Broadening of Cloud Droplet Spectra through Eddy Hopping: Turbulent Entraining Parcel Simulations

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ABSTRACT

This paper discusses the effects of cloud turbulence, turbulent entrainment, and entrained cloud condensation nuclei (CCN) activation on the evolution of the cloud droplet size spectrum. We simulate an ensemble of idealized turbulent cloud parcels that are subject to entrainment events modeled as a random process. Entrainment events, subsequent turbulent mixing inside the parcel, supersaturation fluctuations, and the resulting stochastic droplet activation and growth by condensation are simulated using a Monte Carlo scheme. Quantities characterizing the turbulence intensity, entrainment rate, CCN concentration, and the mean fraction of environmental air entrained in an event are all specified as independent external parameters. Cloud microphysics is described by applying Lagrangian particles, the so-called superdroplets. These are either unactivated CCN or cloud droplets that grow from activated CCN. The model accounts for the addition of environmental CCN into the cloud by entraining eddies at the cloud edge. Turbulent mixing of the entrained dry air with cloudy air is described using the classical linear relaxation to the mean model. We show that turbulence plays an important role in aiding entrained CCN to activate, and thus broadening the droplet size distribution. These findings are consistent with previous large-eddy simulations (LESs) that consider the impact of variable droplet growth histories on the droplet size spectra in small cumuli. The scheme developed in this work is ready to be used as a stochastic subgrid-scale scheme in LESs of natural clouds.

1. Introduction

Grabowski and Abade (2017, hereafter GA17) developed a simple approach to investigate droplet spectral broadening in turbulent clouds through a stochastic condensation mechanism referred to as eddy hopping. The mechanism, suggested about three decades ago by Cooper (1989), supposes that droplets arriving at a given location within a turbulent cloud follow different trajectories and that different supersaturation fluctuations along those trajectories lead to a broadening of the droplet size distribution. The supersaturation fluctuations come from turbulent eddies, with cloud droplets

embedded within those eddies growing in response to local supersaturation fluctuations. GA17 showed that incorporating stochastic effects of unresolved turbulent fluctuations in a simple adiabatic parcel model leads to a significant broadening, with the width of the droplet size distribution increasing from a few tenths of a micrometer for the turbulence-free adiabatic parcel to a few micrometers for the turbulent adiabatic parcel of 100-m horizontal extent and the turbulent kinetic energy (TKE) dissipation rates typical for shallow cumuli (say, between a few tens and a couple hundreds of squared centimeters per cubed seconds). Such an increase is consistent with observations of spectral width in undiluted or weakly diluted cloudy volumes in both cumulus (e.g., Jensen et al. 1985) and stratocumulus (e.g., Pawlowska et al. 2006) clouds.

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FIG. 1. The droplet size spectra predicted by a large ensemble of Lagrangian droplet trajectories driven by a small cumulus LES. (a) The droplet spectrum predicted by an adiabatic ascent from the cloud base. (b) The spectrum without activation of entrained CCN. (c), (d) Spectra with activation of entrained CCN, with (d) assuming a twice-as-large entrained CCN as in (c). The mean droplet diameter and distribution standard deviation are shown in each panel. [Reproduced with permission from Fig. 6 in Lasher-Trapp et al. (2005).]

Lasher-Trapp et al. (2005) investigated the eddy-hopping mechanism in realistic cumulus cloud simulations. They combined a 3D Eulerian large-eddy simulation (LES) cloud model with a Lagrangian trajectory model that included cloud condensation nuclei (CCN) activation and cloud droplet growth. The droplet distribution at a given location within a simulated cloud came from the superposition of distributions predicted by a large ensemble of back trajectories arriving at that location. Lasher-Trapp et al.'s Fig. 6 (reproduced as Fig. 1 here) illustrates key features of simulated droplet spectra. First, there is a peak that corresponds to cloud droplets activated at the cloud base and growing during their ascent inside the cloud. However, the peak is significantly wider than the one representing an adiabatic ascent through the cloud. Arguably, the spread comes at least

partially from the eddy-hopping mechanism discussed in GA17. There is also a significant tail (or shoulder) to the left of the main distribution peak that represents the contributions of smaller droplets. These arguably come from the entrainment of environmental air above the cloud base that leads to either evaporation of cloud droplets arriving from lower levels or activation of new cloud droplets on entrained CCN. As discussed by Lasher-Trapp et al. (2005), all these features of the simulated spectra agree with numerous previous observations of droplet spectra in shallow cumuli.

Although insightful, the Lasher-Trapp et al. (2005) methodology is cumbersome and difficult to apply routinely. GA17 argued that a similar investigation can be conducted in a straightforward way by applying the Lagrangian particle-based thermodynamics such as in

Andrejczuk et al. (2010), Riechermann et al. (2012), Arabas et al. (2015), Hoffmann et al. (2017), and Grabowski et al. (2018), and often referred to as the superdroplet method (Shima et al. 2009). However, to the authors' knowledge, previous applications of the Lagrangian methodology exclude effects of unresolved turbulence; that is, the motion of Lagrangian superdroplets is calculated by applying the flow-model-resolved air velocities, and droplet growth is predicted by applying only resolved thermodynamic variables. Beyond including unresolved fluctuations of velocities and thermodynamic variables as applied in GA17, the stochastic subgrid-scale scheme for a large-eddy simulation model needs to include the effects of entrainment and mixing. The current study fills this gap by expanding GA17's approach to the entraining parcel model and including the effects of turbulent entrainment and entrained CCN activation.

For simplicity, we use an approximated stochastic representation of cloud turbulence and entrainment. Our idealized approach is based on a stochastic Lagrangian model that mimics the general features of turbulence (assumed here to be homogeneous and isotropic). Full assessment of the model will be possible in the future, after applying it as a stochastic subgrid-scale scheme in realistic cloud simulations and comparing the outcome with observations.

We apply a particle-based representation of the microphysics using superdroplets. CCN activation and droplet growth are described explicitly as in Arabas et al. (2015), but here we extend their deterministic approach to a stochastic one. Collisions and coalescence of droplets are not considered.

The paper is organized as follows. Section 2 outlines the stochastic entraining parcel model framework. In section 3 we briefly describe the Köhler theory of droplet activation and condensational growth in the presence of a fluctuating supersaturation. Section 4 presents a Lagrangian model for the supersaturation fluctuation experienced by each cloud particle. Results are presented and discussed in section 5. Concluding remarks are given in section 6.

2. Stochastic entraining parcel model

We consider an air parcel of mass m rising with constant vertical velocity $\langle w \rangle$ and subjected to random entrainment events of the environmental air. The cloud parcel has a linear extent L in the inertial range of three-dimensional turbulence. The environmental temperature $T_e(z)$ and water vapor mixing ratio $q_{ve}(z)$ as a function of the vertical coordinate z are specified by assuming an idealized atmosphere with constant dry

stability β and given relative humidity (RH). The dry stability is defined as

$$\beta \equiv \frac{d \ln \theta_{\text{env}}}{dz}, \quad (1)$$

where $\theta_{\text{env}}(z)$ is the environmental potential temperature. The pressure $p(z)$ within the parcel is assumed to decrease with height z as in the hydrostatically balanced environment.

The time evolution of the temperature T and water vapor mixing ratio q_v of the parcel is governed by (Pruppacher and Klett 1997)

$$c_p \frac{dT}{dt} = -g \langle w \rangle - L_v \frac{dq_v}{dt} - \mu [L_v(q_v - q_{ve}) + c_p(T - T_e)] \quad (2)$$

and

$$\frac{dq_v}{dt} = -\frac{dq_\ell}{dt} - \mu(q_v - q_{ve} + q_\ell), \quad (3)$$

where g is the gravitational acceleration, L_v is the latent heat of vaporization, c_p is the specific heat of air at constant pressure, q_ℓ is the liquid water mixing ratio, and

$$\mu = \frac{1}{m} \frac{dm}{dt} \quad (4)$$

is the entrainment rate. This model assumes homogeneous mixing of the mean-field variables T and q_v inside the parcel. This affects only the mean-field supersaturation $\langle S \rangle$ in the parcel,

$$\langle S \rangle = \frac{e(q_v, p)}{e_s(T)} - 1, \quad (5)$$

where $e(q_v, p)$ is the partial water vapor pressure and $e_s(T)$ is a saturation value given by the Clausius–Clapeyron equation. Supersaturation fluctuations around $\langle S \rangle$ induced by turbulence and experienced by the cloud particles will be introduced later in a Lagrangian manner.

The rate of change of q_ℓ in Eq. (3) is determined by the diffusional growth of the cloud particles inside the parcel. The dispersed liquid phase in the parcel is described by applying Lagrangian particles, the so-called superdroplets [using the terminology introduced by Shima et al. (2009)]. Each superdroplet represents a multiple number of identical cloud particles (either hygroscopic unactivated aerosols or activated cloud droplets) that share the same attributes (such as wet radius and chemical composition) and are evenly distributed throughout the parcel volume (Shima et al. 2009). All real droplets belonging to each superdroplet are exposed to the same temperature,

partial vapor pressure, and hence the same supersaturation. In our Lagrangian treatment of supersaturation variability (to be described later), droplets in a superdroplet experience the same *fluctuations* about the mean-field supersaturation.

Entrainment events are modeled as a stochastic process (see, e.g., [Krueger et al. 1997](#); [Romps and Kuang 2010](#)). Here, we follow [Romps and Kuang \(2010\)](#) and describe the entrainment rate μ as a random variable that is zero except during delta-function bursts corresponding to entrainment events. The integral of μ over the time interval $[t, t + \delta t)$ is given by

$$\int_t^{t+\delta t} \mu(s) ds = XY, \quad (6)$$

that is, the product of two independent random processes X and Y . The former controls the occurrence of an entrainment event and has possible values $\{0, 1\}$ distributed with probabilities $P(X = 1) = \lambda^{-1} \langle w \rangle \delta t$ and $P(X = 0) = 1 - P(X = 1)$, where λ is the mean distance between entrainment events. Process Y controls the entrained mass in an entrainment event. It is assumed to follow an exponential distribution ([Romps and Kuang 2010](#)):

$$f(Y = y) = \frac{1}{\sigma} \exp(-y/\sigma), \quad (7)$$

where σ is the mean ratio of entrained mass to parcel mass in a single entrainment event.

At each entrainment event, entrained unsaturated air carries (deliquesced) hygroscopic CCN from the immediate environment into the parcel. A hygroscopic CCN is characterized by its dry radius r_d and hygroscopicity parameter κ ([Petters and Kreidenweis 2007](#)). Initially dry CCN particles absorb moisture from the air (deliquescence) before the formation of cloud droplets (activation). Subsequently, cloud droplets grow by water vapor diffusion. The processes of droplet activation and subsequent condensational growth are described using Köhler theory (see, e.g., [Pruppacher and Klett 1997](#)). In the next section we present the Köhler equation in a more convenient form for the analysis of droplet activation and growth in turbulent environments.

3. Köhler theory in the presence of supersaturation fluctuations

We assume the time evolution of the radius r of a single cloud particle exposed to supersaturation S is described by Köhler theory (see, e.g., [Pruppacher and Klett 1997](#)):

FIG. 2. (top) Equilibrium supersaturation S_{eq} as a function of the droplet radius r [Eq. (9)]. (bottom) Köhler potential $V_{(S)}$ [Eq. (13)] for $0 < \langle S \rangle < S_c$. The stochastic activation (SA) of haze droplets and stochastic deactivation (SDA) of cloud droplets are schematically shown.

$$r \frac{dr}{dt} = \frac{1}{2} \frac{dr^2}{dt} \approx D \left(S - \frac{C_\gamma}{r} + \frac{C_\kappa}{r^3} \right), \quad (8)$$

where $C_\gamma \approx 1.5 \times 10^{-3} \mu\text{m}$ is the surface tension coefficient (assumed to be a weak function of temperature and independent of the amount of solute) and $C_\kappa = \kappa r_d^3$ is the hygroscopicity coefficient, where r_d is the dry CCN radius and κ the hygroscopicity parameter [following the solute parameterization introduced by [Petters and Kreidenweis \(2007\)](#)]. An expression for the effective diffusion coefficient D is given in the [appendix](#). The approximated form of the growth equation [see Eq. (8)] is used for simplicity of the exposition. A more accurate account of surface tension and solute effects (described in the [appendix](#)) is used in the numerical calculations performed in this work.

A droplet of radius r is in equilibrium ($dr/dt = 0$) when the supersaturation is

$$S_{eq}(r) = \frac{C_\gamma}{r} - \frac{C_\kappa}{r^3}. \quad (9)$$

The curve for $S_{eq}(r)$, referred to as the Köhler curve, is depicted in the top panel of [Fig. 2](#). It has a maximum at the critical radius $r_c = (3C_\kappa/C_\gamma)^{1/2}$ corresponding to the critical supersaturation $S_c = S_{eq}(r_c)$. The critical point (r_c, S_c) depends solely on the CCN parameters (r_d, κ) .

The Köhler curve, as it refers to equilibrium, is very useful in the study of CCN activation when the supersaturation S surrounding each cloud particle varies slowly, that is, at time scales that are long compared

to the relaxation times scales arising from the growth equation [Eq. (8)].¹ In this classical picture (e.g., Rogers and Yau 1989), a CCN is said to be activated when the radius r of the droplet formed on it exceeds the critical value r_c . Once $r > r_c$, the droplet will persist in growing as long as $S > S_{eq}(r)$. The latter condition is easily fulfilled in deterministic adiabatic parcel models, since the decay (due to condensation) of the ambient supersaturation S is slow compared with the fast growth rate of freshly activated droplets.

While accounting here for rapidly changing local supersaturation in a turbulent cloud, we abandon the classical (deterministic) notion of activation. Instead, we separate cloud particles into two statistical populations: interstitial aerosols (or haze droplets), having $r < r_c$, and cloud droplets, having $r > r_c$. Then on the microphysical level, droplets with $r \approx r_c$ in a turbulent cloud may fluctuate between the haze and cloud droplets populations. For convenience we still use an equilibrium parameter, the critical radius r_c , to distinguish the cloud particle populations. Here, we use the term *stochastic activation* of a droplet to designate its transition from a haze to a cloud droplet population driven by turbulent fluctuations in the supersaturation field. Consequently, *stochastic deactivation* refers to the back transition of cloud droplets to the haze population.

In a turbulent environment, the supersaturation S experienced by individual cloud particles may be separated into mean $\langle S \rangle$ and fluctuating S' parts:

$$S = \langle S \rangle + S'. \quad (10)$$

In an LES setting, the mean part $\langle S \rangle$ stands for the resolved or mean-field supersaturation in each grid box and S' for the unresolved fluctuation at subgrid scales.

Defining $x \equiv r^2$ and using the decomposition equation [Eq. (10)], Eq. (8) may be recast into the form

$$\frac{dx}{dt} = -\frac{\partial V_{\langle S \rangle}}{\partial x} + 2DS', \quad (11)$$

where we have defined

$$-\frac{\partial V_{\langle S \rangle}}{\partial x} \equiv 2D \left(-\frac{C_\gamma}{x^{1/2}} + \frac{C_\kappa}{x^{3/2}} + \langle S \rangle \right) \quad (12)$$

as the gradient of the $\langle S \rangle$ -dependent potential function:

$$\frac{1}{4D} V_{\langle S \rangle}(x) = C_\gamma x^{1/2} + \frac{C_\kappa}{x^{1/2}} - \frac{1}{2} \langle S \rangle x. \quad (13)$$

We henceforth refer to $V_{\langle S \rangle}$ as the mean-field Köhler potential. The definition (12) in terms of the resolved (mean field) supersaturation $\langle S \rangle$ is arbitrary, but very convenient for the discussion that follows.²

If we assume that $\langle S \rangle$ is a slow variable compared to S' , then Eq. (11) describes the fluctuating position x of an inertialess “Brownian” particle, moving in an almost constant potential field $V_{\langle S \rangle}(x)$ and acted upon by a randomly fluctuating force proportional to S' . This analogy shall help us to rationalize the role of turbulence [already explored in earlier works (e.g., Zapadinsky et al. 1995; Kulmala et al. 1997; Celani et al. 2008, 2009)] in enabling cloud particles to grow beyond their critical radius in a volume that is unsaturated on average.

The bottom panel of Fig. 2 shows the Köhler potential $V_{\langle S \rangle}$ for the particular case where $0 < \langle S \rangle < S_c$. For $\langle S \rangle$ in this saturated but subcritical range, the mean-field potential exhibits two equilibrium radii: r_1 and r_2 . The former (r_1) corresponds to a stable equilibrium (at the minimum of $V_{\langle S \rangle}$), while r_2 corresponds to an unstable one (at the local maximum of $V_{\langle S \rangle}$). In the absence of fluctuations, an interstitial aerosol particle (whose radius is confined in the potential well of $V_{\langle S \rangle}$) cannot be activated in the classical sense, unless $\langle S \rangle$ increases beyond S_c . In a turbulent environment supersaturation fluctuations S' may aid an interstitial aerosol in climbing the barrier of the mean-field potential $V_{\langle S \rangle}$ and grow beyond r_c (i.e., to *stochastically activate*). A cloud droplet whose radius has just crossed r_c is continuously experiencing supersaturation fluctuations. Such a fresh cloud droplet may grow further if it continues to experience favorable fluctuations S' . Otherwise, if it decorrelates from favorable S' , then its radius may drop back into the potential well (i.e., *stochastically deactivate*). Stochastic activation and deactivation are schematically shown in the bottom panel of Fig. 2.

The mean-field supersaturation $\langle S \rangle$, and consequently its associated potential “landscape” $V_{\langle S \rangle}$, may be varied either by external influences (e.g., parcel updraft and entrainment of unsaturated air) or by the thermodynamic feedback of stochastic condensation/evaporation inside the parcel. The shape of $V_{\langle S \rangle}$ for different values of $\langle S \rangle$ is shown in the bottom panel of Fig. 3. As $\langle S \rangle$ increases

¹ Examples of particle relaxation time scales include the time for haze droplets to shrink or grow to their equilibrium radius and the so-called activation time (Hoffmann 2016; Arabas and Shima 2017), that is, the time needed for a particle to grow beyond its critical radius.

² An alternative picture is provided by Celani et al. (2008). Their Köhler potential is defined in terms of the full supersaturation S experienced by the droplets (i.e., S is not decomposed into mean and fluctuating parts).

FIG. 3. (top) Köhler curve [Eq. (9)]. (bottom) Köhler potential $V_{(S)}$ for different values of the mean-field supersaturation $\langle S \rangle$ (indicated by dashed lines of different colors in the top panel). Dots in both panels show the equilibrium point(s), if any, corresponding to each value of $\langle S \rangle$.

(e.g., due to parcel updraft), the mean-field potential barrier becomes less and less steep and, eventually, disappears (as does the potential well) when $\langle S \rangle$ reaches S_c (red line in Fig. 3). On the other hand, entrainment of unsaturated air (and also the depletion of water vapor due to condensation) reduces $\langle S \rangle$, deepening the mean-field potential well.

Figure 3 also displays the equilibrium point(s), if any, of the mean-field potential corresponding to the constant- $\langle S \rangle$ lines intersecting the Köhler curve. For $\langle S \rangle = S_c$ (red line in Fig. 3), there is a single equilibrium point, which is half stable. For $\langle S \rangle < S_c$ (light blue line), the potential has an infinitely high barrier and a single (stable) equilibrium point. For $\langle S \rangle > S_c$ (dark blue line), there are no equilibrium points. This picture is equivalent to the fixed-point analysis presented by Arabas and Shima (2017) using the terminology of nonlinear dynamics. Their analysis gives insights into the deterministic CCN activation–deactivation in response to fast changes in $\langle S \rangle$ (i.e., when $\langle S \rangle$ varies on a time scale comparable to the activation time). In our stochastic approach, the resolved supersaturation $\langle S \rangle$ sets a background droplet growth environment where the supersaturation fluctuations operate.

To compute the stochastic growth by condensation and transitions between the two droplet populations as described above, we need to model the unresolved supersaturation fluctuations S' . In this work we apply the simple Lagrangian model for S' proposed by GA17. Their model is described in more detail in the next section.

4. Model for supersaturation fluctuations

As in GA17, we assume that the vertical velocity w and the supersaturation S experienced by each cloud particle (either interstitial aerosol or activated cloud droplet) are fluctuating quantities:

$$w = \langle w \rangle + w', \quad S = \langle S \rangle + S', \quad (14)$$

where w' and S' denote fluctuations around the mean values, $\langle w \rangle$ and $\langle S \rangle$, respectively. Here, $\langle w \rangle$ is a fixed external parameter and $\langle S \rangle$ is the resolved supersaturation, diagnosed through Eq. (5) from the predicted temperature T and water vapor mixing ratio q_v inside the parcel.

The supersaturation fluctuation S' is linked to the vertical velocity fluctuation w' via the so-called Lagrangian Twomey model (Twomey 1959) used in other approaches (see, e.g., Celani et al. 2008, 2009; Sardina et al. 2015, 2018; Grabowski and Abade 2017). This yields an evolution equation for the supersaturation fluctuation:

$$\frac{dS'}{dt} = -\frac{S'}{\tau_c} - \frac{S'}{\tau_m} + aw'(t), \quad (15)$$

where

$$a = \frac{L_v g}{R_v T^2 c_p} - \frac{g}{RT}, \quad (16)$$

τ_c is the phase relaxation time scale given by

$$\tau_c = (4\pi\rho_w DN\langle r \rangle C)^{-1}, \quad (17)$$

N is the droplet number concentration, $\langle r \rangle$ is mean droplet radius, and

$$C = \frac{1}{q_v} + \frac{L_v^2}{R_v T^2 c_p}. \quad (18)$$

The second term on the rhs of Eq. (15) mimics the relaxation of S' to zero due to turbulent mixing with time scale τ_m (Pope 2000; Paoli and Shariff 2009). This is the classical relaxation to the mean model (Pope 2000; Paoli and Shariff 2009).

Equation (15) may be cast in a more compact form:

$$\frac{dS'}{dt} = -\frac{S'}{\tau_S} + aw'(t), \quad (19)$$

where

$$\tau_S = \left(\frac{1}{\tau_c} + \frac{1}{\tau_m} \right)^{-1} \quad (20)$$

is the system relaxation time, the harmonic mean of the condensation and mixing time scales. We assume the time scale τ_m to be equal to the eddy turnover time τ_t (Pope 2000), that is, the time needed for homogenization of the supersaturation field by turbulent diffusion over the length scale L of the cloud parcel.

In this stochastic Lagrangian description, the state vector of each cloud particle includes the random variables $x = r^2$, S' , and w' . Variables x and S' evolve according to Eqs. (11) and (19), respectively. To close this model, the statistical properties of $w'(t)$ need to be specified.

We assume the vertical velocity fluctuation w' of each cloud particle is a random stationary process known as the Ornstein–Uhlenbeck (OU) process (e.g., Pope 2000). It is characterized by

$$\langle w'(t) \rangle = 0, \quad (21)$$

and autocorrelation function

$$\langle w'(t)w'(t') \rangle = \sigma_{w'}^2 \exp(-|t - t'|/\tau_t), \quad (22)$$

where $\sigma_{w'}^2$ is the variance of w' and τ_t is the turbulent correlation time assumed equal to the eddy turnover time above. Before moving forward, we would like to comment on the white-noise model for the turbulent velocity fluctuations used in Field et al. (2014) and Furtado et al. (2016). This simplification leads to a reduced description that retains only the variables (x, S') in the cloud particle state vector. We believe that the white-noise model applied in those studies is inappropriate here. This is because the characteristic time scale τ_t for the turbulent decorrelation of w' is not negligible compared to other relevant time scales of the problem (in particular, to the short time scales involved in our detailed description of stochastic droplet activation).

The variance $\sigma_{w'}^2$ in Eq. (22) is determined by equipartition of the turbulent kinetic energy E ,

$$E = \frac{3}{2} \sigma_{w'}^2, \quad (23)$$

under the assumption of isotropic turbulence. Based on the observations, turbulence in atmospheric clouds appears surprisingly close to homogeneous and isotropic down to the scale of a few tens of centimeters (see, e.g., Siebert et al. 2006). The energy E and the turbulent correlation time τ_t may be obtained from L and ε by assuming Kolmogorov's inertial-range scaling as in GA17,

$$E = C_E (L\varepsilon)^{2/3}, \quad (24)$$

with constant $C_E \approx 0.89$ (Schumann 1991), and

$$\tau_t = C_\tau \frac{L^{2/3}}{\varepsilon^{1/3}}, \quad (25)$$

with constant $C_\tau \approx 0.63$ as in Lasher-Trapp et al. (2005).

The parcel extent L increases with time due to the expansion and entrainment of environmental air. Then, E and τ_t increase accordingly. We assume a constant dissipation rate ε along the parcel updraft.

The stochastic differential equation corresponding to the OU process [Eqs. (21) and (22)] is

$$dw' = -\frac{w'}{\tau_t} dt + \sqrt{\frac{2\sigma_{w'}^2}{\tau_t}} dW, \quad (26)$$

where dW is an increment of the Wiener process. In a discrete setting $dW = \psi\sqrt{\delta t}$, where ψ is a random number drawn from the standard Gaussian distribution and δt is the integration time step.

The model described above is an approximation. It reproduces only general features of the turbulent velocity and supersaturation fluctuations experienced by Lagrangian cloud droplets. Supersaturation fluctuations are coupled with the turbulence-induced vertical velocity fluctuations of the droplet-carrying air. This mimics correlations between the droplet trajectories and the supersaturation experienced by them.

There are several more elaborate approaches (see, e.g., Pope 2011) that retrieve intricate aspects of turbulence, such as intermittency. We do not apply those methods here for the sake of simplicity. Investigation into whether more involved schemes definitely improve our model and affect the conclusions derived from it is left for future work.

In Lagrangian simulations using superdroplets, the quantity S must be understood as the supersaturation $S_k = \langle S \rangle + S'_k$ surrounding all real droplets of the k th superdroplet. The same holds true for x and w' , so that the dynamic state vector of the k th superdroplet is (x_k, S'_k, w'_k) . The coupled set of Eqs. (26) and (19) for the jointly distributed random variables w'_k and S'_k is solved numerically using the Euler–Maruyama first-order scheme (Kloeden and Platen 1992). This scheme is sufficiently accurate for the very small time steps used in this work ($\delta t \ll \min\{\tau_S, \tau_t\}$). The stochastic solution for S'_k , together with the mean value $\langle S \rangle$, is used in the (implicit Euler) integration of Eq. (11) for the condensational growth of each superdroplet k . As in GA17, the phase relaxation time of the droplet population [Eq. (17)] is calculated, including all superdroplets present in the parcel, because the superdroplets and real droplets that each superdroplet represents are assumed to be well mixed in the parcel volume.

TABLE 1. Characteristic values of the model parameters.

$\langle w \rangle$	L	ε	λ	σ	κ	N_{CCN}	RH	β
1 m s^{-1}	50 m	$100 \text{ cm}^2 \text{ s}^{-3}$	200 m	0.1	0.61	100 cm^{-3}	80%	10^{-5} m^{-1}

Entrainment injects deliquesced CCN into the parcel. Their state vector is initialized as follows. Supersaturation fluctuations are set as $S' = S_{\text{env}} - \langle S \rangle$, where $S_{\text{env}} = \text{RH} - 1$ denotes the supersaturation of the environment.³ These fluctuations relax due to turbulent mixing and droplet growth or evaporation according to Eq. (19). The radius r of an entrained haze droplet is set equal to the equilibrium radius corresponding to S_{env} and its CCN characteristics (r_d , κ). Initial velocity fluctuations w' of entrained particles are drawn from a Gaussian distribution with zero mean and variance σ_w^2 .

5. Simulations and results

a. Simulation parameters

The external parameters of our turbulent entraining parcel model are 1) the mean updraft velocity $\langle w \rangle$, 2) the parcel linear size L , 3) the dissipation rate ε of the turbulent kinetic energy, 4) the distance between entrainment events λ , 5) the mass fraction σ of entrained air (relative to the parcel mass), 6) the effective aerosol hygroscopicity parameter κ , and 7) the number concentration N of available CCN, 8) the relative humidity (RH) of the environment, and 9) the stability of the environmental atmosphere β . The pair (L, ε) controls the cloud turbulence intensity. The pair (λ, σ) controls the entrainment rate μ . The specific values of these parameters used in the present simulations are listed in Table 1. We performed a large set of simulations spanning a wide range of those parameters, typically varying one of them while keeping the rest unchanged, but our focus here is on only the salient consequences of supersaturation variability due to entrainment and turbulent mixing. Sensitivity simulations are briefly described in section 5d. It is worth noting that turbulence and entrainment parameters are decoupled in the present study. In future applications of the supersaturation fluctuation model as a subgrid-scale (SGS) microphysical scheme in LES simulations, entrainment and turbulence parameters will be provided by the resolved hydrodynamic flow field.

³ In the present cloud parcel model, the histories of the *entrained* superdroplets are assumed to develop entirely in the unsaturated environment. Thus, the supersaturations experienced by the entrained superdroplets have a small dispersion relative to S_{env} .

We assume the CCN dry radii r_d are distributed according to a unimodal lognormal distribution. The concentration, mean radius, and geometric standard deviation (unitless) are 100 cm^{-3} , 75 nm, and 1.6, respectively. Because extremely small time steps are necessary to resolve the stochastic activation of tiny CCNs [small r_d renders Eq. (8) stiff], we use a lower cutoff: $r_{d,\text{min}} = 0.05 \mu\text{m}$.

All simulations are initialized with a temperature of 288.16 K, a pressure of 900 hPa, and a relative humidity of 99% within the parcel. In entraining parcel simulations we use 20 superdroplets per 10^3 kg of air. Typically, simulations start with approximately 2700 superdroplets and finish with approximately 4500 superdroplets on average. The number of superdroplets increases as a result of the entrainment of unsaturated air, which carries environmental aerosols (and hence additional superdroplets) into the parcel. Table 1 lists the initial value of L (which increases with time due to expansion and entrainment). For the typical values of the simulation parameters listed in Table 1, L increases by 20% after a 1-km parcel rise. Equations (2) and (3) for T and q_v are numerically integrated using an Euler forward scheme with a time step $\delta t = 0.1 \text{ s}$. Results presented below come from averaging 1000 independent realizations of the stochastic parcel model calculations.

b. Spectrum broadening

In this section we shall examine the normalized droplet size distribution or probability distribution function (PDF) from different variants of the parcel model. Figure 4 shows the PDFs for the turbulent entraining parcel model in comparison with more simplified models: adiabatic nonturbulent ($\mu = \varepsilon = 0$), adiabatic turbulent ($\mu = 0, \varepsilon \neq 0$), and entraining nonturbulent ($\mu \neq 0, \varepsilon = 0$). Figure 4a depicts the PDF sampled over the population of cloud droplets (i.e., having $r > r_c$) after 1-km parcel ascent for each parcel model. The highly peaked spectrum corresponding to the adiabatic nonturbulent parcel contrasts with broader distributions predicted by other models. Turbulence-induced supersaturation fluctuations produce broadening toward both small and large sizes, as previously shown by GA17 in adiabatic parcel simulation. Entrainment produces broadening of the droplet size distribution toward smaller sizes due to the constant insertion of environmental CCN. The spectra shown in Fig. 4a resemble those obtained by Lasher-Trapp et al. (2005) as shown in Fig. 1.

FIG. 4. Droplet size distribution after a 1-km parcel rise for different parcel models: (a) size spectrum of the cloud droplets population ($r > r_c$) and (b) size spectrum of the interstitial aerosol and cloud droplet population (here the PDF for the highly idealized adiabatic non-turbulent parcel is not shown).

Figure 4b depicts the PDF sampled over all (haze and cloud) droplets applying a logarithmic scale of the vertical axis. It shows more clearly the presence of two populations (haze and cloud droplets) in the entraining parcels. The peaks on the left correspond to the radii of unactivated haze droplets. A nonturbulent entraining parcel exhibits more pronounced haze peaks (compared with turbulent ones) because of the lack of fluctuations that drive stochastic activation in subcritical mean-field supersaturation. Figure 4b also reveals that the PDF for an adiabatic turbulent parcel has an exponential tail toward smaller sizes.

The evolution of mean radius $\langle r \rangle$ and radius standard deviation σ_r (referred to as the spectral width) is shown in Fig. 5. The spectral width of nonturbulent parcels

shows a small bump at the beginning due to the activation of CCN with different critical parameters r_c and S_c (set by the specified lognormal distribution for r_d). In nonturbulent adiabatic parcels, this bump in σ_r is followed by a continuous narrowing of the spectrum as the droplets grow. As already observed by GA17, turbulent adiabatic parcels have significantly broader spectra than their nonturbulent counterparts. The time evolution of σ_r in a rising turbulent adiabatic parcel results from the interplay between deterministic spectral narrowing (driven by $\langle S \rangle$) and stochastic spectral broadening (induced by fluctuations S'). The spectral width of entraining parcels (either turbulent or nonturbulent) increases with time due to the permanent insertion of new CCN, which broadens the spectrum toward smaller sizes. Entraining parcels

FIG. 5. Moments of the cloud droplet size distribution for different parcel models. The evolutions of (a) the mean radius $\langle r \rangle$ and (b) the radius standard deviation σ_r are shown.

FIG. 6. Evolution of mean-field supersaturation $\langle S \rangle$ and the fraction N_c/N_{CCN} of cloud droplets (with radii $r > r_c$) for different parcel models: (a),(b) turbulent and nonturbulent adiabatic parcels and (c),(d) turbulent and nonturbulent entraining parcels. In (a) and (c) the shading around the solid lines (corresponding to $\langle S \rangle$ for turbulent parcels) has a width of twice the standard deviation of the supersaturation experienced by all superdroplets.

with turbulent fluctuations exhibit larger spectral widths due to stochastic activation, which causes broadening also toward larger sizes. As shown in Fig. 5a, the droplet mean radius increases in a steady parcel rise with turbulent parcels having smaller mean radii than their nonturbulent counterparts. This comes from the turbulence effects, which drive the stochastic activation of cloud droplets. We shall discuss them in the next section.

c. Effect of turbulent fluctuations on activation

Turbulence affects the evolution of the mean-field supersaturation $\langle S \rangle$ and the fraction N_c/N_{CCN} of activated cloud droplets N_c with respect to CCN N_{CCN} as shown in Fig. 6. In the particular case presented in Fig. 6 of a nonturbulent adiabatic parcel ascending with $\langle w \rangle = 1 \text{ m s}^{-1}$, all available CCNs at a concentration of 100 cm^{-3} are activated in a short time (i.e., the fraction N_c/N_{CCN} grows sharply). This is not the case of a turbulent adiabatic parcel, as displayed in Fig. 6b.

In the absence of turbulence, interstitial aerosols cross their critical radii only if the mean-field supersaturation $\langle S \rangle$ exceeds the critical value S_c . In a turbulent parcel, supersaturation fluctuations may aid aerosols to stochastically activate even under subcritical mean-field supersaturations (i.e., $\langle S \rangle < S_c$). Those “lucky” activated

droplets start to grow and deplete water vapor, hindering the (updraft driven) increase of $\langle S \rangle$. Turbulent parcels then have less pronounced peaks in $\langle S \rangle$ and leave “unlucky” aerosols unactivated. This is because the feedback on the vapor of the stochastically activated droplets causes the remaining haze droplets to face a steeper slope of the mean-field potential barrier.

The time evolution of $\langle S \rangle$ for a turbulent adiabatic parcel shown in Fig. 6a is in contrast with that obtained in GA17, where the $\langle S \rangle$ -based Twomey activation scheme was used in both turbulent and nonturbulent adiabatic parcels (see Figs. 2 and 3 in GA17). In the Twomey activation, the mean-field supersaturation $\langle S \rangle$ dictates the concentration of cloud droplets that needs to be present inside the cloud parcel. Therefore, Twomey activation (as implemented in GA17) produces the same time evolution for the concentration of activated cloud droplets in both the turbulent and nonturbulent parcels. In contrast, $\langle S \rangle + S'$ drives CCN activation in the case of adiabatic turbulent parcel simulations reported here. As a result, different concentrations of cloud droplets are activated, and consequently different magnitudes of the thermodynamic feedback on the mean supersaturation exist. Apparently, the fluctuations impact the overall CCN activation by reducing the peak supersaturation (Fig. 6a)

FIG. 7. Droplet size spectra (sampled over the cloud droplet population) after a 1-km parcel ascent for different number concentrations N_{CCN} of available CCN (as indicated).

and extending the activation significantly in time (Fig. 6b). With entrainment (cf. Figs. 6c,d) both the fraction of activated droplets and the quasi-equilibrium value of $\langle S \rangle$ is reduced. Droplets in turbulent entraining parcels experience larger supersaturation fluctuations and arguably larger swings of the supersaturation that enhance the stochastic activation and deactivation.

d. Sensitivity simulations

In this section we shall examine the sensitivity of the cloud droplet spectra to some of the model parameters

listed in section 5a. Henceforth we focus on the more elaborate turbulent entraining parcel model. Also we show only the PDF sampled over the population of cloud droplets (since this population is the relevant one for the stage of collision-coalescence in the cloud development).

Figure 7 shows how the cloud droplet size distribution (after 1-km parcel rise) is affected by the concentration N_{CCN} of available hygroscopic CCN. The results display a significant narrowing of the droplet size spectrum as the CCN concentration is increased (polluted conditions).

FIG. 8. Sensitivity of the droplet size distribution (after a 1-km parcel rise) to different simulation parameters: (a) TKE dissipation rate ε for fixed $\tau_t \approx 40$ s, (b) TKE dissipation rate ε for fixed $L \approx 50$ m, and (c) mean updraft velocity $\langle w \rangle$.

FIG. 9. Sensitivity of the droplet size distribution (after a 1-km parcel rise) to different entrainment-related parameters: (a) distance between entrainment events λ , (b) mass fraction σ of entrained air relative to the parcel mass, and (c) relative humidity (RH) of the environment.

This is the so-called (turbulence induced) aerosol indirect effect, already explored by Chandrakar et al. (2016). Keeping the turbulence intensity and autocorrelation time τ_t fixed, an increase in droplet concentration reduces the condensation relaxation time τ_c . It decreases the system relaxation time τ_S [Eq. (20)] as well, since the latter emphasizes the faster of the two time scales, τ_c and τ_t . In this regime of fast microphysics, supersaturation fluctuations are rapidly damped due to droplet growth and evaporation.

Figures 8a and 8b show the effects of the TKE dissipation rate ε on the droplet size distribution. In Fig. 8a, ε is varied by keeping the eddy turn over time τ_t fixed (then the system size L has to be varied accordingly). An increase in ε produces larger vertical velocity fluctuations σ_w^2 , leading to stronger fluctuations of the supersaturation. This intensifies the spectral broadening, as observed by Celani et al. (2008). In Fig. 8b, ε is varied with L fixed (τ_t changes accordingly). As before, an increase in ε intensifies the spectral broadening due to larger supersaturation fluctuations. However, as ε increases, it simultaneously reduces the eddy turnover time τ_t , resulting in faster damping of supersaturation fluctuations by turbulent mixing. The interplay of those

mechanisms leads to the weak dependence on the TKE dissipation rate ε when L is fixed, as shown in Fig. 8b. Figure 8c shows that larger values of updraft velocity $\langle w \rangle$ lead to narrower droplet spectra. Stronger updrafts result in larger quasi-equilibrium values of $\langle S \rangle$, allowing the (deterministic) spectral narrowing to prevail over the (stochastic) spectral broadening.

The effect of entrainment-related parameters on the PDF for the droplets' radii is shown in Fig. 9. Decreasing λ (entrainment events occur more frequently), increasing σ (more unsaturated air is engulfed in an entrainment event), and decreasing RH (entrained air is drier) make the entrainment stronger, resulting in lower quasi-equilibrium values of $\langle S \rangle$. This slows down the droplet growth and restrains the stochastic activation of entrained CCN, since they face a more severe mean-field potential landscape $V_{\langle S \rangle}$ (i.e., with steeper barriers). As a result, the center of the droplet radius PDF moves toward smaller sizes.

6. Conclusions

In this work we have expanded the GA17 approach to include the effects of turbulent entrainment and

entrained CCN activation on droplet spectral broadening. The scheme developed here is ready to be used as a stochastic subgrid-scale (SGS) model of the microphysics in LES of natural clouds. The stochastic model for SGS supersaturation fluctuations requires two external parameters: the SGS turbulent kinetic energy (TKE) and its dissipation rate ε . These quantities are provided (for each grid box of the computational domain) by existing LES schemes that model SGS turbulence (see, e.g., Heus et al. 2010; Riechermann et al. 2012). Also, when Lagrangian stochastic models are coupled with LES, one assumes that ensemble-averaged stochastic quantities can be replaced by their LES-resolved counterparts.

Köhler theory was presented in a form convenient for the study of droplet stochastic activation/deactivation and growth in the presence of turbulence- or entrainment-induced supersaturation fluctuations. This approach provided the framework in which the droplets' radii execute a "random walk" on the mean-field potential landscape $V_{(S)}$ specified by the average supersaturation $\langle S \rangle$. The effects of different model parameters (that control, for example, the entrainment rate, CCN concentration, and turbulent intensity) on the droplet spectrum have been investigated and their influence on the mean-field potential and/or strength of supersaturation fluctuations was explored.

Our parcel simulations have shown the interplay between fluctuating and mean-field thermodynamics. An example is the feedback on $\langle S \rangle$ of the stochastic activation of droplets under subcritical conditions ($\langle S \rangle < S_c$). It is shown that the thermodynamic feedback inside adiabatic and turbulent parcels buffers the increase of the mean-field supersaturation $\langle S \rangle$ driven by cloud updrafts or other external forcings. This can significantly extend the distance over which droplets are activated inside the parcel. A similar mechanism also operates inside the entraining and turbulent parcel, although with significantly larger supersaturation fluctuations. Incorporation of the SGS scheme presented here into a realistic LES cloud model is a subject of ongoing work and will be reported upon in subsequent publications. We note that the present study uses a simple and idealized approach that retrieves only general features of turbulence and entrainment. A comprehensive validation of the conclusions derived from our simple model will be undertaken in the future, after applying the scheme to realistic cloud simulations.

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APPENDIX

The κ -Köhler Theory

For the present calculations, we assume the radius r of a single cloud particle exposed to supersaturation S evolves according to (see, e.g., Pruppacher and Klett 1997)

$$r \frac{dr}{dt} = D \left[1 - a_w(r) \exp\left(\frac{C_\gamma}{r}\right) + S \right], \quad (\text{A1})$$

where C_γ is the surface tension coefficient and $0 \leq a_w \leq 1$ is the water activity in solution. We use the parameterization for a_w introduced by Petters and Kreidenweis (2007):

$$a_w(r) = \frac{r^3 - r_d^3}{r^3 - r_d^3(1 - \kappa)}, \quad (\text{A2})$$

where the solute is characterized by its dry radius r_d and hygroscopicity parameter κ .

The approximated growth equation [Eq. (8)] is valid when $r \gg r_d$, so that

$$a_w(r) \exp\left(\frac{C_\gamma}{r}\right) \approx 1 + \frac{C_\gamma}{r} - \frac{C_\kappa}{r^3}, \quad (\text{A3})$$

with $C_\kappa = \kappa r_d^3$.

The effective diffusion coefficient is

$$D = \left[\left(\frac{L_v}{R_v T} - 1 \right) \frac{L_v \rho_w}{\beta_K(r) K_0 T} + \frac{\rho_w R_v T}{\beta_D(r) D_0 e_s(T)} \right]^{-1}, \quad (\text{A4})$$

where D_0 is the diffusion coefficient of water vapor in air, K_0 is the heat conductivity of the air, ρ_w is the liquid water density, R_v is the vapor gas constant, and $e_s(T)$ is the saturation partial vapor pressure given by the Clausius–Clapeyron equation. Molecular kinetic effects are accounted for by the r -dependent factors $\beta_K(r)$ and $\beta_D(r)$. They are calculated using the expressions derived by Fuchs and Sutugin (1970) in the form suggested by Laaksonen et al. (2005).

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