.0

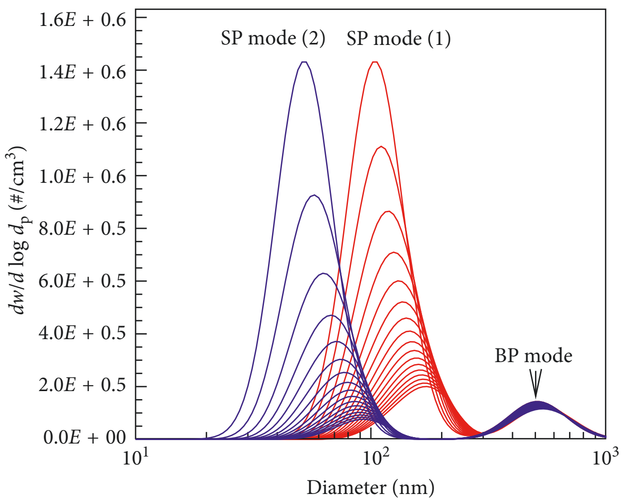

Figure 1: Evolution of NSD with calculated interval time of 11.25 min. Red lines represent particle size distribution of aerosols composed by the BP mode and SP mode (1), and blue lines represent particle size distribution of aerosols composed by the BP mode and SP mode (2).

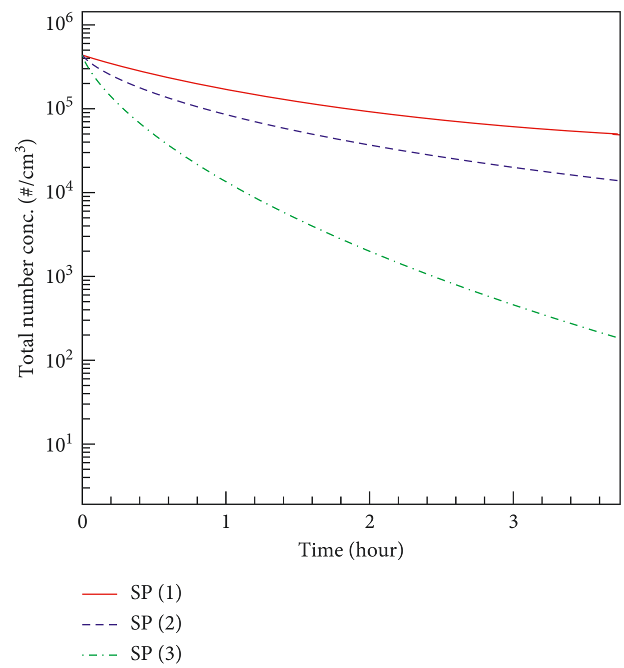

Figure 2: Change of total particle number concentration of small modes in the presence of large mode. At the same large mode, the total particle number concentration decreases more quickly for small modes with smaller size than those with larger size by BP mode and SP mode (2).

more quickly for SP modes with smaller size than those with larger size.

4.2. A Source Injected into a Chamber Filled with Background Particles. There have been some important results available in experimental studies of references for a source injected continuously to background particles [20, 21]. Here, the evolution of NSD in the presence of background particles and the interaction between modes are further investigated based on the simulation method. In this study, both initial background particles and sources take lognormal distributions, and the background particle size distribution is characterized by its GMD of 500 nm, TPN of 4.34×10^{10} #/m^3, and GSD of 1.32. The source acts as an injection to the background particles with constant flow rate of 4.6 lpm, and the source parameters are shown in Table 2. In this study, only coagulation and the injection of the source are considered.

The comparison of NSD with and without background particles is conducted in this work. Figure 3 shows the evolution of particle size distribution with interval time of 225 min for source (1) with and without background particles. It shows that the presence of background particles reduces the particle number concentration in the SP mode but has a negligible effect on the particle size where the SP mode peak is located. In addition, the presence of background particles also has an effect on the appearance of the secondary mode. In this study, it shows that in the presence of background particles, the secondary mode does not appear, while the secondary mode emerges in the case without background particles. The conclusion cannot be obtained using the bimodal TEMOM, in which the details of PSD are lost when calculating [19].

In order to study the evolution of NSD for sources with different GMD at the same background particles, the calculation for sources 1, 2, and 3 is performed, which is shown in Figure 4. From the comparison, it is found that the source GMD is nearer to the size of BP, and the particle number concentration for the SP mode is higher. This should attribute to the coagulation rate which is increased with the increased difference between particles. The above conclusion can be further validated in Figure 5 where the comparison of TPN of SP mode is represented for sources 1, 2, and 3. It shows that the TPN increases with the increase of source GMD. This validates the conclusion that the background particles have larger scavenge effect on smaller particles.

Table 2: Parameters used in (3) for sources.

Source	TPN (#/m^3)	GMD (nm)	GSD	Df	F_{out} (lpm)	F_{in} (lpm)	Deposition (friction velocity, cm/s)
Source (1)	4.34×10^{12}	100	1.32	3.0	0.0	4.6	0.0
Source (2)	4.34×10^{12}	50	1.32	3.0	0.0	4.6	0.0
Source (3)	4.34×10^{12}	24	1.32	3.0	0.0	4.6	0.0

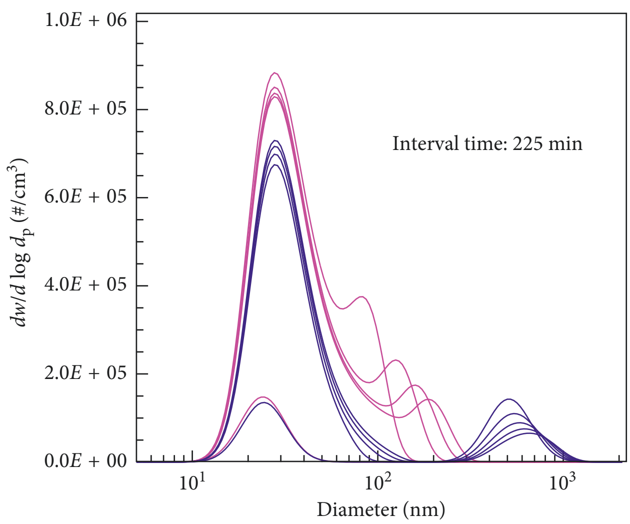

Figure 3: Comparison of NSD with interval time of 225 min for source (1) in Table 2 with (blue) and without background particles (purple). In the calculation, only coagulation is considered.

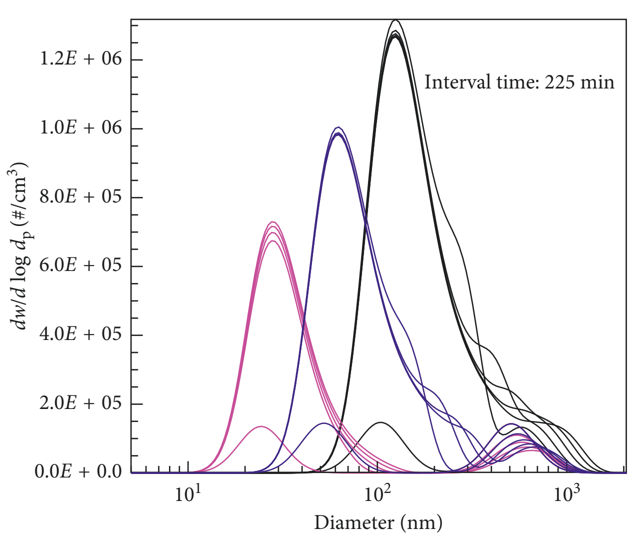

Figure 4: Comparison of NSD with interval time of 225 min for sources (1, 2, and 3) (black, blue, and purple) at the same background particles with its GMD of 500 nm, TPN of 4.34×10^{10} #/m^3, and GSD of 1.32. Although all three sources have the same TPN, the particle number concentration for SP mode at steady state is increased with the increase of source GMD.

4.3. Scavenging Effect of Background Particles on Small Particles. Although it is easy to conclude that background particles have larger scavenge effect on SP from the above calculation, and it is also necessary to know how much time it takes for small particles to be removed by background

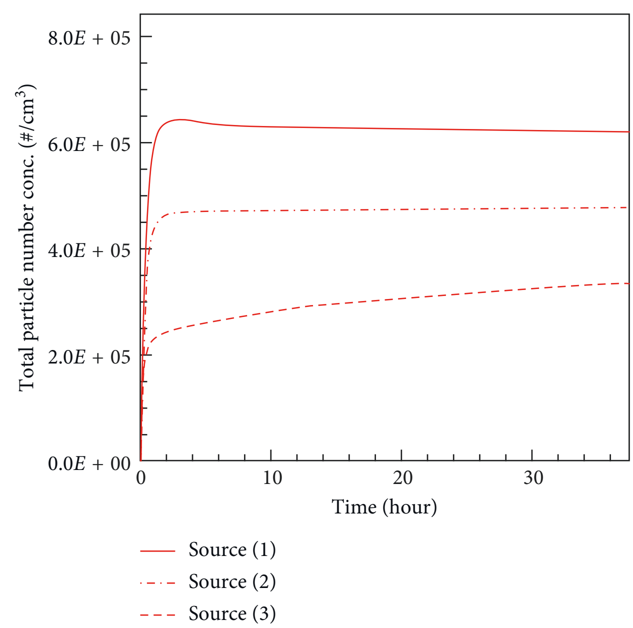

Figure 5: Comparison of total particle number concentration of the SP mode for sources (1–3). The total particle number concentration of the SP mode is increased with the increase of source GMD.

particles. This is especially important in the studies on the establishment of environmental standards. In this study, the BP mode takes a lognormal distribution with GMD of 500 nm, TPN of 4.34×1.0^{11}#/m^3, and GSD of 1.32, and SP modes also take lognormal distributions whose parameters are listed in Table 3. In this study, we study the effect of SP total particle number concentration and GMD on the lag time to be removed. Once the SP total particle number concentration is below 1.00×10^3#/cm^3, the SP nanoparticles are considered to be absolutely removed.

Table 3 shows the lag time for the SP mode to achieve its total particle number concentration of 1.00×10^3#/cm^3. Even at the same SP initial total particle number concentration, there is a large difference in lag time to achieve the number concentration of 1.00×10^3#/cm^3. When the SP initial total particle number concentration is increased from 4.34×1.0^{12} to 4.34×1.0^{13}#/cm^3, the lag time also increases by a factor of 2.38, 3.91, and 3.84 when GMD is 100, 50, and 24 nm. Therefore, the lag time to be removed for small particles is not directly proportional to the initial SP number concentration. The size of small particles is also an important indicator to evaluate how far small particles are removed by background particles.

Figure 6 shows the change of total particle number concentration of SP mode at different initial SP mode and the same BP mode. At the same total source particle number

Table 3: Lag time for SP mode to achieve its total particle number concentration of 1.0^3 #/cm^3.

Mode	TPN (#/m^3)	GMD (nm)	GSD	Df	F_{out} (lpm)	F_{in} (lpm)	Lag time
BP	4.34×10^{11}	500	1.32	3.0	0.0	0.0	—
SP (1)	4.34×10^{12}	100	1.32	3.0	0.0	0.0	9.00
SP (2)	4.34×10^{12}	50	1.32	3.0	0.0	0.0	2.34
SP (3)	4.34×10^{12}	24	1.32	3.0	0.0	0.0	0.44
SP (4)	4.34×10^{13}	100	1.32	3.0	0.0	0.0	21.44
SP (5)	4.34×10^{13}	50	1.32	3.0	0.0	0.0	9.17
SP (6)	4.34×10^{13}	24	1.32	3.0	0.0	0.0	1.69

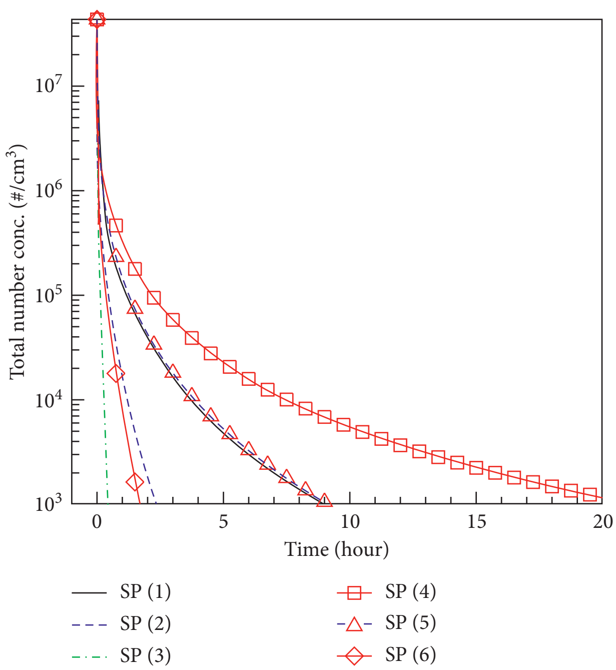

Figure 6: Change of total particle number concentration of SP mode at different initial SP mode and the same BP mode.

concentration, the small particles need less time to be removed with smaller size. For the same GMD, higher initial SP total particle number concentration needs longer time to be removed.

5. Conclusions

In this article, two joint population balance equations are first established for investigating the interaction between modes within nanoparticle-laden aerosol systems. In the joint equations, mass transfer between modes is reserved. The population balance equations are solved by the sectional method. The study mainly involves the effect of background particles on the evolution of small particles and the lag time for the small particles to be removed. It shows that the particles with smaller size are easier to be removed by background particles, and especially, the lag time to be removed is related not only to the number concentration of small particles but also to their sizes. In the presence of background particles, no secondary mode is found for the small particle system during its evolution.

Abbreviations

NSD: Nanoparticle size distribution
PBE: Population balance equation
GSD: Geometric standard deviation
GMD: Geometric mean diameter
PSD: Particle size distribution
NGDE: Nanoparticle general dynamics equation
TPN: Total particle number concentration
SM: Sectional method
ODE: Ordinary differential equation
BPs: Background particles
SPs: Small particles.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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