Impact of Cloud-Base Turbulence on CCN Activation: Single-Size CCN

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ABSTRACT: This paper examines the impact of cloud-base turbulence on activation of cloud condensation nuclei (CCN). Following our previous studies, we contrast activation within a nonturbulent adiabatic parcel and an adiabatic parcel filled with turbulence. The latter is simulated by applying a forced implicit large-eddy simulation within a triply periodic computational domain of 64³ m³. We consider two monodisperse CCN. Small CCN have a dry radius of 0.01 μm and a corresponding activation (critical) radius and critical supersaturation of 0.6 μm and 1.3%, respectively. Large CCN have a dry radius of 0.2 μm and feature activation radius of 5.4 μm and critical supersaturation 0.15%. CCN are assumed in 200-cm⁻³ concentration in all cases. Mean cloud-base updraft velocities of 0.33, 1, and 3 m s⁻¹ are considered. In the nonturbulent parcel, all CCN are activated and lead to a monodisperse droplet size distribution above the cloud base, with practically the same droplet size in all simulations. In contrast, turbulence can lead to activation of only a fraction of all CCN with a nonzero spectral width above the cloud base, of the order of 1 μm, especially in the case of small CCN and weak mean cloud-base ascent. We compare our results to studies of the turbulent single-size CCN activation in the Pi chamber. Sensitivity simulations that apply a smaller turbulence intensity, smaller computational domain, and modified initial conditions document the impact of specific modeling assumptions. The simulations call for a more realistic high-resolution modeling of turbulent cloud-base activation.

KEYWORDS: Aerosols; Aerosol-cloud interaction; Cloud droplets; Condensation

1. Introduction

Formation of cloud droplets on soluble aerosols referred to as the cloud condensation nuclei (CCN) can be understood applying equilibrium thermodynamics theory developed close to a century ago by Hilding Köhler, a Swedish meteorologist from the University of Uppsala (Köhler 1936). The key part of this theory is the so-called Köhler curve that shows the equilibrium water vapor pressure as a function of the wet droplet radius. The Köhler curve combines the change in the equilibrium vapor pressure over a curved droplet surface, the Kelvin effect, with the impact of the dissolved salt that lowers the equilibrium vapor pressure following Raoult’s law. The shape of the curve changes with the dry CCN mass and its chemical composition, together with the environmental temperature and pressure. The peak of the Köhler curve separates unactivated CCN that are in a stable equilibrium with their supersaturated environment from activated CCN that grow rapidly as cloud droplets. The peak defines the activation radius and the activation (critical) water vapor pressure, the latter typically converted to the supersaturation Σ (Σ = e/eᵦ – 1, where e and eᵦ are the water vapor pressure and its saturated value, respectively). The critical supersaturation decreases, and the activation radius increases, with the CCN dry mass. This is illustrated in Fig. 1 for two different salts that are commonly seen as natural CCN: sodium chloride and ammonium sulfate.

As Fig. 1 shows, the critical supersaturation is large for CCN below 10⁻² μm (10 nm), several percent. Such CCN are unlikely to get activated at a cloud base of a natural cloud with a cloud-base updraft ~1 m s⁻¹ and typical CCN concentrations because the maximum supersaturation near the cloud base in such conditions is typically below 1% (Pruppacher and Klett 1997; Reutter et al. 2009; Grabowski et al. 2011; Pinsky et al. 2013). For CCN larger than 1 μm, referred to as the giant and ultra-giant CCN, the critical supersaturation is miniscule (below 0.01%), but the activation radius is large, over 10 μm. Such CCN may never reach their activation radius, they simply grow or evaporate following changes of the in-cloud supersaturation.

Natural CCN typically occur in different sizes and often in different chemical compositions as documented in numerous field projects (e.g., Jiusto 1967; Mészáros 1968; Fitzgerald 1973; Hoppel 1979; Hudson and Yum 2002; Miao et al. 2015). Cloud-base activation of such CCN can be studied by considering an adiabatic parcel rising across the subcloud layer and crossing the cloud base (e.g., Reutter et al. 2009 and references therein; Grabowski et al. 2011, among many others). As the parcel rises, the relative humidity increases within the parcel because the parcel temperature decreases following the dry-adiabatic lapse rate. CCN grow by absorbing water vapor as they try to keep up with the increasing relative humidity. This process is referred to as the deliquescence. Small CCN follow increasing relative humidity well, but large ones (especially giant and ultra-giant) may lag behind, that is, their size can be smaller than the quasi-equilibrium size for the relative humidity at a given height. CCN activation takes place when the parcel passes critical supersaturation for a given dry CCN.

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size. With the increasing supersaturation, progressively smaller CCN become activated. Eventually, latent heat release and water vapor absorption by growing cloud droplets slows down and eventually reverses the supersaturation increase. In this picture, the supersaturation peak near the cloud base determines the smallest CCN that can be activated from the CCN distribution, and it separates growing cloud droplets from small unactivated (interstitial or haze) CCN. Since different CCN are activated at different heights, the spectrum of cloud droplets just above the cloud base has a finite width, typically a few tenths of a micrometer [see Figs. 4 and 5 in Grabowski et al. (2011) that consider activation of CCN observed in two different field projects]. When a single-size CCN is considered in the adiabatic parcel, all CCN are expected to activate once parcel supersaturation reaches the critical supersaturation. Cloud droplets just above the cloud base have a finite width, typically a few tenths of a micrometer [see Figs. 4 and 5 in Grabowski et al. (2011) that consider activation of CCN observed in two different field projects].

When a single-size CCN is considered in the adiabatic parcel, all CCN are expected to activate once parcel supersaturation reaches the critical supersaturation. Cloud droplets just above the cloud base have a finite width, typically a few tenths of a micrometer [see Figs. 4 and 5 in Grabowski et al. (2011) that consider activation of CCN observed in two different field projects]. The purpose of this paper is to show that this is indeed possible when scales of turbulent eddies are at least of few meters, in agreement with scaled-up direct numerical simulations (DNS) in Thomas et al. (2020), stochastic model simulations in Grabowski and Abade (2017) and Abade et al. (2018), and in contrast to small-domain DNS simulations in Chen et al. (2021). As will be shown in this paper, turbulence can have a significant impact on the CCN activation even in the case of a single-size CCN.
activation applying two contrasting dry CCN radii, 10 and 200 nm, in two sets of simulations that feature a small range of mean cloud-base updrafts. Such an approach will help interpreting results from simulations featuring realistic (i.e., wide) CCN spectra. The next section presents results from a traditional approach, that is, idealized adiabatic rising parcel simulations. Section 3 extends those simulations to a case where CCN activation takes place within a rising volume with homogeneous isotropic turbulence. The approach to compare nonturbulent and turbulent rising parcel simulations follows Grabowski and Abade (2017) and Abade et al. (2018). We apply dynamic simulation framework used in Thomas et al. (2020) and Grabowski and Thomas (2021). Details of the framework are given in the appendix (with Fig. A1 illustrating the methodology). As shown in the appendix, supersaturation fluctuations in the adiabatic turbulent parcel prior to CCN activation significantly depend on the parcel spatial extent. At the same time, the turbulence intensity has no impact of the supersaturation fluctuations; however, when cloud droplets grow after activation, turbulence intensity affects the spectral width evolution (e.g., Fig. 4 in Grabowski and Thomas 2021). Because of those considerations, additional sensitivity simulations are conducted, and they are discussed in section 4. Section 5 provides a discussion and summary of model results.

2. The traditional approach to CCN activation: The adiabatic parcel

Based on the discussion of Fig. 1 above, two CCN sizes of the sodium chloride are selected for analyses discussed in this paper: the small CCN with the radius of 0.01 μm (10 nm) and the large CCN with the radius of 0.2 μm (200 nm). As Fig. 1 shows, the small CCN has the activation radius of around 0.6 μm and critical supersaturation of around 1.3%. For the large CCN, these are 5.4 μm and 0.15%. Figure 3 documents evolutions of the supersaturation and droplet radius in six simulations of a rising adiabatic parcel starting at 97% relative humidity (i.e., 2°3% supersaturation) with the temperature of 283 K and pressure of 900 hPa. The parcel rises with the updraft of 0.33, 1 and 3 m s⁻¹. CCN concentration is assumed 200 cm⁻³ in all simulations. CCN are at the equilibrium wet (deliquesced) size at the simulation onset, around 0.03 and 0.7 μm for small and large CCN, respectively. The activation time (i.e., droplet radius being equal to the activation radius) is shown by vertical lines in all panels.
As Fig. 3 shows, the supersaturation increases initially in the same way in all simulations because the dry-adiabatic temperature decrease is independent of the ascent rate. Once saturated conditions are reached, both the updraft strength and the CCN size affect the activation. Small CCN grow slowly initially, but then their growth accelerates when approaching the activation, and they grow rapidly until the droplets reach a couple-micrometer radius. Large CCN grow more uniformly in time and reach their activation radius several tens of meters above the supersaturation maximum, slightly higher for a stronger updraft. Supersaturations have to reach the critical supersaturation to initiate CCN activation in both small and large CCN. It follows that the maximum supersaturations are larger for the small CCN in simulations with the same parcel ascent rates. For the two stronger updrafts, 1 and 3 m s\(^{-1}\), supersaturations continue to increase after passing the critical supersaturation for small and large CCN. When droplets reach several-micrometer radii (e.g., the final few tens of meters of the parcel rise), the dissolved CCN no longer matter for their growth, with the droplet radii evolving similarly in all simulations. Although final supersaturation at 200-m height is different for different updrafts, it matters little for the final droplet radius in all simulations, about 8.1 \(\mu\)m for both CCN and 3 m s\(^{-1}\) versus close to 8.2 \(\mu\)m for both CCN and 0.33 m s\(^{-1}\). These radius values agree with the cloud water content and droplet concentrations at the 200-m height. The key is that CCN concentrations are the same in small and large CCN cases, and all CCN become activated to form cloud droplets.

Changing CCN from sodium chloride to ammonium sulfate in the parcel framework affects only details documented in Fig. 3. As shown in Fig. 1, the critical supersaturation and radius do change when a different CCN chemical composition is considered. As a result, a figure similar to Fig. 3 but for the ammonium sulfate shows larger maximum supersaturations and smaller activation radii, but the overall pattern remains similar (not shown).

### 3. CCN activation in an adiabatic turbulent parcel

The turbulent adiabatic parcel simulations follow implicit large-eddy simulation (ILES) methodology applied in Grabowski and Thomas (2021, hereafter GT21). Details of the model and model simulations are given in the appendix. The homogeneous isotropic turbulence is present within the computational domain at the onset of the rising turbulent parcel simulations and it is maintained throughout the simulations. The triply periodic computational domain (i.e., periodic in all three spatial directions) is 64\(^3\) m\(^3\) with the grid length of 1 m. This is the domain size used in GT21 and one of the domains considered in Thomas et al. (2020). The domain size is close to the turbulent rising parcel extent of 50 m considered in Grabowski and Abade (2017) and Abade et al. (2018). Such a domain size is also similar to the grid volume of LESs of natural clouds (e.g., Siebesma et al. 2003; Stevens et al. 2005; VanZanten et al. 2011; Chandrakar et al. 2021). It follows that the simulations discussed here illustrate the impact of microphysical processes that cannot be resolved in traditional high-resolution cloud simulations. For the fluid flow, we consider low TKE simulations from GT21 that assume the TKE dissipation rate of 10\(^{-3}\) m\(^2\) s\(^{-2}\). Arguably, the low TKE dissipation rate is appropriate for the rising air parcel approaching the cloud base. The low TKE setup corresponds to the rms perturbation vertical velocity (i.e., excluding the prescribed mean ascent) of around 0.2 m s\(^{-1}\) and evolving maximum vertical velocity between 0.5 and 0.8 m s\(^{-1}\). The TKE is around 0.05 m\(^2\) s\(^{-2}\).

As illustrated in Thomas et al. (2020, see Fig. 9 therein) and in GT21 (e.g., Fig. 6 therein), the size of the computational domain and turbulence intensity do affect the turbulence impact on the droplet size distribution. We document the impact of the domain size and turbulence intensity in sensitivity simulations discussed in the next section. The initial temperature and water vapor mixing ratio are taken from moist CCN-free simulations (e.g., no condensation) in GT21 (see also Fig. A2 in the appendix). Such simulations feature spatially uniform water vapor mixing ratio and spatially variable temperature, see section 3 and Fig. 3 in GT21. However, to be consistent with the nonturbulent parcel simulations from the previous section, we adjust water vapor mixing ratio to obtain initially a uniform 97% relative humidity across the entire computational domain. A set of sensitivity simulations with initially uniform water vapor mixing ratio (and thus spatially variable relative humidity) will also be discussed in the sensitivity simulation section. The parcel rises with a constant ascent rate, and three values are considered as in the nonturbulent parcel simulations discussed in the previous section: 0.33, 1, and 3 m s\(^{-1}\). The smallest ascent rate is similar to the rms vertical velocity of the turbulence. As for the parcel simulations in section 2, we consider two CCN sizes, 10 and 200 nm, with concentrations of 200 cm\(^{-3}\). CCN are assumed to be initially at the equilibrium size for the 97% relative humidity.

Before discussing specific model results, we mention in passing that the statistics discussed below are independent of the initial conditions applied in the turbulent parcel simulations. This has been documented in a few test simulations that started with different realizations of the turbulent flow and thermodynamic fields, that is, saving data from the CCN-free ILESs from GT21 at different times. This is important because one may wonder if turbulent parcel simulations discussed here should use small ensembles to ensure statistical convergence. However, such ensembles are not needed because a single simulation provides results that differ in small details when the simulation is repeated with different initial conditions (not shown).

Figures 4 and 5 document evolutions of the mean radius, mean spectral width, mean supersaturation and its spatial distribution standard deviation, and fraction of activated CCN in simulations with small and large CCN, respectively. The mean radius is calculated including either unactivated CCN and

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1 The “adiabatic turbulent parcel” phrase can be considered inappropriate because the simulations do assume an influx of the turbulent kinetic energy (TKE). However, the parcel remains adiabatic from the thermodynamics point of view (i.e., no entrainment). The same term was used in Grabowski and Abade (2017) and Abade et al. (2018).
droplets or only droplets, and the larger one is shown in the figure because not all CCN are activated in some cases. The activation radius is taken as the appropriate one for the small CCN and as 1 μm for the large CCN. The latter is motivated by 1-μm radius often assumed in bin schemes because the activation radius for the large CCN is about 5 μm. The supersaturation spatial-distribution mean and standard deviation come from averaging the gridscale supersaturation fluctuations over the grid.

**FIG. 4.** Evolution of (a) CCN/droplet radius, (b) mean supersaturation and its standard deviation, (c) spectral width, and (d) fraction of the total CCN inside the computational domain that become activated. Symbol colors represent simulation with different ascent rates as shown in (d). Simulations with small CCN. Mean radius and spectral width are calculated including all particles until the mean radius reaches the activation radius, and only activated droplets after that.

**FIG. 5.** As in Fig. 4, but for simulations with large CCN. Only droplets larger than 1 μm are included when calculating the mean radius, spectral width, and activation fraction.
entire computational domain. The horizontal axes in the figure show the distance similarly to the parcel results in Fig. 3. However, the computational domain has extent comparable to the vertical displacement of the domain (64 vs 200 m), and thus the horizontal scale in panels of Figs. 4 and 5 can be thought as representing the height of the computational domain center as it rises through the cloud base.

For small and large CCN (Figs. 4 and 5, respectively), the mean supersaturation increases in the same way, but the supersaturation standard deviation increases from initially zero at a rate that is larger for the smaller ascent rate. The same applies to other simulations discussed later in the paper. This is because the supersaturation standard deviation develops with a characteristic time scale comparable to the turnover time of large eddies that is about 1 min for the 64 m domain size and the assumed low TKE level. Large eddies experience large vertical displacements, and they have the largest impact on the supersaturation field (see section 3 in GT21 and Fig. A2 in the appendix). For that reason, the increase of the supersaturation fluctuations with the distance from the initial level should be approximately inversely proportional to the parcel ascent rate, which seems to be the case in all simulations discussed in this paper. Simply put, supersaturation fluctuations have more time to develop when the parcel ascent rate is small. Maximum supersaturation standard deviation occurs during the activation phase, and then it decreases to values below 0.1% in all cases. Such an evolution is consistent with stochastic model results in Abade et al. (2018, see their Fig. 6). For the small CCN and lower two ascent rates, the supersaturation standard deviation shows some fluctuations after the maximum mean supersaturation is reached; these will be explained in the following analysis.

The CCN activation commences after the local supersaturation surpasses the activation value, 1.3% for small CCN and 0.15% for large CCN. Not all small CCN are activated for 0.33 and 1 m s\(^{-1}\) as activation in a fraction of the domain and subsequent turbulent mixing of activated CCN and droplet growth brings the supersaturation fluctuations down and prevents additional activation. Only about 80% (25%) of the small CCN become cloud droplets for the ascent rate of 1 (0.33) m s\(^{-1}\). This means, for instance, that the mean droplet concentration after activation for the weakest ascent rate is about 50 cm\(^{-3}\), and not 200 cm\(^{-3}\) as in the strongest ascent rate case. This explains the larger mean droplet size at the end of the simulation in this case as shown in Fig. 4a. Overall, activation for the 0.33 m s\(^{-1}\) ascent rate is similar to the fluctuation-dominated regime 3 in Fig. 2, whereas the mean-dominated activation regime 1 describes the case of 3 m s\(^{-1}\) ascent rate. As a result, the increase of the mean radius from below 0.1 \(\mu m\) to over 1 \(\mu m\) (Fig. 4a) takes place over larger distance than in the nonturbulent parcel (Fig. 3). The height corresponding to the rapid mean-radius increase can be referred to as the mean activation height, and it changes depending on the parcel ascent rate. Spectral width that was zero in the parcel simulations in section 2 is between a few tenths and 1.5 \(\mu m\) at the 200-m height, larger for the smaller parcel ascent rates. Small fluctuations in the spectral width for the 0.33 m s\(^{-1}\) ascent rate (Fig. 4c) likely come from CCN activation and deactivation in the locally fluctuating supersaturation (Arabas and Shima 2017; Abade et al. 2018). Their specific pattern changes when different initial conditions are used in the simulation, but the overall evolution remains similar to that shown in the figure.

For large CCN (Fig. 5), all CCN are activated regardless of the parcel ascent rate and the final mean radius agrees among all three simulations at the 200-m height. As for the small CCN, the activation of large CCN seems to cover the entire range from Fig. 2, from the mean-dominated regime 1 for 3 m s\(^{-1}\) parcel ascent rate to the fluctuation-dominated activation regime 3 for 0.33 m s\(^{-1}\) ascent rate. The spectral width changes depending on the ascent rate, with the largest spectral width for the smallest ascent rate simulation. Spectral widths at 200 m are smaller when compared to the corresponding ascent rates for the small CCN cases. Evolutions of the supersaturation standard deviation are similar to the corresponding small CCN cases.

Probability density functions (pdfs; i.e., normalized distributions) of the supersaturation inside the computational domain rising with 1 m s\(^{-1}\) ascent rate at heights of 40, 60, 80, 100, and 120 m for small and large CCN are shown in Fig. 6. At 40 m, before the activation commences, the distributions are wide and not far from Gaussian. At 120 m, when activation is almost completed, the distributions are much narrower, in agreement with the standard deviation evolutions shown in Figs. 4b and 5b. The distributions are asymmetric during activation, especially for the small CCN case. Additional analysis for the small CCN shows that the transition from negatively skewed pdf at the 80-m height (i.e., the distribution wider for smaller supersaturations) into a positively skewed pdf at the 100- and 120-m heights (i.e., the distribution extending more to the right of the maximum) explains standard deviation fluctuations evident in Fig. 4b. The distributions show various modes of activation following the discussion of Fig. 2. The transition from wide pdfs before activation to narrow pdfs after activation, with complex pdfs during activation, is also typical for other parcel ascent rates (not shown).

Droplet spectra at 200-m height in simulations with 0.33 and 3 m s\(^{-1}\) ascent rates and for small and large CCN are shown in Fig. 7. The spectra for the weak ascent rate are amazingly wide, and the spectral peak for the small CCN case is at significantly larger radius, over 10 \(\mu m\) versus around 7 \(\mu m\). This is because of the smaller droplet concentrations activated for the weak ascent rates as shown in Fig. 4. The larger spectral width for small ascent rates can be partially explained by longer times available for the CCN activation and droplet growth through the eddy hopping, that is, individual CCN and droplets experiencing locally different supersaturation fluctuations in response to different vertical velocities (Grabowski and Abade 2017; Abade et al. 2018).

Figure 8 documents evolutions of a small fraction of the simulated CCN in a similar way as in the upper panels of Figs. 3, 4a, and 5a. Clearly, spectral width at the end of simulations comes from different times CCN get activated, with the best illustration for the small CCN 0.33 m s\(^{-1}\) case. In this case, a significant fraction of CCN avoids activation and is carried by the parcel as unactivated (interstitial) CCN. It should be stressed that distance on the horizontal axes represents the height of computational
domain center and not the actual height of a particular CCN or a cloud droplet. Thus, the almost-vertical lines in the upper panels of Fig. 8 represent CCN activated anywhere in the ILES domain at different times.

In summary, including small-scale turbulence into the rising parcel framework significantly affects CCN activation, partitioning between activated and unactivated CCN in particular, and thus droplet spectra just above the cloud base. The next section investigates to what extent this conclusion is affected by specific details of model simulations, the assumed TKE and the domain size in particular.

4. Sensitivity simulations

This section discusses sensitivity simulations that consider reduced turbulence intensity, smaller domain size, and spatially uniform initial water vapor mixing ratio rather than uniform relative humidity. All those can potentially affect specific results from simulations discussed in the previous section. The intensity of cloud-base turbulence is expected to be lower than inside a cloud (e.g., Siebert et al. 2006, 2013) and thus an even lower turbulence intensity should be considered in sensitivity simulations. At the same time, intensity of homogeneous isotropic turbulence is dominated by large eddies and thus TKE depends on the computational domain size in DNS and ILESs. The appropriate scaling between the TKE ($E$), the domain size ($L$), and the TKE dissipation rate ($\epsilon$) is $E \sim (L \epsilon)^{2/3}$ [Pope 2000; Eq. (4) in Thomas et al. 2020]. In this section, we present results of simulations where a lower dissipation rate and a smaller domain size are considered, sections 4a and 4b, respectively. Section 4c briefly discusses results with initially uniform water vapor mixing ratio.

a. Very low TKE simulations

The first set of simulations considers the same domain size ($64^3$ m$^3$) and the TKE dissipation rate of $10^{-5}$ m$^2$ s$^{-3}$, that is, 100 times smaller than in section 3. The turbulence intensity does not affect supersaturation fluctuations in the case without condensation, that is, below the cloud base as the parcel approaches saturation (see Fig. A2 in the appendix). This is because, for simulations featuring initially uniform temperature and moisture as in GT21, the water vapor mixing ratio remains uniform across the computational domain and the temperature changes in agreement with vertical dry-adiabatic displacements. However, the turbulence intensity does matter after droplets are activated because the turbulence intensity impacts local supersaturation fluctuations (see Figs. 4 and 7 in GT21). The TKE dissipation rate of $10^{-5}$ m$^2$ s$^{-3}$ and 64-m domain size implies about factor

Please note that droplet position has no physical meaning because the computational domain is periodic.
0.05 smaller TKE and thus factor-of-0.2-smaller velocities than in section 3. As in section 3, we ran six simulations varying the CCN size and parcel ascent rate. Those simulations are initiated by rescaling velocity fluctuations used to initiate simulations in section 3 and include TKE forcing to maintain the reduced TKE. Results of those simulations in the same format as Figs. 4 and 5 are shown in Figs. 9 and 10. Overall, Figs. 9 and 10 are similar to those in section 3. For small CCN, noticeable differences include a smaller supersaturation standard deviation and thus higher mean supersaturation peaks to allow CCN activation. As a result, there is a larger activated CCN fraction for the 0.33 m s\(^{-1}\) mean ascent and all CCN are activated for the larger ascent rates. Spectral widths are smaller in Fig. 9 when compared to Fig. 4. For large CCN, results are also similar to the higher dissipation rate of Fig. 5 with the exception of the spectral width, smaller in Fig. 10c.

b. Smaller domain simulations

The second set of sensitivity simulations considers CCN activation in 8\(^3\)-m\(^3\) turbulent parcel covered with 64\(^3\)-m\(^3\) grid and the section 3 TKE dissipation rate of 10\(^{-3}\) m\(^2\) s\(^{-2}\). Such simulations have TKE of about a quarter of the simulations in section 3 and a half-rms velocity fluctuations. A dry simulation (i.e., no condensation) ran to provide input to start the ascending turbulent parcel simulations features reduced supersaturation fluctuations because of a smaller domain (see Fig. A2). An adjustment of the initial water vapor mixing ratio to obtain spatially uniform initial relative humidity of 97% (i.e., as in large-domain simulations) is also included. Results of those simulations are illustrated in Figs. 11 and 12 for both small and large CCN, respectively. Overall, results are similar to the larger domain results in section 3 with the exception of smaller supersaturation fluctuations (in agreement with the anticipated scaling) and thus smaller spectral widths.

c. Initially uniform water vapor simulations

The last set of sensitivity simulations uses setup as in section 3 (i.e., 64\(^3\)-m\(^3\) domain and TKE dissipation of 10\(^{-3}\) m\(^2\) s\(^{-2}\)) and removes the initial water vapor adjustment to obtain uniform relative humidity across the computational domain. As a result, the initial relative humidity fluctuates across the computational domain with the statistics shown in Fig. 3a in GT21 (i.e., with the supersaturation between about −4% and −2% rather than uniformly −3% as in section 3). Figure 13 shows evolutions of the spectral width and CCN activated...
fraction for such simulations with small and large CCN. Overall, evolutions shown in Fig. 13 are similar to right panels of Figs. 4 and 5 documenting a rather small impact of the initial conditions for the rising turbulent parcel simulations.

In summary, simulations with a smaller TKE dissipation rate and a smaller ILES domain size, both affecting simulated turbulence intensity and thus CCN activation and cloud droplet growth just above the cloud base, show some impact of
those changes on specific details of the simulated impact. However, the general conclusions from the initial set of turbulent parcel simulations do not change. The same applies to simulations with modified initial conditions for the water vapor mixing ratio.

5. Discussion and summary

This paper presents a modeling study that highlights the impact of small-scale turbulence on the cloud-base CCN activation. We consider a monodisperse (single-size) CCN and contrast simulated impact of the cloud-base turbulence with

![Graphs showing changes in radius and supersaturation over height for different conditions.]

**Fig. 10.** As in Fig. 5, but for simulations with very low TKE and large CCN.

**Fig. 11.** As in Fig. 4, but for simulations with 8³-m³ computational domain and small CCN.
observations and theoretical studies of monodisperse CCN activation in the Pi chamber turbulent airflow (Prabhakaran et al. 2020). We consider CCN at the small and the large ends of the CCN size distribution that are expected to produce cloud droplets at the cloud base of a natural cloud. The small CCN are assumed to have a dry radius of 0.01 \( \mu \text{m} \) (10 nm) and corresponding activation radius and critical supersaturation of 0.6 \( \mu \text{m} \) and 1.3\%, respectively. Smaller CCN require even higher supersaturations to get activated and thus are unlikely to create cloud droplets near the cloud base. The

FIG. 12. As in Fig. 5, but for simulations with 83-m\(^3\) computational domain and large CCN.

FIG. 13. Evolutions of the spectral width and activated fraction for simulations with initially uniform water vapor for simulations with (a),(b) small CCN and (c),(d) large CCN. These need to be compared with right panels of Figs. 4 and 5 form small and large CCN, respectively.
large CCN considered here have a dry radius of 0.2 \( \mu \)m (200 nm) and feature activation radius and critical supersaturation of 5.4 \( \mu \)m and 0.15\%, respectively. Thus, they are easily activated near the cloud base. In numerical calculations presented in this paper, the two CCN sizes are assumed to exist in concentrations of 200 cm\(^{-3}\).

For a reference, we consider in section 2 a nonturbulent adiabatic parcel, a traditional way to study cloud-base activation, see Fig. 3. The parcel initial conditions assume 97\% relative humidity, about 50 m below the height at which the parcel reaches saturation. Parcel displacement of 200 m is considered. Three parcel ascent rates (i.e., cloud-base updraft velocities) are analyzed, 0.33, 1, and 3 m s\(^{-1}\), covering a range typical for a stratocumulus or a small cumulus cloud. In the nonturbulent adiabatic parcel framework, CCN can only become activated after the parcel supersaturation exceeds the critical supersaturation. For the two larger ascent rates, the supersaturation continues to increase after the critical supersaturation is reached until the growth of cloud droplets (the sink of water vapor) and the accompanying latent heat release (increasing the parcel temperature) stop the increase and the supersaturation starts to decline. For the small CCN, supersaturations are larger around the activation height, but they become almost the same for the two CCN after the 200-m parcel rise. Because the same CCN concentration is assumed in all cases and all CCN become activated, the droplet radius is around 8 \( \mu \)m for all cases at the final 200-m height. In the adiabatic nonturbulent parcel with a single-size CCN, all droplets have the same size after activation.

For the CCN activation in the presence of small-scale turbulence, we extend the approach applied in Grabowski and Thomas (2021; GT21) where the homogeneous isotropic turbulence was maintained within a triply periodic computational domain (see Fig. A1 in the appendix for the illustration). GT21 studied broadening of initially monodisperse droplet size distribution following previous numerical and theoretical studies of Vaillancourt et al. (2002), Lanotte et al. (2009), Sardina et al. (2015), and Thomas et al. (2020). Here, we add a prescribed ascent rate of an initially sub-saturated turbulence-filled computational domain that includes deliquesced CCN. Such an approach is an analog of the traditional nonturbulent case considered in section 2. In the main set of simulations, we consider a 64\(^3\)-m\(^3\) computational domain and the parcel mean ascent rate of 0.33, 1 and 3 m s\(^{-1}\) as for the nonturbulent parcel, and follow-up with a set of sensitivity simulations where the assumed turbulence intensity, the computational domain size, and the initial conditions for the humidity field are separately changed from the main set.

The results from the main set of simulations document the key role of turbulence in the CCN activation. Depending on the parcel mean ascent, the simulated activation can cover all regimes illustrated in Fig. 2 (Fig. 1 in Prabhakaran et al. 2020), from the mean-dominated regime 1 for the 3 m s\(^{-1}\) parcel ascent rate to the fluctuation-dominated regime 3 for 0.33 m s\(^{-1}\) ascent rate, regardless of the CCN size. Because small CCN requires 1.3\% supersaturation to activate, not all small CCN get activated into cloud droplets for 0.33 and 1 m s\(^{-1}\) ascent rates. In fact, the mean droplet concentration above the cloud base is only about 25\% (80\%) of the CCN concentration prior to activation for the 0.33 (1) m s\(^{-1}\) in the main set of simulations. Sensitivity simulations that consider lower TKE dissipation rate and smaller computational domain (both affecting the turbulence intensity) show that specific details of the results summarized above do change, but the overall impact of the small-scale turbulence on the cloud-base activation of both small and large CCN remains similar. PDFs of the supersaturation evolve from relatively wide before the activation onset to narrow after CCN activation is completed, with complex (e.g., skewed) distributions during activation.

The impact of the turbulent cloud-base activation on the droplet spectra just above the cloud base documented here is similar to that in Abade et al. (2018, hereafter A18). A18 applied a stochastic model to study the impact of turbulence on the CCN activation and droplet growth, focusing on the impact of entrainment above the cloud base. Results presented in A18 are only for a single updraft strength (1 m s\(^{-1}\)), a single spatial extent of the turbulent volume (\( L = 50 \) m), and a stronger turbulence (TKE dissipation rate of 10\(^{-2}\) m\(^2\) s\(^{-3}\), arguably appropriate for cloudy volumes away from the cloud base). The increase of the spectral width during turbulent activation simulated in A18 is even larger to that shown here (cf. Figs. 4c and 5c here and Fig. 5 in A18), likely because of a relatively wide CCN spectrum considered in A18. The activation fraction in A18 increases gradually compared to the nonturbulent parcel simulations (see Fig. 6b there), again as in weak-updraft simulations here. Finally, supersaturation fluctuations during the activation phase have similar amplitude here and in A18, with the supersaturation standard deviation of several tenth of 1\% (see Figs. 4b and 5b here and Fig. 6a in A18).

Simulations discussed in this paper, without CCN activation in the appendix (Fig. A2) and with CCN activation and initial droplet growth in the main text, suggest the key role of the largest spatial scales for the supersaturation fluctuations. This is consistent with the scaled-up DNS and stochastic model simulations discussed in Thomas et al. (2020) that show larger supersaturation fluctuations and larger spectral widths for simulations applying larger computational domains; see Figs. 7, 9, and 10 therein. This may seem to contradict previous studies (e.g., Lanotte et al. 2009; Li et al. 2019) that emphasize the role of the flow Reynolds number for the diffusional growth of cloud droplets in a turbulent environment. However, in the context of the true DNS, the Reynolds number (i.e., the range of spatial scales resolved in the simulation) typically also implies a larger computational domain as the dissipation scale is always assumed to represent the Kolmogorov microscale [see, for instance, Table 2 in Lanotte et al. (2009)]. For the Pi chamber, its small vertical extent (1 m) eliminates the key physical mechanism discussed here, with the supersaturation fluctuations driven primarily by small-scale (ultimately molecular) mixing between air parcels with contrasting properties, warmer and more humid rising from the chamber lower boundary versus colder and...
drier descending from the upper boundary. This is different from what happens near the cloud base of a natural cloud.

The idealized turbulent parcel framework applied in simulations discussed here calls for more realistic cloud-base activation simulations. One possibility is the framework applied in Clark and Hall (1979) who simulated a forced activation simulations. One possibility is the framework discussed here calls for more realistic cloud-base cloud. From what happens near the cloud base of a natural drier descending from the upper boundary. This is different from what happens near the cloud base of a natural cloud.

The idealized turbulent parcel framework applied in simulations discussed here calls for more realistic cloud-base activation simulations. One possibility is the framework applied in Clark and Hall (1979) who simulated a forced ascent of a three-dimensional turbulent air plume crossing the cloud base and forming a cloud (see Fig. 1 therein for a schematic). Such an approach requires specification of the turbulence intensity as in the current study. Even better would be to include surface buoyancy flux driving turbulence within convective boundary layer and leading to cloud formation above it. Such simulations would include realistic turbulence intensities within the updraft passing through the cloud base. However, such simulations need to feature spatial resolutions like those considered here, that is, on the order of 1 m, and thus would be quite challenging. One possibility is to consider the simulation setup from Grabowski (2020) and Chandrakar et al. (2021) that follow Lasher-Trapp et al. (2005) where a single cumulus cloud developing from a turbulent boundary layer is driven by prescribed surface sensible and latent heat fluxes. We hope to report on such simulations in the future.

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Data availability statement. Data supporting this study are available at https://dashrepo.ucar.edu/dataset/215_grabowski.html (Grabowski 2021).

APPENDIX

The Model, Model Equations, and Model Simulations

This appendix provides details of the rising adiabatic turbulent parcel simulations applying the homogeneous isotropic turbulence approach from GT21. Figure A1 illustrates the modeling methodology: the volume filled with turbulence and deliquesced CCN is assumed to rise with a prescribed ascent rate, reach saturation, and activate some or all of the carried CCN.

The turbulent flow is simulated by the Eulerian–semi-Lagrangian (EULAG) anelastic finite-difference fluid flow model (http://www.mmm.ucar.edu/eulag/) applied in the implicit large-eddy simulation (ILES) mode (Margolin and Rider 2002; Andrejezuk et al. 2004; Margolin et al. 2006; Grinstein et al. 2007). ILES uses no explicit dissipation and removes small-scale velocity and scalar fluctuations through numerical diffusion provided by the monotone Eulerian advection scheme. Model equations for the air velocity \( \mathbf{u} \), air temperature \( T \), and water vapor mixing ratio \( q_w \) are exactly as in GT21:

\[
\frac{\partial \mathbf{u}}{\partial t} + \text{div}(\mathbf{u} \cdot \mathbf{u}) = -(1/\rho) \text{grad} p + \mathbf{f},
\]  
\[
\text{div} \mathbf{u} = 0,
\]  
\[
\frac{\partial T}{\partial t} + \text{div}(\mathbf{u} T) = (L_v/c_p)C_d - (g/c_p)\mathbf{u}_z,
\]  
\[
\frac{\partial q_w}{\partial t} + \text{div}(\mathbf{u} q_w) = -C_d,
\]

where \( \mathbf{u} = (u_x, u_y, u_z) \) is the air velocity, \( p \) is the dynamic pressure (derived applying the incompressibility condition A2), \( \rho = 1 \text{ kg m}^{-3} \) is the air density, \( \mathbf{f} \) is the turbulence forcing term, \( L_v = 2.5 \times 10^9 \text{ J kg}^{-1} \) is the latent heat of condensation, \( c_p = 1005 \text{ J kg}^{-1} \text{ K}^{-1} \) is the air specific heat at constant pressure, \( g = 9.81 \text{ m s}^{-2} \) is the gravitational acceleration, and \( C_d \) is the condensation rate, the rate of change of the cloud water mixing ratio resulting from the diffusional growth of cloud droplets. The forcing term \( \mathbf{f} \) ensures that the turbulence is maintained throughout the simulation with TKE flowing from large scales toward the small-scale dissipation. The forcing term is formulated in such a way that the total TKE and its equal partitioning between the three spatial components do not change during the simulation, and there is no mean flow across the computational domain. See section 2a in GT21 for details. The last term in (A3) represents temperature change due to air vertical motion in the stratified atmosphere. Combining (A3) and (A4) provides local supersaturation that drives CCN activation and droplet growth. Note that the fluid flow equations exclude the
Results from simulations of 643 and 1283 are shown for shows time (green) and left (red) to avoid overlap. The symbol vertical extent of condensation. The symbols are shifted to the right (green color) TKE dissipation rates (10^{-3} and 1 m^2 s^{-3}, respectively) in forced homogeneous isotropic turbulence simulations with no condensation. The symbols are shifted to the right (green color) and left (red) to avoid overlap. The symbol vertical extent shows time fluctuations of the supersaturation standard deviation. Results from simulations of 64^3 and 128^3 are shown for \( L = 64 \) m. The solid line shows the fit for all simulations. Temperature and moisture equations are shown in the following the dry adiabat [given by the last term on the right-hand side of (A3)] provide temperature fluctuations that drive supersaturation fluctuations. Simply put, these fluctuations are larger when the domain vertical extent is larger. For that reason, the Reynolds number that represents the range of scales resolved in a simulation for a given domain size has no role as shown by 64^3 and 128^3 \( L = 64 \) m domain simulations.

For simulations with CCN and droplets, we apply a Lagrangian approach that is similar to the one used in Vaillancourt et al. (2002), Lanotte et al. (2009), GT21, among others. Figure A2 documents the impact of the computational domain size on the simulated supersaturation fluctuations for the case without CCN [i.e., \( C_d = 0 \) in (A3) and (A4)]. Such simulations have been briefly discussed in GT21; see Fig. 3 therein. Figure A2 shows that the domain size \( L \) is the key parameter for the supersaturation fluctuations. As suggested in GT21, this is because vertical air displacements following the dry adiabat [given by the last term on the right-hand side of (A3)] provide temperature fluctuations that drive supersaturation fluctuations. Simply put, these fluctuations are larger when the domain vertical extent is larger. For that reason, the Reynolds number that represents the range of scales resolved in a simulation for a given domain size has no role as shown by 64^3 and 128^3 \( L = 64 \) m domain simulations.

For simulations with CCN and droplets, we apply a Lagrangian approach that is similar to the one used in Vaillancourt et al. (2002), Lanotte et al. (2009), Thomas et al. (2020), and GT21. A set of point particles representing deliquesced CCN and activated droplets is followed in the computational domain. However, for the computational domain considered in this study (\( 10^3 \)–\( 10^5 \) m^3), it is impossible to follow all particles. For that reason, we use the so-called superdroplet method, that is, we apply a computationally feasible total number of Lagrangian particles and assign each of them the multiplicity attribute that tells how many real cloud droplets each superdroplet represents. With the computational grid (64^3) and the number of superdroplets per grid volume (20 for all simulations described here), the total number of particles followed in the computational domain is around 5.2 million. As in GT21, each superdroplet responds to the mean supersaturation within the grid volume corresponding to its location. Condensation rate \( C_d \) is obtained by summing up the mass change of all superdroplets within a given grid volume (see appendix B in GT21). Since the droplet growth requires typically a smaller time step than used in the dynamic flow evolution (especially for small CCN before activation), a simple substepping procedure is used to ensure that the microphysical time step used in the droplet growth calculation is sufficiently small. In this procedure, the time step to calculate droplet growth is not allowed to be larger than 1/20 of the droplet growth characteristic time defined as the ratio of the droplet radius to its rate of change, that is, \( r \) over \( dr/dt \). The microphysical time step cannot be larger than the dynamic model time step.

The equation describing growth rate of a small cloud droplet with radius \( r \) containing a dissolved salt is exactly as in Grabowski et al. (2011). The equation is as follows (e.g., Howell 1949; Fukuta and Walter 1970; Kogan 1991; Pruppacher and Klett 1997):

\[
\frac{dr}{dt} = \frac{G}{r}(S - S_{eq}),
\]

where \( S = e/e_s - 1 \) is the supersaturation (\( e \) and \( e_s \) are the water vapor pressure and its saturated value, respectively), \( S_{eq} = \exp(A/r - B/r^2) - 1 \approx A/r - B/r^2 \) is the equilibrium supersaturation (where \( A \) depends on the surface tension and temperature and represents the Kelvin or curvature effect; and \( B \) depends on the solute properties and represents the Raoult’s or solution effect; Pruppacher and Klett 1997, section 6.5), and

\[
G^{-1} = \rho_w R_v T/(e_s D) + \rho_w L_v/(KT)[L_v/(R_v T) - 1],
\]

where \( \rho_w \) is the density of water, \( R_v \) is the water vapor gas constant, \( D \) is the diffusivity of water vapor in the air, and \( K \) is the air thermal conductivity. Note that \( D \) and \( K \) include kinetic effects (i.e., the effects of condensation and accommodation coefficients; e.g., Fukuta and Walter 1970). The exact formula used in the model follows appendix in Grabowski et al. (2011). In the simulations described here, we assume that the salt is either sodium chloride or ammonium sulfate (Fig. 1). In the simulations described here, we assume that the salt is either sodium chloride or ammonium sulfate (Fig. 1). In the simulations described here, we assume that the salt is either sodium chloride or ammonium sulfate (Fig. 1). In the simulations described here, we assume that the salt is either sodium chloride or ammonium sulfate (Fig. 1).
mean pressure \( p \) in (A1) has also to decrease as the parcel rises. This is in contrast to turbulent simulations discussed in Thomas et al. (2020) and GT21 where the environmental pressure was assumed constant. See (1) in Grabowski and Wang (2009) that provides mathematical description of the rising parcel framework.

The simulations with a rising turbulent parcel are initiated by snapshots of velocity, temperature and moisture fields obtained in simulations without CCN, that is, solving (A1)–(A4) without aerosol and with \( C_d = 0 \) (see Fig. A2). The simulations are initiated with a uniform temperature and water vapor mixing ratio and are run for a few eddy-turnover times to obtain quasi-equilibrium supersaturation statistics, an approach used in GT21. Equations (A3) and (A4) imply that the water vapor mixing ratio in such a simulation remains uniform across the computational domain, whereas the temperature fluctuates in response to the source due to the turbulent vertical velocity. Because these fluctuations are incompatible with the initial 97% relative humidity assumed in the nonturbulent parcel simulations, a simple adjustment is used to create a uniform relative humidity across the computational domain at the simulation onset. The adjustment modifies the local water vapor mixing ratio to match the required 97% relative humidity. We also consider simulations with initially uniform water vapor mixing ratio and spatially variable relative humidity in a set of sensitivity simulations. The dynamic model time step is 0.2 and 0.5 s for \( 64^3 \) m\(^3\) domain simulations with TKE dissipation rate of \( 10^{-3} \) and \( 10^{-2} \) m\(^2\) s\(^{-3}\), respectively, and 0.1 s for \( 8^3 \) m\(^3\) domain simulations. Microphysical calculations include substepping for CCN/droplet growth calculations as explained above.

REFERENCES


