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A statistical-numerical aerosol parameterization scheme

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Abstract

A new modal aerosol parameterization scheme, Statistical-Numerical Aerosol Parameterization (SNAP), was developed for studying aerosol processes and aerosol-cloud interactions in regional or global models. SNAP applies statistical fitting on numerical

- results to generate accurate parameterization formulas without sacrificing details of the growth kernel. Processes considered in SNAP include fundamental aerosol processes, as well as processes related to aerosol-cloud interactions. Comparison of SNAP with numerical solutions, analytical solutions, and binned aerosol model simulations showed that the new method performs well, with accuracy higher than that of the high-order nu merical quadrature technique, at much less computation time. The SNAP scheme has
- been implemented in regional air quality models, producing results very close to those using binned-size schemes or numerical quadrature schemes.

1 Introduction

Aerosol particles may strongly influence air pollution, cloud and precipitation forma-¹⁵ tion, as well as climate and environment changes. Key factors that determine the influence of aerosols are their size spectrum and chemical compositions. However, these factors are highly variable and thus can be difficult to simulate in either regional or global-scale atmospheric chemistry or air pollution models. Moreover, different particulate chemicals may coexist in a specific air parcel by external or internal mixing.

These mixing states have additional influence on the physical and optical properties of particles (Chylek and Wong, 1995; Jacobson, 2000). The large number of possible combinations between aerosols of different origins further complicates their roles in atmospheric processes (Jacobson, 2001; Nenes et al., 2002). Therefore, increasingly sophisticated analytical methods are required to fully understand the roles of aerosols in the atmosphere.





Earlier models for studying aerosol processes, such as RADM2 or CAMx, keep track of only the aerosol mass concentration. Such bulk methods are insufficient in resolving size-sensitive processes, such as dry and wet deposition, cloud drop activation, light scattering and absorption, and impact on health. Therefore, an increasing number of models are adapting size-spectrum schemes. Size-spectrum schemes can be incorporated into regional or global aerosol models in different ways. One approach is to use

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- sectional-size models that categorize the particles into a manageable number of bins according to their sizes (e.g., Gelbard et al., 1980; Wexler et al., 1994; Jacobson, 1997; Russell and Seinfeld, 1998). The accuracy of sectional models very much depends on
- the number of bins applied. Fewer bins inevitably have higher levels of error. Numerical diffusion is a fundamentally challenging problem for the sectional methods when solving the mass transfer among bins. Another problem is cloud droplet activation, for which the size of aerosol particles that can be transformed into cloud drops depends on the ambient supersaturation. This cutoff size lies somewhere within the a particular
- bin, which means that only a portion of the aerosol particles in the particular bin can be activated, yet normally in cloud models they are all activated at the same time. Also, the growth kernel in each bin is often assumed to be constant; in reality, however, the growth kernel usually is very sensitive to aerosol size and thus may vary significantly between bin limits. Using a large number of bins can reduce the numerical diffusion; at
- the same time, however, it results in an increase the computational burden. In particular, the computational time required for particle coagulation processes is proportional to the square of the bin number. Therefore, when computational resources are limited sectional schemes are not suitable for regional or large scale models.

Another frequently used approach for aerosol simulations is the so-called modal schemes, a complete aerosol size distribution is composed of several modes, and each mode is represented by a relatively simple mathematical function. The evolution of the size distribution is solved by deriving analytical solutions for an integral of the size distribution multiplied by the growth kernel. A commonly adopted mathematical function for such a purpose is the log-normal distribution, as it is a rea-





sonably good representation of the observed aerosol size distribution in various environments (Whitby, 1978; Jaenicke, 1993; Hoppel et al., 1994; Brechtel et al., 1998; Bott, 1999). Computation is less intensive for such modal approaches because the number of variables that need to be tracked is significantly reduced. Zhang et al. (1999) showed

- that the modal approach is within reasonable agreement of the sectional model, and requires only about 1 % of the CPU time when calculating coagulation. Because of this, the modal approach has been widely adopted in current aerosol models (e.g., Seigneur et al., 1986; Whitby et al., 1991; Binkowski and Shankar, 1995; Whitby and Mcmurry, 1997; Ackermann et al., 1998; Harrington and Kreidenweis, 1998; Schell et al., 2001;
- Wilson et al., 2001). For example, the modal scheme of Binkowski and Shankar (1995) (hereafter called BS95), or its modified version, is one of the standard schemes in the Community Multiscale Air Quality Modeling System (CMAQ), as well as in the Weather Research and Forecasting (WRF) model coupled with Chemistry (WRF-CHEM). As the modal schemes apply a continuous size spectrum, they may avoid some of the problems associated with the sectional models as mentioned previously. For exam-
- ple, Cheng et al. (2007, 2010) were able to apply the modal aerosol size spectrum to accurately calculate the number of cloud drops formed by the activation process.

The main weakness of modal parameterization is that analytical solutions are needed for calculating the evolution of size distribution, but the exact solutions are not

- ²⁰ always available. Take the Brownian coagulation process for example. Because the collision kernel is too complicated, a general form of its analytical solution is unattainable. In this situation, the coagulation integral must be simplified to get an analytical solution; however, this simplification can lead to large uncertainties. Therefore, we developed in this study a set of aerosol parameterization methods to provide better accuracy and
- ²⁵ computation efficiency for aerosol simulations. These methods are applied to parameterize microphysical processes, such as ice nucleation, condensation, coagulation, sedimentation, as well as to provide diagnostic equations, such as the Kelvin effect on aerosol wet size.





2 Methodology

The basic concept behind our new approach is to perform offline spectral integration for each process and then use the results to fit into parameterization formulas. This approach is somewhat similar to that of Chen and Liu (2004) who developed a moment-

- ⁵ conserving parameterization for warm cloud microphysics by statistically analyzing the results from a detailed sectional model. In their approach, the spectral evolution tendency is solved by in-line numerical integration with the sectional model. Then, the spectral moments (i.e., moments integrated over the whole size spectrum) and their rate change due to each process are stored and later analyzed statistically to produce
- fitting formulas. Apart from a reliable sectional model with full physical and chemical processes, prerequisites of their approach include performing simulations with a wide range of conditions so that all possible variations of the size spectrum are taken into account. These tasks are quite tedious but can be rewarded with accurate and efficient parameterization formulas. Furthermore, unlike the conventional modal methods,
- assumption of the particle size spectrum is not required. But this method has its weaknesses. For example, different bins in the sectional method need to be divided into bulk groups, such as cloud drop versus raindrop in cloud microphysics, or nucleation mode versus accumulation mode in aerosol microphysics. Yet, very often there is no distinct gas between the two populations. Therefore, an artificial and not always appropriate
- ²⁰ cutoff between the groups (or modes) must be applied. For cloud modeling, this might not be a significant problem, because the separation of the cloud drops and raindrop populations is often quite distinct. However, the modal populations of aerosols usually have large overlaps, as shown by Whitby (1978). Another weakness of this approach is in the setup of the conditions that drive the evolution of the size spectra. If not designed
- carefully, the results may be biased toward certain processes or spectral modes. Additionally, there is also the pre-requisite that a full-physics sectional model with good numerical schemes is available.





Here we introduce an alternative approach that not only deals with the above problems, but also simplifies the whole parameterization procedure. Instead of calculating all processes together in a sectional model, as was done by Chen and Liu (2004), we performed numerical integration for each individual process under specified conditions.

⁵ This approach demands less effort in programming and enables control of the variation of size distribution to cover all possible conditions, avoiding the need to assign a cutoff between two modes of particle populations in a sectional model.

2.1 Size distribution function and its moments

The first step of this modal approach is to select a mathematical function that best
represents the number density distribution of each modal population. Note that the need to specify a size distribution function is a disadvantage compared to the offline bin model approach of Chen and Liu (2004). However, the aerosol size distribution can generally be represented well by the multi-mode log-normal function (Whitby, 1978); several studies have indicated that such a distribution is self-preserving (Friedlander, 1960; Hidy, 1965; Liu and Whitby, 1968; Lai et al., 1972). Therefore, we select the log-normal function to represent each modal distribution:

$$n(\ln r) = \frac{N}{\sqrt{2\pi\sigma}} \exp\left[-\frac{\ln^2\left(\frac{r}{\mu}\right)}{2\sigma^2}\right],$$

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where r is the particle size, N is the total number of particles, σ is the standard deviation (in the ln r coordinate), and μ is the modal radius.

However, these modal parameters are not extensive properties and thus cannot be used as prognostic variables in atmospheric models. In practice, the desirable tracking variables are the moments of the size distribution, such as the 0th moment (i.e., number concentration) and 3rd moment (i.e., volume concentration). The *k*th moment is defined



(1)



$$M_k = \int r^k n(r) \, \mathrm{d}r$$

For n(r) in the log-normal form, an analytical solution for Eq. (2) can be solved as

$$M_k = M_0 \mu^k \exp\left(\frac{k^2 \sigma^2}{2}\right)$$

- ⁵ The 0th and 3rd moments are logical choices for tracking variables because of their direct relevance to many physical properties. Yet, the selection of the next moment is optional. For example, in cloud microphysical parameterization, Milbrandt and Yau (2005) used the 0th, 3rd, and 6th moments. The 6th moment represents the radar reflectivity, which is an important characteristic of large precipitation particles. Binkowski
- and Shankar (1995) also selected the 6th moment for their aerosol parameterization because it allows easier derivation of analytical solutions. However, the cross-sectional area, represented by the 2nd moment, is important to light scattering and atmospheric radiation and is consequently more relevant to aerosol studies. Thus, we select the 2nd moment as the third tracking variable for this study. Note that the current CMAQ model does not follow the scheme of Binkowski and Shankar (1995), but instead considers
- the 2nd moment (Binkowski and Roselle, 2003).

The size distribution parameters in Eq. (1) can be diagnosed from the three moments as

$$\sigma^2 = \frac{1}{2} \left[\ln M_0 + 2 \ln M_3 \right] - \ln M_2 \tag{4}$$

²⁰ which can then be used to calculate the modal size:

 $\mu^3 = \frac{M_3}{M_0 \exp\left(4.5\sigma^2\right)}$

(2)

(3)

(5)



Note that the methodology shown in the next section is not restricted to the lognormal size distribution. Another distribution function that is mathematically and numerically attractive for the representation of particle size spectrums is the gamma-type (or modified-gamma) distribution:

$${}^{\scriptscriptstyle 5} \quad n(r) = Nr^i \exp\left(-\lambda r^j\right)$$

Variants of this generic form include the common gamma distribution (j = 1), the Weibull distribution (i = j - 1), the Khrgian-Mazin distribution (i = 2 and j = 1), and the Marshall-Palmer distribution (i = 0 and j = 1). For such distributions, the *k*th moment has a general solution:

$$M_{k} = \frac{N\Gamma(l+1)}{j\lambda^{l+1}}$$

where $l \equiv (k + i - j + 1)/j$ and Γ is the gamma function. Similar to Eqs. (4) and (5), the variables *N*, λ , *i*, and *j* are interchangeable with an equal number of known moments. Take for example the common gamma distribution (*j* = 1 and *l* = *k* + *i*). The so-called shape parameter *i* can be derived as

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$$i = \frac{-(3q-6) + \sqrt{q(q+8)}}{2(q-1)},$$

where $q \equiv \frac{M_0 M_3^2}{M_2^3}$. With this, we can obtain the slope parameter $\lambda = \frac{M_2(i+3)}{M_3}$

and the frequency parameter

$$N = M_0 \frac{\lambda^{i+1}}{\Gamma(i+1)}.$$





(6)

(7)

(8a)

(8b)

(8c)

2.2 Parameterization methods

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After the mathematical form and the key parameters of the size distribution are determined, the evolution of size distribution can be described in terms of the rate change of the moments:

$$\int \frac{\mathrm{d}M_k}{\mathrm{d}t} \equiv I_k = \int K_k n(r) \,\mathrm{d}r = \int \frac{\mathrm{d}r^k}{\mathrm{d}t} n(r) \,\mathrm{d}r$$

where K_k is the growth kernel for the *k*th moment. When the growth kernel is not in a simple form, solving such integrals requires computationally intensive numerical techniques, such as Gauss–Legendre or Gauss–Hermite numerical quadrature. Therefore, the parameterization of Eq. (9), which enables the efficient and accurate calculation of aerosol and cloud microphysical processes, is essential to many meteorological and air pollution models.

Common treatments of Eq. (9) include the use of lookup tables and kernel simplification. The lookup table approach calculates the kernel or the whole integral as a function of their key parameters and then arranges the results in tables which, when applied in ¹⁵ models, can be searched according to the current values of those parameters. This method has the advantage of fast calculation, as it primarily involves searching, and has high accuracy when the tables are large enough. However, the method may become cumbersome to use when the process involves too many parameters that require large table dimensions. In addition, the lookup table method usually cannot be used

- directly for physical interpretation or analysis of the functional dependence on key parameters. Alternatively, the kernel simplification approach is commonly applied in the parameterization of both aerosol and cloud microphysics. Its specific purpose is to allow easy evaluation of Eq. (9) into analytical solutions. However, such simplifications are often too rough and can result in large errors.
- ²⁵ We investigated four methods of parameterization: (A) mean-size approximation, (B) kernel transformation, (C) integral transformation, and (D) optimal-size approximation.



(9)



One of these methods is selected for our final parameterization, based on the accuracy of the analyses. Since the last three methods apply statistical fitting on numerically integrated results, our overall method is named the Statistical-Numerical Aerosol Parameterization (SNAP).

5 2.2.1 Mean-size approximation

Mean-size approximation (hereafter called SNAP-A) is achieved by replacing all or some of the size variable *r* in the growth kernel with a constant size \overline{r} , so that the kernel, or part of the kernel, can be taken out of the integral in Eq. (9). Assuming that the growth kernel can be represented by a polynomial function of *r* (i.e., $I = \int \sum_{i} a_{i} r^{i} \cdot n(r) dr$), the error associated with such a mean-size approximation can be evaluated for each term of order *i* (neglecting the coefficient a_{i})

$$I_i = \int r^i \cdot n(r) \,\mathrm{d}r \tag{10}$$

which has an exact solution M_i as given earlier in Eq. (3). On the other hand, the mean size approximation is

15
$$\widetilde{I}_i = \int \overline{r}^i \cdot n(r) dr = \overline{r}^i M_0$$
 (11)

Several forms of the mean size \overline{r} can be used for SNAP-A. A group of these forms is called the moment-weighted mean size $r_n \equiv (M_n/M_0)^{1/n}$. For example, r_2 and r_3 are the surface- and volume-weighted mean sizes, respectively. According to Eq. (3), r_n can be converted to

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$$r_n = (M_n/M_0)^{1/n} = \mu \cdot \exp(n\sigma^2/2)$$
 (12)

Let us use \tilde{I}_{i,r_n} to represent the approximate solution using these *n*th-moment-weighted sizes. Its ratio to the exact solution I_i can be derived as

$$\frac{\widetilde{I}_{i,r_n}}{I_i} = \exp\left[(n-i)i\sigma^2/2\right]$$

Other forms of the mean size include the modal size μ in Eq. (1) and the effective radius $r_e \equiv M_3/M_2$, which is commonly used for radiation budget calculation. Ratios of the solution using these two mean-size approximations to the exact solution can be derived as

$$\frac{\widetilde{I}_{i,\mu}}{I_i} = \exp\left[-i^2\sigma^2/2\right]$$

$$\int_{0}^{0} \frac{I_{i,r_{e}}}{I_{i}} = \exp\left[(5-i)i\sigma^{2}/2\right]$$
(15)

The approximations using μ and r_e are special cases of Eq. (13), with n = 0 and n = 5, respectively. So, μ and r_e may be called the 0th and 5th moment-weighted sizes, respectively. Figure 1 shows the errors associated with these mean-size approximations, which exhibit the following features: (1) the error increase with the width of the size spectrum (i.e., σ), the order of the kernel (i.e., *i*), and the difference between *n* and *i* (i.e., |n - i|) in Eq. (13). Therefore, the error can be minimized if *n* is set as equal to *i*. (2) The error is positive for n > i and negative for n < i. This indicates that the signs of error may be opposite for the growth of different moments. For growth kernels containing several polynomial terms, it would be best to select *n* that lies between the orders of all of the terms, such that their errors may cancel each other.



(13)

(14)



2.2.2 Kernel transformation

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A complicated growth kernel prohibits the derivation of an analytical solution for Eq. (9). However, it is possible to transform such kernels into manageable mathematic forms. We call this approach SNAP-B. For a log-normal n(r), useful mathematical forms include the power-law function r^a , the exponential function $\exp(b\ln^2 r)$, or their combi-

nations. Each of these functions can be generalized as $r^a \exp(b \ln^2 r)$, which can also be expressed as $\exp(a \ln r + b \ln^2 r)$. This allows Eq. (9) to be expressed as

$$I_{k} = \frac{N}{\sqrt{2\pi\sigma}} \int r^{a} \exp\left(b\ln^{2}r\right) r^{k} \exp\left[-\frac{\ln^{2}\left(r/\mu\right)}{2\sigma^{2}}\right] dr$$

$$= \frac{N}{\sqrt{2\pi\sigma}} \int \exp\left[(a+k)\ln r + b\ln^{2}r\right] \exp\left[-\frac{\ln^{2}\left(r/\mu\right)}{2\sigma^{2}}\right] dr$$
(16)

Its solution can be derived by introducing the variable exchange $x \equiv \alpha \ln r - \gamma$, where 10 $\alpha \equiv \sqrt{\frac{1}{2\sigma^2} - b}, \beta \equiv a + k + \frac{\ln \mu}{\sigma^2}$, and $\gamma \equiv \frac{\beta}{2\alpha}$. We then have

$$I_{k} = \frac{N}{\sqrt{2\pi\sigma}} \frac{1}{\alpha} \exp\left(\gamma^{2} - \frac{\ln^{2}\mu}{2\sigma^{2}}\right) \int \exp\left(-x^{2}\right) dx = \frac{N}{2\sqrt{2}\sigma\alpha} \exp\left(\gamma^{2} - \frac{\ln^{2}\mu}{2\sigma^{2}}\right) \equiv F(a+k,b)$$
(17)

One can verify that Eq. (17) reduces to Eq. (3) when a = b = 0. In other words, Eq. (3) is the special case of F(k, 0). For gamma-type n(r), one can choose kernels in the form of r^a , exp $(-br^c)$, or their combinations, which can be easily incorporated into Eq. (6) and then applied to the general solution Eq. (7) for I_k .

The conversion of growth kernels into such functional forms is done by statistical transformations of numerically solved results. Take for example the gravitational sedi-





mentation velocity:

$$V_{\rm sed} = V_{\rm Stokes} C_{\rm C} = \frac{2g\rho_{\rm p}}{9\eta} \left\{ 1 + K_N \left[1.246 + 0.42 \cdot \exp\left(-\frac{0.87}{K_N}\right) \right] \right\}$$
(18)

were V_{Stokes} is the Stokes' law fall speed, C_{C} (term expressed in the braces above) is the Cunningham slip-flow correction, g is the normal gravitational acceleration, ρ_{p} is the particle density, η is the dynamic viscosity of air, $K_N \equiv \lambda/r$ is the Knudsen number, and λ is the mean-free path of air molecules. Note that C_{C} may take a form somewhat different from Eq. (18) (cf. Seinfeld and Pandis, 2006, p. 407), but our parameterization procedure works the same with both forms. Sedimentation flux for the whole size distribution (also termed the group sedimentation flux) is expressed as

10
$$I_k = \int V_{\text{sed}}(r) r^k n(r) dr$$
 (19)

As the analytical solution for this equation cannot be readily obtained the BS95 scheme simply ignores the exponential term in Eq. (18) to obtain the following solution:

$$I_{k} = \frac{2g\rho_{\rm p}}{9\eta} \int r^{2} \left(1 + 1.246\frac{\lambda}{r} \right) r^{k} n(r) dr = \frac{2g\rho_{\rm p}}{9\eta} \left(M_{k+2} + 1.246\lambda M_{k+1} \right)$$
(20)

Under standard atmospheric conditions, omitting the exponential term in $C_{\rm C}$ would ¹⁵ cause an underestimation in sedimentation speed by 4 % and 26 % for particles of 0.1 and 0.01 µm radii, respectively (Fig. 2). Such underestimations actually contribute to a small absolute error in the group sedimentation flux; the percentage error is significant only for small particles whose gravitational fall speed is low. However, an accurate description of $C_{\rm C}$ may still be important for other calculations. For example, $C_{\rm C}$ is an ²⁰ important parameter in the Brownian coagulation kernel (see Sect. 2.4).

If one wants to consider the exponential term for better accuracy, we can apply SNAP-B by calculating $C_{\rm C}$ for a realistic range of K_N then apply statistical fitting of





the results using commercially available software. For example, after calculating $C_{\rm C}$ for a range of K_N values, their relation can be curve-fitted into the following:

$$C_{\rm C} \approx 1 + a_1 K_N^{a_2} = 1 + a_1 (\frac{\lambda}{r})^{a_2}$$
 (21)

where $a_1 = 1.43089$ and $a_2 = 1.0295$ are the fitting coefficients. From Fig. 2, one can see that the above fitting is quite accurate, with less than 5% error (R^2 of fitting = 0.9999) for all relevant values of K_N . Adding more terms to Eq. (21) may give even higher accuracy but is not necessary for practical purposes. This transformation allows Eq. (19) to be evaluated analytically as

$$I_{k} \approx \frac{2g\rho_{\rm p}}{9\eta} \left(M_{2+k} + a_{1}\lambda^{a_{2}}M_{2+k-a_{2}} \right)$$
(22)

¹⁰ Whitby et al. (1991) applied a similar transformation but used different a_2 values for different K_N regimes to gain better accuracy. The above parameterization might not be of practical use, as the error is relatively large only for very small aerosol particles whose motion in the air is controlled not by gravitational sedimentation but by Brownian motion. Nevertheless, a more accurate representation of C_C may become important in ¹⁵ some situations, such as the calculation of Brownian collision coefficients (see details in Sect. 2.3).

2.2.3 Integral transformation

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SNAP-B formulations, such as Eq. (22), are computationally efficient. In fact, SNAP-B formulations are usually more efficient than the next two approaches. Yet, satisfactory fitting of the growth kernel, as discussed above, is not always available. When this is the case, we can turn to the integral transformation method (hereafter called SNAP-C) which involves two steps: (1) solving Eq. (9) numerically by discretizing the size spectrum into fine bins (as fine as possible) for a wide range of ambient conditions and size



spectrum parameters (e.g., μ and σ); (2) analyzing the results by statistical fitting to obtain a transformed formula. However, a technical problem may arise while performing the fitting. Besides the three moments, the growth equation often contains other dependent variables, such as air temperature and pressure. Few statistical software

- ⁵ packages can handle nonlinear fitting on multiple variables. For example, the commercial software we are using can handle only two variables at a time. Processing all of the variables may require intensive trial by error or iteration before a satisfactory parameterization formula can be acquired. Consequently, a conversion of the growth kernel for the purpose of variable separation before performing the numerical integration is still
- ¹⁰ necessary. For instance, the ambient parameter λ in Eq. (21) becomes a coefficient and can be taken out of the integral. However, such separation is not always easy, and this greatly limits the application of this approach. We overcome this deficiency by taking advantage of the mean-size method in which the dependence on ambient parameters is largely retained in the simplified kernel. We obtain SNAP-C first by rewriting Eq. (9)
- 15 **as**

$I_k \cong \widetilde{I}_k \cdot g_{1,k}$

where \tilde{I}_k is the modal-value approximation of I_k , and g_1 is a correction factor that brings \tilde{I}_k closer to I_k . The corrector g_1 should be a strong function of the spectral width σ because \tilde{I}_k is calculated by assuming a monodisperse size distribution (and thus $\sigma = 0$). We derive g_1 by using the integral transformation method, i.e., by integrating Eq. (9) numerically (with finely discretized size bins) for a range of σ , as well as other size distribution parameters and ambient parameters to obtain the "true" value of I_k . Each I_k value is then divided by \tilde{I}_k , and their ratios are fitted to obtain g_1 as a function of σ and other parameters. In this way, the ambient-parameter dependence is largely

retained in *K*, while the dependence on the spectral width σ is largely contained in g_1 . Note that some computational efficiency is lost by keeping the details of the growth kernel in \tilde{I}_k , as compared with a direct integral transformation, such as SNAP-B. This loss in computational efficiency is well compensated by the accuracy that is gained.



(23)



2.2.4 Optimal-size approximation

In SNAP-A, we assume that $I_k \cong \tilde{I}_k(\mu)$, and in SNAP-C, we find a correction factor to improve this approximation. The deviation of $\tilde{I}_k(\mu)$ from I_k indicates that the modal value μ (or any other mean size) may not be the best representative size. In fact, we showed in Eq. (13) that this "best size" is actually a function of the order of the kernel and spectral width σ , and potentially some ambient parameters as well. Thus, instead of using a specific mean size (i.e., μ) and then correcting the whole integral with g_1 , as done in SNAP-C, it may be possible to find in advance an optimal mean size, which can be adjusted with the imposed conditions to provide an accurate value of \tilde{I}_k directly, according to the following relationship:

 $I_k \cong \widetilde{I}_k(\mu \cdot g_{2,k}) \equiv \widetilde{I}_k(\mu')$

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25

To determine the formula for the optimal size μ' for this SNAP-D method, we first calculate I_k for a range of relevant parameters. For each I_k value, we search by iteration a value of μ' which, when placed into \tilde{I}_k , gives an exact value of I_k . Afterward, the ratios of μ' to μ (i.e., g_2) under various conditions are analyzed statistically to fit into a function of the key parameters, such as σ or μ .

In summary, SNAP-A can be considered as a no-skill parameterization. We will demonstrate that the other three SNAP parameterization methods are all significantly more accurate and thus have high skills. Among them, SNAP-C is suitable for parameterizing more accurate and thus have high skills.

²⁰ terizing most of the aerosol processes, whereas SNAP-D can occasionally be applied to provide somewhat better parameterizations than SNAP-C.

3 Parameterization of microphysical processes

In this section, we apply the above methods to various aerosol microphysical processes and analyze the parameterization accuracy by comparison with the numerical solutions. The numerical solutions for I_{μ} are obtained by discretizing the size spectrum into



(24)



10 bins per decade change in radius, and then summing the rates from individual bins. Higher resolutions are also tested; the differences are less than 0.1 %, which can be regarded as the precision of the numerical solutions. In this study, the error is defined as $\operatorname{abs}(\exp\left\{\sum_{i=1}^{j}\left[\operatorname{abs}\left(\ln \widetilde{I}_{k}/I_{k}\right)\right]/j\right\}-1\right)$, where *j* is the number of conditions selected for evaluation.

3.1 Ice nucleation

Heterogeneous ice nucleation from insoluble aerosol particles (which are thus called ice nuclei) such as mineral dust, soot or bio-aerosols is an important factor in the glaciation of clouds. This process is usually not considered in traditional aerosol models, but

is included here because more models are considering detailed aerosol-cloud interactions for which ice nucleation is a critical mechanism (cf. Tao et al., 2012). According to the classical theory, heterogeneous ice nucleation rate can be generalized into the following form for several modes of nucleation (cf. Chen et al., 2008):

$$J_{\rm HN} = 4\pi r^2 A \sqrt{f} \exp\left(\frac{-\Delta g_{\rm a} - f \cdot \Delta g_{\rm g}}{k_{\rm B}T}\right)$$

where *r* is the radius of the ice nuclei, *A* is a parameter that depends on the ambient conditions only, *f* is a size-dependent geometric factor, Δg_a is the activation energy, Δg_g is the homogeneous germ formation energy, and k_B is the Boltzmann constant. The overall nucleation rate for a population of ice nuclei is then expressed as

$$J_{k} = \int J_{\text{HN}} \cdot r^{k} \cdot n(r) \,\mathrm{d}r \tag{26}$$

which represents the rate of decrease in ice nuclei or increase in cloud ice number concentration. This integral cannot be solved analytically, as the geometric factor f,



(25)

which appears twice in the kernel J_{HN} , has a very complicated form:

$$f = \frac{1}{2} \left\{ 1 + \left(\frac{1 - mq}{\phi}\right)^3 + q^3 \left[2 - 3\left(\frac{q - m}{\phi}\right) + \left(\frac{q - m}{\phi}\right)^3 \right] + 3mq^2 \left(\frac{q - m}{\phi} - 1\right) \right\}$$
(27)

where $m \equiv \cos(\theta)$, θ is the contact angle, $q \equiv r/r_q$ is the ratio of the nuclei size to the nucleation-germ size, and $\phi \equiv \sqrt{1 - 2mq + q^2}$. There are several modes of heteroge-⁵ neous ice nucleation. Here, we take the immersion freezing nucleation as an example. Its key ambient parameters include temperature and saturation vapor pressure over water (with solute and curvature effects) of the supercooled droplet wherein the ice nuclei are immersed.

Applying SNAP-A to Eq. (26) is straightforward:

$$I_{k} \approx \widetilde{J}_{HN} \int r^{k} \cdot n(r) dr = \widetilde{J}_{HN} M_{k}$$
(28)

where \widetilde{J}_{HN} is Eq. (25) calculated with the modal size. One may also take out the prefactor r^2 in $J_{\rm HN}$ and let $I_k \approx \widetilde{J}_{\rm HN}' M_{k+2}$ where $\widetilde{J}_{\rm HN}'$ is calculated with $J_{\rm HN}' = J_{\rm HN}/r^2$. For the parameterization using SNAP-B, the parameter f in Eq. (25) should be transformed in order to derive a semi-analytical solution for Eq. (26). The following is a readily available formula from Chen et al. (2008):

$$\ln f \approx a_1 + a_2 \ln(1 - m) + a_3 \ln \frac{r}{r_a}$$

where a_1 , a_2 , and a_3 are constants. This formula is suitable for converting the first term that contains f in Eq. (25) into

 $\sqrt{f} \approx a_{\Lambda} r^{a_3/2}$

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(29)

(30)

where $a_4 \equiv \sqrt{\exp(a_1 + a_2 \ln(1 - m) - a_3 \ln r_g)}$ is independent of r. However, this formula is not useful for simplifying f in the exponential term. Thus, we produced another transformation formula:

$$\ln f \approx \ln \left(b_1 + b_2 \ln \frac{r}{r_g} \right) + b_3 + b_4 \ln \left(1 - m \right) + b_5 \left(1 - m \right)$$
(31)

s where $b_1 = 4.5102915$, $b_2 = -0.1130082$, $b_3 = -1.6013035$, $b_4 = 2.0058907$, and $b_5 = -1.6013035$ -0.45839176. With this approximation, we have

$$\exp(Bf) \approx \exp(c_1) r^{c_2} \tag{32}$$

where $B = \frac{\Delta g_g}{k_0 T}$, $c_1 = B \exp(b'_3) (b_1 - b_2 \ln r_g)$, and $c_2 = B \exp(b'_3) b_2$ are all independent of r. The R^2 of fitting for Eqs. (29) and (31) both reached 0.9998 for θ in the range of 1° to 110° and q from 10 to 400; it could be more accurate if the ranges are divided into a few sectors, each with its own fitting coefficients. With Eqs. (30) and (32), the overall nucleation rates for a spectrum of ice nuclei can be derived as

$$I_{k} = \int J_{\rm HN} r^{k} n(r) \, \mathrm{d}r \approx 4\pi A a_{4} \exp\left(c_{1} - \frac{\Delta g_{\rm a}}{k_{\rm B}T}\right) M_{2+k+c_{2}+a_{3}/2} \tag{33}$$

15

For SNAP-C, we first perform numerical integration on Eq. (26) and then compare the results with the modal approximation Eq. (28) to obtain a fitting on g_1 . The selection of the fitting parameters is not a trivial task. After trial by error, we found that the most relevant parameter, besides the spectral width σ , is the parameter q in Eq. (27), and we represented it with $\overline{q} \equiv \mu / r_{q}$, since it might remove some of the size dependence. Here is one of the better fitting formulas:

²⁰
$$g_1 = \exp\left[a_1 \cdot \sigma^2 + a_2 \cdot \exp\left(-\overline{q}\right)\right]$$

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From this point onward, we will not show the fitting coefficients, as they are listed in the Appendix. This adjustment only needs to be applied to $\mu < 0.1 \,\mu\text{m}$. Whereas for SNAP-D, we obtain a formula for g_2 :

$$g_2 = \exp\left(a_1 \cdot \sigma^2 + \frac{a_2}{\overline{q}^2}\right)$$

Figure 3 shows that these two formulas provide reasonably good fittings. It also reveals that large corrections are necessary when q is small and, at least in the case of g_2, σ is large. Note that there are numerous fitting formulas for our selection, and we often select those that are easier to use or can reflect physical meanings, but are not of the highest accuracy. For example, we select Eqs. (34) and (35) to warrant a unit value toward the extreme conditions of $\sigma \rightarrow 0$ and $\overline{q} \rightarrow \infty$. Recalling Eq. (13) we showed an " $\exp(\sigma^2)$ " dependence between the mean-size approximation and the exact solution. Such a relationship is reflected in Eq. (34).

Next, we compare the four parameterization approaches against the detailed numerical solution. The results for immersion freezing are shown in Fig. 4, for which the ranges of values tested are the following: 6 modal sizes (μ) between 0.02 and 4.0 μ m, 15 10 modal widths (σ) between 0.26 and 0.95, 8 temperatures between -5 and -40 °C, and 4 water activities between 0.82 and 1.0. Visually, one can tell that the SNAP-D method performs significantly better than the other methods do. The mean errors in I_0 are 39% for SNAP-A, 21% for SNAP-B, 22% for SNAP-C, and 5% for SNAP-D. Additionally, the error increases toward higher moments. For example, the error values 20 calculated for I_2 and I_3 when applying the SNAP-D method are 15% and 24%, respectively. Fortunately, large deviations occur only when the absolute values are close to negligible. Note that there exist feather-like features in the left panel of Fig. 4, and each filament represents a set of values with different σ values. The SNAP-A points with the largest σ values are indicates by black circles. The largest error is associated 25 with the highest σ , and the error approaches zero for a monodisperse distribution (i.e.,



(35)



methods. We will omit similar details when discussing the parameterization for other processes.

3.2 Condensation

Under the assumption of a steady-state diffusion process, the kernel of condensation growth following the two-stream Maxwellian kinetic theory with a steady-state assumption is commonly expressed as (cf. Pruppacher and Klett, 1997)

 $\frac{\mathrm{d}m}{\mathrm{d}t} = 4\pi r D f_{\mathrm{g}} f_{\mathrm{v}} \left(\rho_{\mathrm{v},\infty} - \rho_{\mathrm{v},\rho} \right)$

where *D* is the diffusion coefficient, f_g is the modification due to the gas kinetic effect (Fuchs, 1959, 1964), f_v is the ventilation coefficient, which can be ignored for small aerosol particles; $\rho_{v,\infty}$ is the ambient vapor density, and $\rho_{v,p}$ is the surface vapor density. The parameters D, $\rho_{v,\infty}$, f_g , and $\rho_{v,p}$ are species-dependent, whereas f_g and $\rho_{v,p}$ are also size dependent. Furthermore, $\rho_{v,p}$ is influenced by latent heating/cooling during condensation/evaporation. A quasi-analytical solution can be obtained to account for this effect (cf. Pruppacher and Klett, 1997, p. 510), but the details will not be elabo-15 rated here.

Equation (36) can be generalized for the simultaneous condensation of multiple species. Let the volume change due to condensation be $dv = dm/\rho_L$, where $v = 4\pi r^3/3$ and ρ_L is the density of the condensate. From this, the bulk growth rate of the *k*th moments can be expressed as

$${}_{20} I_{k} = \int \frac{\mathrm{d}r^{k}}{\mathrm{d}t} n(r) \,\mathrm{d}r = \int \frac{k}{4\pi} r^{k-3} \frac{\mathrm{d}v}{\mathrm{d}t} n(r) \,\mathrm{d}r = \int \frac{k D f_{g} \left(\rho_{v,\infty} - \rho_{v,p}\right)}{\rho_{L}} r^{k-2} n(r) \,\mathrm{d}r \tag{37}$$

Note that in this formula the rate change of the total number (k = 0) for the condensation process necessarily equals zero. If we assume that $Df_{g}(\rho_{v,\infty} - \rho_{v,p})$ is size-



(36)

independent, then its analytical solution can be derived as

$$I_{k} = \frac{kDf_{g}(\rho_{v,\infty} - \rho_{v,\rho})}{\rho_{L}} \int r^{k-2}n(r)dr = \frac{kDf_{g}(\rho_{v,\infty} - \rho_{v,\rho})}{\rho_{L}}M_{k-2}$$
(38)

However, in reality the size dependence of f_g cannot be ignored, particularly for small aerosol particles. This parameter is generally expressed as

$${}_{5} \quad f_{g} = \frac{1}{\left[\frac{r}{r+\Delta} + \frac{4D}{\alpha v r}\right]}$$

10

where Δ is the vapor jump distance and is on a scale similar to that of the mean free path λ , α is the mass accommodation coefficient, and ν is the mean thermal velocity of the gas molecules (cf. Fuchs, 1959; Pruppacher and Klett, 1997). Considering the dependence of Δ on λ , Fuchs and Sutugin (1970) provided an empirical formula for f_g as a function of K_N and α :

 $f_{\rm g} = \frac{0.75\alpha(1 + K_N)}{K_N^2 + K_N + 0.283K_N\alpha + 0.75\alpha}$

It is difficult to arrive at analytical solutions to Eq. (37) with the formulas for f_g given in Eqs. (39) and (40). An approach to resolving such a problem, as suggested by Pratsinis (1988) and adopted by the BS95 method, is to consider the harmonic mean of growth in the free-molecular regime and continuum regime:

$$I_k \approx \frac{I_{\mathrm{M},k} I_{\mathrm{C},k}}{I_{\mathrm{M},k} + I_{\mathrm{C},k}} \tag{41}$$

where $I_{M,k}$ is calculated with the free-molecular regime growth kernel $K_M = \pi r^2 \alpha v (\rho_{v,\infty} - \rho_{v,p})$, and $I_{C,k}$ with the continuum regime kernel $K_C = 4\pi r D (\rho_{v,\infty} - \rho_{v,p})$. Since $I_{M,k}$ and $I_{C,k}$ can be solved analytically as a function 12054



(39)

(40)

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of M_k and M_{k-1} , respectively, Eq. (37) can be evaluated analytically. Although Pratsinis (1988) indicated that the harmonic mean can approximate the results well using f_g from the equation developed by Fuchs and Sutugin (1970) shown in Eq. (40), it inevitably contains some inaccuracy, which we will evaluate below. Fukuta and Walter (1970) suggested a slightly different form of f_g , which, in effect, excludes the term Δ in Eq. (20), and is, for practical purposes, a harmonic mean of K_M and K_C

$$f'_{g} = \frac{1}{\left[1 + \frac{4D}{\alpha v r}\right]}.$$

5

Below we omit the application of the SNAP-B method, because the fitting formula becomes too cumbersome for practical purposes. Additionally, we omit the SNAP-D method because the SNAP-C method is sufficient. The SNAP-C fitting formula that we derived is as follows:

$$g_1 = \exp\left\{\sigma\left[a_1 + a_2 \exp\left(-\ln\overline{K_N}\right) + a_3\sigma\right]\right\}$$
(43)

where $\overline{K_N} \equiv \lambda/\mu$ represents a mean Knudsen number. Figure 5 shows the comparison between various parameterization methods for the condensation growth process. Note that the number concentration does not change during the condensation process (i.e., $I_0 = 0$), so only I_2 and I_3 are presented. SNAP-A gives good results only when σ is small but it is biased toward lower values for increasingly larger σ values (i.e., the true value increases with σ , but SNAP- A does not). The overall error for SNAP-A is 17 % in I_2 and 92 % in I_3 . SNAP-C performed rather well, with 0.74 % and 1.3 % error in I_2 and I_3 ,

²⁰ respectively. The BS95 method produced significantly larger discrepancies, with 10.7 % and 57.1 % error in M_2 and M_3 , respectively. However the BS95 computation time is 21 % less than that of the SNAP-C method. In Fig. 5 we also plotted the numerical solutions using f'_g from Fukuta and Walter (1970). The strong positive biases (around 83 %) indicate the error associated with the harmonic mean approximation.



(42)



3.3 Brownian coagulation

Calculation of the rate change of moments caused by collision-coagulation processes involves double integrals over the size spectra of the two aerosol modes involved. For coagulation between two particles of sizes r_A and r_B , the coagulated particle has a size $r_C = (r_A^3 + r_B^3)^{1/3}$. It follows that the changes in their *k*th moments are $-r_A^k$ and $-r_B^k$, respectively, for each original particle, and $+r_C^k$ for the coagulated particle. With these parameters defined, the fundamental equation for coagulation between particles in the collector mode *A* and the contributor mode *B* can be expressed as

$$I_{k,A} = \iint \left[r_C^k - r_A^k \right] \mathcal{K}(r_A, r_B, C_{air}) n_A(r_A) n_B(r_B) dr_A dr_B$$
(44)

$$I_{k,B} = \iint \left[-r_B^k \right] \mathcal{K}(r_A, r_B, C_{\text{air}}) n_A(r_A) n_B(r_B) dr_A dr_B$$
(45)

where the coagulation kernel *K* is usually a nonlinear function of the two particle sizes and environment properties denoted by the parameter C_{air} . Note that the coagulated particle is placed back into mode *A* as indicated in Eq. (44). In these generalized equations one can easily verify that the number concentration (M_0) in the collector mode remains unchanged (i.e., $I_{0,A} = 0$) and that the total volume is conserved (i.e., $I_{3,A} = -I_{3,B}$). So, a total of four conversion rates are needed, i.e., $I_{0,B}$, $I_{2,A}$, $I_{2,B}$, and $I_{3,A}$ (or $-I_{3,B}$). For the intra-modal coagulation (i.e., A = B), the number of rates reduces to two, and all coagulation rates in should be divided by 2 to correct for double counting.

- Processes contributing to aerosol coagulation include Brownian diffusion, convective Brownian diffusion enhancement, gravitational collection, turbulent inertial motion, and turbulent shear flow (Jacobson, 1997). Brownian diffusion is the dominant coagulation process for fine aerosol particles with radii typically in the range 0.01–1 µm. Here, we take this most complicated kernel as an example for parameterization, starting with
- the intra-modal coagulation, which involves only its own moments. By analogy of gas diffusion formulation, Fuchs (1959) expressed the Brownian coagulation kernel K_{Br}





between particles A and B as

$$K_{\rm Br} = 8\pi \overline{r} \overline{D}_{\rm p} \beta$$

5

where $\overline{r} = (r_A + r_B)/2$, \overline{D}_p is the mean particle diffusion coefficient, and β represents the modification due to concentration discontinuity near the surface of the receiving particle. The mean particle diffusion coefficient is defined as $\overline{D}_p = (D_{p,A} + D_{p,B})/2$, where $D_{p,i} = \frac{k_B T C_{C,i}}{6\pi r_i \eta}$, C_C is the Cunningham slip flow correction factor, as shown in Eq. (19); k_B is the Boltzmann constant, and η is the dynamic viscosity of air. The conventional form of β is

$$\beta = \left(\frac{4\overline{D}_{\rm p}}{\alpha_{\rm p}\overline{v}_{\rm p}\overline{r}} + \frac{\overline{r}}{\overline{r} + \overline{\delta}}\right)^{-1} \tag{47}$$

¹⁰ where $\alpha_{\rm p}$ is the sticking probability (usually assumed to be unity) when two particles collide, $\overline{v}_{\rm p} = \sqrt{v_{{\rm p},A}^2 + v_{{\rm p},B}^2}$ in which $v_{{\rm p},i} = \sqrt{\frac{8k_{\rm B}T}{\pi m_i}}$ is the particle thermal velocity, *m* is particle mass, $\overline{\delta} = \sqrt{\delta_A^2 + \delta_B^2}$ in which $\delta_i = \frac{(2r_i + \lambda_{{\rm p},i})^3 - (4r_i^2 + \lambda_{{\rm p},i}^2)^{3/2}}{6r_i\lambda_{{\rm p},i}} - 2r_i$ represents a mean coagulation distance, $\lambda_{{\rm p},i} = \frac{2D_{{\rm p},i}}{\pi v_{{\rm p},i}}$ is the mean free path of the particle, and *i* is either *A* or *B*. The factor β has a similar form as Eq. (39). However, the variables that it contains, namely $\overline{\delta}, \overline{D}_{\rm p}$, and $\overline{v}_{\rm p}$, are all complex functions of the particles sizes, and this makes the SNAP-B method unfeasible to use. For this coagulation process, Pratsinis (1988) applied the harmonic-mean approximation. This approximation was also applied in the BS95 method:

$$I_{\rm Br} \approx \frac{I_{\rm Br,M} \cdot I_{\rm Br,C}}{I_{\rm Br,M} + I_{\rm Br,C}}$$

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where $I_{\text{Br,M}}$ and $I_{\text{Br,C}}$ are the results with kernels $K_{\text{Br,M}} = 2\pi \overline{r}^2 \alpha_p \overline{\nu}_p$ and $K_{\text{Br,C}} = 8\pi \overline{r} \overline{D}_p$, respectively. However, the complex forms of $C_C, \overline{\nu}_p$, and $\overline{\delta}$ still prevent the derivation of analytical solutions for $I_{\text{Br,M}}$ and $I_{\text{Br,C}}$. Thus, following Whitby et al. (1991), BS95 made a few algebraic manipulations and combined them with lookup tables to solve the harmonic mean.

For a similar reason, our parameterization for Brownian coagulation focuses on SNAP-A and SNAP-C, but ignores SNAP-B and SNAP-D. There is a complication in using SNAP-A here, because the two modal sizes used for the calculation are the same for intra-modal coagulation. We found it helpful to offset the modal radius and assign $r_A = \mu \cdot \sigma^2$ and $r_B = \mu / \sigma^2$ in Eqs. (44) and (45) for calculating \tilde{l}_k in Eq. (23). With this treatment, the correction factor for l_0 is obtained as

$$g_{1,0} = \exp\left[\frac{a_1 + a_2\sigma + a_3\ln\overline{K}_N}{a_4 + a_5\sigma + a_6\sigma^2 + a_7\ln\overline{K}_N + a_8\ln\overline{K}_N^2}\right]$$

which is used further to get the correction factor for I_2

$$g_{1,2} = g_{1,0} \cdot (a_1 + a_2 \ln \mu + a_3 \sigma^3)$$

- Figure 6 shows the results using SNAP-A and SNAP-C for intra-modal coagulations. Also compared is the harmonic-mean approximation of BS95, as well as the numerical solutions calculated with the 5th order Gauss–Hermit quadrature (GHQ), which is an accurate but computationally expensive option in the CMAQ model. Note that the amount of data for Brownian coagulation is much larger than that for the previous processes, so only a selected amount of data is shown to avoid clutter. One can see that
- BS95, GHQ, and SNAP-C all perform reasonably well. SNAP-C produces 3.7% and 5.9% errors in I_0 and I_2 , respectively, which are similar to those in GHQ (4.5% and 4.0%). The error in BS95 is about the same in I_0 (4.5%) but somewhat larger in I_2 (22%). The computation time used for SNAP-C and BS95 are 12% and 10% of that for GHQ, respectively.

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The inter-modal Brownian coagulation involves two size distributions, so one would imagine its parameterization must be more complicated than the intra-modal coagulation. However, using the SNAP-C method, we found a rather simple but accurate formula for all inter-modal rates:

$$5 \quad g_1 = \exp\left[a_1\sigma_A^2 + a_2\sigma_B^2\right] \tag{51}$$

It turns out that g_1 is mainly dependent on the two spectral widths (i.e., σ_A and σ_B), whereas the effects of other parameters, such as λ , have been largely reflected in the modal-mean, \tilde{I}_k , and thus play little role in g_1 . Also, this formula agrees with the exp (σ^2) dependence shown in Eq. (13). The two coefficients vary with the moments (i.e., the *k* value), but a_1 is consistently much smaller than a_2 (see Appendix B), indicating that inter-modal coagulation is more sensitive to the spectral width of the contributor mode (σ_B) than that of the collector mode (σ_A) .

Figure 7 shows the accuracy of various evaluation methods for these rates. The SNAP-A method again deviates from the numerical solution more pronouncedly at larger σ , and the mean error ranges from 18.1% to 74.1% for various moments. The SNAP-C method is rather accurate, having errors ranging from 2.6% to 4.5% for the four conversion rates, which are a little better than the errors of 4.8% to 5.4% produced by GHQ, and 4.8% to 7.4% produced by BS95. The computation time required for SNAP-C and BS95 are 7.8% and 7.0%, respectively, of that for GHQ.

20 3.4 Other processes and diagnostic parameters

10

A rate process that has not been discussed earlier is aerosol scavenging by cloud drops or raindrops, which is also a type of inter-modal coagulation. The mechanisms that control aerosol scavenging include Brownian diffusion, collection by phoretic forces, and gravitational collection. For the two former mechanisms, Wang et al. (1978) provided

²⁵ a mathematical solution that combines the two kernels, which is adopted for our parameterization. For the gravitational collection, we used the kernel in Slinn (1977). Pa-





rameterization procedures for these processes are quite similar to that for the Brownian coagulation, so only the final results are listed in Appendices A and B.

Another important microphysical process is aerosol nucleation (new aerosol production). The mechanisms that control aerosol nucleation include homogeneous binary or ternary nucleation (Nair and Vohra, 1975; Coffman and Hegg, 1995) and ion-enhanced nucleation (Yu, 2006). Although this process does not depend on the size distribution of existing aerosols and thus requires no modal parameterization, our SNAP approach can still be applied to speed up the calculation. Here we discuss briefly the concept of such parameterizations, using the homogeneous binary nucleation from water and sulfuric vapors as an example. The rate of binary nucleation depends mainly on the temperature and the saturation ratios of water vapor and sulfuric vapor. We will not focus on the details of the binary nucleation rates, which can be found in textbooks such as that written by Seinfeld and Pandis (2006, p. 514). Instead, we will focus on a key parameter that needs to be solved by iteration: the water-sulfuric acid mixing proportion in the critical embryo. Once this parameter is obtained, the calculation of

- ¹⁵ proportion in the critical embryo. Once this parameter is obtained, the calculation of nucleation rate is straightforward. In brief, we pre-calculated this mixture fraction numerically for various ambient conditions and then fit the results into certain formulas, as was done earlier using the SNAP methods. By applying this formula, the time required for iteration can be saved. A similar approach was applied by Kulmala et al. (1998) and
- Vehkamäki et al. (2002). Note that, although some studies suggest that the classical binary nucleation rate may be too weak to explain observed new particle formation (e.g., Covert et al., 1992), Chen et al. (2011) indicated that earlier studies may have significantly underestimated the nucleation rates because they omitted the size dependence of surface tension. Therefore, for the binary nucleation formula given in Appendix A, we adopted the method of Chen et al. (2011) for calculating the rate parameters.

There are several time-independent but size-dependent parameters that play important roles in aerosol processes. SNAP can be used to derive parameterization formulas for these so-called diagnostic parameters. In fact, we have already shown parameterizations for the parameter $C_{\rm C}$, which is used to derive the group sedimentation velocity,





 V_{sed} , in Sect. 2.2. Here, we first look at an effect often ignored in aerosol models: the Kelvin effect, which affects the equilibrium vapor pressure of the droplet. The equilibrium radius r_{eq} , and thus the water content of a hygroscopic particle, can be described by the Köhler theory, which is a combination of the Raoult (or solute) effect and Kelvin

- ⁵ (or curvature) effect. With the Kelvin effect, the particles absorb less water and thus have smaller sizes (Fig. 8). The size difference due to the Kelvin effect increases with humidity, reaching about 50% at 95% relative humidity and near infinity as the relative humidity approaches 100% for the case shown in Fig. 8. Apparently this effect cannot be ignored, especially at high humidity conditions. Normally, the calculation of r_0
- ¹⁰ requires numerical iteration. Here, we apply the SNAP approach to parameterize r_{eq} as a function of the ambient humidity and temperature, particle dry size, and a kappa parameter which was introduced by Petters and Kreidenweis (2007) to represent particle composition. Note that for aerosol mixtures (soluble or insoluble), the overall kappa parameter can be obtained by a volume weighting of individual kappa parameters.
- ¹⁵ A similar formula is obtained for calculating the wet volume of a whole aerosol mode. See Appendix A for the details of these formulas.

Another useful diagnostic parameter related to the Köhler curve is the activation cutoff size, which determines the smallest aerosol particles that can be activated into cloud drops under a certain supersaturation. Exact calculation of this cutoff size is even more todiaus then obtaining r ... Hence, it is after derived by simplifying the Köhler equation

- ²⁰ tedious than obtaining r_{eq} . Hence, it is often derived by simplifying the Köhler equation to obtain an approximate but direct relationship between the cutoff size and ambient supersaturation (cf. Pruppacher and Klett, 1997; p. 173). Our SNAP approach is well suited for parameterizing the cutoff size with high accuracy (< 0.5 % error) in a way similar to that for obtaining r_{eq} . As given in Appendix A, the cutoff size is expressed as a function of the supersaturation, temperature, particle dry size, and the kappa param-
- eter.

Other diagnostic parameters which we have provided in Appendix A include the modal extinction coefficient and absorption coefficient, which are important for calculating aerosol radiation effects. Another important parameter for radiation calculation



is the effective radius which, under the modal assumption, has an analytical solution $r_e \equiv M_3/M_2$, and thus does not need parameterization. Coefficients for the parameterization formulas in Appendix A are given in Appendix B.

4 Numerical verifications

⁵ In the previous section, we obtained fairly accurate modal-type parameterizations for aerosol microphysical processes. Additional checking of the reliability of these formulas is necessary when performing time integration, as errors may accumulate with time, which could cause numerical instability in extreme cases.

4.1 Verification with the binned parcel model

Verification of the time evolution of the size spectrum is not an easy task, especially for collision processes. A commonly accepted verification method is to use a detailed bin model that truly resolves the size distribution. The binned aerosol model used in this study is modified from the detailed cloud microphysical model of Chen and Lamb (1994), which applies a moment-conserving numerical scheme that ensures accuracy and conservation of mass and number concentration. This model has been applied to

various aerosol studies (cf. Chen et al., 2011; Tsai et al., 2012).

Another verification method is to obtain analytical solutions for the spectral time evolution. Such analytical solutions exist for simple collision kernels, such as the constant kernel (Bleck, 1970) and the simple mass-dependent Golovin's kernel (Berry, 1967), which have been used in warf time cloud mismaphysical solutions of a part 1007.

which have been used in verifying cloud microphysics schemes (e.g., Berry, 1967; Tzivion et al., 1987; Chen and Lamb, 1994). However, there is no need to develop modal parameterization for these simple kernels because exact analytical solutions exist. Thus, time-evolving analytical solutions are typically used to verify the performance of bin models. The performance of the model we are using has been verified against these time-evolving analytical solutions for cloud microphysical processes (Chen and



Lamb, 1994). We re-conducted the verification for aerosol size scales and found similar results. Note that the analytical solutions mentioned above are for gamma-type size distributions. For the log-normal size distribution that we applied here, Park and Lee (2000) provided an analytical solution for constant kernel collision process. Hence, we

⁵ conducted an additional verification by comparing with their analytical solution for a lognormal size distribution. The bin model produced 0.1 % and 0.3 % errors in M_0 and M_2 , respectively, after a 12 h time integration. These smaller errors indicate the robustness of our bin model.

We selected Brownian coagulation (including intra-modal and inter-modal) for testing the time integration for its complexity. The simulations were run in parcel mode to avoid complications from other processes, such as transport and sedimentation. Results obtained using the GHQ and BS95 methods were also compared. Figure 9 shows the initial bimodal aerosol size distribution (nucleation mode and accumulation mode) and the evolved size distributions. The size distributions of the modal approaches (i.e.,

- BS95, GHQ, and SNAP-C) are retrieved from the three moments by assuming lognormal distribution for each mode. All modal calculations give results similar to those of the binned calculation, showing that the nucleation mode decreased significantly after 1 h and essentially disappeared after 6 h, whereas the accumulation mode evolved rather slowly. When looking into the details, one can find visible differences between
- the modal distributions and the binned calculation. The BS95 and GHQ distributions deviate more obviously at the small end of the nucleation mode, whereas the SNAP-C distribution deviates more at somewhat larger sizes to the left of the modal size. All modal methods show fewer particles at the larger end of the accumulation mode, especially for the BS95 and GHQ methods and for the higher moments. However, such
- differences are not totally due to the inaccuracy of the parameterization formulas. The modal approaches retrieve the size distribution by assuming a fixed log-normal shape, which is symmetrical about the mode. However, the binned solution indicates that the true shape is not perfectly symmetrical.





A more appropriate comparison is done by examining the evolution of the overall moments M_0 and M_2 (while M_3 is conserved). As shown in Fig. 10, M_0 of all modal calculations closely follows the binned results, with errors of 1.8%, 2.1%, and 2.1% in SNAP-C, BS95, and GHQ, respectively, after 12 h of integration. The superiority of

⁵ the SNAP-C method is more obvious in the evolution of M_2 , with a final error of 0.8%, compared with the 2.0% error in either BS95 or GHQ. Note that the total errors are relatively small because the accumulation mode varies rather slowly. Another simulation with nucleation mode only (i.e., intra-modal coagulation) shows that the errors in BS95 becomes three times larger than those in SNAP-C (figures omitted).

4.2 Verification with regional models

More laborious verifications of the SNAP method are performed here using regional models. We first incorporate the SNAP scheme into a regional atmospheric dust model of Chen et al. (2004), which originally applied 12 size bins for mineral dust. The modified dust model applies two modes of mineral dust particles. The physical processes relevant to dust are emission, dry and wet depositions, and transport. We demonstrate the performance of the SNAP scheme by simulating an East Asian dust storm event that occurred on 19 May 2005, and comparing the simulation with the binned approach. Figure 11 shows the near-surface M_2 distribution (other moments are very similar and thus omitted). The differences between the binned and SNAP calculations are barely

- noticeable. We further examine the size distributions at a location near the dust source (110° E, 40° N) and a downstream location between Korea and Japan (130° E, 35° N). As shown in Fig. 12, the SNAP size distributions are generally in good agreement with the binned results. For this regional model simulation, the SNAP scheme requires significantly shorter computation time, about 1/3 less including all other overheads, to
- ²⁵ produce a very similar result than the binned calculation. Most of the time savings is due to the reduced computation time required for particle advection because the SNAP scheme uses 6 variables (3 moments for each mode) to describe the size distributions, as compared with the 12 variables (bins) used for the binned scheme.



A second test was conducted using the CMAQ model, in which we incorporated the SNAP scheme only for the Brownian coagulation process. Three levels of nesting with 81, 27, and 9 km resolutions are applied to simulate particulate pollution over the Taiwan area during early December 2007. The simulation was conducted for eight days including spin up time, and only the last five days' results of the innermost domain were analyzed. However, verification is difficult, as there is no high-resolution binned scheme in CMAQ for verification. Nevertheless, from the analyses shown earlier in Sects. 3 and 4, we know that the GHQ method is fairly accurate, so it was used as a benchmark for this comparison. Figure 13 shows the 5 day average aerosol dry mass loading simulated with SNAP, and the percent difference comparison against the GHQ method. The two schemes produced similar results. The differences are mostly less than 1 %, and reached 3 % in limited areas. This suggests that the SNAP scheme's

performance is close to that of the GHQ scheme in CMAQ.

An additional test was conducted for the same case to demonstrate the Kelvin effect on aerosol processes, which is often ignored in aerosol models. As discussed in Sect. 3.4, the Kelvin effect reduces the water content and thus the wet size of hygroscopic aerosol particles, and this effect influences essentially all aerosol processes. Therefore, this simulation included the diagnostic formula for the equilibrium wet size, with the Kelvin effect taken into consideration. Figure 14 shows that when the Kelvin

- ²⁰ effect is included, aerosol number concentration varies by less than 2%. However, changes in the higher moments are significant, with a reduction of over 30% in the cross-section area (M_2) and total volume (M_3). Most of the changes in M_2 and M_3 were simply due to differences in water content, but the dry aerosol mass loading also changed significantly, with up to a 10% increase or decrease at various locations.
- Mechanisms that may contribute to the decrease in dry aerosol volume include less solute uptake as a result of less water content and enhanced Brownian diffusional deposition due to reduced particle size. A mechanism that may increase dry aerosol volume is reduced gravitational sedimentation, especially for large particles at high humidity. There are certainly many details worthy of discussion that are beyond the





scope of this study. The purpose of the simulations here is simply to demonstrate the importance of including the Kelvin effect in the parameterization of aerosol wet size.

5 Conclusions

An innovative three-moment modal parameterization scheme was developed for ac-⁵ curate simulation of aerosol microphysical processes. Numerical calculations for the growth of a population of aerosol particles, represented by log-normal size distributions, were first performed and then the results were analyzed by statistical fitting to generate parameterization formulas. Four different approaches were devised for this Statistical-Numerical Aerosol Parameterization (SNAP), namely, the mean-size approx-

- imation, kernel transformation, integral transformation, and optimal-size approximation. The mean-size approximation was taken as a no-skill reference. Other approaches might be optimal for a certain process; however, we found that the integral transformation approach is suitable for most of the processes. These approaches provide parameterization formulas without simplifying the growth kernels, and only a minor inaccuracy
- resulted from the statistical fitting. Rate processes being parameterized include aerosol condensation, Brownian coagulation, sulfuric acid-water binary nucleation, and dry deposition. Special attention was given to processes related to aerosol-cloud interactions, and we provided formulas for heterogeneous ice nucleation, wet scavenging, and a diagnostic formula for aerosol activation into cloud drops. Other diagnostic formulas pro-
- ²⁰ vided in this work include considerations for aerosol equilibrium wet size and the Kelvin effect, as well as considerations for the group extinction and absorption coefficients.

The SNAP schemes were verified in various ways, including comparison against numerical solutions, analytical solutions, and results from a binned aerosol parcel model. All comparisons show that SNAP scheme is more accurate than the modal scheme used in CMAQ and WRF-Chem models, including the option that solves the growth

²⁵ used in CMAQ and WRF-Chem models, including the option that solves the growth integrals with a 5th order Gauss–Hermit numerical quadrature technique. The computational efficiency of the SNAP scheme is slightly lower (10% to 20%) than that of the





fast scheme in CMAQ, which utilizes lookup tables to speed up calculation; however, it is about 15 times faster than CMAQ's numerical quadrature option.

The SNAP scheme has been implemented in an atmospheric dust regional model, and the results (including the total moments and the dust size distribution) are very

- close to those simulated using a binned scheme. With such modal parameterization, much computation time is saved, mainly because of the reduced number of variables that need to be considered in advection calculation. We also utilized the CMAQ model to test the integrity of the SNAP scheme, with focus on the Brownian coagulation process. The results indicate that our scheme is as reliable as the 5th order Gauss-Hermit
 numerical quadrature scheme. In this model, we further showed that the commonly ignared Kahin effect, which reduces the water content of accessing a sculd paths in the imported to be integrated.
- nored Kelvin effect, which reduces the water content of aerosols, could not be ignored in aerosol modeling.

The parameterization scheme we developed is based on log-normal size distribution. However, detailed bin model simulations indicate that the size distribution may deviate from the log-normal form. It might be worthwhile to revise the scheme based on the gamma-type function, which is suitable for describing skewed size distributions. Because it has no restriction to the number of moments used, the SNAP method can even be applied to the modified gamma distribution, which requires four moments to solve. The SNAP method also has the potential to be used for the modal parameterization of cloud microphysical processes and even other types of physical or chemical processes.

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Table A1. Formulas for SNAP.

ltem #	Process/Parameter	Adjustment factor for the prognostic formulas	R^2	
1	Ice nucleation- deposition nucleation	$g_2 = \exp\left(a_1 \cdot \sigma^2 + a_2/\overline{q}\right)$	0.9109	
2	Ice nucleation- immersion freezing	$g_2 = \exp\left(a_1 \cdot \sigma^2 + a_2 / \overline{q}^2\right)$	0.9790	
3	Condensation	$g_{1,2} = \exp \left\{ \sigma \cdot [a_1 + a_2 \cdot \exp \left(-\ln \overline{K}_N \right) + a_3 \cdot \sigma] \right\}$	0.9989	
		$g_{1,3} = \exp\left\{\sigma \cdot [a_1 + a_2 \cdot \exp\left(-\ln \overline{K}_N\right) + a_3 \cdot \sigma]\right\}$	0.9961	
4	Intra-model coagulation 1	$g_{1,0} = \exp\left(\frac{\frac{a_1 + a_2 \cdot \sigma + a_3 \cdot h\overline{K}_N}{1 + a_4 \cdot \sigma + a_5 \cdot \sigma^2 + a_6 \cdot h\overline{K}_N + a_7 \cdot h\overline{K}_N^2}\right)$	0.9947	
		$g_{1,2} = g_{1,0} \cdot \exp\left(a_1 + a_2 \cdot \ln\mu + a_3 \cdot \sigma^3\right)$	0.9991	
5	Inter-modal coagulation	$g_{1,0} = \exp\left(a_1 \cdot \sigma_A^2 + a_2 \cdot \sigma_B^2\right)$	0.9997	
		$g_{1,2A} = \exp\left(a_1 \cdot \sigma_A^2 + a_2 \cdot \sigma_B^2\right)$	0.9999	
		$g_{1,2B} = \exp\left(a_1 \cdot \sigma_A^2 + a_2 \cdot \sigma_B^2\right)$	0.9999	
		$g_{1,3} = \exp\left(a_1 \cdot \sigma_A^2 + a_2 \cdot \sigma_B^2\right)$	0.9998	
6	Scavenging- Brownian diffusion and phoretic forces	$g_{1,0} = \exp\left(a_1 \cdot \sigma_A^2 + a_2 \cdot \sigma_B^2\right)$	0.9706	
	P	$g_{1,2} = g_{1,0} \cdot \exp\left(a_1 + a_2 \cdot \ln^2 \mu + a_3 \cdot \sigma^2\right)$	0.9989	
		$g_{13} = g_{10} \cdot \exp(a_1 + a_2 \cdot \ln^2 \mu + a_3 \cdot \sigma^2)$	0.9979	
7	Scavenging- Impaction	$g_{1,0} = \left(\ln^2 \mu_A + \ln^2 \mu_A \right) \cdot \exp\left(a_1 \cdot \sigma_A^2 + a_2 \cdot \sigma_B^2 \right)$	0.9914	
		$g_{1,2} = g_{1,0} \cdot \exp\left(a_1 + a_2 / \ln^2 \mu + a_3 \cdot \sigma^2\right)$	0.9923	
		$g_{1,3} = g_{1,0} \cdot \exp\left(a_1 + a_2 / \ln^2 \mu + a_3 \cdot \sigma^2\right)$	0.9947	
Diagno	ostic formulas	$X \rightarrow R = C \rightarrow C = C \rightarrow D = C^2 C \rightarrow C = C^2 C \rightarrow C = C$	0.0000	
8	embryo composition ²	$A = A + B \cdot IIIS_W + C \cdot IIIS_A + D \cdot III S_W + E \cdot III S_A + F \cdot IIIS_W \cdot IIIS_A$ $A = a_1 + a_2 \cdot T + a_3 \cdot T^2 + a_4 \cdot T^3 + \frac{a_5}{T} + \frac{a_6}{T^2}, B = a_1 + a_2 \cdot T^3, C = a_1 + a_3 \cdot T^2 + a_4 \cdot T^3 + \frac{a_5}{T^2} + \frac{a_6}{T^2}, B = a_1 + a_2 \cdot T^3, C = a_1 + a_3 \cdot T^2 + a_4 \cdot T^3 + \frac{a_5}{T^2} + \frac{a_6}{T^2}, B = a_1 + a_2 \cdot T^3, C = a_1 + a_3 \cdot T^2 + a_4 \cdot T^3 + \frac{a_5}{T^2} + \frac{a_6}{T^2}, B = a_1 + a_2 \cdot T^3, C = a_1 + a_3 \cdot T^2 + a_4 \cdot T^3 + \frac{a_5}{T^2} + \frac{a_6}{T^2}, B = a_1 + a_2 \cdot T^3, C = a_1 + a_3 \cdot T^2 + a_4 \cdot T^3 + \frac{a_5}{T^2} + \frac{a_6}{T^2}, B = a_1 + a_2 \cdot T^3, C = a_1 + a_3 \cdot T^2 + a_4 \cdot T^3 + \frac{a_6}{T^2} + \frac{a_6}{T^2}, B = a_1 + a_2 \cdot T^3, C = a_1 + a_3 \cdot T^2 + a_4 \cdot T^3 + \frac{a_6}{T^2} + \frac{a_6}{T^2}, B = a_1 + a_2 \cdot T^3, C = a_1 + a_3 \cdot T^2 + a_4 \cdot T^3 + \frac{a_6}{T^2} + \frac{a_6}{T^2}, B = a_1 + a_2 \cdot T^3, C = a_1 + a_3 \cdot T^2 + a_4 \cdot T^3 + \frac{a_6}{T^2} + \frac{a_6}{T^2}, B = a_1 + a_2 \cdot T^3, C = a_1 + a_3 \cdot T^2 + a_4 \cdot T^3 + \frac{a_6}{T^2} + \frac{a_6}{T^2}, B = a_1 \cdot T^3 + \frac{a_6}{T^2}, C = a_1 + a_3 \cdot T^3 + \frac{a_6}{T^2} + \frac{a_6}{T^2}, C = a_1 + a_3 \cdot T^3 + \frac{a_6}{T^2} + \frac{a_6}$	0.9999	
		$a_2 \cdot T^3, D = a_1 + a_2 \cdot T^3, E = a_1 + a_2 \cdot T^3, F = \exp\left(a_1 + a_2 \cdot T^2\right)$		
9	Cunningham slip flow cor- rection	$C_{\rm C} = 1 + a_1 \cdot \left(\lambda/r\right)^{a_2}$	0.9999	
10	Equilibrium wet size ³ (with Kelvin effect)	$r_{\rm eq} = r_d \cdot \left[1 + \kappa / (a_1 + a_2/S_W + a_3/r_d) \right]^{1/3}$	0.9960	
11	Modal equilibrium wet vol- ume (with Kelvin effect) ³	$M_{3,\text{eq}} = M_{3,\text{dry}} \cdot \left(1 + \kappa / (a_1 + a_2 / S_W + a_3 / \mu_d)\right) \cdot \exp\left(a_4 \cdot \sigma + a_5 \cdot \sigma^2\right)$	0.9998	
12	Activation cutoff size ⁴	$r_{\text{cut}} = \exp\left(a_1 + a_2 \cdot \ln\Delta s + a_3 \cdot \ln\kappa + a_4 \cdot \Delta T + a_5 \cdot \Delta s / \kappa\right)$	0.9976	
13	Modal extinction coefficient	$Q_{e,\text{bulk}} = Q_{e,r=\mu} \cdot \exp\left(a_1 + a_2 \cdot \ln\mu + a_3 / \ln\mu + a_4 / \ln^2\mu + a_5 / \sigma^2\right)$	0.9736	
14	Modal absorption coefficient	$Q_{a,\text{bulk}} = Q_{a,r=\mu} \cdot \exp\left(a_1 + a_2 \cdot \ln\mu + a_3 / \ln\mu + a_4 / \ln^2\mu + a_5 / \sigma^2\right)$	0.9882	

Note: $g_{1,j}$ and $g_{2,j}$ are SNAP-C and SNAP-D adjustment factors for the j^{th} moment (see Sect. 2.2); all μ and r are in m. When combined with SNAP-A to get the full prognostic equations (i.e., \tilde{I}), their R^2 are usually higher than those shown in the last column.

¹ \tilde{I} should be calculated with $r_A = \mu \cdot \sigma^2$ and $r_B = \mu / \sigma^2$ (see Sect. 3.3).

 2 X is sulfuric acid mass fraction of the critical embryo, S_W is relative humidity, and S_A is relative acidity.

³ Applicable at $S_W < 100\%$; r_d is dry radius; μ_d is dry modal value; $\kappa = \frac{\sum_i \kappa_i V_i}{\sum_i V_i}$, where $\kappa_j \equiv \frac{i_i \rho_i M_w}{\rho_w M_j}$, *j* is species index, *V* is volume fraction, *i* is van't Hoff factor, ρ is bulk density, *M* is molecular weight, and $\kappa_j = 0$ for insoluble species.

⁴ $\Delta s \equiv S_W - 1$, and $\Delta T \equiv T - 273.15$ K.



Item #	Identifier	a ₁	a ₂	a ₃	<i>a</i> ₄	a ₅	a ₆	a ₇
1		1.70943	0.92581					
2		1.55255	5.24148					
3	12	-0.70413	-0.12431	0.96979				
	<i>I</i> ₃	0.66771	-0.0415	0.97147				
4	Ĭ ₀	0.06784	-1.36133	-0.02511	-1.50752	0.99634	-0.02649	0.004882
	I_2	-0.78168	2.00439	-0.36568				
5	$\bar{I_0}$	0.67867	1.73118					
	1 _{2.A}	0.64602	-0.00638					
	I _{2.B}	-0.05216	0.62269					
	I_3	0.63212	0.60635					
6	<i>I</i> ₀	0.58113	1.98301					
	I_2	-18.86178	-0.05279	-1.98356				
	/ ₃	-25.28709	-0.079187	-1.51129				
7	<i>I</i> ₀	0.02450	-0.00041					
	I_2	-34.97740	1579.1761	2.62242				
	/ ₃	-52.55848	2381.8451	5.36787				
8	A	-6.81904	0.02574	-4.184E-5	3.004E-8	1070.618	-61228.947	
	В	-0.02095	-1.59219					
	С	0.0041	2.68278E-10					
	D	0.00174	-3.3858E-10					
	E	8.43580E-5	-1.3835E-11					
	F	-8.11399	2.46303E-5					
9		1.43089	1.0295					
10		-1.02733	1.02654	6.07891E-10				
11		-1.02733	1.02654	6.07891E-10	-8.98388	4.50074		
12		-6.35130	-0.66654	-0.33351	-0.00560	-0.08657		
13		919.62123	16.09653	17 486.858	110 668.73	0.50533		
14		-109.55798	-1.88697	-2219.1591	14696.404	0.21686		

Table A2. Coefficients for SNAP Formulas.





Fig. 1. Ratios of different mean-size approximation \tilde{l}_i to the true moment l_i as a function of the size distribution spread (σ) for various *i* values in Eq. (13).











Fig. 3. Fitting surface for the correction factors. Left: g_1 for SNAP-C; Right: g_2 for SNAP-D. The dots are the original values, and the vertical bars indicate their deviation from the fitting surface. The degree of deviation is also indicated by the color of the dots (increasing from blue, yellow to red).







Fig. 4. Comparing parameterized immersion freezing rates (ordinate) against the numerical solutions (abscissa). Panels from left to right are rates of the zeroth, second, and third moments, respectively. Results from SNAP-A to SNAP-D are represented by the blue diamonds, green squares, red crosses and black triangles, respectively. At the lower right corner of each panel is a zoom up of the central section. In the left panel, SNAP-A points with the largest σ are highlighted with circles.



















Fig. 7. Same as Fig. 6, but for inter-modal coagulation. Upper left: $I_{0,B}$; upper right: $I_{3,A}$ or $-I_{3,B}$; lower left: $I_{2,A}$; lower right: $I_{2,B}$.







Fig. 8. The Köhler curve (red dashed curve) which describes the equilibrium surface saturation ratio of a solution drop as a function of ambient relative humidity, S, for an ammonium sulfate particle with 0.01 µm dry radius. The blue curve is the equilibrium surface saturation ratio without considering the Kelvin effect.







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Fig. 10. Evolution of M_0 (left panel; in logarithmic scale) and M_2 (right panel; in linear scale) due to Brownian coagulation according to the SNAP-C (red dashed curve), BS95 (blue dotted curve), GHQ (green dash-dotted curve) and binned (thick black curve) calculations.













Fig. 12. Dust particle size distribution calculated with SNAP (red curve) and binned (blue dots connected by solid lines) schemes at two selected locations in Fig. 11.







Fig. 13. Left: simulated aerosol mass loading over the Taiwan area using SNAP for Brownian coagulation in the CMAQ model. Other panels from left to right: percentage difference between SNAP and GHQ in M_0 , M_2 and M_3 , respectively.







Fig. 14. Changes in aerosol moments due to the inclusion of Kelvin effect. Panels from left to right are percent change in number (ΔM_0), surface area of wet particles (ΔM_2 wet), volume of wet particles (ΔM_3 wet), and volume of dry particles (ΔM_3 dry).

