Metamodeling of Droplet Activation for Global Climate Models

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(Manuscript received 4 August 2015, in final form 13 November 2015)

ABSTRACT

The nucleation of cloud droplets from the ambient aerosol is a critical physical process that must be resolved for global models to faithfully predict aerosol-cloud interactions and aerosol indirect effects on climate. To better represent droplet nucleation from a complex, multimodal, and multicomponent aerosol population within the context of a global model, a new metamodeling framework is applied to derive an efficient and accurate activation parameterization. The framework applies polynomial chaos expansion to a detailed parcel model in order to derive an emulator that maps thermodynamic and aerosol parameters to the supersaturation maximum achieved in an adiabatically ascending parcel and can be used to diagnose droplet number from a single lognormal aerosol mode. The emulator requires much less computational time to build, store, and evaluate than a high-dimensional lookup table. Compared to large sample sets from the detailed parcel model, the relative error in the predicted supersaturation maximum and activated droplet number computed with the best emulator is $2.0\%$ and $4.9\%$ (one standard deviation), respectively. On average, the emulators constructed here are as accurate and between 10 and 17 times faster than a leading physically based activation parameterization. Because the underlying parcel model being emulated resolves size-dependent droplet growth factors, the emulator captures kinetic limitations on activation. The results discussed in this work suggest that this metamodeling framework can be extended to accurately account for the detailed activation of a complex aerosol population in an arbitrary coupled global aerosol-climate model.

1. Introduction

Interactions between aerosol and clouds yield one of the largest sources of uncertainty in understanding climate and future climate change on regional and global scales (Boucher et al. 2013). Within Earth’s atmosphere, homogeneous liquid water droplet formation is not thermodynamically favorable (Pruppacher and Klett 1997); instead, the pathway to nucleating cloud droplets is aided by the presence of ambient aerosol, a subset of which possess physical and chemical characteristics that allow them to serve as cloud condensation nuclei (CCN). These CCN provide a linkage between the physiochemical processes of atmospheric particles and cloud microphysics.

Changes in the background aerosol population can directly affect the properties of a nascent cloud droplet population. For instance, holding liquid water content constant, an increase in the number of CCN would tend to increase the total cloud droplet number concentration (the “Twomey” effect) while necessarily reducing the average size of the droplets (Twomey 1974). Such a change could enhance a cloud’s albedo, an effect that could be further amplified through microphysical feedbacks since smaller droplets impede the production of drizzle and thus lengthen cloud lifetime (Albrecht 1989). Mechanisms whereby aerosol influence the properties of clouds (and ultimately climate) are generally known as “aerosol indirect effects” (Haywood and Boucher 2000; Lohmann and Feichter 2005) and provide a path for changes in the ambient aerosol to produce cascading effects up to progressively larger scales of atmospheric motion (e.g., Wang 2005; Ekman et al. 2011; Morrison et al. 2011; Tao et al. 2011; Fan et al. 2012; Altaratz et al. 2014).

Aerosol indirect effects can either warm or cool the climate, but they all fundamentally depend on a subset of the ambient aerosols that function as CCN. The theory describing the dependency of cloud droplet nucleation
(also known as aerosol activation) on CCN availability and ambient aerosol has been rigorously developed using adiabatic and entraining parcel theory (Seinfeld and Pandis 2006; Pruppacher and Klett 1997) and depends on details of the heterogeneous chemical composition, number, size distribution(s), and mixing state of the background aerosol (Mefiggans et al. 2006) as well as local meteorology (Morales and Nenes 2010). Under polluted conditions, effects relating to chemical composition could produce a climatic effect as large as the basic Twomey effect (Nenes et al. 2002; Lance et al. 2004).

The development of activation parameterizations was pioneered by Twomey (1959) and Squires and Twomey (1961), who derived a relationship between the number of activated particles and the environmental supersaturation based on an aerosol size distribution approximated by a power law. Ghan et al. (2011) presented a thorough overview of subsequent developments over the past five decades and an intercomparison of several modern parameterizations. However, there is still an active effort to improve these parameterizations, as they are increasingly called upon to mediate between ever more complex aerosol models and the climate models to which they are coupled. For instance, the parameterization initially developed by Nenes and Seinfeld (2003) has seen continuous development, including modifications to handle condensation onto insoluble but wettable particles using adsorption activation theory (Kumar et al. 2009), environmental entrainment (Barahona and Nenes 2007), and numerical improvement of the population-splitting technique (Barahona et al. 2010; Morales Betancourt and Nenes 2014). Similarly, Ghan et al. (2011) extended the parameterization of Abdul-Razzak and Ghan (2000) to account for nonunity values of the accommodation coefficient $a_e$. Beyond idealized testing and droplet closure studies (Meskhidze 2005; Fountoukis et al. 2007), these modern parameterizations have been implemented in coupled climate–aerosol models such as the Community Earth System Model (CESM) to predict online cloud droplet number concentrations, where they have been shown to correct biases in global-average cloud droplet number concentrations and improve agreement with cloud properties measured from satelliteborne instruments (Gantt et al. 2014). Furthermore, adjoints of these parameterizations have been derived and coupled to chemical transport and global models in order to study the sensitivities of cloud droplet number to aerosol, chemical, and microphysical factors (Karydis et al. 2012; Moore et al. 2013).

Additionally, following the original integral/geometric approach by Twomey (1959), analytical representations of supersaturation evolution from adiabatic parcel theory have been progressively generalized to relate aerosol distributions to activation kinetics (Cohard et al. 1998; Khvorostyanov and Curry 2006, 2008; Shipway and Abel 2010; Shipway 2015). Although fundamentally analytical parameterizations, schemes of this class typically must rely on expensive numerical operations, such as in the evaluation of hypergeometric functions and iterative loops.

While most of these recent efforts toward improving activation parameterizations have focused on building highly generalized, “physically based” tools, there is still an application for other parameterization approaches. Saleeby and Cotton (2004) parameterized droplet nucleation for a cloud-resolving model, the Regional Atmospheric Modeling System (RAMS), by constructing a four-dimensional lookup table based on temperature, vertical velocity, aerosol number concentration, and the median radius of a lognormal aerosol mode with assumed chemical composition. Ward et al. (2010) added a fifth dimension representing chemical composition via aerosol hygroscopicity [following $\kappa$–Köhler theory (Petters and Kreidenweis 2007)] to the lookup table and later generalized this dimension to aerosol soluble fraction (Saleeby and van den Heever 2013). Constructing lookup tables of detailed parcel model results can be considered a form of model emulation combining a cache of known results and local polynomial (linear) approximation.

As the degrees of freedom and number of parameters describing a given aerosol population in a model increase, the burden of saving enough known points to interpolate through the parameter space via lookup table to some reasonable accuracy increases algebraically. For instance, the CESM features a modal aerosol population with three predefined, internally mixed lognormal modes, each with a fixed geometric standard deviation (Liu et al. 2012). Each mode is uniquely described by two moments (total number and total mass concentration) and the chemical composition of the mode by a single prognostic hygroscopicity term. Thus, the entire aerosol population has $N = 9$ degrees of freedom—too many to build a lookup table of activation statistics. Physically based parameterizations were designed to accommodate these sorts of arbitrary mixtures of aerosol but have a tendency to systematically underestimate activated fractions and subsequently cloud droplet number (Simpson et al. 2014). This is because of the parameterizations’ use of a set of assumptions that become increasingly likely to be violated as the aerosol population becomes more complex, specifically 1) that protodroplets grow in equilibrium with environmental changes in relative humidity and 2) that there are no kinetic or inertial limitations to droplet growth. The
presence of giant CCN (Barahona et al. 2010) and weak updrafts or excessively polluted conditions (Nenes et al. 2001) exacerbates this problem.

The goal of this study is to apply a surrogate modeling or emulation technique commonly used in the uncertainty quantification literature to a detailed parcel model capable of describing aerosol activation; this yields an efficient parameterization optimized for the high-dimensional parameter space affecting droplet nucleation in coupled aerosol–climate model. In essence, employing the derived emulator as an activation parameterization would be akin to directly coupling a detailed parcel model to a global model. Such a parameterization would be directly physically based but rely on fewer assumptions that affect the condensational growth of aerosol into CCN. However, it would also incorporate the efficiency of a lookup table, since the emulator would be designed to require a scarce amount of cached information and to be computationally cheap to evaluate. In this way, the emulator would improve upon the framework of a lookup table and be extensible to a very high-dimensional parameter space and thus be compatible with aerosol–climate models of increasing complexity.

Additionally, this study aims to better understand which parameters and inputs into droplet nucleation calculations are key to determining the resulting activated number concentration. Morales Betancourt and Nenes (2014) supplemented traditional error metrics by computing local sensitivities of the number concentration activated to key aerosol size distribution parameters using a detailed parcel model and comparing them to the joint of their parameterization. This study applies a related approach instead using global sensitivity analysis, which is suitable for identifying how uncertainty in inputs and parameters contributes to uncertainty in a model response. This analysis has not previously been applied to droplet activation and can provide additional metrics for evaluating parameterizations and their potential biases.

Section 2 describes the parcel model and the probabilistic collocation method (PCM) used to build its emulator. Section 3 presents results from applying the PCM to build a parcel model emulator designed to simulate the activation of a single lognormal aerosol mode under a wide variety of background environments and compares the new emulator to existing activation parameterizations. Section 4 motivates an extension of the technique to an emulator suitable of mediating aerosol activation in a coupled aerosol–climate model.

2. Methodology

a. Parcel model

Adiabatic parcel models are commonly used to study droplet activation and its sensitivity to factors such as environmental conditions and ambient aerosol properties. For this work, a novel parcel model based on previous studies (Leaitch et al. 1986; Nenes et al. 2001; Seinfeld and Pandis 2006) was designed and implemented to accommodate diverse, chemically heterogeneous, and polydisperse aerosol populations. The model simulates droplet growth on the initial aerosol population due to condensation within a constant-speed adiabatic updraft.

Although an arbitrary aerosol size distribution function can be supplied as an input to the model, for the purposes of this study the initial aerosol distribution is assumed to be lognormal and described by the equation

\[ n_n(r) = \frac{dN}{dm} = \frac{N_t}{\sqrt{2\pi ln\sigma_g}} \exp\left[-\frac{ln^2(r/\mu_g)}{2ln^2\sigma_g}\right], \]  

where the parameter set \( (N_t, \mu_g, \sigma_g) \) corresponds, respectively, to the total aerosol number concentration, the geometric mean radius, and the geometric standard deviation of the distribution. Within the model, this distribution is discretized into 200 size bins equally spaced over the logarithm of particle radius \( r \) and covers the size range \( \text{min}[0.1 \text{ nm}, \mu_g/10\sigma_g], \mu_g \times 10\sigma_g \). The mean radius in each bin grows as a result of condensation so that the activation of wetted aerosol into droplets is calculated in a Lagrangian sense. To relate size-dependent droplet growth to its embedded aerosol’s chemical composition, each bin is prescribed a hygroscopicity following \( \kappa \)-Köhler theory (Petters and Kreidenweis 2007).

To simulate droplet activation, the parcel model first computes an equilibrium wet-size distribution from the given initial aerosol population and initial environmental temperature, pressure, and relative humidity. Then, a set of conservation equations that describe the evolution of the parcel temperature, supersaturation, liquid/vapor water content, and pressure are integrated forward in time using a solver suitable for stiff systems [variable-coefficient ordinary differential equation solver (VODE); Brown et al. (1989)]. The complete system of equations and further details on the parcel model can be found in appendix A.

b. Polynomial chaos expansion

We construct an emulator of the parcel model in order to assess droplet activation by applying the probabilistic collocation method (PCM; Tatang et al. 1997). The PCM maps a set of input parameters to an output from the parcel model by building a response surface using a polynomial chaos expansion. The polynomial that results from this process is a computationally efficient, high-fidelity reproduction of the detailed parcel model.
simulation. Although often used for conducting global sensitivity analyses (Pan et al. 1997; Calbó et al. 1998; Mayer et al. 2000; Lucas and Prinn 2005; Anttila and Kerminen 2007) chaos expansion-based emulators have also been used to build deterministic parameterizations (Cohen and Prinn 2011). To apply and build the chaos expansions discussed here, the open-source Design Analysis Kit for Optimization and Terascale Applications (DAKOTA; Adams et al. 2014), version 6.1, was used, which automates the sampling of the PCM collocation points and the computation of the coefficients of the polynomial chaos expansion given a user-generated interface to a numerical model (the parcel model described in section 2a) and a description of the inputs and outputs to and from that interface.

A review of the theoretical basis of polynomial chaos expansion and its potential applications is provided by Sudret (2008); here, we highlight the important details of the technique as applied via PCM for the benefit of the reader. PCM is a nonintrusive polynomial chaos expansion technique; rather than require complex, significant modifications to the model being emulated, PCM instead considers the model to be a black box and constructs a map from an input parameter space to the model output parameter space. To accomplish this, PCM recasts the input parameters to a model as a set of independent input parameters, $X = X_1, \ldots, X_M$, each with an associated probability density function. For each input in $X$, the associated PDF is used as a weighting function to derive an orthogonal polynomial that adds to the bases for the polynomial chaos expansion $\phi_j$. Using a finite number of these bases, the chaos expansion for a given model response $R$ is then

$$R \approx \sum_{j=0}^{p} a_j \phi_j(X). \quad (2)$$

The complete basis of polynomials up to a fixed total-order $p$ is retained in the expansions computed here. For such a total-order expansion, Eq. (2) has $N_t = p + 1 = (M + p)!/(M!(p!)$ terms as it contains each of the $p + 1$ orthogonal basis polynomials for each input parameter. PCM provides an experimental design for determining the coefficients of the expansion $a_j$ by evaluating the model response for a set of $N_t$ total input parameter sets, $X^1, \ldots, X^{N_t}$, corresponding to the roots of $\phi_j$ and solving a regression problem

$$\Phi \alpha = R. \quad (3)$$

where $R$ is the vector of model responses, $\alpha$ is the vector of expansion coefficients, and the matrix $\Phi$ contains rows for each of the polynomial terms $\phi_j$, evaluated for a given input parameter set $X^i$.

A practical consideration in applying the PCM to a particular problem is what subset of the $N_t$ potential points to use in solving for the coefficients. In general, there exists a full factorial design of size $N_s = (p + 1)^M$ available for use (all the roots of the orthogonal basis polynomials for all inputs). However, for even moderately sized $p$ and $M$, the number of potential model evaluations grows very rapidly. In our application we choose a subset of $N'_s$ parameter sets when applying the PCM by using two rules of thumb:

1) choose parameter sets with roots closest to the origin (Sudret 2008) and
2) cross validate the regression result using $3N_t$ parameter sets chosen according to rule 1.

These rules will always produce an overdetermined system for Eq. (3). The accuracy of the resulting emulators derived in this study were not sensitive to increasing $N'_s$, and $3N_t$ does not produce an excessive number of required parcel model simulations. Three different techniques were tested for solving this system: typical linear regression by ordinary least squares (OLS), least angle regression (LARS; Efron et al. 2004), and least absolute shrinkage and selection operator (LASSO; Tibshirani 2011). Both LARS and LASSO involve computing

$$\alpha = \arg \min \|\Phi \alpha - R\|_2^2 \quad \text{such that} \quad \|\alpha\|_l \leq \tau \quad (4)$$

in an iterative, greedy fashion with the potential to yield sparse solutions with some coefficients $\alpha_i = 0$. This would be desirable for high-order chaos expansions for many input parameters, as it would reduce the number of coefficients necessary to save for reusing the expansion as an emulator. Additionally, this greedy characteristic helps to avoid overfitting the chaos expansions. Each time a potential term is added to the trial expansion, an error estimate is calculated based on leave-one-out sampling (Blatman and Sudret 2011); if the error estimate increases, the potential term is rejected. Inspecting the resulting terms gives a metric to compare the OLS-derived chaos expansion. The same error calculation using leave-one-out sampling can be applied using the larger, independent sampling dataset used to evaluate the chaos expansions in section 2a for each higher-order chaos expansion to help identify when overfitting is occurring.

c. Emulation of parcel model

The PCM was applied to emulate the activation of a single, lognormal aerosol mode embedded in a constant-speed adiabatic updraft as simulated by the parcel model described in section 2a. Specifically, the model was used to predict the maximum supersaturation $S_{\text{max}}$
The mechanics of the PCM—and polynomial chaos expansion more generally—permit the use of arbitrary PDFs to describe the input parameters over their physically relevant values. In this application, uniform distributions were chosen to emphasize that the derived chaos expansion should perform equally well anywhere within the input parameter space. Each uniform distribution is defined by minimum and maximum permissible bounds for each input parameter, $a$ and $b$, such that its probability distribution is just given as $f(x) = 1/(b - a)$ for $a < x < b$.

To utilize the PCM, the uniform distribution for each parameter must be rescaled to the range $[-1, 1]$. This produces a new set of random variables for each parameter $X_i$:

$$Z_i = \frac{2(X_i - a_i)}{b_i - a_i} - 1. \quad (5)$$

The orthogonal polynomials used in the basis of the chaos expansion that correspond to a uniform PDF over the interval $[-1, 1]$ are the canonical Legendre polynomials that follow the three-term recurrence relation:

$$P_0(Z) = 1, \quad (6)$$

$$P_1(Z) = Z, \quad \text{and} \quad (7)$$

$$P_{n+1}(Z) = \frac{(2n + 1)ZP_n(Z) - nP_{n-1}(Z)}{n + 1}. \quad (8)$$

The roots of these Legendre polynomials can be inverted using Eq. (5) to determine values in the original, physical parameter space to use in sampling the parcel model.

The bounds for the physical parameters supplied to the PCM were chosen in order to characterize activation near cloud base (Table 1). The logarithm of several variables (aerosol number concentration, aerosol geometric mean radius, and updraft velocity) is used because the supersaturation maximum computed by the parcel model is sensitive to changes in these parameters over several orders of magnitude. Updraft velocity is permitted to vary between 0.01 and 10.0 m s$^{-1}$; over this range (which covers a spectrum from weakly convecting, stratiform clouds to strong, deeply convecting ones) and the range of aerosol number concentration (which includes clean and very polluted regimes), activated fraction can range from virtually nothing to complete activation of the entire aerosol population. The lower bound of updraft speeds considered here is less than the minimal value allowed in many climate models (Golaz et al. 2011). The aerosol mode geometric mean radii $\mu_g$ span a variety of smaller Aitken-type modes to large, coarse aerosol modes and potentially giant CCN (Seinfeld and Pandis 2006). The mode geometric standard deviation is fixed in some global model aerosol schemes (e.g., Kim et al. 2008; Liu et al. 2012) and the range chosen here covers many potential values. Hygroscopicity values are based on Table 1 of Petters and Kreidenweis (2007) and span values for materials ranging from organic aerosol to highly hygroscopic salts. The accommodation coefficient was limited to a globally representative range based on observations of CCN activation kinetics from many campaigns (Raatikainen et al. 2013). Temperature and pressure ranges were chosen to reflect typical lower-troposphere values. Note that the bounds on the parameters considered here are expanded from those considered by Ghan et al. (2011) in their intercomparison of activation schemes.

Many parameterizations of droplet nucleation diagnose activation directly by applying equilibrium Köhler theory. To do this, the maximum supersaturation achieved by a cloudy parcel is used as a threshold; particles with a Köhler-predicted critical supersaturation lower than this maximum environmental supersaturation are considered to be activated. However, physically, for a droplet to activate it must grow beyond a critical size corresponding to this critical supersaturation. Because of kinetic limitations on droplet growth, this may

| Symbol | Name | Units | Bounds |
|--------|------|-------|--------|
| $\log_{10}N$ | Mode number concentration | $\log_{10} \text{cm}^{-3}$ | [1, 4] |
| $\log_{10}\mu_g$ | Mode geometric mean radius | $\log_{10} \mu_m$ | $[-3.0, 1.0]$ |
| $\sigma_g$ | Mode standard deviation | — | $[1.2, 3.0]$ |
| $\kappa$ | Mode hygroscopicity | — | $[0.0, 1.2]$ |
| $\log_{10}V$ | Updraft velocity | $\log_{10} \text{ms}^{-1}$ | $[-2.0, 1.0]$ |
| $T$ | Air temperature | K | [240, 310] |
| $P$ | Air pressure | Pa | $[50000, 105000]$ |
| $\alpha_a$ | Accommodation coefficient | — | $[0.1, 1.0]$ |

Table 1. Input parameters and bounds used in computing chaos expansion for emulating droplet activation from a single, lognormal aerosol mode embedded in a constant-speed updraft.
not be realized for droplets growing on very large CCN (Nenes et al. 2001). Ghan et al. (2011) suggests particles with radius larger than 0.1 μm or those whose critical supersaturations are close to the environmental maximum supersaturation are likely to suffer from this effect. By directly considering a detailed parcel model, the emulators constructed here consider kinetic limitations on droplet growth and their feedback on the evolving parcel supersaturation. In existing, physically based parameterizations in the literature, an instantaneous growth-rate assumption must be applied. This assumption causes parameterizations to underpredict supersaturation maximum because instantaneous growth will tend to condense water from the vapor phase too quickly and release surplus latent heat, which suppresses the increase of the supersaturation.

Because the computed supersaturation maximum in a parcel model activation simulation can also vary over several orders of magnitude, we use \( \log_{10}(S_{\text{max}}) \) as the response function emulated by the PCM. However, in order to apply this transform to the response function, it must be assumed that the cloudy parcel always super saturates with respect to water vapor (i.e., \( S_{\text{max}} > 0 \)). To ensure this, all simulations performed during sampling by the PCM start with an aerosol population equilibrated to 100% relative humidity and an initial environmental supersaturation of 0. Many existing parameterizations in the literature implicitly make this same assumption by representing the aerosol population with respect to a coordinate derived from the critical supersaturation for a given size [Ghan et al. (2011); Eqs. (12)–(17)]; in this case the integral over the size distribution spans \( 0 \leq S \leq S_{\text{max}} \) and, thus, considers the same situation with respect to the growth of the nascent droplet population as Eq. (3) of Ghan et al. (2011).

For the eight-parameter input space governing single-mode activation considered here, the third- and fourth-order chaos expansions produced by the PCM have 165 and 495 terms, respectively. The number of terms is equivalent to the number of coefficients one must store in order to reuse a given chaos expansion. This small memory footprint affords chaos expansions a huge advantage over similar parameterizations based on detailed lookup tables. An isotropic lookup table with \( M \) parameters and \( n \) sample points for each parameter would require \( n^M \) values to be stored—a value that for even small numbers of parameters can be several orders of magnitude larger than even a high-order chaos expansion. A more detailed description of how the chaos expansions are saved and later evaluated is given in appendix B.

The parcel model emulator yields \( \log_{10}(S_{\text{max}}) \) as a function of an input parameter set drawing from the terms defined in Table 1:

\[
\log_{10}(S_{\text{max}}) = f(\log_{10}N, \log_{10}\mu, \sigma, \kappa, \log_{10}V, T, P, a_e).
\]

(9)

From this value of \( \log_{10}(S_{\text{max}}) \) the number concentration of cloud droplets activated, \( N_{\text{act}} \), can be obtained by integrating over the original lognormal aerosol size distribution reexpressed as a function of critical supersaturation rather than droplet radius (Ghan et al. 2011), yielding the expression

\[
N_{\text{act}} = \frac{N}{2} \left \{ 1 - \text{erf} \left [ 2 \ln \left ( \frac{S_m}{S_{\text{max}}} \right ) / \left ( 3 \sqrt{2} \ln \sigma_g \right ) \right ] \right \}.
\]

(10)

where \( S_m \) is the critical supersaturation for the geometric mean radius \( \mu_g \).

d. Global sensitivity analysis

We supplement the assessment of our new droplet activation emulator by calculating a set of global sensitivity metrics not previously applied to this problem. The method deployed here is a variance-based decomposition, which seeks to assign uncertainty in a model response to uncertainty in both individual model input parameters and their interactions with one another. Two different quantities, called Sobol’ indices (Sobol’ 2001), are produced by this method: main \((S_i)\) and total \((T_i)\) effect indices. Sobol’ indices can be used to rank the relative importance of model inputs in influencing its response and for identifying potential inputs that are unimportant and also candidates to be held fixed without grossly biasing the accuracy of a model emulator (Sobol’ 2001).

The main effect index indicates what fraction of the uncertainty in a given model response \( R \) is attributable to a single member of the model parameter set \( X_i \) by comparing the variance of the model response conditioned on \( X_i \) against the total variance in \( R \). That is,

\[
S_i = \frac{\text{Var}_X(E[R | X_i])}{\text{Var}(R)}.
\]

(11)

This is in contrast with the total effect index, which instead compares the variance of \( R \) conditioned on all the input parameters save for \( X_i \) (notated as \( X_{-i} \)). The index \( T_i \) quantifies the variance of \( R \) attributable to \( X_i \) and the sum of its interaction with other input terms. Similar to the main effect index,

\[
T_i = \frac{\text{Var}(R) - \text{Var}(E[R | X_{-i}])}{\text{Var}(R)}.
\]

(12)

Sudret (2008) derives alternative equations that further clarify the meanings of these terms in the context of studying model emulators. Critically, these terms can be
expressed as multidimensional integrals over model input parameters that can be approximated via Monte Carlo or other sampling techniques, although it is very computationally expensive to do so. We adopt the column swap-out sampling method of Weirs et al. (2012), which combines Latin hypercube sampling with perturbed combinations of parameters to efficiently approximate $S_i$ and $T_i$ [see also Saltelli et al. (2010)]. However, the sampling procedure employed still requires a large number of model evaluations; for an initial $n$-size sample of the $M$ parameters being studied, the method requires $(2^M) \times n$ evaluations of the full-complexity model. We found that the computed $S_i$ and $T_i$ converged to stable values for the parcel model (and other parameterizations studied here) for $n \approx O(10^3)$ and used $n = 1280$ to derive the values reported here.

Although the sampling procedure can be repeated for the emulators derived via polynomial chaos expansion, the orthogonality of the basis terms that constitute each expansion lends itself to a more direct computation of $S_i$ and $T_i$. Following Sudret (2008), we compute these indices directly from the coefficients of the derived chaos expansions. The sampling technique used to derive Sobol’ indices for the parcel model, applied to the chaos expansions, produces similar estimates to those computed from the coefficients.

3. Results

a. Evaluation of emulators

To assess the performance of the emulator, two sets of $n = 10000$ samples were drawn using maximum Latin hypercube sampling from the parameter space defined in Table 1. This randomized design ensured that representative, equal numbers of samples were drawn from across the multidimensional parameter space. In the first set, variables whose logarithms were used to build the emulator were sampled in logarithmic space; in the second set, these variables were transformed back to their original values (e.g., from $\log_{10} N$ to $N$) before the sample was constructed. The two independent sets were blended together to assess the emulator. This helps ensure that both very high and very low values of the log transformed are thoroughly represented within the sample. The set of sample parameter sets were run through all the derived chaos expansions of all orders, as well as the detailed parcel model as a reference benchmark for activation dynamics.

Figure 1 illustrates the performance of a fourth-order expansion whose coefficients were derived using ordinary least squares. The large range of initial temperatures, pressures, aerosol populations, and updraft speeds sampled here leads to a very large range of supersaturation maxima achieved by the ascending parcel. Weaker updraft speeds are generally associated with lower maximum supersaturations and corresponding to lower aerosol activated fraction; the opposite is true when strong updrafts are present, although there are some cases where a strong updraft activates a small fraction of aerosol. This typically occurs when initial aerosol size distribution is shifted toward larger radii and under polluted conditions with aerosol number concentrations greater than $3000 \text{ cm}^{-3}$. However, over the large parameter space sampled, the chaos expansion accurately reproduces the parcel model’s determination of $S_{\text{max}}$ and corresponding activated fraction. This is even true for predictions of small $S_{\text{max}}$, which could potentially have a larger bias since the predicted error is expected to be uniform in $\log_{10}(S_{\text{max}})$.

![Fig. 1. One–one plot comparing (a) predicted supersaturation maximum and (b) diagnosed equilibrium droplet activated fraction between parcel model and a polynomial chaos expansion of order $p = 4$, with coefficients computed using ordinary least squares. Black lines denote a factor-of-2 difference between predicted values using parcel model and those computed with the parameterization. Glyph shading denotes updraft velocity $V$ with corresponding scale in (a).](image-url)
For parameter sets leading to a large activated fraction of 0.8–1.0, the relative error of the chaos expansion (compared to the parcel model) rarely exceeds 5% and on average (for all activated fractions) is 5.7%. While the mean relative error in each activated fraction decile is close to 0, the standard deviation in the relative error tends to increase for the lower ones; the standard deviation in relative error decreases from 19.7% for activated fractions in the range 0.1–0.2 to 3.5% for those in the range 0.8–0.9. This suggests that there is a nonlinear component in the mapping from the input parameter space to the emulated maximum supersaturation and diagnosed droplet number concentration that is prevalent in the weak droplet activation regime; the predicted activated fraction is more sensitive to small changes in the input parameters in this regime than in others.

Increasing the order of the chaos expansion tends to improve the accuracy of the predicted $S_{\text{max}}$, as recorded in Table 2. However, there is not much difference between the methods used to compute the coefficients of the expansion beyond expansion order. For example, for the fourth- and fifth-order expansions, the expansions perform equally well regardless of what method (OLS, LARS, or LASSO) was used to compute the coefficients when considering the mean and spread of the relative error to the parcel model reference simulations. In all cases, the chaos expansions produce very large $r^2$ values and small normalized fractional root-mean-square errors [$\text{RMSE}/\sum_{i=1}^{m}(X_i^2/n)$], which decrease as the order of the expansion increases.

These same statistics, computed for the diagnosed droplet number concentration given the predicted supersaturation maximum, are summarized in Table 3. Here, the trend is similar to before; increasing the order of the expansion tends to improve the accuracy of the diagnosed number concentration in terms of mean relative error and also tends to decrease spread around that value. Third-order expansions tend to produce more accurate results with respect to the mean relative error, but this is overshadowed by the fact that there is far more variance in their predicted values as indicated by the standard deviation of their relative errors, which are almost twice as large as those of the higher-order expansions.

**b. Comparison with other parameterizations**

We compare the performance of the chaos expansion-based emulators to two existing parameterizations from the literature. The scheme by Abdul-Razzak and Ghan (2000) (ARG)—which is widely used in global models—utilizes a pseudoanalytical solution to an integro-differential equation derived from the adiabatic parcel system with embedded aerosol growing via condensation. This is in contrast to the scheme by Morales Betancourt and Nenes (2014) (MBN), which is based on an iterative scheme to separate the aerosol population into subsets whose growth is inertially limited or not and uses this information to derive a maximum supersaturation for a given parcel system. The MBN scheme is generally more expensive to evaluate than the ARG scheme because of its iterative nature but is often more accurate owing to its consideration of the potentially important effect of large albeit unactivated aerosol particles (Simpson et al. 2014). In contrast with these schemes, our emulators simulate the activation process based on the explicit numerical solution obtained from a detailed parcel model, which is similar to the one used to build and evaluate the MBN scheme (e.g., Nenes and Seinfeld 2003).

The parameter sets used in section 3a were also used to compute droplet activation with the ARG and MBN parameterizations. The relative errors
the supersaturation maximum and droplet number predicted by these schemes and the chaos expansions compared to the parcel model are illustrated in Fig. 2. Both the ARG and MBN parameterizations are more accurate than the second- and third-order chaos expansions. The ARG scheme tends to underpredict the maximum supersaturation, which is consistent with previous investigations into its performance (Abdul-Razzak and Ghan 2000; Ghan et al. 2011; Simpson et al. 2014). This tends to produce a bias toward underprediction of droplet number. The MBN scheme tends to yield more accurate predictions of both maximum supersaturation and droplet number. However, both schemes are outperformed by the fourth- and fifth-order chaos expansions, both on average and in terms of the variance of the predictions; for instance, the OLS-derived fourth- and fifth-order expansions yield relative error in predicted $S_{\text{max}}$ with a mean plus or minus one standard deviation of $4.2\% \pm 14.4\%$ and $-0.32\% \pm 10.4\%$, whereas the ARG and MBN schemes yield $-13.7\% \pm 18.2\%$ and

### Table 3

As in Table 2, but for predicted droplet number concentration.

| Method | Expansion order | NRMSE  | $r^2$     | MRE    | MRE std dev |
|--------|-----------------|--------|-----------|--------|-------------|
| LASSO  | 2               | 0.165 043 | 0.929 439 | 5.697 161 | 49.813 356 |
|        | 3               | 0.108 790 | 0.969 342 | -0.930 886 | 20.270 639 |
|        | 4               | 0.074 406 | 0.985 659 | 2.545 998  | 19.834 428 |
|        | 5               | 0.058 872 | 0.991 022 | 1.441 665  | 14.497 129 |
| LARS   | 2               | 0.167 168 | 0.927 611 | 3.524 631  | 39.031 227 |
|        | 3               | 0.121 896 | 0.961 510 | -0.030 537 | 34.613 813 |
|        | 4               | 0.075 984 | 0.985 044 | -0.280 769 | 16.430 850 |
|        | 5               | 0.058 961 | 0.990 995 | 0.884 227  | 17.817 749 |
| OLS    | 2               | 0.174 550 | 0.921 076 | 7.640 377  | 43.978 774 |
|        | 3               | 0.125 045 | 0.959 496 | 0.380 943  | 28.562 715 |
|        | 4               | 0.079 971 | 0.983 434 | 2.556 756  | 20.929 598 |
|        | 5               | 0.061 762 | 0.990 119 | 1.295 857  | 17.903 202 |

### Fig. 2

Box plots illustrating mean relative error between (a) supersaturation max and (b) droplet number concentration predicted by chaos expansions and parameterizations vs detailed parcel model. The chaos expansions have been grouped by expansion order (x axis) and method for computing their coefficients (OLS, LARS, and LASSO; hue). “ARG” refers to the scheme of Abdul-Razzak and Ghan (2000); “MBN” refers to the scheme of Morales Betancourt and Nenes (2014).
−2.4% ± 15.8%, respectively. This is larger than other studies have reported, but we explore a much larger parameter space in our sampling for the purposes of deriving the chaos expansion.

As a consequence of tending to slightly underpredict \( S_{\text{max}} \), both the ARG and MBN schemes underpredict the number of activated droplets in the framework considered here. The mean relative error in droplet number predicted by the ARG and MBN schemes for the samples here are −9.6% ± 23.4% and −4.9% ± 16.8%, respectively. All of the chaos expansions outperform the mean relative error of the ARG scheme, and those of order \( p \geq 3 \) do so with less variance.

In addition to producing low mean relative error in predicted \( S_{\text{max}} \) and droplet number activated, the chaos expansions also reproduce the dependence of activation dynamics on aerosol physical properties and updraft speed, as illustrated in Fig. 3. In response to increasing aerosol number concentration, \( S_{\text{max}} \) reached by an ascending parcel tends to decrease because there is a larger surface area available where condensation can occur, producing a larger source of latent heat release that limits the production of supersaturation. Overall, though, the droplet number concentration increases despite this effect as the aerosol activated fraction only decreases by a factor of 4 when the total number of initial aerosol increases by an order of magnitude (Figs. 3a, b). Shifting the aerosol population to larger sizes (Figs. 3c, d) produces a similar effect in inhibiting the increase in a parcel’s supersaturation; however, Köhler theory predicts that these larger particles will more easily activate, which offsets the increase in \( S_{\text{max}} \) and yields larger droplet number concentrations. A similar effect occurs as aerosol hygroscopicity increases (Figs. 3e, f).

The chaos expansions, as well as both the ARG and MBN schemes, capture these subtleties of activation dynamics as well as the detailed parcel model. More importantly, the expansions reproduce the sensitivity of activation to updraft speed (Figs. 3g, h), which is an important factor controlling \( S_{\text{max}} \) and setting the droplet number. At the largest updraft speeds of a few meters per second—indicative of deep, vigorous convection—the MBN scheme outperforms both the chaos expansions and the ARG scheme. However, for the aerosol population considered in Fig. 3g, h (with \( N = 1000 \text{ cm}^{-3} \), \( \mu = 0.05 \mu \text{m} \), and \( \sigma = 2.0 \)), the relative error in predicted \( S_{\text{max}} \) by the chaos expansions at high updraft speeds does not substantially affect the diagnosed droplet number concentration, since in this case all but the smallest aerosol particles activate under equilibrium considerations. Note that this \( S_{\text{max}} \) overprediction coupled with an accurate assessment of activated fraction occurs for many different single-mode, lognormal aerosol populations.

Although all the chaos expansion results in Fig. 3 appear biased high compared to the parcel model, this bias does not hold true in general. Note that the ARG scheme is biased low in this particular analysis; this is generally just an artifact of fixing seven of the eight parameters and analyzing one-dimensional transects. Fixing the nonvarying parameters at different values tends to shift the bias positive or negative in a non-systematic way. Critically, the choice of values for these parameters does not affect the chaos expansion’s ability to reproduce the sensitivity to the varying parameter, which lends confidence that the expansions accurately reproduce the behavior of the parcel model.

Since Fig. 3 highlights the fact that different schemes potentially perform better in different parts of the parameter space governing droplet activation, we stratified the sampling results based on level of pollution and updraft-speed strength and computed activated fraction relative error statistics in each of these bins as shown in Fig. 4. All of the schemes are accurate in clean and lightly polluted conditions (with aerosol number concentration \( N < 1000 \text{ cm}^{-3} \)). However, there is a tendency for both the ARG and MBN schemes to underpredict droplet number in heavily polluted conditions (\( N > 2500 \text{ cm}^{-3} \)).

The fourth-order OLS-derived chaos expansion is plotted in Fig. 4a as a representative example of the chaos expansions, and it retains its accuracy across the pollution level–updraft strength spectrum. The combination of light updrafts and heavy pollution tends to produce the largest underprediction in activated droplet number, ranging from 10% to 30% for the ARG and MBN schemes. Unsurprisingly, relative error in activated fraction tends to be least sensitive to increasing aerosol number concentration in the strong updraft regime. In this case, the vigorous updraft produces strong adiabatic cooling that overwhelms latent heat release from condensation as the droplets in the parcel grow, contributing to a rapid and large \( S_{\text{max}} \) [Eq. (A1)] and thereby activating a significant fraction of the aerosol.

It should be noted that parts of the parameter space we considered in applying the PCM, evaluating its output, and comparing to existing parameterizations may not be typical of real atmospheric cases. We chose a large parameter space in order to derive the most general emulator possible for this particular single-mode aerosol case. In the real world, there should be some correlation between the ambient temperatures, pressures, and updraft speeds used when diagnosing aerosol activation, while we sample these factors as if they were independent from one another. The output from applying the parcel model
to nonrealistic activation scenarios could tend to inflate the computed relative errors and exacerbate differences between the parcel model and the existing, physically based parameterizations.

c. Global sensitivity analysis

Total Sobol’ indices $T_i$ for each of the input parameters summarized in Table 1 corresponding to the prediction of

Fig. 3. Sensitivity of (a)–(d) parameterized and simulated maximum supersaturation and (e)–(h) activated number fraction to changes in mode number concentration, mode geometric mean radius, mode hygroscopicity, and updraft speed with all other parameters held fixed at the values $T = 283$ K, $P = 850$ hPa, $V = 0.5$ m s$^{-1}$, $a_c = 1.0$, $\mu = 0.05$ $\mu$m, $\kappa = 0.54$, $N = 1000$ cm$^{-3}$, and $\sigma = 2.0$. “MBN” and “ARG” correspond to the schemes of Morales Betancourt and Nenes (2014) and the update by Ghan et al. (2011) to Abdul-Razzak and Ghan (2000), respectively; the curves correspond to fourth-order chaos expansions with coefficients derived using the named method.
$S_{\text{max}}$ are plotted for each of the chaos expansions, parameterizations, and parcel model in Fig. 5. The order of the chaos expansion does not impact the relative importance of each term between different schemes or for higher-order expansions of the same scheme. Both updraft speed and the aerosol distribution size parameter most strongly contribute to the variance in $S_{\text{max}}$, followed by the total number of aerosols. This is in contrast with the chemical parameters in the model, $a_c$ and $k$, which provide the weakest constraints, suggesting the importance of the total aerosol surface area in dominating the potential for droplet activation by controlling $S_{\text{max}}$.

Indices derived using the chaos expansions very closely approximate those derived by sampling the full parcel model. This is expected since they simply provide an alternative framework for calculating the indices from the parcel model. However, the differences between the ARG and MBN schemes and the parcel model highlight the potential for biases in these schemes due to oversensitivity to particular model parameters. For instance, the ARG scheme is less sensitive to variations in $\sigma_\epsilon$ than the full parcel model; however, the dependence of $S_{\text{max}}$ in that scheme on $\sigma_\epsilon$ is tuned to their own numerical calculations, which may differ from ours (Abdul-Razzak et al. 1998). Furthermore, the ARG scheme is more sensitive to variations in $a_c$ than our parcel model and emulators, which parameterize the dependence of the condensational growth coefficient by a simple rescaling against a reference value computed for $a_c = 1$ rather than explicitly account for it.

Maximum supersaturations produced by the MBN scheme are more sensitive to the geometric mean size of the aerosol than those from the parcel model or ARG scheme and generally less sensitive to the strength of the updraft speed. The relative importance of each term for both the ARG and MBN scheme generally agrees with the estimates from the parcel model and chaos expansions, although the ARG scheme is most sensitive to

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**FIG. 4.** Mean relative error in activated fraction for (a) fourth-order OLS-derived chaos expansion, (b) ARG, and (c) MBN schemes relative to detailed parcel model. Updraft speeds are light (10–50 cm s$^{-1}$), moderate (0.5–2.0 m s$^{-1}$), and strong (2.0–10.0 m s$^{-1}$); pollution levels are clean (10–250 cm$^{-3}$), light (250–1000 cm$^{-3}$), moderate (1000–2500 cm$^{-3}$), and heavy (2500–10 000 cm$^{-3}$). Error bars denote 95% confidence interval on mean relative error from in-bin samples; samples with $\mu < 10$ nm were omitted from these calculations.

**FIG. 5.** Total Sobol’ indices corresponding to the prediction of $S_{\text{max}}$ for each parameter in Table 1 for each chaos expansion (OLS, LARS, and LASSO), parameterization (ARG and MBN), and parcel model. For the chaos expansions, the lighter colors indicate successively higher-order expansions.
The main terms dominate the higher-order ones. The only
ings of the top eight terms do not change relative order, and
With the exception of the second-order scheme, the rank-
order of magnitude less important than those in the table.
( second- through fifth-order) chaos expansions summa-
ized, all the terms with rank greater than eight were an

| Rank | Term       | Main $S_i$ | Term       | Main $S_i$ | Term       | Main $S_i$ | Term       | Main $S_i$ |
|------|------------|------------|------------|------------|------------|------------|------------|------------|
| 1    | $\log_{10}\mu_g$ | 0.379      | $\log_{10}\mu_g$ | 0.358      | $\log_{10}\mu_g$ | 0.360      | $\log_{10}\mu_g$ | 0.359      |
| 2    | $\log_{10}V$    | 0.331      | $\log_{10}V$    | 0.311      | $\log_{10}V$    | 0.315      | $\log_{10}V$    | 0.312      |
| 3    | $\log_{10}N$   | 0.122      | $\log_{10}N$   | 0.129      | $\log_{10}N$   | 0.127      | $\log_{10}N$   | 0.129      |
| 4    | $\sigma_g$    | 0.049      | $\sigma_g$    | 0.058      | $\sigma_g$    | 0.055      | $\sigma_g$    | 0.056      |
| 5    | $T$          | 0.042      | $T$          | 0.052      | $T$          | 0.045      | $T$          | 0.047      |
| 6    | $\log_{10}V\log_{10}\mu_g$ | 0.016 | $\log_{10}N\log_{10}\mu_g$ | 0.022 | $\log_{10}N\log_{10}\mu_g$ | 0.021 | $\log_{10}N\log_{10}\mu_g$ | 0.021 |
| 7    | $\log_{10}N\log_{10}\mu_g$ | 0.016 | $\kappa$    | 0.017      | $\kappa$    | 0.019      | $\kappa$    | 0.019      |
| 8    | $\kappa$     | 0.015      | $\log_{10}V\log_{10}\mu_g$ | 0.017 | $\log_{10}V\log_{10}\mu_g$ | 0.018 | $\log_{10}V\log_{10}\mu_g$ | 0.017 |

On average, the second- and fifth-order OLS-derived
chaos expansion was 10–17 times faster than the MBN
scheme given the same single-mode aerosol population.
The exact speedup depended on the background ve-
locity; for weak updraft speeds, the performance of the
MBN scheme fared better, although it became much
worse for updrafts where $V < 2$ m s$^{-1}$. The ARG scheme
was consistently 1–3 times faster than those same chaos
expansions. Since the pathway for evaluating either the
ARG or chaos expansion schemes do not change de-
pending on the input parameters, their performance was
the same regardless of what inputs were provided.

4. Summary and conclusions
An efficient parameterization of droplet activation
for a single aerosol model under a wide variety of dif-
ferent physico-chemical properties and thermodynamic
conditions was developed via statistical emulation of a
detailed parcel model using polynomial chaos expan-
sions. The emulators predict the maximum supersatura-
tion achieved by a parcel, which is then used to diagnose
activated droplet number using Köhler theory in a
similar framework to existing activation parameteriza-
tions. The fourth- and fifth-order chaos expansions de-
derived from the detailed parcel model are more accurate
on average than two commonly used, physically based
parameterizations from the literature (Abdul-Razzak
and Ghan 2000; Morales Betancourt and Nenes 2014).
Additionally, the chaos expansions are all at least 10
times faster to evaluate than the MBN scheme and only
about twice as expensive as the ARG scheme. A simple
algorithm was suggested for evaluating a chaos expan-
sion that requires a minimal amount of data about the
expansion (such as the basis polynomials and the co-
efficients of the expansion terms) to be saved; in this
way, the chaos expansions offer a method for extending
lookup tables to very high dimensionalities without
suffering from exponentially rising storage costs.
Based on the large set of aerosol properties and thermodynamic conditions we sampled in order to derive and evaluate the chaos expansions, we observed that our emulators particularly outperform the existing schemes in conditions where a light updraft and heavy aerosol pollution (with respect to number concentration) are present. Because the ultimate goal of an activation parameterization is to couple the aerosol physics and chemistry to the cloud microphysics of a global-scale model, this deficiency in the existing parameterizations could be particularly important. Few global models have aerosol–cloud microphysics connections in their deep convection parameterizations, but many source potential cloud droplet formation based on a detailed aerosol activation calculation for their shallow convection and stratiform cloud microphysics schemes. These schemes sometimes artificially restrict the lowest possible updraft speed available for estimating droplet activation, but as a consequence they ensure that weak updrafts make up a large portion of the activation conditions considered during a model run. In regions of the world with heavy anthropogenic aerosol pollution—such as southern and eastern Asia—this provides a recipe for systematically underpredicting droplet number and potentially impacting either a global model’s simulated aerosol indirect effect on climate or the modeled aerosol–cloud interaction’s sensitivity to changes in anthropogenic aerosol emissions.

The global sensitivity analysis framed on the input parameter set used to derive the new chaos expansion emulators provides an additional, new check on the performance of existing activation schemes compared to the detailed parcel model. The breakdown of main Sobol’ indices calculated using the chaos expansions provides insight into the importance of interactions between the dominant first-order terms (updraft speed, number concentration, and geometric mean size of the aerosol distribution), which are further summarized by the total Sobol’ indices derived for the parcel model and both ARG and MBN schemes. The oversensitivity of the MBN scheme to the geometric mean radius and the undersensitivity of the ARG scheme to the updraft speed contribute to their disagreement with the parcel model across the range of pollution levels and updraft speeds studied here. Further, such sensitivity analyses could shed additional light on the potential biases of activation schemes and could provide useful metrics for evaluating the improvement of parameterizations more generally than simple ones based on relative or absolute error alone.

Critically, the framework from which the chaos expansions reported here are derived is extensible to the case where a complex, multispecies/multimodal aerosol population is tracked by a global model; in that case, the number of parameters describing the aerosol size distribution and chemical composition simply increases. Future work will derive chaos expansions emulating activation for a multimodal aerosol distribution specific to a particular global aerosol–climate model. Additionally, physical processes not considered here can also be introduced into the chaos expansion framework. For instance, entrainment can be incorporated into the parcel model following Seinfeld and Pandis (2006) and Barahona and Nenes (2007). Subgrid-scale variability in updraft speeds due to the coarse resolution of global model grids and the distribution of these updrafts can be represented either by a characteristic value (Morales and Nenes 2010) or by numerical integration over a distribution (Lohmann et al. 1999; Golaz et al. 2011). In the latter case, many activation calculations must be performed, incurring a large computational cost. However, the entire integration over a spectrum of droplet speeds could be parameterized in the chaos expansion framework, greatly reducing the cost of this calculation and potentially improving the accuracy of diagnosed cloud droplet number.

As the complexity of global aerosol–climate models increases with respect to the number of aerosol modes and species tracked by the model, there is a pressing need to understand how biases in activation calculations across the high-dimensional parameter spaces defining the aerosol–climate model affect cloud properties and ultimately impact modeled climate. This work highlights a novel way to build efficient, accurate activation schemes for this purpose akin to customized lookup tables, which cannot themselves extend to cover the necessary parameters. Employing such schemes should help improve simulated cloud microphysical properties and constrain modeled aerosol indirect effects on climate.

Acknowledgments. The work in this study was supported by the National Science Foundation Graduate Research Fellowship Program under NSF Grant 1122374, NSF Grant AGS-1339264, the National Research Foundation Singapore through the Singapore–MIT Alliance for Research and Technology and the interdisciplinary research group of the Center for Environmental Sensing and Modeling, and the U.S. Department of Energy, Office of Science (DE-FG02-94ER61937). We thank Steve Ghan (PNNL) and Athanasios Nenes (Georgia Tech) for reference implementations of their activation parameterizations.

APPENDIX A

Parcel Model Description

The adiabatic cloud parcel model implemented for this study follows the basic equations of Pruppacher and Klett (1997) and adopts the framework used by Nenes et al. (2001) to account for kinetic limitations on droplet growth. Fundamentally, the model integrates a system
of coupled ordinary differential equations that describe the thermodynamic evolution of an adiabatically lifted, nonentraining parcel. In all the simulations described here, we use the variable-coefficient ordinary differential equation solver (VODE; Brown et al. 1989) to integrate the system forward in time.

The model tracks the evolution of supersaturation $S$ with respect to water as

$$\frac{dS}{dt} = \alpha(T, P)V - \gamma(T, P) \frac{dw_c}{dt}, \quad (A1)$$

where $\alpha(T, P) = (gM_w L/c_p RT^2) - (gM_a/RT)$ and $\gamma(T, P) = (PM_a/\varepsilon sM_w) + (M_w L^2/c_p RT^2)$ are functions that are weakly dependent on temperature and pressure (Leaitch et al. 1986), $M_w$ and $M_a$ are the molecular weights of water and air, $L$ is the latent heat of evaporation of water, $c_p$ is the specific heat of dry air at constant pressure, $R$ is the universal gas constant, $g$ is the acceleration due to gravity, $\varepsilon s$ is the saturation vapor pressure, and $w_c$ is the liquid cloud water mass mixing ratio. Equation (A1) expresses the supersaturation as a balance between production due to adiabatic cooling and loss due to latent heat release. This same framework describes the parcel’s change in temperature over time,

$$\frac{dT}{dt} = \frac{gV}{c_p} - \frac{L}{c_p} \frac{dw_v}{dt}, \quad (A2)$$

where $V$ is the updraft velocity and $w_v$ is water vapor mass mixing ratio. Water mass is conserved as vapor condenses into cloud water,

$$\frac{dw_v}{dt} + \frac{dw_c}{dt} = 0. \quad (A3)$$

Equations (A1)–(A3) are linked through the growth of the cloud droplet population from the initial aerosol. Given $n$ bin sizes, each associated with a number concentration $N$ and a radius $r$, the change in cloud water can be written as

$$\frac{dw_c}{dt} = \frac{4\pi}{\rho_w} \sum_{i=1}^{n} N r_i^2 \frac{dr_i}{dt}, \quad (A4)$$

where $\rho_w$ and $\rho_a$ denote the density of water and air, respectively.

The diffusional growth rate for droplets in the $i$th bin is calculated by

$$\frac{dr_i}{dt} = \frac{G}{r_i} (S - S_{eq}), \quad (A5)$$

where $S$ is the environmental supersaturation, $S_{eq}$ is the Köhler-predicted equilibrium supersaturation of the droplet, and $G$ is a growth coefficient which is a function of both the physical and chemical properties of the particle receiving condensate,

$$G = \left\{ \frac{\rho_w RT}{e \gamma D_v M_w} \left[ (LM_w/RT) - 1 \right] \right\}^{-1}. \quad (A6)$$

Noncontinuum effects on the diffusivity $D_v$ and thermal conductivity $k'_a$ factors are accounted for with the corrections

$$D'_v = D_v \left( 1 + \frac{D_v}{a_c} \sqrt{\frac{2\pi M_w}{RT}} \right) \quad (A7)$$

and

$$k'_a = k_a \left( 1 + \frac{k_a}{a_T \rho_a c_p} \sqrt{\frac{2\pi M_w}{RT}} \right). \quad (A8)$$

In these correction terms, the thermal accommodation coefficient $a_T$ is assumed to be 0.96; the condensation coefficient $a_c$ is allowed to vary as observations suggest it could take values between 0.1 and 1.0 (Raatikainen et al. 2013). The instantaneous droplet growth rate is further modulated by the difference between the environmental supersaturation $S$ and the saturation ratio over the surface of the aqueous droplet $S_{eq}$. We treat the droplet-dependent $S_{eq}$ following Petters and Kreidenweis (2007), who employ a single-term $\kappa$ to parameterize particle hygroscopicity; values of $\kappa$ can be derived from laboratory experiments. Under the framework of $\kappa$–Köhler theory the curvature effect term remains the same, while the solute effect term is rewritten such that

$$S_{eq} = \frac{r^3 - r_d^3 \kappa}{r^3 - r_d^3 (1 - \kappa)} \exp \left( \frac{2M_w \sigma_w}{RT \rho_w} \right) \quad (A9)$$

where $r$ and $r_d$ are the droplet radius and the dry radius of its embedded aerosol particle (which is tracked for each initial aerosol size in the model) and $\sigma_w$ is the droplet surface tension, which we take to be independent of the droplet solution composition and described following the recommendation of Pruppacher and Klett (1997), $\sigma_w = 0.0761 - 1.55 \times 10^{-4} (T - 273)$. A limitation of this approach for computing $S_{eq}$ is that it is not convenient to derive analytical expressions for the critical supersaturation and radius; they must be computed numerically by finding the value $r_{crit}$ such that

$$\frac{dS_{eq}}{dr} \bigg|_{r_{crit}} = 0 \quad (A10)$$

and then computing $S_{crit} = S_{eq}(r_{crit})$ for a given $\kappa$ and $r_d$. This is accomplished using Brent’s method (Brent 1973) and by bounding $r_{crit}$ from below with the observation that $r_{crit} > r_d$. 
Finally, the parcel thermodynamic description is closed by predicting the pressure change within the ascending parcel following the hydrostatic relationship, which can be written using the ideal gas law as

\[
\frac{dP}{dt} = \frac{gPV}{R_d T_v},
\]

where \(T_v\) is the virtual temperature, which is employed to account for changes in air density due to loss of water vapor to condensate. Equations (A1)–(A4), (A11), and (A5) applied to each \(n\) droplet size bins form a closed system which conserves total water mass.

**APPENDIX B**

**Chaos Expansion Emulator Evaluation**

The PCM as applied here produces two outputs: a \(P\)-length vector of coefficients \(\alpha\) comprising real values and a \(P \times M\) matrix of orthogonal polynomial orders \(\Phi\) comprising integers. Each term in matrix \(\Phi\) contains a multi-index component for each term in the chaos expansion and indicates the order of the orthogonal polynomial corresponding to term 1 \(< j \leq M\) for expansion term 0 \(i \leq P\). For any expansion, \(\max(\Phi) = p\), the desired order of the chaos expansion. Algorithm 1 describes the evaluation of a chaos expansion.

**Algorithm 1:** Pseudocode for evaluating a polynomial chaos expansion of the form given in Eq. (2), applied to the computation of \(S_{\text{max}}\)

1: **for all** \(X_j\) **do**
2: \(Z_j \leftarrow \text{project } X_j\)
3: **end for**
4: \(\hat{S} \leftarrow 0\)
5: **for** row \(i = 0; i \leq P\) **do**
6: \(\hat{S}_i \leftarrow 1\)
7: **for** column \(j = 0; j \leq M\) **do**
8: \(k \leftarrow \Phi(i, j)\)
9: \(P_{i,k} \leftarrow \phi_j[Z(j)]\)
10: \(\hat{S}_i = \hat{S}_i \times P_{i,k}\)
11: **end for**
12: \(\hat{S} = \hat{S} + \alpha(i) \times \hat{S}_i\)
13: **end for**
14: \(S_{\text{max}} \leftarrow 10^5\)

Evaluating the chaos expansion involves two parts. First, the input parameters must be projected to conform to the space supported by the PDFs associated with each basis polynomial type, producing a set of parameters \(Z_j\). In general, a set of mixed orthogonal polynomials could be used to derive a chaos expansion, but here only Legendre polynomials were used, and each parameter can be projected using Eq. (5). Second, the polynomial can be evaluated by treating \(\Phi\) as a lookup table for the orders of each basis orthogonal polynomial. In practice, the evaluation of these orthogonal polynomials at \(Z_j\) for orders up to \(k\) can be efficiently precomputed (before the polynomial evaluation loop) by existing orthogonal polynomial libraries (Gautschi 1994).

In general, the only computationally complex part of the chaos expansion algorithm is the projection from \(X_j\) to \(Z_j\); given certain basis orthogonal polynomials and their associated PDFs, this procedure could involve numerical integration or otherwise complicated function evaluations. In the case of simple uniform PDFs, though, the process is achieved entirely by rescaling the parameters, with little computational overhead. Furthermore, although evaluating a chaos expansion requires looping over each of its terms, each term can be computed independently from one another and efficiently optimized. This is in contrast with an iterative scheme that could involve numerical integration or other costly operations and must be performed in sequence.

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