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Technical Note

A New Mathematical Scheme for Approximating the Overall Aerosol Extinction Coefficient during Brownian Coagulation

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ABSTRACT

The approximation of the overall aerosol extinction coefficient is conventionally achieved through integrating the single particle extinction efficiency over the whole size distribution, which requires much computational time. In this work, a new approximation scheme with higher efficiency than the conventional scheme is proposed, in which the combination of polynomials for fitting Mie's solution and the method of moments produces the overall extinction coefficient of evolving nanoparticles. The closure of arbitrary moments is achieved by implementing the Taylor-series expansion method of moments. The new approximation scheme was verified by comparing it to a more exact referenced scheme for two different typical aerosol modes, namely the nucleation mode and the accumulation mode. This study verifies that the new scheme is a reliable method for approximating the overall extinction coefficient during aerosol evolution with acceptable efficiency and accuracy; thus, it is suitable for use in some atmospheric aerosol models.

Keywords: Overall extinction coefficient; Method of moments; Polynomial; Taylor-series expansion; Aerosol evolution.

INTRODUCTION

Aerosol light scattering plays a major role in the evolution of earth surface temperature, visibility and also radiation transfer through the atmosphere (Friedlander, 2000; Rosenfeld, 2006; Quaas, 2009; Qureshi *et al.*, 2009; Kim *et al.*, 2016). The light scattering by aerosols, whose size is from nanometer to micrometer, can be formulated as the sum of light scattered over all sizes (Friedlander, 2000). There is an extensive literature on the optical properties of single particles, whereas the study on the optical property of aerosols during evolution is few. To predict the effect of aerosols on the visibility and environment temperature, it is necessary to establish a predictive model, which enables to produce the index of visibility, such as overall particle extinction coefficient in the air (Jung and Kim, 2007). However, the aerosol light scattering is determined by several major aspects, including the aerosol size distribution and shape, and the wavelength of the incident light. In atmospheric environment, even only coagulation mechanism dominates, the aerosol size distribution has a polydisperse property and especially it evolves over time, which leads to the difficulty of the establishment of predicative models.

When light passes through an aerosol, its interaction with the aerosol particles produces scattering and absorption effects. The combination of these two effects is called the extinction effect of aerosol particles. If the single particle extinction coefficient is a quantitative representation of the single particle extinction effect, then the integral of the single particle extinction coefficient over particle size distribution represents the overall extinction coefficient of the aerosol (Seinfeld and Pandis, 1998). A numerical method can be used to calculate the overall extinction coefficient to develop an optical particle counter and simulate aerosol visibility (Koschmieder, 1925). If the frequency of light does not change before and after scattering, the particle scattering can be further divided into types according to the particle size; if the particle size is much smaller than the wavelength of the incident light (i.e., $d_p \ll 623$ nm), the light-scattering pattern is called Rayleigh scattering, but if the particle size is much larger than the wavelength of the light (i.e., $d_p \ll 623$ nm), the particle scattering theory of geometrical optics applies; if the particle size is close to

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the wavelength of the incident light, the scattering theory proposed by Mie applies (Mie, 1908; Bohren and Huffman, 1983), which involves deriving an analytical solution for the extinction coefficient of spherical particles. Mie scattering is a general theory that addresses the scattering and absorption of plane waves by uniformly spherical particles.

The overall extinction effect of the aerosol in the atmospheric environment is expressed as the sum of the scattering and absorption effects (Bohren and Huffman, 1983). The resulting extinction coefficient is given by:

$$b_{ext} = b_{sg} + b_{ag} + b_{sp} + b_{ap} \quad (1)$$

where b_{sg} and b_{ag} are the scattering and absorption coefficients of the air on the light, and b_{sg} and b_{ag} are the scattering and absorption coefficients of the aerosol on the light.

In a clean atmosphere which contains a small number of aerosol particles, the scattering and absorption effects of visible light are mainly caused by molecules in the air such as nitrogen, oxygen, and carbon dioxide; thus, Rayleigh scattering theory can be applied (van de Hulst, 1957). However, in a polluted atmosphere replete with lots of aerosol particles, where the main contribution to the scattering and absorption effects of visible light comes from aerosol particles, Rayleigh scattering theory is not applicable and Mie scattering theory is required instead. The main task for numerically calculating the extinction coefficient of aerosol particles is to calculate the single-particle extinction coefficient and the evolution of aerosol particles dominated by Brownian coagulation.

The single particle extinction coefficient at a specific incident light wavelength can be calculated from Mie's solution (Bohren and Huffman, 1983). From the Mie scattering theory, the single particle extinction coefficient depends mainly on the wavelength of the incident light, the particle size, and the refractive index. Unfortunately, the Mie's solution involves several expressions composed of series, which have slow convergence and low calculation efficiency (Jung and Kim, 2008).

The particle size distribution of an aerosol evolves over time; this can be described theoretically by the population balance equation (PBE) (Friedlander, 2000). A common method for solving the PBE is the sectional method (SM) (Gelbard *et al.*, 1980; Kostoglou, 2007), which is a direct numerical calculation of the PBE that facilitates calculation by not incorporating any induced assumptions. This method has the advantage of high precision, because it need not presuppose a certain statistical distribution in each calculating step, or a simplified coagulation kernel, but at the expense of low computational efficiency. The SM can be used to produce the particle size distribution of the aerosol at any time in the evolutionary process, and with it the overall extinction coefficient can be obtained by implementing the integral of the single particle extinction coefficient on each particle size (Seinfeld and Pandis, 1998). Thus, the scheme involves coupling the SM and the extinction model to obtain the extinction effect of the aerosol with time. However, the low efficiency of the SM and Mie's solution hinders the wide application of the scheme because considerable computation time is required. Thus, it is significant to propose a scheme to obtain the extinction effect of the aerosol with acceptable efficiency. Monte Carlo method has some similar property to the SM for solving PBE (Kruis *et al.*, 2012; Wei, 2017), but the application of the Monte Carlo method in the field of visibility is few.

In the study of aerosols, statistical quantities including the particle number concentration of the aerosol, average particle size, and particle dispersion are usually central for calculation and analysis (Hulburt and Katz, 1964; Qureshi *et al.*, 2009; Wei, 2017). In theory, these quantities in terms of the moments of particle size distribution with respect to the particle size. To solve the PBE, it is possible to convert it into moment equations and then solve the ordinary differential equations with respect to moments rather than the particle size distribution function. On the basis of this idea, the method of moments (MOM) was proposed by Hulburt and Katz (1964). The advantage of MOM over SM is that the degree of freedom of MOM can be largely reduced, raising its efficiency. MOM has become the most efficient method for solving PBE, especially in engineering problems, and several versions of it exist, including the quadrature-based MOM (QMOM) (McGraw, 1997; Marchisio and Fox, 2005; Yuan *et al.*, 2012), the lognormal method of moments (log MM) (Lee *et al.*, 1984), the method of moments with interpolation closure (MOMIC) (Frenklach and Harris, 1987), the Taylor-series expansion method of moments (TEMOM) (Yu *et al.*, 2008, 2016), and the gamma MOM (Williams, 1986). Jung and Kim (2006, 2007) successfully applied the log MM to calculate the overall extinction coefficient of aerosols, and further verified that the coupling between the MOM and the extinction model is a feasible method to trace the evolution of the aerosol extinction property. Jung and Kim's groundbreaking work provides us a conventional way to predict aerosol visibility property. However, the log MM has a shortcoming that it has to be implemented with an assumption for the size distribution, which limits its scope of application. TEMOM was proposed by Yu *et al.* (2008), and has been verified as a promising method for solving the PBE when multiple aerosol dynamics are involved, such as coagulation, condensation, and nucleation (Yu and Lin, 2018). Although some promising properties have been revealed, especially it has no assumption for the size distribution, the TEMOM has never been used to predict the overall extinction coefficient of particles.

In this work, TEMOM was applied for the first time to study the overall extinction coefficient of aerosols. To capture the evolution of the extinction coefficient of particles with time, the polynomial fitting method proposed by Jung and Kim (2006) was employed, and to verify the new scheme, the coupling between the SM and Mie scattering was used as a reference. The reliability and precision of the new scheme were also evaluated. To highlight the difference between the two schemes in this work, the coupling between the TEMOM and polynomial solution is called the *polynomial & TEMOM* scheme, while the coupling between the SM and Mie's solution is called the *Mie & SM* scheme.

MATHEMATICAL MODELS FOR AEROSOL DYNAMICS

In this work, the mechanism affecting the evolution of particle size distribution is Brownian coagulation. Other aerosol dynamics, such as condensation, nucleation and deposition, are not included for shortening the length of this note even though they can be also conventionally introduced here. Brownian coagulation theory and the solution of PBE are briefly explained here.

Particle Balance Equation

Aerosols in the air usually undergo internal dynamics, such as Brownian coagulation, condensation, and nucleation, as well as external dynamics, such as convection and diffusion (Crowe *et al.*, 2011). In the atmospheric environment, Brownian coagulation plays a key role in determining the evolution of particle size distribution, and it affects the extinction coefficient of aerosols (Jung and Kim, 2008). In this work, only Brownian coagulation is considered. The PBE can be expressed as follows (Friedlander, 2000):

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

here, $n(v, t)$ is the particle number density for particle volume v and time t, and $\beta(v, v')$ is the particle coagulation kernel between two particles with size v and v'. Eq. (2) is a nonlinear integral-differential equation whose analytical solution cannot be obtained if the kernel for Brownian coagulation for polydispersed particles is employed. In this work, the TEMOM was applied to solve Eq. (2), which is discussed in detail in a later section (Yu *et al.*, 2017).

Brownian Coagulation

The Brownian motion of particles results in Brownian coagulation, which is closely related to the Knudsen number (Kn), defined as the ratio of the mean free path to the particle radius ($Kn = 2\lambda/d_p$). Accordingly, the mechanism of Brownian coagulation varies for different regimes, which include the free molecular, continuum, continuum-slip, and transient regimes (Otto *et al.*, 1999; Park *et al.*, 1999). In the free molecular regime, the kernel for Brownian coagulation can be derived from gas kinetic theory:

$$\beta_{\mathrm{FM}}(v,v') = \left(\frac{3}{4\pi}\right)^{1/6}\left(\frac{6\mathrm{k_b}T}{\rho}\right)^{1/2}\left(\frac{1}{v}+\frac{1}{v'}\right)^{1/2}\left(v^{1/3}+v'^{1/3}\right)^2 \tag{3a}$$

Whereas in the continuum-slip regime, the kernel for Brownian coagulation is obtained by mass diffusion theory:

$$\beta_{\mathrm{Co}}(v,v') = \frac{2\mathrm{k_b}T}{3\mu}\left(\frac{C_{\mathrm{c}}(Kn,v)}{v^{1/3}}+\frac{C_{\mathrm{c}}(Kn,v')}{v'^{1/3}}\right)\left(v^{1/3}+v'^{1/3}\right) \tag{3b}$$

where k_b is the Boltzmann constant; T is the temperature of the air; ρ is the particle density, μ is the gas viscosity; and $C_c(Kn, v)$ is the slip-correction factor, which is determined by both Kn and v.

Brownian coagulation leads to increased particle size. Even for a closed aerosol system without any mass exchange with its surrounding environment, the particle size might scan from a free molecular regime to a continuum regime. In this case, the kernel at a particular regime cannot be used for the whole size regime. Although Fuchs (1964) proposed a kernel to cover the entire size regime, the complexity of the kernel's mathematical form makes it unsuitable for the log MM and TEMOM. In this work, we accepted the concept of the harmonic mean solution for this kernel, which was verified as reliable by Otto *et al.* (1999). However, we need to note the harmonic mean solution has limitation in accuracy as compared to other solutions, such as Dahneke's solution (Otto *et al.*, 1999).

Taylor-series Expansion Method of Moments

Multiple methods exist for solving the PBE numerically, including the SM, MOM, and Monte Carlo method (MC) (Yu and Lin, 2018). Among these, the MOM is the most efficient, providing statistical information on aerosols including the particle number concentration, mean particle diameter, and polydispersity. As a promising MOM proposed in 2008 (Yu *et al.*, 2008), the TEMOM achieves the closure of moment differential equations by implementing the Taylor-series expansion technique. The TEMOM is briefly presented here.

When TEMOM is implemented, Eq. (2) can be converted to the following expression through multiplication of both sides by v^k and subsequent integration over the whole size range:

$$\frac{dm_k}{dt} = \frac{1}{2}\int_0^\infty\int_0^\infty \left[\left(v+v'\right)^k - v^k - v'^k\right]\beta(v,v')\,n(v,t)\,n(v',t)\,dv\,dv' \tag{4}$$

here, the k^{th} moment is defined as:

$$m_k = \int_0^\infty v^k\,n(v)\,\mathrm{d}v \tag{5}$$

Because Eq. (2) cannot usually be closed by itself, a general closure function for k^{th} moments must be established. This can be achieved by expanding v^k around u, implementing the Taylor-series expansion technique as follows:

$$v^k = u^k + u^{k-1}k(v-u) + \frac{u^{k-2}k(k-1)}{2!}(v-u)^2 + \frac{u^{k-3}(k-1)(k-2)}{3!}(v-u)^3 + \ldots \tag{6a}$$

In the TEMOM, $u = m_1/m_0$. If Eq. (6a) is introduced into

Eq. (5) with the first 3 terms reserved, this results in:

$$m_k = \left(\frac{u^{k-2}k^2}{2} - \frac{u^{k-2}k}{2}\right)m_2 + \left(-u^{k-1}k^2 + 2u^{k-1}k\right)m_1 + \left(u^k + \frac{u^2k^2}{2} - \frac{3u^k k}{2}\right)m_0 \quad (6b)$$

Eq. (6b) is the closure model for arbitrary order moment. According to the selection of Taylor-series expansion orders and moment sequence, several versions of closure model have been proposed (Yu and Lin, 2018). Once the specific mathematical expression of $\beta(v, v')$ is introduced into Eq. (2), it can be integrated out. The details about the TEMOM can be found in the reviewed article (Yu and Lin, 2018).

AEROSOL EXTINCTION COEFFICIENT

Single Particle Extinction Coefficient

The scattering efficiency of the particles represents the ratio of the energy intercepted by the cross-section of the particles to the total energy of the incident light. Similarly, the absorption efficiency of the particles represents the ratio of the energy absorbed by the particles to the total energy of the incident light. The ratio of energy that is removed from the incident light, namely the sum of the scattered and absorbed energy, relative to the total energy of the incident light, is called the extinction coefficient of particles. The extinction coefficient has the following expression (Seinfeld and Pandis, 1998):

$$Q_{\text{ext}}(m, p) = Q_{\text{sca}}(m, p) + Q_{\text{abs}}(m, p) \quad (7)$$

where m is the refractive index and the parameter size p is defined as the diameter of the particles divided by the wavelength of the incident wave:

$$p = \frac{\pi d_{\text{p}}}{\lambda} \quad (8)$$

The extinction coefficient for any particle size can be obtained by Mie theory (Mie, 1908). The theoretical Mie's solution can be appropriately reduced to the formula only for the refraction index (m) and the size parameter (p) when used in the calculation of the single particle extinction coefficient (Jung and Kim, 2007).

In Fig. 1, we present the extinction coefficients for the various particle sizes of five typical aerosols (Hinds, 1999; Garcia-Nieto, 2002). The refractive index, which is scattering particle relative to surrounding medium, is a complex number; the real part is a quantification of light scattering by particle; and the imaginary part is a quantification of absorption. Fig. 1 indicates that the refractive index has a notable effect on the extinction coefficient of the particles. When the refractive index is not imaginary (i.e., only the scattering effect exists), the peak of the location and magnitude of the single particle extinction coefficient curve is affected by the real part of the refractive index. In the absence of an absorption effect, the curve exhibits clear oscillation. When the refractive index exists in the imaginary part (i.e., the scattering and absorption effects occur together), the curve becomes smoother as the imaginary part increases.

Given the single particle extinction coefficient, the overall extinction coefficient of the aerosol can be obtained using the following integral formula (Seinfeld and Pandis, 1998):

$$b_{\text{ext}} = \int_0^{\infty} \frac{\pi d_{\text{p}}^2}{4} Q_{\text{ext}}(m, p)\, n(d_{\text{p}})\, \mathrm{d}d_{\text{p}} \quad (9)$$

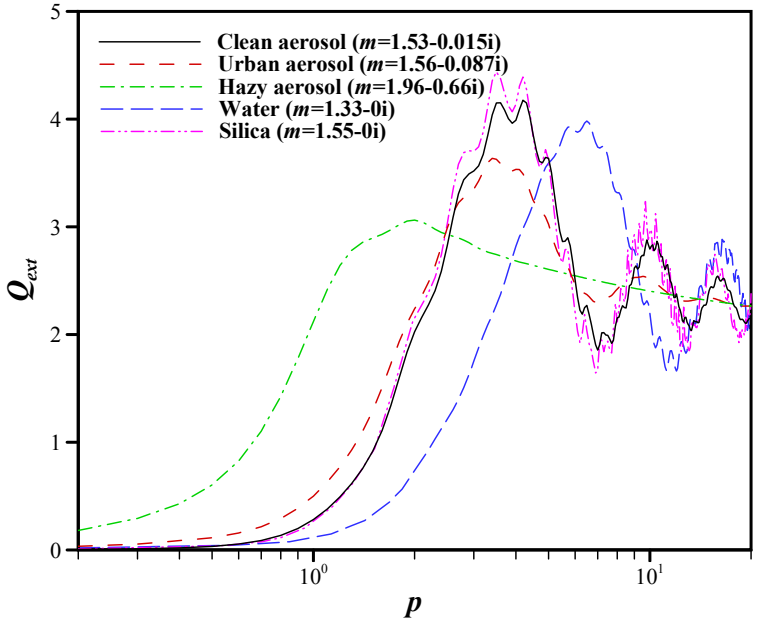

Fig. 1. Single particle extinction coefficient for typical aerosol conditions (λ = 598 nm).

The differences in light intensities are particularly important and are the principal basis for the classical theory of visibility. Visibility is described as the maximum distance at which an object can be distinguished from the background by human eyes. Therefore, visibility is one of the quantitative indexes to assess air quality, and Koschmieder (1925) proposed this formula:

$$C = -e^{-b_{\text{ext}} L_{\text{v}}} \tag{10a}$$

The parameter C is sometimes called the threshold constant or "psychophysical constant", because it depends on human perception. Based on data averaged over responses of a group of individuals, its value is usually taken to be 0.02 (Friedlander, 2000), and then the following formula can be obtained:

$$L_v = \frac{3.912}{b_{ext}} \tag{10b}$$

where L_v is the visibility, b_{ext} is the overall extinction coefficient.

In this way, the visibility of the aerosol can be calculated by the obtained aerosol overall extinction coefficient b_{ext}.

Polynomial Fitting

The original extinction coefficient for a single particle size can be obtained by Mie's solution, but the complex formula of Mie scattering cannot be directly applied with the MOM. To apply Mie's solution to the MOM, for the first time, Jung and Kim (2006) proposed to fit the single particle extinction coefficient curves using a polynomial scheme, which is expressed as a function of the power of the particle size parameter. With Jung and Kim's promising scheme, the log MM was successfully used to approximate the evolving overall extinction coefficient of a polydispersed aerosol. In this study, we further extend Jung and Kim's scheme to the TEMOM, and we verify whether the TEMOM has the similar capability to the log MM in this aspect.

The accuracy of TEMOM and the support of the decimal moment calculation enable the application of higher-order polynomial fittings to approximate the single particle extinction coefficient curve, to make the approximate result closer to the theoretical result. As discussed in the previous section on TEMOM, the maximum moment supported by the TEMOM is raised to the fourth moment by preserving five terms for Eq. (6a). In addition, we used the sixth order and ninth order polynomials to fit the single particle extinction coefficient and then compare their accuracy. An approximation of the ninth order polynomial can be defined as follows:

$$\begin{aligned} Q(p,m) &= \sum_{k=0}^{9} A_k p^k = \sum_{k=0}^{9} A_k \left[\frac{\pi}{\lambda} \left(\frac{6v_{\text{p}}}{\pi} \right)^{1/3} \right]^k \\ &= \sum_{k=0}^{9} A_k \left(\frac{\pi}{\lambda} \right)^k \left(\frac{6v_{\text{p}}}{\pi} \right)^{k/3} \end{aligned} \tag{11}$$

where A_k is the approximated coefficient of the ninth order polynomial and λ is the wavelength of incident light.

Bringing Eq. (11) into Eq. (9) can generate the approximate expression of the overall extinction coefficient. However, whereas the integral formula Eq. (9) uses particle diameter as an argument, this study uses particle volume. Before the equations are combined, the particle diameter argument in Eq. (9) must be converted into volume, as follows:

$$\begin{aligned} b_{ext} &= \int_0^\infty \frac{\pi}{4} \left(\frac{6v_p}{\pi} \right)^{\frac{2}{3}} Q_{ext}(m,p) n(v_p) dv_p \\ &= \sum_{k=0}^{9} A_k \frac{\pi}{4} \left(\frac{\pi}{\lambda} \right)^k \left(\frac{6}{\pi} \right)^{(2+k)/3} \int_0^\infty v_p^{(2+k)/3} n(v_p) dv_p \\ &= \frac{\pi}{4} \sum_{k=0}^{9} A_4' m_{(2+k)/3} \end{aligned} \tag{12a}$$

here, A_k' is an approximate coefficient of the ninth order polynomial, which is defined as:

$$A_k' = A_k \left(\frac{\pi}{\lambda} \right)^k \left(\frac{6}{\pi} \right)^{(2+k)/3} \tag{12b}$$

Through this approximation, we can quickly calculate the overall extinction coefficient of an aerosol.

RESULTS AND DISCUSSIONS

In this work, we followed Jung and Kim's scheme to propose a new approximated overall extinction coefficient model (Jung and Kim, 2006, 2007), in which the TEMOM rather than the log MM was used. To verify the new model in accuracy, we implemented the SM to achieve the size distribution of the studied aerosol, which enables the achievement of the most exact overall extinction coefficient through integrating Mie's single particle extinction efficiency over the widespread polydispered aerosol.

Polynomial Solution

As an example, an aerosol composed of carbon nanoparticles is used as the simulation object in this paper, and the refractive index is $m = 2 - 0.66\text{i}$ when incident light wavelength is 0.623 μm (i.e., $\lambda = 0.623$ μm).

Fig. 2(a) shows the curve obtained by polynomial fitting of the single particle extinction coefficient for carbon nanoparticles in the range of $p \in (0, 13)$. As the reference, the curve obtained from the Mie & SM scheme is present. Compared with the referenced Mie & SM scheme, the newly proposed polynomial & TEMOM has acceptable precision, especially $p \geq 1.5$ where the relative errors of the polynomial & TEMOM scheme to the referenced scheme are always close to zero. The relative errors of the polynomial & TEMOM scheme to the referenced scheme can be found in Fig. 2(b). Although it is found that the fitting in the range $p < 1.5$ is not as promising as that in the range $p \geq 1.5$, what we see from Fig. 2(b) is the error of

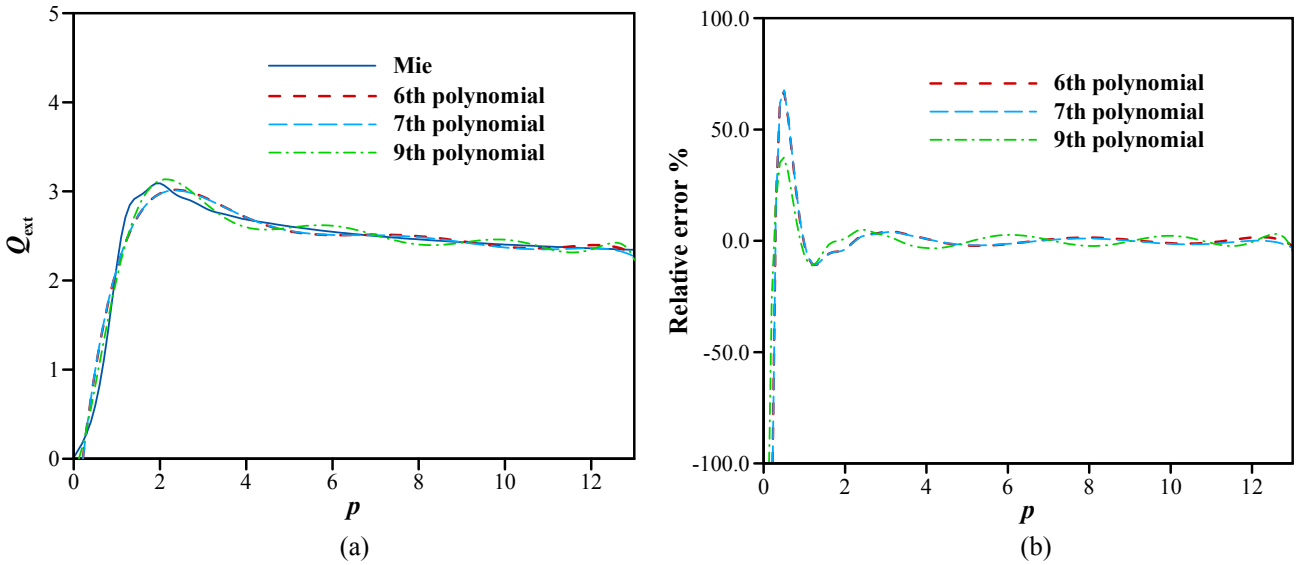

Fig. 2. (a) Comparison of the polynomial & TEMOM scheme and the Mie & SM scheme and (b) Relative error of the polynomial & TEMOM to the Mie & SM scheme.

the ninth order polynomial fitting is significantly lower than that of the low order fitting. This demonstrates that the accuracy of the fitting using the polynomial solution can be increased with an increase of polynomial order when p is small (i.e., the particle size is small because of $p = (\pi d_p)/\lambda$). In this study, the modulus of residual on the fitting range for the sixth, seventh, and ninth order polynomials are 1.3231, 1.3217, and 0.93259, respectively. The accuracy of the polynomial fitting is only slightly higher from the sixth to seventh order, but considerably improved when fitted in the ninth order. It needs to be addressed here that in Jung and Kim's work, the sixth order polynomial was used for the coupling with the log MM (Jung and Kim, 2006), which produces reliable results for the overall extinction coefficient.

Evolving Overall Extinction Coefficient Using the Polynomial & TEMOM Scheme

Under any conditions, the size distribution of aerosols evolves over time (Friedlander, 2000). Thus, it is necessary to assess the capability of the newly proposed polynomial & TEMOM scheme for capturing the evolving overall extinction coefficient. As discussed in the previous sub-section, the accuracy of the polynomial fitting varies with p, which might lead to different accuracy of the polynomial & TEMOM scheme under the different initial aerosol mean size. To verify the polynomial & TEMOM scheme, we applied this scheme to approximate overall extinction coefficient of aerosols with two different aerosol mean sizes. The first is 3 nm, which is a typical size of freshly nucleated particles in air (nucleation mode), and the second is 0.3 μm, which accounts for grown particles (accumulation mode) (Seinfeld and Pandis, 2012).

In the study, the initial particle size distribution is assumed to be lognormal (Pratsinis, 1988; Otto *et al.*, 1999). With carbon nanoparticles defined as the simulation object, the overall extinction coefficient was numerically simulated with different initial values of mean particle diameter, distribution standard deviation, and particle number concentration. Fig. 3(a) shows the aerosol evolution under the Brownian coagulation process. The initial mean particle diameter was set to 3 nm, the particle size distribution standard deviation to 1.20, and the initial particle number concentration to 300,000 cm^{-3} The particles' Brownian coagulation process increases the particle volume but decreases the particle number concentration. The SM code comes from Yu and Lin (2009), whose reliability has been verified (Yu and Lin, 2009). Fig. 3(b) shows the first 3 order moments obtained by SM and TEMOM with the same initial condition; in order to facilitate the comparison, the moments are converted into dimensionless (Yu and Lin, 2009). Fig. 3(c) is the relative error curves for the moments obtained by TEMOM relative to that by SM. From above figures, we can see TEMOM has a less than 4% error in 10,000 steps of calculation, and first 1000 steps' error is less than 0.1%. When the simulation steps are constrained within 10,000 steps, the relative error of the overall extinction coefficient is mainly contributed by polynomial fitting.

Fig. 4 shows the evolution of the polynomial & TEMOM and Mie & SM schemes over time for the overall extinction coefficient, corresponding to the respective initial conditions. The overall extinction coefficient at this initial particle size rises, which is consistent with the finding in Fig. 2 that the single particle extinction coefficient increases in the range of $p \in (0, 2)$ (i.e., about $d_p \in (0,0.4\ \mu m)$ for carbon nanoparticles). In Fig. 4, we found a relatively large deviation between the polynomial & TEMOM and Mie & SM schemes at initial time. This is because in early stage of the simulation for small size particles, the fitting error mainly causes a deviation of the overall extinction coefficient. Nevertheless, with the dynamic evolution going, particle size will increase and the range of p will be larger than 1, and then the fitting error will reduce to less than 5%. As the fitting error further converges after p larger

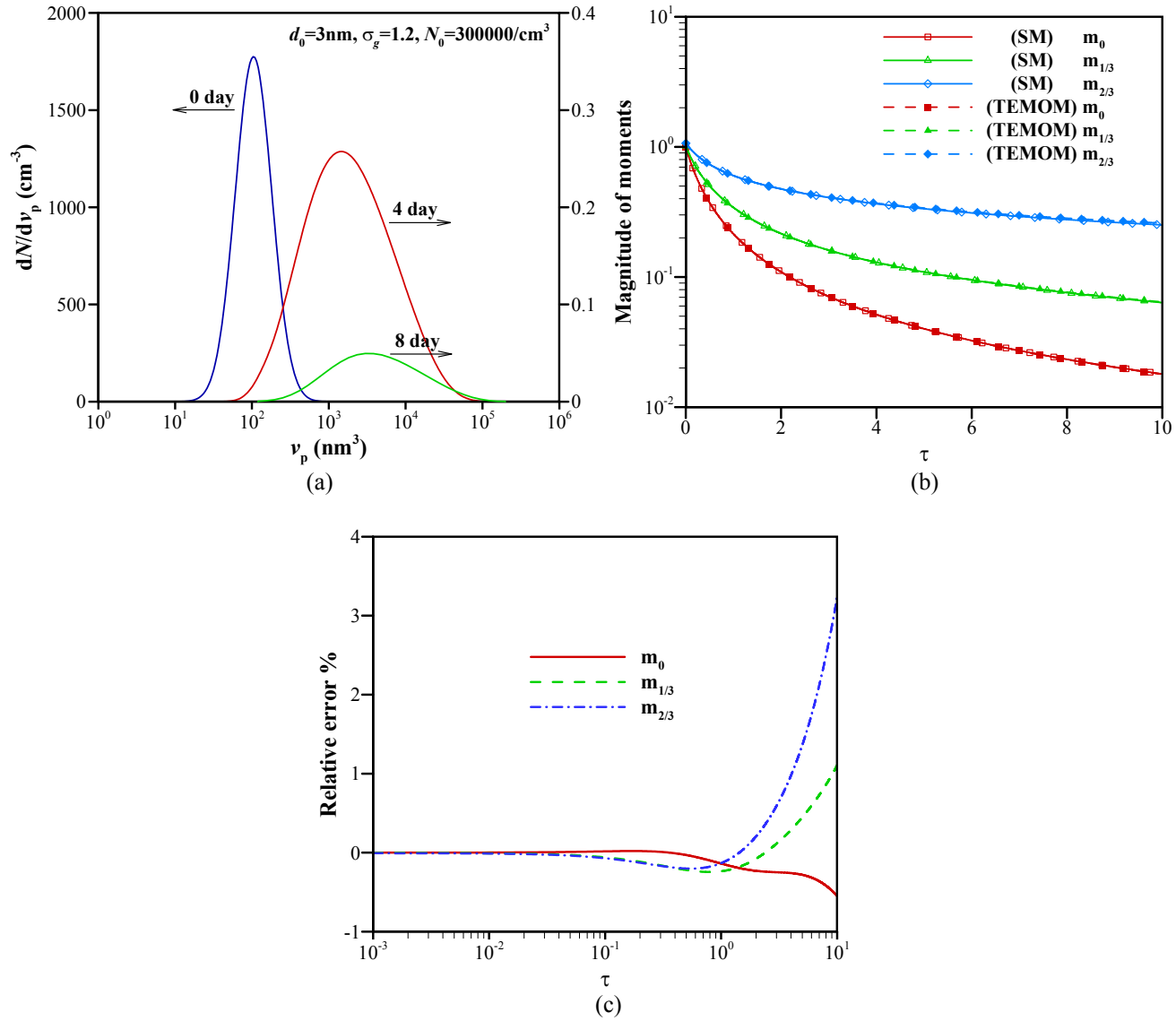

Fig. 3. (a) Evolution of the aerosol particle size distribution over time under the Brownian coagulation process (d_p = 3 nm, σ_g = 1.20, N_0 = 300,000 cm^{-3}), (b) The first three order moments obtained by SM and TEMOM with the same initial condition, and (c) The relative error for the moments obtained by TEMOM relative to that by SM.

than 2, the result curves obtained by two schemes will overlap. In the polynomial & TEMOM scheme, the fitting order set as sixth and ninth exhibit little difference, and the two curves coincide at three different σ_g values. As compared to the Mie & SM scheme in Fig. 4(d), the polynomial & TEMOM shows different accuracy under different σ_g, indicating the accuracy of the newly proposed polynomial & TEMOM scheme is related to the initial size distribution. In Jung and Kim's work, the study on the effect of σ_g on the extinction coefficient is not involved.

Similar to Fig. 4, we also verified the polynomial & TEMOM scheme for approximating overall extinction coefficient for relative larger particles during evolution. Here, the initial mean size of the studied aerosol is 0.3 μm, which corresponds to particles within accumulation mode. Three different initial geometric standard deviations, namely σ_g = 1.10, 1.20, and 1.40, are considered. Fig. 5 depicts the evolution of the polynomial & TEMOM and Mie & SM schemes over time for the overall extinction coefficient, corresponding to the respective initial conditions. Because the size parameter was in the range of $p \in (2, \infty)$, the single particle extinction coefficient decreased from the maximum and reached a steady value. When the initial mean particle diameter was 0.3 μm, most of the particle size parameters were approximately 2. With the aerosol internal processing, the single particle extinction coefficient decreased, and eventually the overall extinction coefficient decreased. Fig. 5 also indicates that the polynomial & TEMOM scheme obtained by two polynomial fittings is approximately equal to the Mie & SM scheme, and their relative errors can finally converge. Unlike the case with small initial particle size (nucleation mode, 3 nm), the accuracy of the polynomial & TEMOM obtained by the ninth order polynomial is superior to that obtained by the sixth order polynomial in

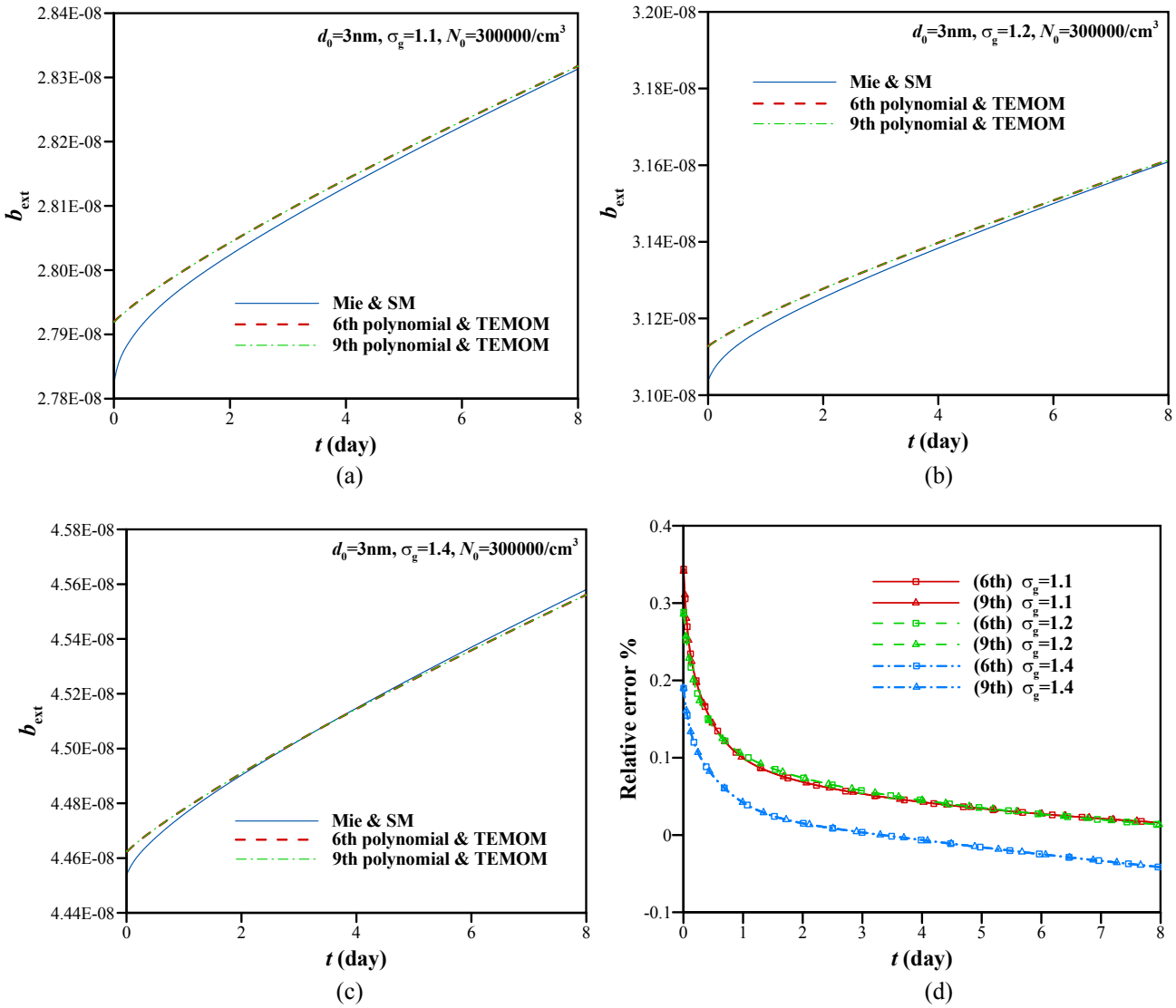

Fig. 4. (a), (b), and (c) Comparison of the evolution of the polynomial & TEMOM and Mie & SM schemes over time with three different σ_g, when the initial mean particles are within nucleation mode. (d) The relative error for the overall extinction coefficient obtained by the polynomial & TEMOM scheme relative to that by the Mie & SM scheme.

Fig. 5(d). Especially in the early period of evolution, the error of the sixth order scheme is larger than 4%, but the error of the ninth order scheme is less than 2% in the overall evaluation of the coagulation process.

Although this work is mainly to establish a new scheme for approximating overall extinction coefficients of aerosols with acceptable accuracy, the efficiency of the scheme should also be considered. The efficiency of any numerical method or scheme is reflected by its computational time. In the polynomial & TEMOM scheme, the computational cost is mainly determined in two aspects. One aspect is the computational cost on the solution of ordinary differential equations of TEMOM. The second aspect of the efficiency is the computational cost for solving Eq. (12a), in which the closure model of arbitrary order moment needs to be implemented. As compared to the Mie & SM scheme, the polynomial & TEMOM is an economical scheme. Both the implementation of Mie's solution and SM consumes huge computational time (Jung and Kim, 2007). As a comparison, we used both Mie & SM and polynomial & TEMOM schemes to produce overall extinction coefficients with times up to 1 day in Fig. 4(b); the Mie & SM, polynomial & TEMOM (9^{th}), and polynomial & TEMOM (6^{th}) consume computational times of 4.10 hour, 39 seconds and 26 seconds, respectively. It is obvious the polynomial & TEMOM largely reduces the computational time as compared to the conventional Mie & SM scheme. In addition, the polynomial & TEMOM (6^{th}) consumes only 66.67% of computational time of the polynomial & TEMOM (9^{th}). Thus, under the precondition that the accuracy is met, the lower the order of polynomials, the higher the advantage of using the polynomial & TEMOM scheme. If the accuracy is primarily required, larger order of polynomials and TEMOM is recommended for implementation, since both polynomial and TEMOM solutions are more economical methods as compared to Mie's solution and SM.

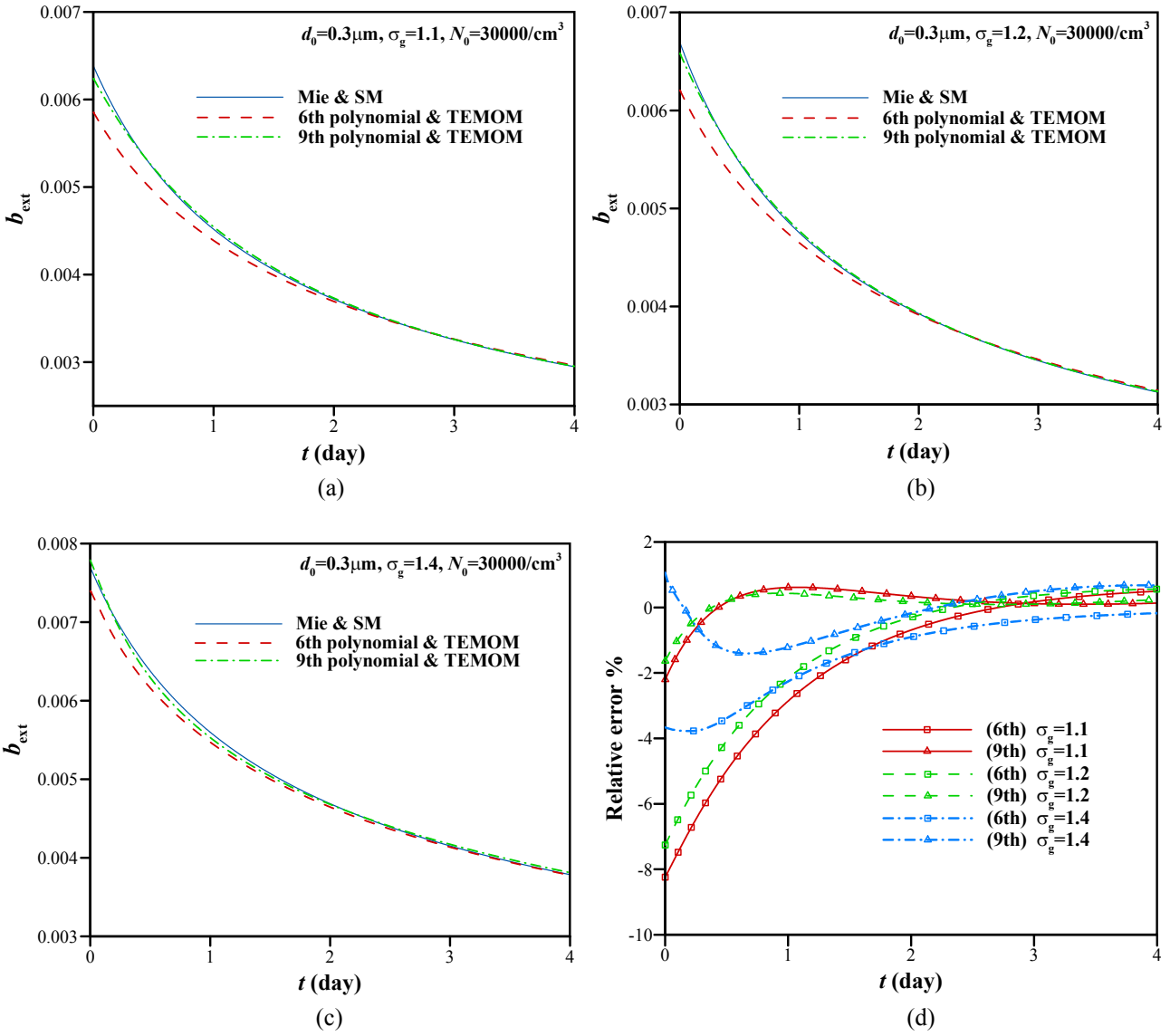

Fig. 5. (a), (b), and (c) Evolution of the polynomial & TEMOM and Mie & SM schemes over time with three different σ_g, when the initial mean particle is within accumulation mode. (d) The relative error for the overall extinction coefficient obtained by the polynomial & TEMOM scheme relative to that by the Mie & SM scheme.

CONCLUSION

In this work, we applied polynomial fitting to estimate the coefficient for a single aerosol particle size and TEMOM to obtain the moments of the particles; this combination produced an approximate model of the overall extinction coefficient of an aerosol. Numerical results were calculated for two different initial particle sizes of carbon nanoparticles using the polynomial & TEMOM scheme and compared with results using the Mie & SM scheme. From the results, the accuracy of the result obtained by the polynomial & TEMOM scheme converges and remains at a low relative error compared to the referenced Mie & SM scheme, validating the newly proposed scheme in this work. In this study, we also used two different orders of polynomials to fit the single particle extinction coefficient. We found that the accuracy of the polynomial & TEMOM scheme is related to the initial size distribution and the order of polynomials. As the results indicate, because of the high computational accuracy and efficiency of TEMOM, it is feasible to obtain more accurate results by using higher-order polynomials without sacrificing computational efficiency. Other dynamical processes, such as condensation, deposition, and nucleation, can also be included in this method. The model is limited by the assumption that aerosol particles are spherical, which might cause deviation from the real values when the environmental aerosols are agglomerates or aggregates.

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