Characteristic updrafts for computing distribution-averaged cloud droplet number, and stratocumulus cloud properties

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Abstract. A computationally-effective framework is presented that addresses the contribution of subgrid-scale vertical velocity variations in predictions of cloud droplet number concentration (CDNC) in large-scale models. Central to the framework is the concept of a "characteristic updraft velocity", $w^\star$, which yields CDNC value representative of integration over a probability density function (PDF) of updraft (i.e., positive vertical) velocity. Analytical formulations for $w^\star$ are developed for computation of average CDNC over a Gaussian PDF using the Twomey droplet parameterization. The analytical relationship also agrees with numerical integrations using a state-of-the-art droplet activation parameterization. For situations where the variabilities of vertical velocity and liquid water content can be decoupled, the concept of $w^\star$ is extended to the calculation of cloud properties and process rates that complements existing treatments for subgrid variability of liquid water content. It is shown that using the average updraft velocity, $\bar{w}$, (instead of $w^\star$) for calculations of N_d, r_e and A (a common practice in atmospheric models) can overestimate PDF-averaged N_d by 10%, underestimate r_e by $10\% - 15\%$, and significantly underpredict autoconversion rate between a factor of 2 to 10. The simple expressions of $w^\star$ presented here, can account for an important source of parameterization "tuning" in a physically-based manner.

1. Introduction

The direct microphysical link between aerosol and clouds is the process of activation [*Köhler*, 1936] during which a fraction of aerosol particles (termed Cloud Condensation Nuclei; CCN) experience unconstrained growth and form cloud droplets. Increases in precursor aerosol concentration can augment cloud droplet number concentration (CDNC), cloud albedo [*Twomey*, 1977] and lifetime [*Albrecht*, 1989], with important implications for climate. Even though droplet activation is well understood (e.g., *Pruppacher and Klett* [1997]; *Conant et al.* [2004]; *Fountoukis et al.* [2007]), its representation in global climate models (GCMs) is far from trivial. The current practice is to use "mechanistic parameterizations" (simplified but accurate relationships based on ascending cloud parcel theory) that provide CDNC as a function of the precursor aerosol and the parcel (cloud-base) updraft velocity w. Since the pioneering work of Twomey [*Twomey*, 1959], a number of prognostic, physically based parameterizations of aerosol activation have been developed and implemented in GCMs (e.g., *Abdul-Razzak et al.* [1998]; *Nenes and Seinfeld* [2003]; *Fountoukis and Nenes* [2005]; *Ming et al.* [2006]), some of which have been evaluated against observations of CDNC in stratocumulus and cumulus clouds (e.g., *Fountoukis et al.* [2007]; *Meskhidze et al.* [2005]).

CDNC predicted by mechanistic parameterizations is sensitive to updraft velocity; this poses a challenge in their implementation in GCMs, because cloud-scale updraft velocity is not resolved. This issue is currently addressed by either prescribing the updraft velocity from observations (e.g., *Sotiropolou et al.* [2007]; *Pringle et al.* [2009]), or, diagnosing it from grid-resolved quantities, such as the grid-cell scale turbulent kinetic energy (K), $w^\star = \hat{w} + 0.7\sqrt{K}$ [*Lohmann et al.*, 1999], where $\hat{w}$ is the average (resolved) vertical velocity. CDNC is then computed from this "characteristic velocity" and applied to all cloud processes, mainly calculation of cloud optical depth, effective radius and autoconversion rate of cloud water to rain. These treatments for updraft velocity carry an important assumption: that a single updraft can be used to compute the "representative" CDNC for all cloud processes in the grid cell. In reality, a distribution of updrafts exist in each grid cell, each of which could be associated with its own droplet number. To account for this sub-grid variability, a probabilistic approach towards cloud properties can be used, in which each cloud forms in a grid cell with a characteristic updraft w occurring with a probability $P(w)$. Assuming that a continuous probability distribution function (PDF) can be used to describe $P(w)$, grid-averaged cloud properties can be derived. For example, the average droplet number in the grid cell is given by $\bar{N}_d = \int_0^\infty N_d(w)P(w)\mathrm{d}w / \int_0^\infty P(w)\mathrm{d}w$, where $N_d(w)$ denotes the CDNC that corresponds to w.

The probabilistic approach has been shown to successfully predict cloud-base CDNC in warm clouds in a number of field studies (e.g., *Conant et al.* [2004]; *Peng et al.* [2005]; *Meskhidze et al.* [2005]; *Fountoukis et al.* [2007]). Despite its conceptual strength, numerically integrating droplet number over a PDF is computationally expensive. *Peng et al.* [2005], *Meskhidze et al.* [2005] and *Fountoukis et al.* [2007] explored the possibility of replacing $\bar{N}_d$ with a single CDNC calculation at a "characteristic" updraft velocity, $w^\star$, so that $N_d(w^\star) = \bar{N}_d$. These studies found that, within measurement uncertainty, $w^\star$ is given by the PDF-average updraft velocity, $\bar{w} = \int_0^\infty wP(w)\mathrm{d}w / \int_0^\infty P(w)\mathrm{d}w$.

Although insightful, the aformentioned studies focused on a limited range of aerosol types and updraft velocity spectra, so the general applicability of $w^\star$ needs to be established. Furthermore, calculation of CDNC in climate models is not an endpoint, but rather an intermediate step for computing cloud processes (e.g., autoconversion rate) and radiative properties (e.g., effective radius) that impact the simulated

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hydrological cycle and climate. Given that cloud properties depend nonlinearly on N_d and the cloud liquid water content, q_c, correctly accounting for their subgrid variability is crucial for unbiased representation of clouds in large-scale models. GCM schemes have been developed to account for subgrid-scale variability in q_c (e.g., *Morrison and Gettelman* [2008]), as using grid-scale values of q_c were known to induce biases in nonlinear cloud processes (e.g., *Pincus and Klein* [2000]). Climate models however do not account for the subgrid-scale (SGS) variability of N_d. This is an especially important oversight for indirect effect studies, since the aerosol-N_d link (and its subgrid variability) is at the heart of the aerosol-cloud-climate interactions.

This study aims to provide a computationally-effective framework to address the issue of PDF-averaging of CDNC that arise from subgrid scale variations in vertical velocity. The optimum characteristic velocity, $w^\star$, is determined for computation of average droplet number concentration over a Gaussian PDF of updraft velocity. For situations where the joint distribution of q_c and w can be decoupled, we develop expressions of characteristic velocity that accounts for sub-grid variability of CDNC in processes such as autoconversion rate and effective radius, that complement existing treatments for subgrid variability of liquid water content.

2. Probability Distributions

2.1. The problem of joint PDFs between q_c and w.

Cloud microphysical processes depend on several quantities (the two most important being cloud liquid water content, q_c and CDNC, N_d) that exhibit large sub-grid scale (SGS) variations. The functional form of the PDFs that express their SGS variability has been the subject of intense study (e.g., *Pincus and Klein* [2000]; *Golaz et al.* [2002]; *Morrison and Gettelman* [2008]; *Cheng and Xue* [2009]; *Zhu and Zuidema* [2009]). The problem of determining these distributions is usually approached by prescribing a joint PDF of the variables under consideration. Although a number of PDF functions have been proposed with a wide range of complexity, a universally-accepted form remains elusive. The parameters of the PDFs are either prescribed (obtained by fits to observational data or cloud-resolving models), diagnosed from resolved quantities in the GCM simulation (e.g., turbulent kinetic energy) or determined from prognostic equations that describe the higher-order moments of the distribution (e.g., *Golaz et al.* [2002]).

The joint-distribution approach can be described as follows. Consider a cloud microphysical property or process rate (e.g., effective radius, CDNC, autoconversion rate), $F(q_c, N_d)$, that depends on q_c and N_d. If the joint probability distribution $\mathcal{P}(q_c, N_d)$ is known, the average property, $\bar{F}$, is given by

$$\bar{F} = \iint \mathcal{P}(q_c, N_d) F(q_c, N_d) \mathrm{d}q_c \, \mathrm{d}N_d \quad (1)$$

Using mean values of N_d ($\bar{N_d}$) and q_c ($\bar{q_c}$) to estimate $\bar{F}$ is equivalent to neglecting their SGS variability, as it implies that $\mathcal{P}(q_c, N_d) \simeq \delta(q_c - \bar{q_c})\delta(N_d - \bar{N}_d)$, where δ is the well-known Dirac function. This approach induces biases in calculations of $\bar{F}$, because in general $\bar{F} \neq F(\bar{q_c}, \bar{N}_d)$.

Some GCMs cloud schemes (e.g., *Pincus and Klein* [2000]; *Morrison and Gettelman* [2008]) partially overcome the subgrid variability problem by accounting for variations in q_c. This is equivalent to assuming $\mathcal{P}(q_c, N_d) = Q(q_c)\delta(N_d - N_0)$, where $Q(q_c)$ is a PDF describing the SGS variability of liquid water content (e.g., a gamma distribution; *Morrison and Gettelman* [2008]). N_0 is a "characteristic" value of droplet number in the grid, typically assumed to correspond to $N_d(\bar{w})$ or prescribed to a fixed value (e.g., *DelGenio et al.* [1996]). *Morrison and Gettelman* [2008] proposed using a characteristic $q_c^\star$, such that the integration over $Q(q_c)$ is equivalent to evaluating F at $q_c^\star$, i.e.,

$$\overline{F} = F(q_c^\star, N_0) = F_0 N_0^x \int q_c^y Q(q_c) \mathrm{d}q_c \quad (2)$$

where we have assumed that $F(q_c, N_d)$ has a power law dependence on N_d and q_c, i.e., $F(q_c, N_d) = F_0 N_d^x q_c^y$, where F_0, x, y are coefficients. From Equation 2, $q_c^\star = \left[\int q_c^y Q(q_c) \mathrm{d}q_c\right]^{1/y}$

This kind of approach, although in the right direction of addressing the issue of SGS variability, still neglects the variability of cloud properties on N_d hence will induce biases in the computation of $\bar{F}$. This is particularly important for indirect effect assessments, given that a key sensitivity (i.e., of cloud processes to changes in N_d) is not resolved correctly.

2.2. Joint PDF for stratocumulus in well-mixed boundary layers

Despite many conceptual advantages of the joint PDF approach, its main limitation is the need to predict the distribution moments (*Pincus and Klein* [2000]), often from a set of prognostic equations that need to be solved at much higher temporal resolution that of the parent model (*Golaz et al.* [2002]; *Zhu and Zuidema* [2009]). As a result, an explicit dynamic PDF solution requires significant computational resources. However, in-situ observations and reanalysis of large-eddy simulations of clouds suggest that simpler, prescribed forms of PDFs may capture much of the q_c-w variability for specific cloud regimes.

For cumulus clouds, q_c and w are correlated so that the functional form of the joint PDF is complex (e.g., *Golaz et al.* [2002]; *Guo et al.* [2008]). Stratocumulus clouds in well-mixed non-precipitating boundary layers however exhibit a single mode updraft velocity PDF with little skewness that can often be described with a Gaussian distribution (*Golaz et al.* [2002]; *Kogan* [2005]; *Guo et al.* [2008]). Another important characteristic is that the distribution of updrafts tends to be weakly-coupled with the distribution of thermodynamic variables (e.g., q_c or equivalent potential temperature θ_l) (e.g., C̆urry [1985]). Because of this, we can assume that the distribution of q_c is (to first order) decoupled from the distribution of w, or, $\mathcal{P}(q_c, w) = Q(q_c)P(w)$, where $Q(q_c)$, $P(w)$ are the respective PDFs of q_c and w. It is interesting to note that each of the Gaussian distributions in the double-Gaussian cloud scheme proposed in *Golaz et al.* [2002] also exhibit a similar decoupling between dynamic and thermodynamic variables within each Gaussian.

If $P(w)$ is known, the one-to-one correspondence between w and N_d (e.g., provided by cloud drop parameterizations) suggests that $P(w)$ can be remapped onto the N_d domain to provide a PDF of N_d, $p(N_d)$. This means that the decoupling between q_c, w variabilities implies a decoupling between q_c, N_d variabilities, hence $\mathcal{P}(q_c, N_d) = Q(q_c)p(N_d)$. Accounting for SGS variability in cloud processes can therefore be addressed as follows. Assuming $F = F_0 N_d^x q_c^y$, and $\mathcal{P}(q_c, N_d) = Q(q_c)p(N_d)$, Equation 1 becomes

$$\bar{F} = F_0 \int N_d^x p(N_d) \mathrm{d}N_d \int q_c^y Q(q_x) \mathrm{d}q \quad (3)$$

Equation 3 allows the definition of characteristic values of q_c and N_d such that $q_c^\star = \left[\int q_c^y Q(q_c) \mathrm{d}q_c\right]^{1/y}$, $N_d^\star = \left[\int N_d^x p(N_d) \mathrm{d}N_d\right]^{1/x}$. Equation 3 then becomes

$$\bar{F} = F(q_c^\star, N_d^\star) = F_0 q_c^{\star y} N_d^{\star x} \quad (4)$$

Existing approaches for SGS variability of q_c can be used to compute $q_c^\star$ (e.g., *Morrison and Gettelman* [2008]). $N_d^\star$ can then be related to a characteristic velocity, $w^\star$, so that an application of a cloud droplet parameterization can give $N_d^\star = N_d(w^\star)$. The value of $w^\star$ will depend on the cloud process parameterization, and is detailed in sections 3,4. If $w^\star$ is known however, application of Equation 4 constitutes a *substantial* acceleration of calculating PDF-averaged properties, as a numerical integration over the joint PDF (Equation 1) is replaced with a single function evaluation (Equation 4).

2.3. PDFs for calculating CDNC

PDF-averaged CDNC is an important quantity useful for evaluating GCM simulations. Its calculation, compared to other cloud microphysical properties and processes (e.g., autoconversion and effective radius) is also much simpler, given that it requires only knowledge of the PDF of vertical velocity $P(w)$ (it also depends on aerosol properties, but these are assumed known). For the purpose of this work, we will assume that $P(w)$ follows a Gaussian distribution, $P(w) = (\sqrt{2\pi}\sigma)^{-1}\exp\left\{-(w-\hat{w})^2/(2\sigma^2)\right\}$, where $\hat{w}$ is the mean (resolved) vertical velocity, and σ is the standard deviation of the velocity PDF. Given the scale of GCM grid cells ($\sim 100km$), $\hat{w}$ is very small compared to the magnitude of fluctuations, so that $P(w) \approx (\sqrt{2\pi}\sigma)^{-1}\exp\left\{-w^2/(2\sigma^2)\right\}$.

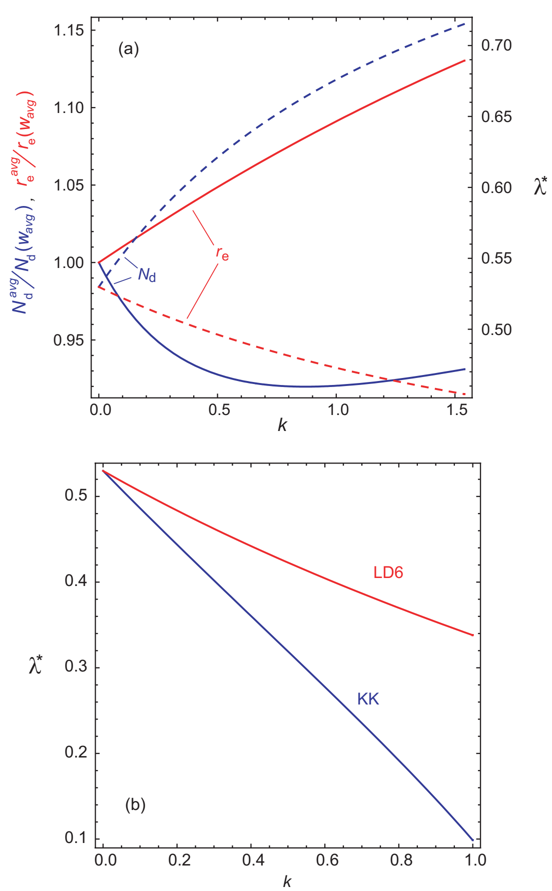

Figure 1. (a) $\bar{N}_d/N_d(\bar{w})$ (solid blue line) and $\bar{r_e}/r_e(\bar{w})$ (solid red line) and respective characteristic non-dimensional velocity $\lambda^\star = w^\star/\sigma$ (dashed lines) as a function of the CCN spectrum steepness parameter k. (b) $\lambda^\star$ for KK (blue) and LD6 (red) parameterization as a function of the parameter k. CDNC is computed from *Twomey* [1959] parameterization.

In boundary layers, velocity fluctuations are mainly associated with turbulence, so that the turbulent kinetic energy, K, scales with the sub-grid velocity fluctuations, $K \sim \sigma^2$. Therefore, from the grid-scale resolved K, a Gaussian PDF can be diagnosed (with $\sigma \sim K^{1/2}$) that is consistent with the large-scale simulation and suitable for computing velocity averaged cloud processes and properties. Outside of the boundary layer, a distribution can still be diagnosed from other sources of variability (such as gravity waves) but is outside of the scope of this study. For a Gaussian distribution with $\hat{w} \approx 0$, the average updraft velocity, $\bar{w}$, (i.e., the average over the positive part of the vertical velocity distribution) is given by $\bar{w} = (2/\pi)^{1/2}\sigma \approx 0.79\sigma$ [*Fountoukis et al.*, 2007].

2.4. Impact of CDNC variability on cloud processes and properties

Let $F(w)$ denote any cloud microphysical property that depends on the updraft velocity w. F averaged over the positive vertical velocities, $\bar{F}$, is then given by

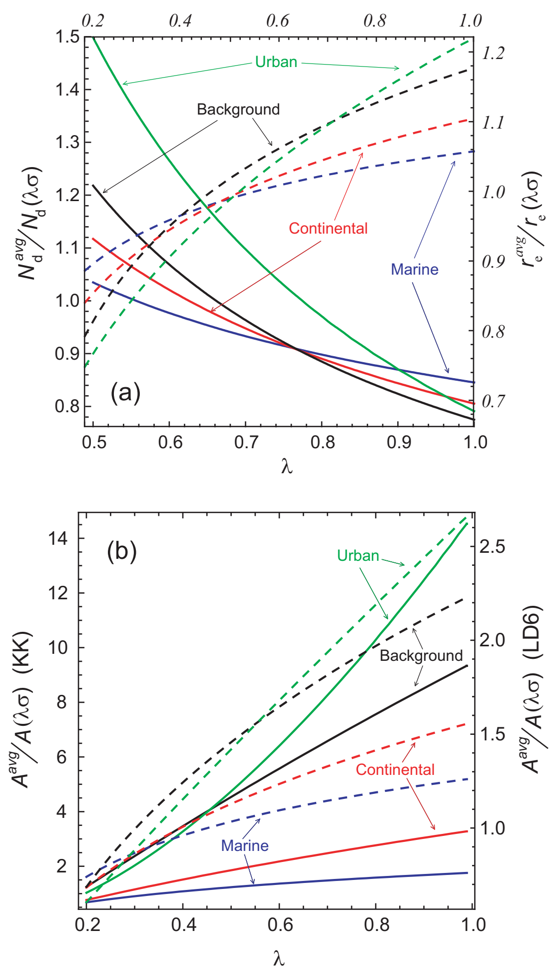

Figure 2. (a) $\bar{N}_d/N_d(\lambda\sigma)$ (solid lines) and $\bar{r_e}/r_e(\lambda\sigma)$ (dashed lines) computed with the *Fountoukis and Nenes* [2005] parameterization as a function of λ. (b) $\bar{A}/A(\lambda\sigma)$ using N_d computed with *Fountoukis and Nenes* [2005], as a function of λ for the four types of aerosol considered. Solid lines correspond to the *Khairoutdinov and Kogan* [2000] (KK) parameterization, and, dashed lines show calculations for *Liu and Daum* [2004] (LD6) expression.

$$\bar{F} = \int_0^\infty F(w)P(w)\mathrm{d}w / \int_0^\infty P(w)\mathrm{d}w \quad (5)$$

Often, $F(w)$ can be approximated with a power law, $F(w) = aw^b$, where a, b are coefficients that do not depend on w. In this case, $\bar{F}$ for a Gaussian $P(w)$ is computed from Equation 5,

$$\bar{F} = \frac{a}{\sqrt{\pi}} 2^{b/2} \sigma^b \Gamma\left(\frac{b+1}{2}\right) \quad (6)$$

where $\Gamma(z) \equiv \int_0^\infty t^{z-1}e^{-t}\mathrm{d}t$ is the Gamma function.

The only natural velocity scale present in a Gaussian distribution $P(w)$ is σ; a non-dimensional velocity, λ, can therefore be defined as $\lambda \equiv w/\sigma$. $\bar{F}$ (Equation 6) normalized with $F(\lambda\sigma) = a\lambda^b\sigma^b$ is given by

$$\frac{\bar{F}}{F(\lambda\sigma)} = \frac{2^{b/2}}{\sqrt{\pi}} \lambda^{-b} \Gamma\left(\frac{b+1}{2}\right) \quad (7)$$

which only depends on b and λ. Equation 7 can be used to determine the characteristic non-dimensional velocity, $\lambda^\star$, for which $F(\lambda^\star\sigma) = \bar{F}$,

$$\lambda^\star = 2^{1/2}\pi^{-1/2b} \Gamma\left(\frac{b+1}{2}\right)^{1/b} \quad (8)$$

which depends solely on b.

3. Application of PDF to calculation of droplet number

The extensively used *Twomey* [1959] parameterization was developed assuming a power law expression for the CCN spectrum, $N_{CCN}(s) = cs^k$, (i.e., the number of CCN at supersaturation s), from which $N_d(w) = f(c,k)w^{3k/(2k+4)}$, where $f(c,k) = c^{2/(k+2)}\left[1.63 \times 10^{-3}/\{k\mathrm{B}(3/2, k/2)\}\right]^{k/(k+2)}$ and $\mathrm{B}(a,b) = \Gamma(a)\Gamma(b)/\Gamma(a+b)$ is the Euler beta function. Using this simple parameterization, the PDF-averaged droplet number $\bar{N_d}$ over the CDNC computed at $w = \lambda\sigma$, $N_d(\lambda\sigma)$, is given by setting b equal to $3k/(2k+4)$ in Equation 7,

$$\frac{\bar{N_d}}{N_d(\lambda\sigma)} = \frac{1}{\sqrt{\pi}} 2^{\frac{3k}{4k+8}} \lambda^{-\frac{3k}{2k+4}} \Gamma\left(\frac{5k+4}{4k+8}\right) \quad (9)$$

The characteristic updraft, $\lambda^\star\sigma$, so that $N_d(\lambda^\star\sigma) = \bar{N_d}$, is given by Equation 8

$$\lambda^\star = 2^{1/2}\pi^{\frac{k+2}{3k}} \Gamma\left(\frac{5k+4}{4k+8}\right)^{\frac{2k+4}{3k}} \quad (10)$$

Therefore the characteristic updraft $w^\star = \lambda^\star\sigma$ is solely determined by the steepness of the CCN spectrum (i.e, k) and σ. Over the atmospherically relevant values of k (0.1-1.5) [*Twomey and Wojciechowski*, 1968], $\lambda^\star$ ranges between 0.55 for $k = 0.1$ (clean conditions) and 0.74 for $k = 1.5$ (polluted). An average value for $\lambda^\star$, $\lambda^\star_{avg} = 0.65$, is 17% lower than the average updraft ($\lambda = 0.79$). $\bar{N_d}/N_d(\bar{w})$ is numerically close to unity, ranging between 0.92 and 1 (Figure 1a). All together, this implies that using $\bar{w}$ to estimate average droplet number would tend to overestimate $\bar{N_d}$ by at most 10%.

More sophisticated activation parameterizations also exhibit a power-law like dependence on w, hence we expect $\lambda^\star$ to be closely approximated by Equation 10. To assess this, we use the *Fountoukis and Nenes* [2005] parameterization, which is based on the framework of an air parcel rising at constant speed. Droplets are classified by the proximity to their critical diameter ("population splitting"), allowing calculation of the cloud maximum supersaturation from the numerical solution of the balance of water vapor availability from cooling and depletion from the condensational growth. The CDNC is then equal to the CCN that activate at the cloud maximum supersaturation. The parameterization allows for the accurate treatment of complex aerosol size distribution, chemical composition and droplet growth kinetics. It has been expanded to treat entrainment effects on CDNC (*Barahona and Nenes* [2007]), adsorption activation (*Kumar et al.* [2009]) and Giant CCN (*Barahona et al.* [2010]). The accuracy of the parameterization has been evaluated with detailed numerical simulations [*Nenes and Seinfeld*, 2003; *Fountoukis and Nenes*, 2005; *Barahona and Nenes*, 2007; *Barahona et al.*, 2010] and in-situ data for cumuliform and stratiform clouds of marine and continental origin [*Meskhidze et al.*, 2005; *Fountoukis et al.*, 2007].

The *Fountoukis and Nenes* [2005] parameterization provides a numerical relation $N_d(w)$ rather than a explicit functional form; PDF averaging is therefore done numerically. Calculations are carried out for ammonium sulfate aerosol with size distribution characteristics of marine, clean continental, average background and urban environments given by *Whitby* [1978] (listed in Table 1). PDF averages of N_d are computed for a set of Gaussian vertical velocity distributions with $\hat{w} = 0$, and σ ranging from 0.05 to 0.75 ms^{-1}. Since the $N_d(w)$ relation provided by this parameterization does not follow a strict power law dependence, $\lambda^\star$ will exhibit some dependence on the parameter σ defining the PDF. Therefore, $\bar{N_d}$ calculated with *Fountoukis and Nenes* [2005] is averaged over the range of σ under consideration in this study.

Figure 2a presents the resulting $\bar{N_d}/N_d(\lambda\sigma)$ (solid lines) as a function of λ for all aerosol considered. When $\bar{w}$ is used (i.e., $\lambda = 0.79$), $\bar{N_d}$ is overestimated on average by 9.8%. The average $\lambda^\star$ for all aerosol distributions and σ considered is $\lambda \approx 0.676$ (close to the Twomey value of 0.65), which if used for calculating N_d, overestimates $\bar{N_d}$ by as little as 0.03%. Using the characteristic λ derived from the *Twomey* [1959] parameterization ($\lambda = 0.65$) gives N_d that approximates $\bar{N_d}$ to within 2.2%.

4. CDNC variability effects on Autoconversion rate and Effective radius

4.1. Autoconversion rate

Assuming that the joint distribution of q_c and w can be expressed as in Equation 3, characteristic values of q_c and N_d (w) can be defined that allow evaluation of cloud processes and properties (with a single function evaluation) that are representative of their PDF-averaged values (Equation 4). *Morrison and Gettelman* [2008] have already determined $q_c^\star$ assuming that $Q(q_c)$ follow a gamma distribution. Here we determine $w^\star$ that can be used for computing $N_d^\star$ in Equation 4.

Most autoconversion parameterizations sensitive to CDNC exhibit a power-law dependence on N_d; $w^\star$ thus would depend on how autoconversion rate, A, scales with N_d. This is demonstrated with two autoconversion parameterizations, by *Khairoutdinov and Kogan* [2000] (denoted KK hereafter), where $A \sim N_d^{-1.79}$, and, the R_6 formulation of *Liu and Daum* [2004] (denoted here LD6), where $A \sim N_d^{-1}$. Application of Equation 7 to each parameterization provides the updraft-average autoconversion rate.

Table 1. Dry aerosol size distributions used in this study (*Whitby* [1978]). D_{pgi}, σ_{gi}, N_i is the geometric mean diameter (μm), spectral width, and number concentration (cm^{-3}), respectively, of mode "i".

Aerosol Type	D_{pg1}	σ_{g1}	N_1	D_{pg2}	σ_{g2}	N_2	D_{pg3}	σ_{g3}	N_3
Marine	0.010	1.6	340	0.070	2.01	60	0.620	2.70	3.1
Continental	0.016	1.6	1000	0.068	2.10	800	0.920	2.20	0.7
Background	0.016	1.7	6400	0.076	2.01	2300	1.020	2.16	3.2
Urban	0.014	1.8	106000	0.054	2.16	32000	0.860	2.21	5.4

For KK, $A = 1350 q_c^{2.47} N_d^{-9/5}$, with q_c being the cloud water mixing ratio. Introducing the *Twomey* [1959] relation for $N_d(w)$,

$$A(q_c, w) = 1350 q_c^{2.47} f(c,k)^{-9/5} w^{-27k/(10k+20)} \quad (11)$$

Thus, if $A(w, q_c)$ is evaluated at a characteristic $q_c^\star$ that account for the integration over the PDF of q_c (Equation 4), then autoconversion follows a power law dependence on w; the formalism developed in section 2.4 can then be applied to calculate the PDF-average A, where $a = 1350(q_c^\star)^{2.47} f(c,k)^{-9/5}$ and $b = -27k/(10k+20)$.

For LD6, $A = 3\rho_a^3 \kappa_2 \beta_6^6 q_c^3 N_d^{-1} H(R_6 - R_{6c})/(4\pi\rho_w)$, where $\kappa_2 = 1.0 \times 10^{17} m^{-3} s^{-1}$, H is the Heaviside function, R_6 is the 6$^{\text{th}}$ moment of the size distribution, R_{6c} is the "critical" threshold radius, and β_6 is a non-dimensional parameter depending on the spectral shape of the cloud droplet size distribution [*Liu and Daum*, 2004]. LD6 is a product of two functions: a collection function that gives the total coalescence rate, and, a threshold function that expresses the fraction of coalescence attributed to autoconversion. LD6 implement the Heaviside threshold function, which may introduce biases (e.g., *Wood and Blossey* [2005]; *Liu and Daum* [2005]). *Liu et al.* [2006a] addressed this issue by developing a kinetically-defined threshold function of the form $T(q_c, N_d) = 1 - exp\left[-(1.03 \times 10^{16} N_d^{-3/2} q_c^2)^\mu\right]$ where $\mu \sim 0.36$ (e.g., *Hsieh et al.* [2009]) is an empirical parameter. Autoconversion rates using this threshold function exhibits two regimes: i) one where the argument in the exponential of $T(q_c, N_d)$ is large, so that $T(q_c, N_d) \simeq 1$, and A scales with N_d^{-1}, and, ii) one where the argument in the exponential of $T(q_c, N_d)$ is small, so that upon expansion yields $T(q_c, N_d) \simeq 1 - \left[1 - (1.03 \times 10^{16} N_d^{-3/2} q_c^2)^\mu\right] = (1.03 \times 10^{16} N_d^{-3/2} q_c^2)^\mu$. Therefore, A scales with $q_c^{3+2\mu} N_d^{-(1+3\mu/2)}$, which for $\mu \sim 0.36$ carries a strong $N_d^{-1.54}$ dependence, close that seen for the KK parameterization. Given this, we carry out calculations only with the N_d^{-1} dependence, to determine an "upper" limit in $w^\star$ (the lower given by KK), knowing that it lies between the limits defined by the two parameterizations.

After introducing the Twomey relation for $N_d(w)$ into LD6,

$$A(q_c, w) = \frac{3\kappa_2 \beta_6^6 \rho_a^3}{4\pi\rho_w f(c,k)} H(R_6 - R_{6c}) q_c^3 w^{-3k/(2k+4)} \quad (12)$$

hence A follows a power-law dependence on updraft with $a = 3\kappa_2 \beta_6^6 \rho_a^3 q_c^3 H(R_6 - R_{6c})/\{4\pi\rho_w f(c,k)\}$ and $b = -3k/(2k+4)$.

Equation 8 can then be applied to $A(q^\star, w)$, and from Equations 11 and 12 determine the characteristic updraft $\lambda^\star$, for autoconversion (i.e, $\lambda^\star$ for which $A(q_c^\star, \lambda^\star\sigma) = \bar{A}$). This is presented in Figure 1b, which shows $\lambda^\star$ for KK (blue line) and LD6 (red line) as a function of k. Averaging $\lambda^\star$ over the atmospheric range of k gives $\lambda_{avg}^\star = 0.33$ for KK and $\lambda_{avg}^\star = 0.44$ for LD6. These characteristic λ values are about 50% smaller than the characteristic updraft for estimating $\bar{N}_d$.

$\bar{A}/A(q_c^\star, \lambda\sigma)$ is also numerically computed with respect to λ using the *Fountoukis and Nenes* [2005] parameterization (for the same aerosol types and σ as of section 3) and presented in Figure 2b. $\lambda^\star$ ranges between 0.2 and 0.35 for KK (with $\lambda_{avg}^\star = 0.26$ for the aerosol considered), and, from 0.3 to 0.45 for LD6 (with $\lambda_{avg}^\star = 0.37$). These values are on average between 33% to 45% of the average updraft. If CDNC computed at $\bar{w}$ is used (instead of $\lambda^\star\sigma$) for autoconversion rate calculations (a current practice in climate models), A will be substantially underestimated. The worst underestimation occurs for the Urban aerosol, where the factor is ~ 10 for KK and ~ 2 for LD6 (Figure 2). The deviation seen for KK is related to the strong sensitivity of A to N_d; given that the parameterization was developed for "clean" drizzling clouds (i.e., N_d <150 cm^{-3}), it is possible that its usage for polluted clouds may be outside of its region of applicability. This agrees with *Hsieh et al.* [2009], whom found that deviation of KK from observation-derived autoconversion was largest when clouds were far from a drizzling state. Averaged over all aerosol types considered, the underestimation factor is 5.5 for KK, and 1.7 for LD6.

4.2. Effective radius

Droplet effective radius, r_e, is a key parameter of the cloud droplet distribution used for calculating cloud optical depth. r_e depends on N_d as,

$$r_e = \beta \left(\frac{3\rho_a}{4\pi\rho_w}\right)^{1/3} q_c^{1/3} N_d^{-1/3} \quad (13)$$

where β is a spectral dispersion parameter ranging from 1.1 to 1.6 (*Liu and Daum* [2002]). β exhibits a weak dependence on q_c and N_d (i.e., w) (*Liu and Daum* [2002]; *Liu et al.* [2006b, 2008]). We will therefore first consider β as a constant during the PDF integration (equal to the value for $q_c = q_c^\star$ and $N_d = N_d(w^\star)$).

By substituting N_d into Equation 13 with the Twomey parameterization, r_e and w are related by a power law, $r_e = \{3\beta\rho_a q_c/(4\pi\rho_w f(c,k))\}^{1/3} w^{-k/(2k+4)}$. Equation 8 can then be used, with a and b equal to $\{(3\beta\rho_a q^\star)/(4\pi\rho_w f(c,k))\}^{1/3}$ and $-k/(2k+4)$ respectively, to determine the dependence of the relevant characteristic velocity (such that $r_e(\lambda^\star\sigma) = \bar{r_e}$) on the slope of the CCN spectrum. Figure 1a shows that $\lambda^\star$ for r_e ranges between 0.46 and 0.52; averaged over the atmospherically relevant range of k, $\lambda_{avg}^\star = 0.48$, corresponding to 60% of $\bar{w}$.

Numerically calculating $\lambda^\star$ with the *Fountoukis and Nenes* [2005] parameterization (Figure 2a, dashed line) indicates that $\lambda^\star$ ranges between 0.50 and 0.70 (with $\lambda_{avg}^\star = 0.54$). Using $N_d(\bar{w})$ to estimate effective radius leads to an average 10% underestimation (13% for urban aerosol) relative to r_e^{avg}. When the Twomey-derived $\lambda_{avg}^\star = 0.48$ is used, r_e^{avg} is underestimated on average by only 2%.

Liu et al. [2008] proposed an empirical parameterization to account for the dependence of β on q_c and N_d, in which β scales with $(q_c/N_d)^{-0.14}$. The r_e expression still maintains its power law dependence on q_c and w, but the exponent in the latter changes from $-k/(2k+4)$ to $-0.57k/(2k+4)$. With this modification, $\lambda_{avg}^\star$= 0.50 using the Twomey parameterization and 0.62 with the *Fountoukis and Nenes* [2005] parameterization.

5. Summary - Conclusions

This study presents a computationally-effective framework to address the issue of PDF-averaging of CDNC that arise from subgrid scale variations in vertical velocity. Central to the framework is the concept of a "characteristic velocity", $w^\star$, which if introduced into a mechanistic CDNC parameterization provides a droplet number concentration characteristic of the value averaged over a PDF of updraft velocity. The concept of characteristic velocity is then extended for calculation of cloud properties and process rates, such as autoconversion and effective radius.

The approach of using characteristic values of q_c and N_d in place of integrating over a PDF requires knowledge of its functional form; this may be possible for certain climatically important cloud types, such as stratocumulus in well-mixed boundary layers. Based on the weak correlation between w and q_c for these clouds, we propose the usage of a joint PDF that is the product of two functions, each representing the PDF of q_c and w, respectively. This, together with the power-law dependence on q_c and N_d (often characterizing cloud process parameterizations) allows the determination of $w^\star$ that complement existing treatments for subgrid variability of liquid water content.

Analytical expressions for $w^\star$ (or its equivalent nondimensional form $\lambda^\star = w^\star/\sigma$) are determined assuming a Gaussian PDF of updrafts and CDNC from the parameterization of *Twomey* [1959]. $\lambda^\star$ was also numerically determined with the *Fountoukis and Nenes* [2005] parameterization for a wide range of aerosol size and updraft velocity distributions. Both approaches give $\lambda^\star$ that are in close agreement (e.g., for N_d, both agree to within 3%), ensuring that the analytical expressions are accurate approximations of $\lambda^\star$. For CDNC, $\lambda^\star$ is $\approx 15\%$ lower than $\bar{w}$, but within the 20% experimental uncertainty associated with the "optimal" $\lambda = 0.8$ determined using in-situ cloud observations (*Peng et al.* [2005], *Conant et al.* [2004], *Meskhidze et al.* [2005] and *Fountoukis et al.* [2007]). Using $\bar{w} = 0.79\sigma$ to compute N_d overestimates $\bar{N}_d$ by about 10%. In calculations of effective radius, using $\bar{w}$ underestimates PDF-averaged values by $10 - 15\%$ ($\sim 1 - 2\mu$m); $\lambda^\star$ for this microphysical parameter is on average 68% of the average updraft. Owing to its strongly nonlinear dependence on N_d, $\lambda^\star$ for autoconversion rate calculation is $\sim 30 - 40\%$ of $\bar{w}$; depending on the parameterization used, estimating autoconversion rate with $\bar{w}$ underestimates A on average by a factor of $2 - 5$.

This study presents a methodology that maps, in a simple way, the subgrid-scale variability of w onto subgrid-scale variability of N_d. We demonstrate that using $\bar{w}$ for calculations of N_d, r_e and A (a common practice in GCMs) leads to biases which are especially significant for autoconversion rates. This bias can be corrected if an appropriate $w^\star$ (specific to each cloud property or microphysical parameterization) is used. The method still needs to be evaluated against detailed cloud simulations and in-situ observations, especially when CDNC in calculations of effective radius and autoconversion is strongly affected by microphysical processes above cloud base (e.g., collision-coalescence or entrainment). Nevertheless, the method of characteristic properties carries much potential, as it addresses a source of parameterization "tuning" in a physically-based way. Even GCMs that use two-moment cloud schemes (and account for the effect of cloud microphysical processes on the droplet distribution) will substantially benefit from the methodology presented here, as it points to how one should calculate the droplet activation term (and other processes when cloud are far from a precipitating state). Therefore, the work presented here constitutes an important step forward for GCM studies of the aerosol indirect effect, which ignore any treatment of subgrid-scale variability of N_d.

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