



## Technical note

# A new analytical solution for solving the population balance equation in the continuum-slip regime



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## ABSTRACT

A new analytical solution is first proposed to solve the population balance equation due to Brownian coagulation in the continuum-slip regime. An assumption for a novel variable  $g$  ( $g = m_0 m_2 / m_1^2$ , where  $m_0$ ,  $m_1$  and  $m_2$  are the first three moments, respectively) is successfully applied in executing a separate variable method for ordinary differential equations of the Taylor expansion method of moments. The sectional method is selected as a reference to verify the new solution. The accuracy between the new solution and Lee et al. analytical solution (Lee et al., 1997, *Journal of Colloid and Interface Science*, 188, 486–492) is mainly compared. The geometric standard deviation of number distribution for the new analytical solution is revealed to be limited to 1.6583. Within the valid range of the geometric standard deviation, the new analytical solution is confirmed to solve the population balance equation undergoing Brownian coagulation with the very nearly same accuracy as Lee et al. analytical solution. For the total particle number concentration, the new solution usually yields higher accuracy. The new solution and Lee et al. analytical solution approximately become one solution as the Knudsen number is smaller than 0.1000. The new solution has the potential to become a competitive analytical solution for solving population balance equation regarding its accuracy and very straightforward derivation.

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## 1. Introduction

A reliable prediction for aerosol properties including the total particle number concentration, the geometric mean size and the geometric standard deviation of number distribution has received much attention in emerging fields such as the risk evaluation of aerosols at workplace, the development of realistic exposure scenarios and the nanoparticle synthesis process (Buesser & Pratsinis, 2012; Vogel et al., 2014; Yu et al., 2008a, 2008b). For these aerosols, the evolution of particle size distribution due to Brownian coagulation is unavoidable (Lee & Wu, 2005; Upadhyay & Ezekoye, 2003), which usually leads to unsteady systems and has been confirmed to determine the aerosol characteristics in almost all ultrafine and nanoparticle processes (Crowe et al., 2011; Friedlander, 2000). When these processes are theoretically investigated, the evolution of the size distribution must be captured in mathematical models (Buesser & Pratsinis, 2012; Xie & Wang, 2013;

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Nomenclature			
$A$	constant (= 1.591)	$N$	kinematic viscosity, $\text{m}^2 \text{s}^{-1}$
$r$	particle radius, m	$B$	particle collision kernel
$N$	particle number concentration density, $\text{m}^{-3}$	$M$	gas viscosity $\text{kg m}^{-1} \text{s}^{-1}$
$B_2$	collision coefficient for the continuum-slip regime	$\lambda$	mean free path of the gas, m
$C$	Cunningham correction factor	$\sigma_g$	geometric mean deviation of size distribution
$k_b$	Boltzmann constant, J K	$\tau$	dimensionless coagulation time, $tN_0B_2$
Kn	particle Knudsen number	<i>Abbreviation</i>	
$m_k$	$k$ th moment of particle size distribution	PBE	population balance equation
$g$	$m_0m_2/m_1^2$	TEMOM	Taylor expansion method of moments
$M_k$	dimensionless $k$ th moment of size distribution	GSD	geometric standard deviation of number distribution
$T$	time, s	ODE	ordinary differential equation
$T$	temperature, K	SM	sectional method
$U$	the point of Taylor-series expansion ( $m_1/m_0$ )	QMOM	quadrature method of moments
$v$	particle volume, $\text{m}^3$	SPSD	self-preserving size distribution
$v_g$	geometric mean particle volume, $\text{m}^3$	PSPSD	pseudo-self-preserving size distribution
$N$	initial total particle number concentration, $\text{m}^{-3}$	Log-normal AMM	log-normal analytical method of moments
<i>Greek letters</i>		Log-normal NMM	log-normal numerical method of moments

Santos et al., 2013; Singh et al., 2013). To meet the requirement, Müller established the integral-differential equation for the dynamical process in 1928 based on the ground-breaking work of Smoluchowski (Müller, 1928; Smoluchowski, 1917), which was later called Population Balance Equation (PBE). The PBE has become a basic governing equation to study aerosol dynamics from then on. However, the analytical solution of this equation, especially in terms of a particle size dependent coagulation kernel, still remains a challenging issue.

The PBE is a strong non-linear equation with the same mathematical structure as Boltzmann's transport equation. Thus, an exact analytical solution for it cannot be achieved (Lee et al., 1997; Yu et al., 2008a, 2008b). To solve it analytically, the group of Prof. Lee in Kwangju Institute of Science and Technology, Korea has performed a series of ground-breaking works in different specific-size regimes with a log-normal distribution assumption (Lee et al., 1997, 1984; Otto et al., 1999; Park et al., 1999). These works received much attention because of their ability to capture the evolution of the size distribution. Another solution deserved to be mentioned to solve the PBE was proposed in 1964 by introducing a similarity transformation in the size distribution function (Swift & Friedlander, 1964), which is actually an asymptotic solution independent of the initial size distribution. The idea in the asymptotic solution was currently accepted in studies on Brownian coagulation processes using the Taylor expansion method of moments (TEMOM) (Chen et al., 2014a; Xie & Wang, 2013). In both the free molecular and continuum regimes, asymptotic solutions exist because the asymptotic status for the size distribution, i.e., self-preserving size distribution (SPSD), has been verified in both the regimes (Friedlander, 2000). However, in the continuum-slip regime, especially as the Knudsen number ranges from  $\sim 0.1000$  to  $\sim 5.0000$  (also called the near-continuum regime), the geometric standard deviation (GSD) of number distribution always varies with the Knudsen number (Otto et al., 1994; Park et al., 1999; Yu et al., 2011). In this case, the asymptotic solution will no longer exist. In fact, the asymptotic solution has a fatal shortcoming in that it is not able to capture the evolution of size distribution for the time period before the self-preserving size distribution is achieved (Lee et al., 1997). Therefore, an alternative solution beyond the asymptotic status and without the requirement for the pre-defined size distribution becomes necessary.

The TEMOM exhibits a huge potential to achieve the time-dependent analytical solution for the PBE due to its very simple mathematical structure of equations (Chen et al., 2014b; Xie et al., 2012; Yu et al., 2011; Yu & Lin, 2009a, 2009b). The key of the TEMOM is that fractal moments in the ordinary differential equations (ODEs) for moments can be replaced by the following expression (Yu et al., 2008a, 2008b):

$$m_k = \left( \frac{u^{k-2}k^2}{2} - \frac{u^{k-2}k^2}{2} \right) m_2 + \left( -u^{k-1}k^2 + 2u^{k-1}k \right) m_1 + \left( u^k + \frac{u^k k^2}{2} - \frac{3u^k k}{2} \right) m_0 \quad (1)$$

where  $u$  is the Taylor expansion point,  $k$  is the fractal number, and  $m_0$ ,  $m_1$  and  $m_2$  are the first three moments. The zeroth and the first moments represent the total particle number concentration and the total volume concentration, respectively, while the second moment is a polydispersity variable. In the continuum-slip regime, the TEMOM (Yu et al., 2011) can be used for the Knudsen number up to 5.0000 (Lee et al., 1997). This method is valid for particles with diameter larger than  $\sim 27$  nm; thus, this method can be used to resolve almost all Brownian coagulation issues, as shown in Fig. 1. The TEMOM ODEs have been successfully numerically solved using a highly reliable Runge–Kutta algorithm (Yu et al., 2011).

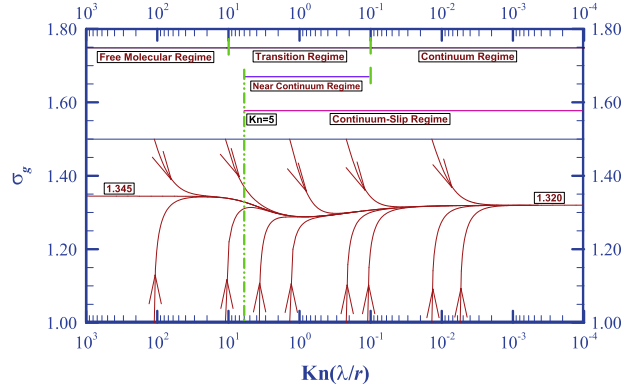


Fig. 1. Variance of geometric standard deviation of number distribution ( $\sigma_g$ ) with Knudsen number over the entire size regime.

For an aerosol solely dominated by Brownian coagulation, the SPSSD with constant GSDs can be quickly achieved in both the free molecular regime and the continuum regime (Friedlander, 2000; Otto et al., 1999). Even in the entire size regime, the GSD of aerosol size distribution only varies in a notably small range (Park et al., 1999). It is believed that the novel property of GSD was the key in the derivation of the asymptotic solutions and the log-normal analytical solutions. The TEMOM ODEs in the continuum-slip regime revealed that these equations can be further represented by the first three moments along with a new variable  $g (= m_0 m_2 / m_1^2)$ . Based on a mathematical analysis, the variable  $g$  was verified to vary in a notably small limited range too, which makes it possible for treating  $g$  as a constant to simplify the TEMOM ODEs. In this case, the PBE in the continuum-slip regime is expected to be solved analytically.

In conclusion, in this work, a new analytical solution will be proposed to solve the PBE due to Brownian coagulation in the continuum-slip regime. The newly proposed analytical solution is notably different from the existing asymptotic solutions and the log-normal analytical method of moments. The newly proposed analytical solution will be verified by selecting the reliable sectional method (SM) solution as a reference. Since the analytical solution proposed by Lee et al. (1997) is usually regarded as the most suitable and reliable analytical solution for solving PBE in the continuum-slip regime, it is introduced in this work for a comparison. In addition, three numerical methods, including the TEMOM (Yu et al., 2008a, 2008b), the quadrature method of moments (QMOM) (McGraw, 1997), and the log-normal numerical method of moments (Lee et al., 1997), are also performed for comparisons. To make a clear difference, in this work, the log-normal numerical method of moments is called *log-normal NMM*, while the log-normal analytical method of moments is called *log-normal AMM*.

## 2. Theories

The integral-differential PBE was first proposed by Müller in 1928 based on the Smoluchowski ground-breaking work for coagulation dynamical process (Müller, 1928; Smoluchowski, 1917), and it takes the following expression:

$$\frac{\partial n(v, t)}{\partial t} = \frac{1}{2} \int_0^v \beta(v-v', v') n(v-v', t) n(v', t) dv' - n(v, t) \int_0^\infty \beta(v, v') n(v', t) dv' \quad (2)$$

where  $n(v, t)dv$  is the particle number 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  $v'$ . In the continuum-slip regime, the collision kernel has the following form:

$$\beta(v, v') = B_2 \left( \frac{C(v)}{v^{1/3}} + \frac{C(v')}{v'^{1/3}} \right) (v^{1/3} + v'^{1/3}) \quad (3)$$

where  $B_2 = 2k_b T / 3\mu$ ,  $k_b$  is the Boltzmann constant,  $T$  is the gas temperature, and  $\mu$  is the gas viscosity. The slip correction factor,  $C(v) = 1 + A \text{Kn}$ , is used to accommodate the gas slip effects on small particles, where  $A = 1.591$  and  $\text{Kn} = \lambda/r$ , which is expected to make the collision kernel to be valid for the Knudsen number up to about 5.0000 (Otto et al., 1999). Here,  $\lambda$  is the mean free path of gas and  $r$  is the particle radius. In this case, the ODEs for moments, which are obtained using a Taylor expansion technique, take the following form:

$$\begin{cases} \left( \frac{dm_0}{dt} \right)_{cs} = B_2 \left\{ \frac{(-151m_1^4 + 2m_2^2 m_0^2 - 13m_2 m_1^2 m_0) m_0^2}{81m_1^4} + \frac{\phi m_0^{7/3} (5m_2^2 m_0^2 - 64m_2 m_1^2 m_0 - 103m_1^4)}{81m_1^{13/3}} \right\} \\ \left( \frac{dm_1}{dt} \right)_{cs} = 0 \\ \left( \frac{dm_2}{dt} \right)_{cs} = -B_2 \left\{ \frac{2(-151m_1^4 + 2m_2^2 m_0^2 - 13m_2 m_1^2 m_0)}{81m_1^2} + \frac{4\phi m_0^{1/3} (m_2^2 m_0^2 - 2m_2 m_1^2 m_0 - 80m_1^4)}{81m_1^{7/3}} \right\} \end{cases} \quad (4)$$

Here,  $\varnothing = A\lambda(4\pi/3)^{1/3}$ . Equation (4) takes the following form when introducing  $g = m_0 m_2 / m_1^2$ :

$$\begin{cases} \left(\frac{dm_0}{dt}\right)_{cs} = B_2 \left\{ \frac{2g^2 - 13g - 151}{81} m_0(t)^2 + \frac{\varnothing m_1(t)^{-1/3} \{5g^2 - 64g - 103\}}{81} m_0(t)^{7/3} \right\} \\ \left(\frac{dm_1}{dt}\right)_{cs} = 0 \\ \left(\frac{dm_2}{dt}\right)_{cs} = B_2 \left\{ -\frac{2\{2g^2 - 13g - 151\}}{81} m_1(t)^2 - \frac{4\varnothing m_1(t)^{7/3} g^{1/3} \{g^2 - 2g - 80\}}{81} m_2(t)^{-1/3} \right\} \end{cases} \quad (5)$$

If Eq. (5) is further treated with a dimensionless solution,  $m_k = M_k m_{k0}$  and  $m_{k0} = N v_{g0}^k$ , then it has the following expression:

$$\begin{cases} \left(\frac{dM_0}{d\tau_2}\right)_{cs} = \left\{ \frac{2g^2 - 13g - 151}{81} M_0(\tau_2)^2 + \frac{(A \text{Kn}_0) M_1(\tau_2)^{-1/3} \{5g^2 - 64g - 103\}}{81} M_0(\tau_2)^{7/3} \right\} \\ \left(\frac{dM_1}{d\tau_2}\right)_{cs} = 0 \\ \left(\frac{dM_2}{d\tau_2}\right)_{cs} = \left\{ -\frac{2\{2g^2 - 13g - 151\}}{81} M_1(\tau_2)^2 - \frac{(4A \text{Kn}_0) M_1(\tau_2)^{7/3} g^{1/3} \{g^2 - 2g - 80\}}{81} M_2(\tau_2)^{-1/3} \right\} \end{cases} \quad (6)$$

where  $\tau_2 = B_2 N t$ ,  $\text{Kn}_0 = \lambda(4\pi/3)^{1/3} (v_{g0})^{-1/3}$ , and  $v_{g0}$  is the initial geometric mean volume of the investigated aerosol,  $N$  is the total initial particle number concentration. The GSD in the continuum-slip regime is only valid in the range from 1.0000 to 1.6583, as shown in Appendix A. By assuming  $g$  to be a constant, Eq. (6) can be analytically solved using a Separate Variable Technique for both  $M_0$  and  $M_2$  as follows:

(i)  $M_0$

$$\begin{aligned} t_0 = & -\frac{1}{A_c} \left( \frac{1}{M_0} - \frac{1}{M_{00}} \right) + \frac{3A_c}{2A_c^2} \left( \frac{1}{M_0^{2/3}} - \frac{1}{M_{00}^{2/3}} \right) - \frac{3A_{cm}^2}{A_c^3} \left( \frac{1}{M_0^{1/3}} - \frac{1}{M_{00}^{1/3}} \right) \\ & + \frac{A_{cm}^3 \left\{ -\ln \left( M_0^{2/3} A_{cm}^2 - A_{cm} A_c M_0^{1/3} + A_c^2 \right) + \ln \left( M_0 A_{cm}^3 + A_c^3 \right) + 2 \ln \left( M_0^{1/3} A_{cm} + A_c \right) - \ln(M_0) \right\}}{A_c^4} \\ & - \frac{A_{cm}^3 \left\{ -\ln \left( M_{00}^{2/3} A_{cm}^2 - A_{cm} A_c M_{00}^{1/3} + A_c^2 \right) + \ln \left( M_{00} A_{cm}^3 + A_c^3 \right) + 2 \ln \left( M_{00}^{1/3} A_{cm} + A_c \right) - \ln(M_{00}) \right\}}{A_c^4} \end{aligned} \quad (7.1)$$

(ii)  $M_0$

$$\begin{aligned} t_2 = & \frac{1}{B_c} (M_2 - M_{20}) - \frac{3B_{cm}}{2B_c^2} (M_2^{2/3} - M_{20}^{2/3}) + \frac{3B_{cm}^2}{B_c^3} (M_2^{1/3} - M_{20}^{1/3}) \\ & - \frac{3B_{cm}^3 \left\{ \ln \left( B_c M_2^{1/3} + B_{cm} \right) - \ln \left( B_c M_{20}^{1/3} + B_{cm} \right) \right\}}{B_c^4} \end{aligned} \quad (7.2)$$

where  $t_0$  and  $t_2$  are the time, and

$$\begin{cases} A_c = \frac{2g^2 - 13g - 151}{81} \\ A_{cm} = \frac{A M_1(\tau_2)^{-1/3} \{5g^2 - 64g - 103\}}{81} \text{Kn}_0 \\ B_c = -\frac{2\{2g^2 - 13g - 151\}}{81} M_1(\tau_2)^2 \\ B_{cm} = -\frac{4A M_1(\tau_2)^{7/3} \{g^2 - 2g - 80\}}{81} \text{Kn}_0 \end{cases}$$

The above derivation is straightforward and does not involve any assumption for the particle size distribution. In particular, the zeroth moment and the second moment in Eq. (7) are both functions of time implicitly, making the new solution to be able to capture the evolution of aerosol dynamical process before approaching its asymptotic status, which is the same as the log-normal AMM. Thus, the newly proposed solution has the same ability as the log-normal AMM which extends beyond the asymptotic solution.

If the Knudsen number is smaller than 0.1000 where the slip correction factor,  $C(v)$ , can be approximated to be 1.0000, Eq. (7) automatically degrades to a much simpler form

$$\begin{cases} (M_0(\tau_2))_c = \frac{1}{-A_c \tau_2 + (1/M_{00})} \\ (M_1(\tau_2))_c = M_{10} \\ (M_2(\tau_2))_c = B_c \tau_2 + M_{20} \end{cases} \quad (8)$$

Here, the zeroth and second moments are explicit functions of time; thus, it is much easier to be used in practice. However, Eq. (8) has to be used for aerosols in the continuum regime rather than in the continuum-slip regime.

### 3. Computations

The numerical computations are all performed using an Intel (R) Core i7-3820 CPU 3.6 GHz computer with 4 GB of memory. The fourth-order Runge–Kutta method with a fixed time step of 0.0010 is used to numerically solve the set of ODEs, including the TEMOM ODEs, the log-normal NMM ODEs, and the QMOM ODEs. Note that the newly proposed analytical solution is derived from the TEMOM ODEs (Yu et al., 2011), while the log-normal AMM was derived from the log-normal NMM ODEs (Lee et al., 1997). For all numerical and analytical solutions, the total time is up to 100. All the programs are written using the C Programming language and performed with the Microsoft Visual Studio 2008 compiler. In all simulations, the temperature and the pressure of the surrounding air are assumed to be 300.0000 K and  $1.0130 \times 10^5$  Pa, respectively. In this case, the viscosity and the mean free path of gas molecules are  $1.8508 \times 10^{-5}$  Pa s and 68.4133 nm, respectively. The relative error of any variable in the investigated solutions to that in the referenced SM is calculated as follows (Yu et al., 2008a, 2008b):

$$\text{Error}\% = \frac{\phi_{NM} - \phi_{SM}}{\phi_{SM}} \times 100\% \quad (9)$$

Here,  $\phi_{NM}$  is the variable obtained from methods of moments, and  $\phi_{SM}$  is the referenced SM variable. In the calculation, all initial dimensionless moments take the same expressions as shown in Eq. (13) in Yu et al. (2014). The program code of the SM used in this work has been verified in our previous works (Anand et al., 2012; Yu & Lin, 2009b), which is generally considered as a very accurate solution for solving the PBE (Otto et al., 1999). In the SM, the section spacing factor is 1.025 and the bin number is 360, which ensures the accuracy of the SM as a reference. For the QMOM, four specific models with node 3, 4, 5 and 6 are performed and employed in comparisons.

### 4. Results and discussion

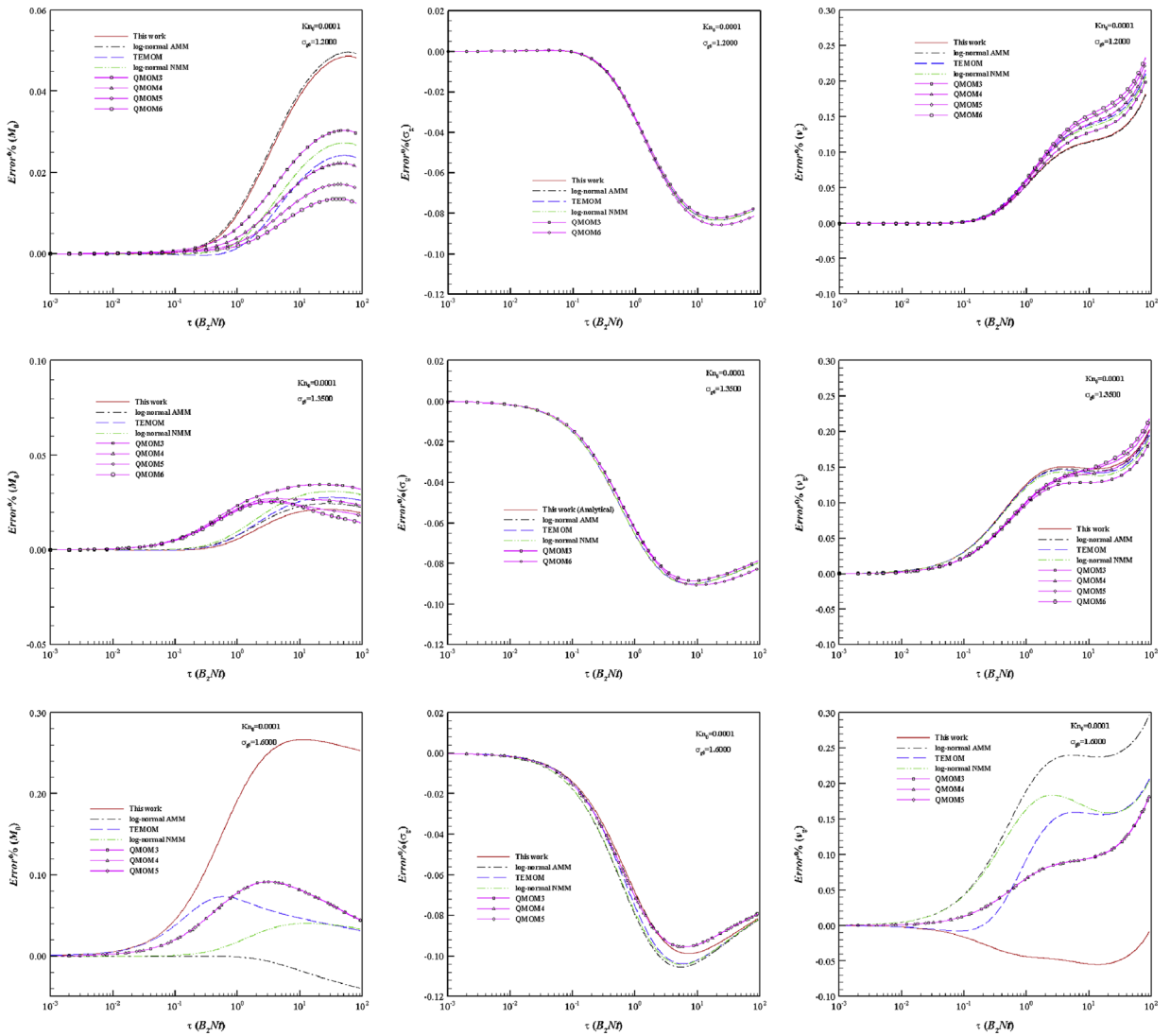
In theory, the geometric standard deviation of an aerosol can be an arbitrary value which is equal to or larger than 1.0000 (Yu et al., 2008a, 2008b). However, a drawback inheriting from the TEMOM ODEs makes the new solution to be confined from 1.0000 to 1.6583 for GSD, as presented in Appendix A. Therefore, the ability of the new analytical solution to solve the PBE must be evaluated, or the errors generated by the analytical solution must be specified. The accuracy of the newly proposed analytical solution can be characterized by the relative errors of the analytical solution to the referenced SM solution for the three key quantities, including  $M_0$ ,  $\sigma_g$  and  $v_g$ . To make the study much more representative, aerosols with three representative initial GSD, i.e., 1.2000, 1.3500, and 1.6000, and three representative initial Knudsen numbers, i.e., 0.0001, 0.1000 and 5.0000, are selected and investigated. The Knudsen number 5.0000 corresponds to the high limit of the continuum-slip regime in terms of aerosol geometric mean size, while 0.0001 corresponds to its low limit.

To verify the newly proposed analytical solution with very small Knudsen number, three aerosols with different initial GSDs, i.e., 1.2000, 1.3500, and 1.6000, are investigated, which correspond to the cases 1, 2 and 3 shown in Table 1. To make a clear comparison, other solutions including the log-normal AMM, the TEMOM, the QMOM and the log-normal NMM are also performed. Figure 2 shows the relative errors of  $M_0$ ,  $\sigma_g$  and  $v_g$  of the investigated solutions to the referenced SM solution, respectively. As the initial GSDs are selected to be 1.2000 and 1.3500, the new analytical solution yields higher accuracy than or nearly the same accuracy as the log-normal AMM for the three investigated key quantities. Especially, the newly proposed analytical solution shows a little advantage in accuracy for  $M_0$ . However, as the initial GSD is selected to be a larger value, i.e., 1.6000, the new solution shows disadvantage over all of other solutions. Fortunately, the newly proposed analytical solution yields clear higher accuracy than the log-normal AMM for the other two key quantities, i.e.,  $\sigma_g$  and  $v_g$ . It is thus concluded that the log-normal AMM and the new solution approximately become one solution when the aerosol is in the continuum regime and the GSD is not a high value. In fact, as the Knudsen number is 0.0001, the analytical solution shown in Eq. (7) automatically degrades to the form shown in Eq. (8) where the moments are explicit functions of time, which has been presented in Section 2.

When the Knudsen number is selected to be 0.1000, the investigated aerosol should be in the near continuum regime where the slip correction for coagulation kernel cannot be ignored (Yu & Lin, 2009b). In this regime, three representative

**Table 1**  
Characteristics of investigated size distribution cases with specific geometric standard deviation and Knudsen number.

Case number	Geometric standard deviation	Knudsen number
1	1.2000	0.0001
2	1.3500	0.0001
3	1.6000	0.0001
4	1.2000	0.1000
5	1.3500	0.1000
6	1.6000	0.1000
7	1.2000	5.0000
8	1.3500	5.0000
9	1.6000	5.0000

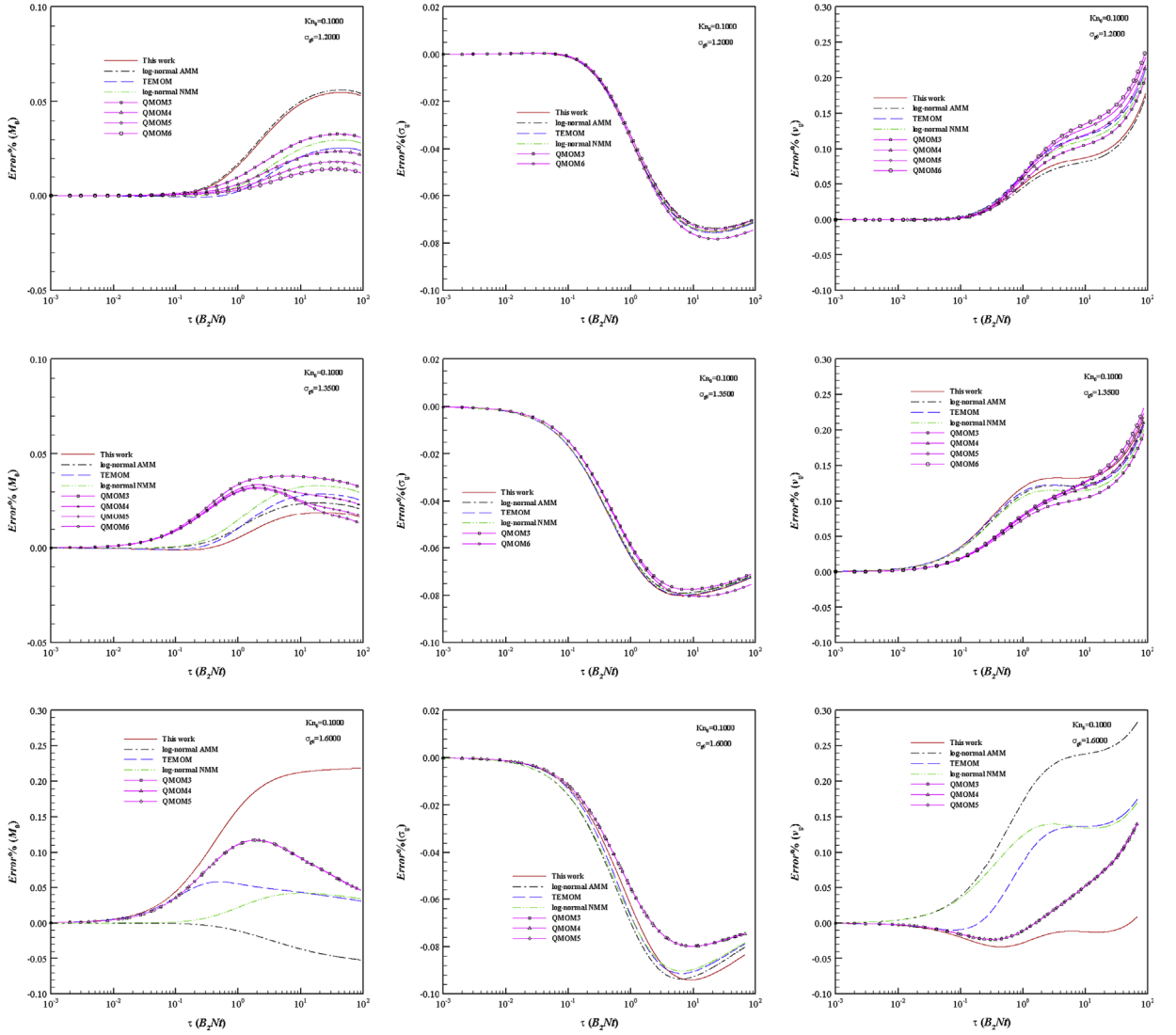


**Fig. 2.** The comparison of relative errors of  $M_0$ ,  $\sigma_g$  and  $v_g$  among the new solution, the log-normal AMM, the TEMOM, the QMOM and the log-normal NMM with initial Knudsen number 0.0001.

aerosols with different initial GSDs, i.e., 1.2000, 1.3500 and 1.6000, are selected, which correspond to the cases 4–6 shown in Table 1. Figure 3 shows the comparison of relative errors of  $M_0$ ,  $\sigma_g$  and  $v_g$  of different solutions to the referenced SM solution. It is clear when the initial GSDs are 1.2000 and 1.3500, the newly proposed analytical solution yields higher accuracy than the log-normal AMM for  $M_0$ . The newly proposed analytical solution has the same accuracy as the log-normal AMM for  $\sigma_g$  and has lower accuracy for  $v_g$ . As the initial geometric standard deviation is selected to be a larger value, 1.6000, the new analytical solution is found to yield a lower accuracy for  $M_0$  than the log-normal AMM. However, for the same numerical condition, the new solution yields higher accuracy for both  $\sigma_g$  and  $v_g$ .

Because of the coagulation kernel used in this work, the newly proposed analytical solution has to be applied to aerosols whose Knudsen number must be smaller than 5.0000 (Lee et al., 1997). Here, three representative aerosols with different initial GSDs, i.e., 1.2000, 1.3500 and 1.6000, are investigated for verifying the newly proposed analytical solution at the high Knudsen number limit. The selected aerosols correspond to the cases 7–9 shown in Table 1. The comparison of the relative errors of  $M_0$ ,  $\sigma_g$  and  $v_g$  of different solutions to the referenced SM solution is performed as shown in Fig. 4. For  $M_0$ , it is shown that the newly proposed analytical solution yields higher accuracy than the log-normal AMM, as the initial GSD is selected to be 1.2000 and 1.6000, whereas it yields lower accuracy as the initial GSD is 1.3500. For  $\sigma_g$  and  $v_g$ , the accuracy of the newly proposed analytical solution is found to be nearly same as or lower than the log-normal AMM.

In conclusion, from the joint evaluation of the three key quantities including  $M_0$ ,  $\sigma_g$  and  $v_g$  for the new solution and the log-normal AMM, it is not possible to determine which analytical solution yields higher accuracy. For any solution investigated in this work, its accuracy only prevails in one or two of the three key quantities. Actually, we also obtained the

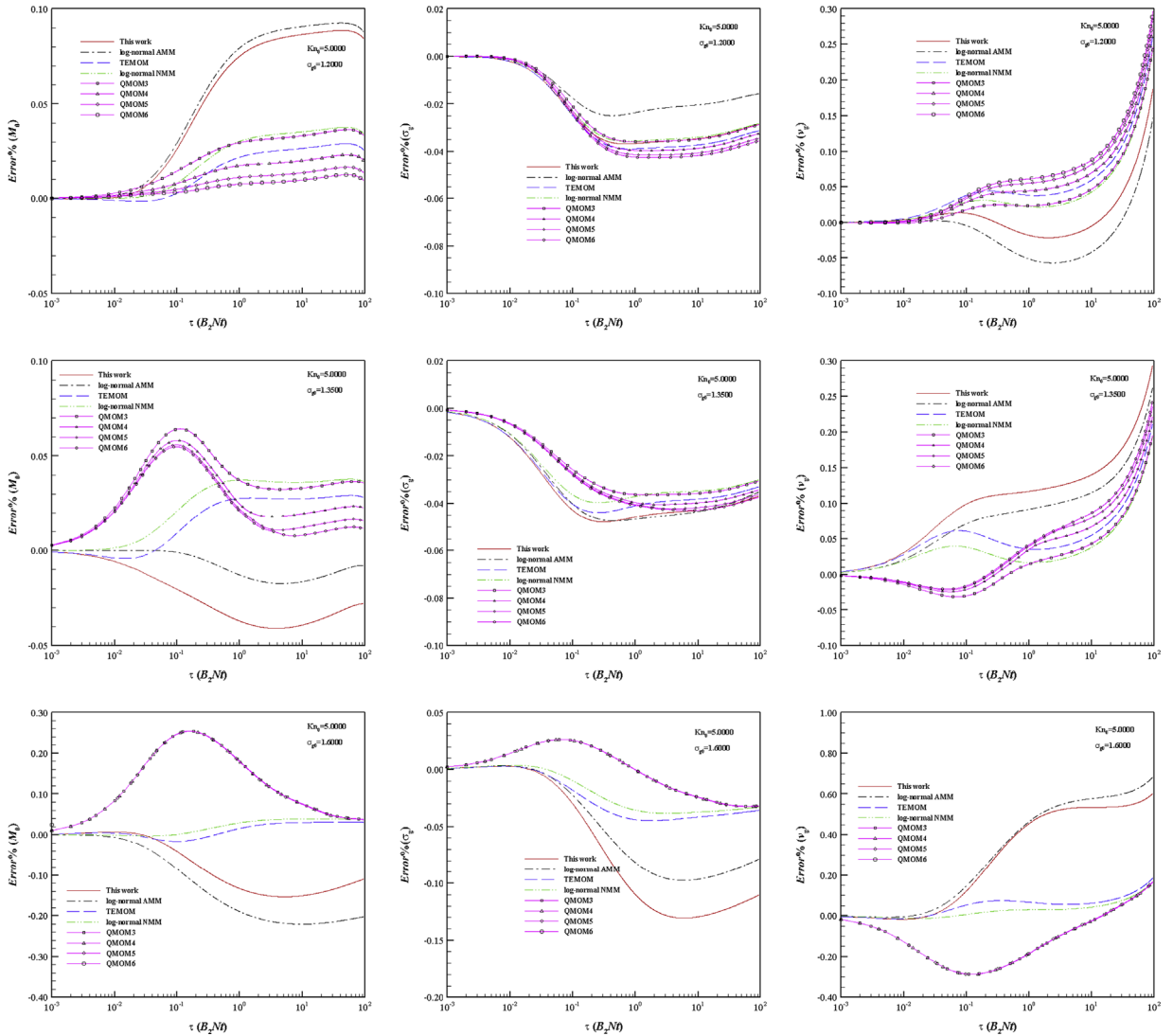


**Fig. 3.** The comparison of errors of  $M_0$ ,  $\sigma_g$  and  $v_g$  among the new solution, the log-normal AMM, the TEMOM, the QMOM and the log-normal NMM for three cases with initial Knudsen number 0.1000.

same conclusion for pseudo-preserving-size-distribution aerosols which is not presented here. With a decrease of the Knudsen number, the new solution and the log-normal AMM yield much closer relative errors, indicating both solutions might become one solution in the continuum regime. In addition, it is found that in the new solution the total particle number concentration usually achieves a higher accuracy than in its competitor.

## 5. Discussions

Although the integral-differential PBE has been proposed for nearly 100 years (Müller, 1928), an exact analytical solution for this equation with a size-dependent coagulation kernel is not yet known (Vogel et al., 2014). It attributes to the strong nonlinear property of the PBE and also the common characteristics of Boltzmann equations which cannot be analytically solved using the current mathematical techniques (Yu et al., 2008a, 2008b). Up to now, there are only two main approximate analytical solutions for the PBE, i.e., the asymptotic solution (Friedlander, 2000) and the log-normal NMM. The log-normal NMM is prior to the asymptotic solution due to its ability to resolve time-dependent aerosol dynamical processes, thus, it is acknowledged as the mostly suitable analytical solution for the PBE. Unlike the above two solutions, the present work provides an alternative way to solve the PBE. In the derivation, there is no assumption for the size distribution, thus, it might be much more rigorous. More importantly, the derivation in this work is much straightforward where only the two mathematical techniques, i.e., the Taylor expansion technique and the separate variable technique, need to be



**Fig. 4.** The comparison of errors of  $M_0$ ,  $\sigma_g$  and  $\nu_g$  among the new solution, the log-normal AMM, the TEMOM, the QMOM and the log-normal NMM for three non-self-preserving size distributed cases with initial Knudsen number 5.0000.

employed. Here, the key to analytically solve the PBE is to assume the novel variable  $g$  to be a constant, which is verified to be reasonable in this work.

For any solution to be used for solving the PBE, both the accuracy and the scope of application are primarily important. An ideal solution is required to not only yield high accuracy, but also to have the scope of application as wide as possible. In the continuum-slip regime, the geometric standard deviation of number concentration and the Knudsen number are both key physical quantities (Lee et al., 1997). Thus, the ideal solution is required to have an ability to cover the entire valid Knudsen number and geometric standard deviation. Here, the expected Knudsen number ranges from an infinite small value to 5.0000 and the geometric standard deviation ranges from 1.0000 to an infinite large value, which has been discussed in Section 4. It has been verified that the log-normal AMM has no limitation for the above two key quantities. However, the new solution inherits a drawback from the TEMOM ODE whose solution has to be limited from 1.0000 to 1.6583 for geometric standard deviation. Fortunately, in almost all atmospheric environment and particle process engineering, the geometric standard deviation is usually a value smaller than 1.6583 because aerosols have to quickly move into a self-preserving size distribution status as only Brownian coagulation is involved. It has been verified that the geometric standard deviations of number distributions are 1.3200 and 1.3550 in the continuum regime and free molecular regime, respectively, by executing both the log-normal NMM and the TEMOM (Park et al., 1999; Yu et al., 2008a, 2008b). In addition, the geometric standard deviation of number distribution in the continuum-slip regime has been verified to be smaller than 1.3550 when achieving pseudo-self-preserving size distribution status (Otto et al., 1999). Therefore, the newly



proposed analytical solution can be applied in both basic study and engineering studies, although its scope of application is not as wide as the log-normal AMM.

The selection of the SM as a reference to verify the newly proposed analytical solution as well as other solutions is reasonable and feasible. This is because the SM is now regarded as the most accurate solution for solving PBE as the size distribution dependent coagulation kernel is involved (Otto et al., 1999). In theory, the verification of the newly proposed analytical solution should be performed for all valid Knudsen numbers and geometric standard deviations. This is because in the continuum-slip regime only the PSPSD distribution rather than SPSSD distribution exists. This is absolutely different from the free molecular and continuum regimes where individual constant geometric standard deviation exists (Lee et al., 1997; Otto et al., 1994). However, it is not possible to go through all the Knudsen numbers and geometric standard deviations for verification in practice, thus, some representative cases have to be selected and investigated, just as the performance in this work.

In Figs. 2–4, as compared to the analytical solutions, no significant advantage in accuracy for the numerical solutions is found, although a very highly reliable fourth-order Runge–Kutta method with a very small time step was used. A much important conclusion is drawn from the joint evaluation of three key quantities as the Knudsen number is 0.0100 and less, i.e., the newly proposed analytical solution and the log-normal AMM approximately become one solution. This attributes to a fact in both the analytical solutions that the effect of slip correction factor on the equation can be removed in the continuum size range. Although the effect of Knudsen number on the accuracy of analytical solutions is found, it is not possible to obtain a general conclusion, e.g., the accuracy of analytical solutions increases or decreases with the variance of Knudsen number. When the newly proposed analytical solution is verified for non-self-preserving aerosols in Figs. 2–4, it is found that both the initial geometric standard deviation of number distribution and the Knudsen number have an effect on the accuracy. The same properties are also found in other numerical and analytical solutions. However, it is not possible to determine which solution is a better one, especially for the newly proposed solution and the log-normal AMM.

## 6. Conclusions

A new analytical solution is first proposed to solve the population balance equation due to Brownian coagulation in the continuum-slip regime with the Knudsen number up to 5.0000. The analytical solution is achieved based on the performance of the Taylor expansion method of moments together with an assumption for a novel variable  $g$  ( $g = m_0 m_2 / m_1^2$ , where  $m_0$ ,  $m_1$  and  $m_2$  are the first three moments), which is verified to capture the time-dependent evolution of aerosol dynamical process without an assumption for the size distribution. The sectional method is selected as a reference to verify the accuracy of the new solution. The new solution is compared to the log-normal AMM as well as other three numerical solutions, namely the TEMOM, the QMOM and the log-normal NMM, whereas the comparison between the new solution and the log-normal AMM is mainly analyzed. It is revealed that as the three key aerosol quantities including  $M_0$ ,  $\sigma_g$  and  $v_g$  are jointly concerned, it is not possible to determine which solution yields higher accuracy, but the new solution usually yields higher accuracy for the total number concentration. As the Knudsen number is smaller than 0.1000, the new solution and the log-normal AMM are verified to approximately become one solution. Within the valid geometric standard deviation, the new solution is verified to be a reliable solution and it has potential to become a competitive solution in the future.

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## Appendix A. TEMOM model in the continuum-slip regime

The TEMOM model in the continuum regime can be written as the following expression with  $g = m_0 m_2 / m_1^2$  (Yu et al., 2008a, 2008b):

$$\begin{cases} \left( \frac{dm_0(t)}{dt} \right)_c = \frac{B_2 \{2g^2 - 13g - 151\}}{81} m_0(t)^2 \\ \left( \frac{dm_1(t)}{dt} \right)_c = 0 \\ \left( \frac{dm_2(t)}{dt} \right)_c = -\frac{2B_2 \{2g^2 - 13g - 151\}}{81} m_1(t)^2 \end{cases} \quad (A1)$$

For an aerosol, the zero moment must decrease while the second moment must increase with the time when its dynamical process is solely dominated by Brownian coagulation mechanism, thus,

$$2g^2 - 13g - 151 < 0 \quad (A2)$$

The term of right hand of (A1) is the same as in Eq. (4) for both  $m_0$  and  $m_2$  shown in this work, thus, Eq. (A2) is also valid for Eq. (4). In addition, the coagulation process will be enhanced in the continuum-slip regime as compared to that in the continuum regime, thus, the following criterion must be satisfied:

$$\begin{cases} 5g^2 - 64g - 103 < 0 \\ g^2 - 2g - 80 < 0 \end{cases} \quad (\text{A3})$$

In order to obtain the validate scope of application of TEMOM equations in the continuum-slip, it needs to solve Eqs. (A2) and (4) simultaneously. Finally, the scope of  $g$  is

$$g \in (0.0000, 10.0000)$$

As the equation  $\ln^2(\sigma_g) = (1/9)\ln(g)$  is applied, then

$$\sigma_g \in (1.0000, 1.6583)$$

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