Understanding aerosol-cloud interactions through modelling the development of orographic cumulus congestus during IPHEx

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Abstract. A new cloud parcel model (CPM) including activation, condensation, collision-coalescence, and lateral entrainment processes is presented here to investigate aerosol-cloud interactions (ACI) in cumulus development prior to rainfall onset. The CPM was applied with surface aerosol measurements to predict the vertical structure of cloud development at early stages, and the model results were compared with airborne observations of cloud microphysics and thermodynamic conditions collected during the Integrated Precipitation and Hydrology Experiment (IPHEx) in the inner region of the Southern Appalachian Mountains (SAM). Sensitivity analysis was conducted to examine the model response to variations in key ACI physiochemical parameters and initial conditions. The sensitivities of the CPM used here mirror those found in parcel models without entrainment and collision-coalescence included, except for the evolution of the droplet spectrum and liquid water content with height. Simulated CDNC spectra evolution exhibits high sensitivity to variations in initial aerosol concentration at cloud base, but weak sensitivity to aerosol hygroscopicity. The condensation coefficient a_c plays an important role in determining the cloud droplet number concentration (CDNC), liquid water content (LWC), and droplet size distribution. Lower values of a_c lead to higher cloud droplet number concentrations, broader droplet spectra, and higher maximum supersaturation near cloud base. The simulated vertical structure of CDNC spectra exhibits strong nonlinear sensitivity to entrainment strength and condensation efficiency indicating significant competitive interference among turbulent dispersion and activation, which impacts droplet growth (condensational growth and collision-coalescence processes) and spectral width. This explains at least in part the heterogeneity of CDNC spectra from aircraft measurements in different regions of the cloud and at different heights. Comparison of model predictions with data suggest that the application of the CPM with classical parameterizations of lateral entrainment remains challenging due to the spatial heterogeneity of the convective boundary layer and the intricate 3D circulation in mountainous regions. Nonetheless, the model simulations provide new constraints of the determinant factors of convective cloud formation leading to mid-day warm season rainfall in complex terrain.

1 Introduction

Atmospheric aerosols produced by dramatically increased industrialization and urbanization exert a large impact on the climate system and the hydrological cycle (Koren et al., 2008;Ramanathan et al., 2001;Tao et al., 2012). Aerosols influence the earth-atmosphere system primarily via two mechanisms: a radiative (direct) effect and a microphysical (indirect) effect (Rosenfeld et al., 2008). The direct effect on the Earth's energy budget occurs via scattering and absorbing of shortwave and longwave radiation in the atmosphere, hence modulating the net radiation and climate (Haywood and Boucher, 2000;Ramanathan et al., 2001). The indirect effect is related to aerosols as cloud condensation nuclei (CCN) or ice nuclei (IN) that alter microphysical properties and consequently affect cloud radiative properties and precipitation efficiency (Jiang et al., 2008;Lohmann and Feichter, 2005;McFiggans et al., 2006). In particular, an increase in aerosol concentration results in enhanced cloud droplet number concentration (CDNC), smaller average drop size, and increased cloud albedo (Twomey, 1977). Smaller cloud droplets are associated with lower collection and coalescence efficiency, slower drop growth and reduced precipitation, thus leading to longer cloud lifetimes (Albrecht, 1989;Andreae and Rosenfeld, 2008;Khain et al., 2005). Over complex terrain in California and Israel, Givati and Rosenfeld (2004) attributed a reduction in annual precipitation of 15–25% to air-pollution aerosols from upwind urban areas. By comparing two scenarios of maritime and continental aerosols, Lynn et al. (2007) found that simulations with maritime aerosols with relatively lower aerosol number concentration yielded 30% more precipitation than continental aerosols over a mountain slope. Such local effects can translate into large spatial shifts in clouds and precipitation as aerosol-cloud interactions (ACI) inducing suppression of precipitation upwind could give rise to the enhancement of precipitation downwind (Muhlbauer and Lohmann, 2008), thus modifying the spatial distribution of orographic precipitation, transferring precipitation from one watershed to another and thus strongly influencing the local and regional hydrology. Yang et al. (2016) examined the reasons for warm rain suppression due to increased air pollution in the Mt. Hua area in central China. They demonstrated that weakened valley-ridge circulations as a result of aerosol-radiation interactions and lower water vapor concentrations in the valley led to the suppression of convection and precipitation in the mountain. A study of thermally driven orographic clouds over a tropical island during the Dominica Experiment (DOMEX) field campaign found that atmospheric moisture was the predominant constraint in cloud and precipitation formation over the aerosol effect, and the surface aerosol source has the strongest influence on precipitation under unfavourable environmental condition for cloud growth (Nugent et al., 2016). Barros et al. (2018) showed that model simulations using aerosol activation spectra from local sources and activation spectra from remote aerosol sources resulted in significant spatial and temporal redistribution of precipitation in the central Himalayas including changes in cloud dynamics and the vertical distribution of hydrometeors. The latter is the basis for remote sensing measurements of precipitation, and therefore understanding how ACI and ACPI modify precipitation structure is key to improving retrievals in mountainous regions (e.g. Duan et al. 2015)

In the Southern Appalachian Mountains (SAM, Fig. 1), persistent low-level clouds and fog (LLCF) play a governing role in warn season rainfall by increasing the frequency and duration of light rainfall and drizzle, and by enhancing storm rainfall via seeder-feeder interactions (SFI; Wilson and Barros, 2014, 2015 and 2017; Duan and Barros, 2017). Albeit with large spatial variability, microphysical observations and idealized model simulations of the dynamical evolution of raindrop size distribution (RDSD) with height show that SFI in the lower atmosphere can explain up to one order magnitude increases in rainfall rate at low elevations similar to orographic enhancement at higher elevations in the SAM. Understanding and modelling the spatial variability of the vertical microstructure of clouds in complex terrain is therefore key to understand precipitation processes toward improving rainfall estimation and prediction. Whereas previous studies linked LLCF in the SAM to high biogenic aerosol loading produced locally with occasional influx from remote pollution sources (Link et al., 2015; Lowenthal et al., 2009), quantitative understanding of the indirect effect of aerosols on clouds with implications for precipitation dynamics including SFI is lacking. The purpose of this study is to investigate ACI in the SAM integrating models and observations collected during IPHEx (Integrated Precipitation and Hydrology Experiment; Barros et al., 2014) with a focus on the evolution of cloud droplet spectra with height. This is an important first step toward understanding the spatial variability in the vertical structure of cloud microphysics from one location to another toward capturing the observed spatial and temporal heterogeneity of SFI.

The representation of physical and chemical processes related to clouds and precipitation in numerical models relies on parameterizations with varying degrees of uncertainties depending on space-time model resolution because of multiscale and complex physics, (Khairoutdinov et al., 2005;Randall et al., 2003). For example, the characteristic time-scale of condensational growth of submicron-size droplets is on the order of 1 ms, and length scales of individual drops range from µm to cm (Pinsky and Khain, 2002), that is a scale gap of five to nine orders in magnitude with respect to the spatial resolution of cloud-resolving models (kms). Although detailed 2-D and 3-D models that explicitly resolve cloud formation and microphysical evolution to varying degrees of completeness have been reported in the literature (Fan et al., 2009;Leroy et al., 2009;Muhlbauer et al., 2010), the wide range of length (µm–m) and time (ms–s) scales associated with aerosol-cloud-rainfall interactions poses significant challenges for model spatial and temporal resolution. For instance, analysis of high resolution (~ 1 km) numerical weather prediction (NWP) simulations in the SAM for various hydrometeorological regimes using different Weather Research and Forecasting (WRF) physical parameterizations concluded that the prediction of cloud development and cloud vertical microphysical structure are inadequate to capture the spatial and temporal resolution of precipitation rate and precipitation microphysics at the ground (Wilson and Barros, 2015, 2017). In particular, six microphysical parameterization schemes available in WRF were examined to investigate the spatiotemporal evolution of low level moisture fields in the SAM under weak and strong synoptic conditions. However, the simulations could not capture persistent low-level clouds and fog (LLCF) and in particular the mid-day rainfall peak observed in this region (Duan and Barros, 2017b;Wilson and Barros, 2015). Furthermore, simulations exploring the use of different planetary boundary layer (PBL) parameterizations in WRF could not replicate the observed vertical structure of LLCF, thus failing to reproduce the reverse orographic enhancement linked to seeder-feeder interactions (SFI), and consequently resulting in significantly lower rainfall intensities as compared to the surface observations (Duan and Barros, 2017b;Wilson and Barros, 2014, 2015, 2017).

An alternative modelling approach to investigate ACI at fine spatial resolution is the cloud parcel model (CPM) to simulate aerosol activation and cloud droplet growth, as well as thermodynamic adaptation of ascending air parcels at μm and ms scales (Abdul-Razzak et al., 1998;Cooper et al., 1997;Flossmann et al., 1985;Jacobson and Turco, 1995;Kerkweg et al., 2003;Nenes et al., 2001;Pinsky and Khain, 2002;Snider et al., 2003). A synthesis of model formulation including spectral binning strategy, principal physical processes (i.e., condensational growth, collision-coalescence, entrainment), and key aspects of their numerical implementation is presented in Table 1 for CPMs frequently referred to in the peer-reviewed literature. The CPM used in this study (the Duke CPM, DCPM) solves explicitly the cloud microphysics of condensation, collision-coalescence, and lateral entrainment processes (first reported by Duan et al. 2017, Duan 2017). The DCPM was developed to be coupled to an existing rainfall microphysics column model describing the dynamic evolution of raindrop size distributions (bounce, collision-coalescence, and breakup mechanisms) (Prat and Barros, 2007b; Prat et al., 2012;Testik et al., 2011), which in turn is coupled to a radar model. The overarching motivation for the coupled process model is to simulate end-to-end ACPI from CN activation to raindrop at the ground and investigate what is the impact of the space-time heterogeneity of aerosol on the vertical structure of warm season precipitation in the SAM, and ultimately how this affects radar reflectivity measurements and consequently quantitative precipitation estimation (QPE). Here, the focus is strictly on ACI leveraging HYPHEx observations to drive, constrain and evaluate the model.

The manuscript is organized as follows. The mathematical formulation of the cloud parcel model is briefly described in Sect. 2. Section 3 presents the IPHEx measurements relevant for the modelling study. In Sect. 4, model sensitivity tests are conducted by varying key parameters of ACI within their physically-meaningful ranges and are compared against in situ observations to identify major contributors to cloud formation over the complex terrain of the SAM. Finally, a discussion of the main findings and a brief outlook of ongoing and future research are presented in Sect. 5.

2 Model description

To investigate the evolution of cloud droplet spectra originating from aerosol distributions of uniform chemical composition, a new cloud parcel model (hereafter DCPM, or Duke CPM for specificity) was developed to explicitly solve key cloud microphysical processes (see the last row of Table 1 for details). The model synthesizes well-established theory and physical parameterizations in the literature. In particular, condensation and lateral homogeneous entrainment follow the basic formulations of Pruppacher and Klett (1997) and Seinfeld and Pandis (2006) albeit modified to incorporate the single parameter representation of aerosol hygroscopicity (Petters and Kreidenweis, 2007). The representation of collision-coalescence processes takes into account the variation of collision efficiencies with height (Pinsky et al., 2001), and the effects of turbulence on drop collision efficiency as per Pinsky et al. (2008). Droplet sedimentation is not simulated explicitly in the DCPM because the hydrometeors are very small, and thus terminal velocities are negligible compared to the parcel updraft, although an Eulerian-Lagrangian framework is available from Prat and Barros (2007a,b) when terminal velocities must be considered.

The model discretizes the droplet spectra on a finite number of bins (*nbin*) using a discrete geometric volume-size distribution, spanning a large size range with fewer bins and very fine discretization in the small droplet sizes to improve computational efficiency (Kumar and Ramkrishna, 1996; Prat and Barros, 2007a,b). The characteristic single-particle volumes in adjacent bins are expressed as $v_{i+1} = V_{rat}\, v_i$, where V_{rat} is a constant volume ratio (Jacobson, 2005). When condensation and coalescence are solved simultaneously, a traditional stationary (time-invariant) grid structure often introduces artificial broadening of the droplet spectrum by reassigning droplets to fixed bins through interpolation, that is numerical diffusion (Cooper et al., 1997;Pinsky and Khain, 2002). To eliminate numerical diffusion artefacts, a moving grid structure is implemented so that an initial size distribution based on a fixed grid discretization can change with time according to the condensational growth. This approach allows particles in each bin to grow by condensation to their exact transient sizes without partitioning between adjacent size bins. Subsequently, collision and coalescence are resolved on the moving bins that evolve from condensation. The DCPM predicts number and volume concentrations of cloud droplets and interstitial aerosols, liquid water content (LWC), effective drop radius, reflectivity and other moments of DSD. It also tracks thermodynamic conditions (e.g., supersaturation, temperature, pressure) of the rising air parcel. The flowchart in Fig. 2 graphically describes the key elements and linkages in the parcel model, including microphysical processes, and main inputs and outputs. A detailed description of the formulation of key processes in presented next. A glossary of symbols as well as additional formulae are summarized in Appendix A. The performance of the DCPM was first evaluated by comparing its dependence on different parameters with the results from the numerical simulations reported by Ghan et al. (2011) as shown in Sect. S1 of the Supplemental information. Specifically, Figs S1-S6 demonstrate that the simulated maximum supersaturation and number fraction activated from the DCPM are in good agreement with the numerical solutions in Ghan et al. (2011) for a wide range of updraft velocities, aerosol number concentrations, geometric mean radii, geometric standard deviations, hygroscopicity, and condensation coefficients.

2.1 Condensation growth with entrainment

The time variation of the parcel's temperature (T) can be written as

$$-\frac{dT}{dt} = \frac{gV}{c_p} + \frac{L}{c_p}\frac{dw_v}{dt} + \mu\left[\frac{L}{c_p}(w_v - w_v{}') + (T - T')\right]V \tag{1}$$

where the first two terms on the right-hand side represent adiabatically cooling of a rising parcel and the third term describes the modulation by entraining ambient dry air with entrainment rate μ. The vertical profiles of ambient temperature (T') and water vapour mixing ratio (w_v') can be interpolated from input sounding data from atmospheric model simulations or radiosonde observations.

The change of the water vapour mixing ratio (w_v) in the parcel over time is described by

$$\frac{dw_v}{dt} = -\frac{dw_L}{dt} - \mu(w_v - w_v' + w_L)V \tag{2}$$

The change of the parcel's velocity (V) is given by

$$\frac{dV}{dt} = \frac{g}{1+\gamma}\left(\frac{T - T'}{T'} - w_L\right) - \frac{\mu}{1+\gamma}V^2 \tag{3}$$

Where the parameter $\gamma = 0.5$ to include the effect of induced mass acceleration introduced by Turner (1963).

Due to significant uncertainties and complexities of entrainment and turbulent mixing (Khain et al., 2000), only lateral entrainment that mixes in ambient air instantaneously is considered in the DCPM. Based on observations from McCarthy (1974), the entrainment rate (μ) is represented by an empirical relationship that describes the influx of air and ambient particles into the parcel as varying inversely with cloud radius R. To predict the evolution of cloud radius, two classical conceptual models of lateral entrainment are available in the DCPM: the bubble model (Scorer and Ludlam, 1953) and the jet model (Morton, 1957).

For the bubble model, the change of the radius of a thermal bubble (R_B) over time is given as

$$\frac{dlnR_B}{dt} = \frac{1}{3}\left(\mu_B V - \frac{dln\rho_a}{dt}\right) \tag{4}$$

where $\mu_B = C_B/R_B$ and $C_B = 0.6$ as per McCarthy(1974).

For the jet model, the time variation of the radius of a jet plume (R_J) is expressed by

$$\frac{dlnR_J}{dt} = \frac{1}{2}\left(\mu_J V - \frac{dln\rho_a}{dt} - \frac{dlnV}{dt}\right) \tag{5}$$

where $\mu_J = C_J/R_J$ and $C_J = 0.2$ as per Squires and Turner (1962).

The condensational growth rate of droplets in the i^{th} bin ($i = 1, 2, \ldots, nbin$) is represented as

$$\frac{dr_i}{dt} = \frac{G}{r_i}\left(S - S_{eq,i}\right) \tag{6}$$

where droplet growth via condensation is driven by the difference between the ambient supersaturation (S) and the droplet equilibrium supersaturation ($S_{eq,i}$, see Eq. A4 in Appendix A). The growth coefficient (G) depends on the physicochemical properties of aerosols (see Eq. A1 in Appendix A). Microscale perturbations in supersaturation due to air flow around individual hydrometeors, that is ventilation of the drop boundary layer, are implicitly parameterized by the modified diffusivity parameter used in the growth factor equation, which depends on particle size (Eq. A2, Appendix A).

Assuming $S \ll 1$, then $(1+S) \approx 1$, and the time variation of the supersaturation in the parcel can be expressed as

$$\frac{dS}{dt} = \alpha V - \gamma\left(\frac{dw_L}{dt} + \mu V w_L\right) + \mu V\left[\frac{LM_w}{RT^2}(T - T') - \frac{pM_a}{e_s M_w}(w_v - w_v')\right] \tag{7}$$

where α and γ depend on temperature and pressure (see Eq. A5 and A6 in Appendix A; see also Korolev and Mazin, 2003).

During the parcel's ascent, entrainment mixes out cloud droplets and interstitial aerosols inside the parcel and brings in dry air and aerosol particles from the environment. Entrained aerosols are exposed to supersaturated conditions in the parcel. Some of them become activated and continuously grow into cloud droplets. The rate of change in droplet number in the i^{th} bin (i = *1, 2,…, nbin*) due to entrainment is

$$\left(\frac{dN_i}{dt}\right)_{ent} = -\mu V(N_i - N_i') \tag{8}$$

where *N′(z)* is the number concentration of ambient aerosol particles (i.e. outside the cloud) at altitude z.

The rate of change in liquid water mixing ratio (w_L) in the parcel is calculated as follows

$$\frac{dw_L}{dt} = \frac{4\pi\rho_w}{3\rho_a}\sum_{i=1}^{nbin}\left(3N_i r_i^2 \frac{dr_i}{dt} + r_i^3 \frac{dN_i}{dt}\right) \tag{9}$$

2.2 Collision-coalescence growth

To describe droplet growth by collision-coalescence process, the stochastic collection equation (SCE) that solves for the time rate of change in the number concentration is written following Hu and Srivastava (1995)

$$\frac{\partial N(v)}{\partial t} = \frac{1}{2}\int_0^v N(v-v',t)N(v',t)C(v-v',v')dv' - N(v,t)\int_0^\infty N(v',t)C(v,v')dv' \tag{10}$$

where the first integral on the right-hand side of the equation describes the production of droplets of volume *v* resulting from coalescence of smaller drops, and the second integral accounts for the removal of droplets of volume *v* due to coalescence with other droplets. The continuous SCE is discretized and numerically solved by a linear flux method as outlined by Bott (1998). This method is mass conservative, introduces minimal numerical diffusion, and is highly computationally efficient (Kerkweg et al., 2003;Pinsky and Khain, 2002). As noted before, the collision-coalescence process is calculated on a moving grid with bins modified by condensational growth at each time step.

For two colliding drops of volume of *v* and *v′*, the coalescence kernel *C(v, v′)* in Eq. (10) is computed as the product of the gravitational collision kernel *K(v, v′)* and the coalescence efficiency $E_{coal}(v, v')$,

$$C(v,v') = K(v,v')E_{coal}(v,v') \tag{11}$$

$$K(v,v') = (9\pi/16)^{\frac{1}{3}}\left(v^{\frac{1}{3}} + v'^{\frac{1}{3}}\right)^2 |V_t - V_t'|E_{coll}(v,v') \tag{12}$$

where $V_t(V_t')$ is the terminal velocity of drop volume *v (v′)* and $E_{coll}(v, v')$ is the corresponding collision efficiency.

E_{coal} is parameterized following Seifert et al. (2005), who applied Beard and Ochs (1995) for small raindrops (d_S < 300 µm), Low and List (1982) for large raindrops (d_S > 600 µm), and used an interpolation formula for intermediate drops (300 µm < d_S < 600 µm) where d_S is the diameter of the small droplet. A simpler and faster option suggested by Beard and Ochs (1984) is also available in the model. The terminal velocity of cloud drops is estimated following Beard (1976) in three ranges of the particle diameter (0.5 µm–19 µm, 19 µm–1.07 mm, 1.07 mm–7 mm). Another approximation by Best (1950) is also available as an option in the model. The table of drop-drop collision efficiencies at 1-µm resolution developed by Pinsky at al. (2001) is used for E_{coll}. This table was created based on simulations of hydrodynamic droplet interactions over a broad range of droplet radii (1–300 μm), including collisions among small cloud droplets as well as between small cloud droplets and small raindrops. Moreover, E_{coll} was derived at three pressure levels of 1,000-, 750-, and 500-mb and can be interpolated at each level of a rising cloud parcel, thus taking the increase of E_{coll} with height into account. Turbulence can significantly enhance collision rates especially for small droplets (below 10 μm in radii) as it increases swept volumes and collision efficiencies, and influences the collision kernels and droplet clustering (Khain and Pinsky, 1997;Pinsky et al., 1999;Pinsky et al., 2000). Considering different turbulent intensities for typical stratiform, cumulus, and cumulonimbus clouds, detailed tables of collision kernels and efficiencies in turbulent flow, created by Pinsky et al. (2008) for cloud droplets with radii below 21 μm, are also incorporated in the model.

2.3 Numerical formulation

The equations in Sect. 2.1 constitute a stiff system of non-linear, first-order ordinary differential equations and involve state variables at very different scales. For the numerical integration of condensation growth, a fifth-order Runge-Kutta scheme with Cash-Karp parameters (Cash and Karp, 1990) using adaptive time steps (Press et al., 2007) is employed. At each time step, the error is estimated using the fourth-order and the fifth-order Runge-Kutta methods. Because dependent variables differ by several orders of magnitude, a fractional error (ε) is defined to scale the error estimate by the magnitude of each variable. Specifically, the step size is adaptively selected to satisfy a fractional tolerance of 10^{-7} for all variables. The initial time step to calculate condensational growth is 5×10^{-4} s. The maximum time step is set as 10^{-3} s to ensure the diffusional growth of drops is precisely simulated and non-activated particles reach equilibrium with the parcel supersaturation at each time step. For the collision-coalescence processes in Sect. 2.2, a simple Euler method is applied to integrate forward in time. The flux method for solving the discrete SCE was demonstrated to be numerically stable for various grid structures and integration time steps when the positive definiteness is maintained (Bott, 1998). Thus, a time increment of 0.2s is chosen to assure that the available mass in each bin is much larger than the change of mass in the bin during the redistribution of the mass at one time step. Relying on separate numerical integration methods for calculating condensation and collision-coalescence allows us to either include or exclude each process easily to examine its role individually in cloud formation.

3 IPHEx data

The intense observing period (IOP) of the IPHEx field campaign took place during 01 May–15 June, 2014. The study region was centred on the SAM extending to the nearby Piedmont and Coastal Plain regions of North Carolina (see maps in Fig. 1). IPHEx was one of the ground validation campaigns after the launch of NASA's Global Precipitation Mission (GPM) core satellite, and details about this campaign can be found in the science plan (Barros et al. 2014). Surface measurements in the inner region of the SAM were conducted in the Pigeon River Basin (PRB, Fig. 1b) including a dense network of raingauges and disdrometers. During the IPHEx IOP, measurements of aerosol concentrations and size distributions ranging from 0.01 to 10 μm were collected at the ground level in the Maggie Valley (MV), a tributary of the Pigeon River. Collocated with aerosol instruments at the MV supersite, the ACHIEVE (Aerosol-Cloud-Humidity Interaction Exploring & Validating Enterprise) platform was also deployed, equipped with W-band (94 GHz) and X-band (10.4 GHz) radars, a ceilometer, and a microwave radiometer. Two aircraft were dedicated to the IPHEx campaign. The NASA ER-2 carried multi-frequency radars (e.g., a dual-frequency Ka-/Ku-, W-, X-band) and radiometers, and functioned as the GPM core-satellite sampling simulator from high altitude. The University of North Dakota (UND) Citation aircraft was instrumented to characterize the microphysics and dynamical properties of clouds, including LWC and DSDs from cloud to rainfall drop sizes. Therefore, this data set offers a great opportunity to investigate ACI tied to warm season moist processes in complex terrain. Detailed description of the specific measurements relevant to this study is provided below and in Sect. S2.

3.1 Surface measurements

Aerosol observations were carried out at the MV supersite (marked as the yellow star in Fig. 1b) in the inner mountain region during the IPHEx IOP. The elevation of the MV site is 925 m mean sea level (MSL). This data set provides a clear characterization of the size distribution and hygroscopicity of surface aerosols in this inner mountain valley, which was not available previously. Nominal dry aerosol size distributions at the surface were measured by a scanning mobility particle counter system (SMPS) for particles from 0.01 to 0.5 μm in diameter, and a passive cavity aerosol spectrometer (PCASP; manufactured by Droplet Measurement Technologies, Inc., Boulder, CO, USA) for particle diameters in the size range of 0.1–10 μm. The SMPS consists of an electrostatic classifier (TSI Inc. 3081) and a condensation particle counter (CPC, TSI 3771). Note that the relative humidity (RH) of the differential mobility analyser (DMA) column is well controlled and the average RH (± one standard deviation) of the sheath and sample flows are 2.0±0.8% and 3.2±0.5%, respectively. In addition, a co-located ambient CPC (TSI 3772), which measures aerosol particles greater than 10 nm without resolving their size distributions, shows very close agreement with the SMPS measurements with regard to total number concentrations of aerosol particles (N_{CN}). Size-resolved CCN concentrations (N_{CCN}) were sampled by a single column CCN counter (manufactured by Droplet Measurement Technologies, Inc., Boulder, CO, USA) that was operated in parallel to the SMPS-CPC. The CCN instrument cycles through 6 levels of supersaturation (S) in the range of 0.09–0.51%. At a given S level, each CCN measurement cycle took approximately 8 min, corresponding to one SMPS-scan and buffer time to adjust supersaturation. On average 178 measurement cycles were collected daily during the IPHEx IOP, except for occasional interruptions due to instrument maintenance. CN and CCN distributions were inverted as described previously (Nguyen et al., 2014;Petters and Petters, 2016). Supersaturation was calibrated using dried ammonium sulfate and a water activity model (Christensen and Petters, 2012;Petters and Petters, 2016). The midpoint activation diameter (D_{50}) is derived from the inverted CN and CCN distributions (Petters et al., 2009). The hygroscopicity parameter (κ) is obtained from D_{50} and instrument supersaturation (Petters and Kreidenweis, 2007). Detailed time series and diurnal cycles of CN and CCN measurements are presented in in Supplementary Sect. S2 (Figs. S7 – S9). The data show that the average total number concentration (± one standard deviation) of dry aerosol particles is 2,487±1,239 cm^{-3} for particles with diameters between 0.01 to 0.5 μm, and 1,106±427 cm^{-3} for particles with diameters between 0.1 and 10 μm in diameter. No significant diurnal variability in number concentration and hygroscopicity were observed. In addition, a co-located Vaisala weather station (WXT520) was continuously recording local meteorological conditions (e.g., wind speed, wind direction, relative humidity, temperature, and pressure) at 1-s interval. Diurnal cycles of these local meteorological variables during the IPHEx IOP are displayed in Fig. S10. The average meteorological conditions at the sampling site are 0.8±0.6 m s^{-1} in wind speed, 172±115° in wind direction, 77±18% in relative humidity, 19±4 °C in ambient temperature (arithmetic mean ± one standard deviation).

3.2 Aircraft measurements

Airborne observations from the UND Citation, equipped with meteorological (e.g., temperature, pressure, humidity) sensors and microphysical instruments, are used in this study (Poellot, 2015). Vertical velocity was obtained from a gust probe and bulk LWC values were retrieved from two hot-wire probes (a King-type probe and a Nevzorov probe). Size-resolved concentrations were measured from three optical probes, covering droplet diameter from 50 μm to 3 cm: a PMS two-dimensional cloud (2D-C) probe, a SPEC two-dimensional stereo (2D-S) probe, and a SPEC high volume precipitation spectrometer 3 (HVPS-3) probe. The cloud droplet probe (CDP) measures cloud drop concentrations and size distributions for small particles with diameters from 2 to 50 μm in 30 bin sizes. The droplet sizes are determined by measuring the forward scattering intensity when droplets transit the sample area of the CDP. Coincidence errors have been found to cause CDP measurement artefacts, which tend to underestimate droplet concentrations and broaden droplet spectra. This type of error occurs when two or more droplets pass through the CDP laser beam simultaneously, and is highly dependent on droplet concentrations (Lance et al., 2010). Correction of the CDP observations is described in Sect. S2.2 (Fig. S11). The corrected CDP measured cloud droplet distributions were used in this study mainly to compare with model simulated cloud microphysics. The corrected CDP spectra slightly shift the measurement to smaller drop sizes (not shown here), thus providing confidence in the performance of the CDP probe during the IPHEx campaign.

3.3 IPHEx case-study: 12 June 2014

On 12 June 2014, the W-band radar observations at MV (see Fig. S12) indicate the formation of cumulus congestus clouds before 12:30 local time (LT) and further growth into cumulonimbus clouds. Near the MV site, a coordinated aircraft mission of both the UND Citation and NASR ER-2 was conducted from 12:14 to 15:51 LT on 12 June. Cloud droplet concentrations and size distributions were sampled by conducting successively higher constant-altitude flight transects through clouds. Droplet spectra were sampled at 1-Hz resolution (corresponding to approximately 90 m in flight distance) by the CDP and coincidence errors were corrected as described in Sect. S2.1. In particular, the lowest horizontal leg (see the flight track in Fig. 3a, altitude around 2,770–2,800 m MSL) through the cloud is investigated to avoid the influence of substantial mixing in the upper portion of the cloud. The flight period of the first horizontal leg (~ 2,800 m MSL) is from 12:17 to 12:28 LT. In rising updrafts, in-cloud samples (white plus signs in Fig. 6a)) are defined with a minimum LWC of 0.25 g m^{-3} from the CDP. The drop number concentrations from the 2-DC probe (measuring hydrometeors with diameter between 105 μm and 2 mm) indicate negligible amount of precipitation-sized drops in these cloudy regions, indicating the sampling of cumulus congestus clouds development by the aircraft. Significant topographic heterogeneity (terrain transect indicated by the thick black line in Fig. 3b) can exert a considerable influence on cloud formation across this region. As shown in Figs. 3c and d, a pronounced variability in drop number distributions is manifest in the in-cloud samples clustered by low (0–1 m s^{-1}) and high (1–2 m s^{-1}) updrafts. Along the first leg, three cloudy regions are identified near the eastern ridges (ER, highlighted in the blue dashed box in Fig. 3), over the inner valley region (IC, highlighted by the blue circle), and near the Eastern Cherokee Reservation (ECR, highlighted in the blue dashed box). Measurements of in-cloud samples for three regions are discussed in Sect. S2.2.

A total of 11 samples was collected over approximately 1 km flight distance in cloud region IC (circled in Fig. 3a, vertical velocities shown as blue bars in Fig. 3b). The droplet spectra in stronger updrafts (see Fig. 3d) have higher number concentrations and narrower size range compared to the samples in the weaker updrafts at the edge of the cloud (see Fig. 3c). This is because in the stronger (faster) updrafts the time-scale of vertical motion is very short, thus thwarting entrainment and collision-coalescence processes with condensation alone governing droplet growth. In the slower updrafts, the longer time-scale of vertical motion enhance entrainment leading to replenishment of CN, and more importantly collision-coalescence processes to produce larger droplets, thus broadening the distribution. Aerosol size distributions are not resolved in the CPC measurements from UND Citation, and thus surface measurements at MV (marked as the black asterisk in Fig. 3a) are used as model input at IC.

4. Modelling Experiments

4.1 Model initialization and reference simulation

Dry aerosol concentrations measured by the SMPS and PCASP at MV were averaged over the first 10 mins (averaging interval: 12:14 LT–12:24 LT) of the 12 June flight, and then merged into a single size distribution as shown in Fig. 4. The combined aerosol distribution at the surface is fitted by the superimposition of four lognormal functions using least-squares minimization. Table 2 summarizes parameters (total number concentration, geometric mean diameter, and geometric standard deviation) that characterize the four lognormal distributions. Notice that aerosol number concentrations below 0.03 μm are underestimated by the fitted cumulative distribution (cyan curve in Fig. 4). The CPM numerical configuration parameters are presented in Table 3a, and the model physiochemical parameters and initial conditions are highlighted in Table 3b.These particles in such small sizes mostly remain non-activated under the supersaturated conditions typical of the atmosphere, thus, underestimation of their concentrations does not affect cloud development in the model. The initial aerosol distribution is discretized into up to 1,000 bins covering the size range of 0.01–10 μm. The grid evolves with new bins added as larger particles form by condensational growth. The bin grid at such a high resolution is sufficient to precisely simulate the partitioning of growing droplets and interstitial aerosols in the parcel. It is also assumed that the aerosol is internally mixed so that the hygroscopicity does not vary with particle size. Thus, we prescribe a κ value of 0.14 for each aerosol bin, deriving from the average κ during the first 10 mins of the 12 June flight.

During the IPHEx IOP, daytime radiosondes were launched every 3-hours at Asheville, NC (red star in Fig. 1b). This location is on the eastern slopes of the SAM in the French-Broad valley outside of the inner mountain region far away from the targeted cloudy region (IC). In addition, the closest sounding (11 LT) was launched much earlier than the flight take-off time on 12 June 2014. To address the lack of sounding observations needed for CPM input, high-resolution (0.25-km grid size) WRF simulations were conducted to extract model soundings in the IC region (highlighted in Fig. 3a). Detailed configuration of the WRF model for these simulations (see Fig. 5a for nested grid domains) is described in Sect. S3. Upon inspection of model results 15 min prior to the flight time, the ensemble mean of six-simulated soundings in valley locations within the IC region at 12:15 LT was used the process-study (Fig. 5b). The cloud base height (CBH) is chosen as the level where simulated RH is approximately 100%. As marked by the horizontal black line in Fig. 5b, CBH = 1,270 m AGL at 12:15 LT when the parcel is released from cloud base. The vertical distribution of simulated horizontal winds along the aircraft flight path is highly heterogeneous and anisotropic due to the complex 3D structure of winds in the complex terrain of the inner mountain region including shallow thermal upslope winds between the main valley and surrounding ridges and ridge-valley circulations with multiple orientations in lateral tributary valleys as illustrated by the supplementary animations SA1 (near surface), SA2 (at ridge level), SA3 (at CBH) and SA4 (along the first aircraft flight leg). The animations show southerly mesoscale horizontal transport above ridges, upslope flows along the topography in the inner region, and a mesoscale honeycomb like structure of weak to moderate updrafts and downdrafts with short-lived intensification linked to overturning processes across the entire region.

At the cloud base of IC, aerosol size distributions are estimated by assuming that total number concentrations at the surface decay exponentially with a reference scale height (H_S) of 1,000 m (representative of the effectiveness of the vertical venting mechanism), and geometric mean diameters and corresponding geometric standard deviations remain constant with height. The reference H_S selected for the control simulation is the height above ground level where the Lifting Condensation level (LCL) and the Convective Boundary Layer (CBL) are the same (i.e. $H_S \sim$ CBL~ LCL) in the SAM inner valley region under the flight path. The dry aerosol distribution at cloud base is calculated as the sum of four lognormal distributions with fitted parameters indicated in the last three columns of Table 2 and is taken as initial input to the model. The number concentration of entrained ambient aerosol particles (see Eq. 8) is also calculated based on the assumption that the initial aerosol distribution at the surface $N(0)$ decays exponentially with height: $N'(z)=N(0)exp(-z/H_S)$, where z is the height above ground level (AGL) and H_S is the scale height, depending on aerosol types (Kokhanovsky and de Leeuw, 2009).

The temperature excess of the air parcel over the environment is initialized as 1.0 K, and the initial pressure and RH of the parcel at cloud base adapt to cloud surroundings. As vertical velocities were not sampled at cloud base, the initial updraft velocity (V_0) at cloud base is assumed to be uniformly distributed and equal to 0.5 m s^{-1}, consistent with vertical velocities observed by the W-band radar (see Fig. S12b) and simulated by the model around the same altitude (2.5 km MSL). Therefore, the air parcel is launched at cloud base with an initial parcel radius (R) of 500 m, an initial updraft of 0.5 m s^{-1}, and initial aerosol particles that are in equilibrium with the humid air at cloud base. When the parcel is rising, the lateral entrainment is treated as the bubble model parameterization with the characteristic length scale R = 500 m (see Eq. 4 in Sect. 2.1). Ambient aerosol particles penetrate through lateral parcel boundaries and their number concentrations also decrease exponentially with height (H_S = 1,000 m). The turbulent kinetic energy dissipation rate is chosen as 200 cm^2s^{-3}, typical of cumulus clouds at early stages. The parcel reaches cloud top when vertical velocity is near zero. Note that despite specified as stated above for the reference simulation, sensitivity to parcel radius R, scale height H_S, and hygroscopicity κ will be explored in Sect. 4.2.

4.2 Parameter sensitivity analysis

In the past, process studies using CPMs targeted principally aerosol-CDNC closure between model simulations and field observations. For example, Conant et al. (2004) conducted an aerosol-cloud droplet number closure study against observations from NASA's Cirrus Regional Study of Tropical Anvils and Cirrus Layers–Florida Area Cirrus Experiment (CRYSTAL-FACE) using the adiabatic CPM by Nenes et al. (2001;2002) that solves activation and condensation processes only (see Table 1 for details). Using a condensation coefficient (a_c) value of 0.06, they reported that predicted CDNC was on average within 15% of the observed CDNC in adiabatic cloud regions. Fountoukis et al. (2007) used the same CPM as Conant et al. under extremely polluted conditions during the 2004 International Consortium for Atmospheric Research on Transport and Transformation (ICARTT) experiment. They found that the optimal closure of cloud droplet concentrations was achieved when the condensation coefficient was about 0.06. For marine stratocumulus clouds sampled during the second Aerosol Characterization Experiment (ACE-2), Snider et al. (2003) applied the UWyo parcel model (http://www.atmos.uwyo.edu/~jsnider/parcel/) to simulate condensation processes in adiabatic ascent (see Table 1) and experimented with various condensation coefficients in the range of 0.01–0.81. They hypothesized that the lower CDNC overestimation errors (20 to 30% for a_c = 0.1) in their CPM simulations could be mitigated by varying the condensation coefficient as a function of dry particle size instead of using one value for the entire distributions, but did not actually demonstrate this was the case. The condensation coefficient of water is a key ACI physical parameter in parcel models and has a strong influence on activation and droplet growth by condensation as it expresses the probability that vapour molecules impinge on the water droplet when they strike the air-water interface (McFiggans et al., 2006). Experimental measurements reviewed by Marek and Straub (2001) exhibit a strong inverse relationship between pressure and a_c values ranging from 1000 hPa to 100 hPa and from 0.007 to 0.1, respectively (their Fig. 4). Chodes et al. (1974) measured condensation coefficients in the range of 0.02–0.05 with a mean of 0.033 from measurements of individual droplets grown in a thermal diffusion chamber for four different supersaturations. Ganier et al. (1987) repeated Chodes et al.'s experiments and found that the average condensation coefficient is closer to 0.02 after correcting their supersaturation calculations. Shaw and Lamb (1999) conducted extensive simultaneous measurements of the condensation coefficient and thermal accommodation coefficients (a_T) for individual drops in a levitation cell and reported values for a_c and a_T in the ranges of 0.04–0.1 and 0.1–1 with most probable values of 0.06 and 0.7, respectively. Using adiabatic CPMs with laboratory based condensation coefficients, the resulting errors in aerosol-cloud droplet number closure studies are still well above 10% and often around 20–30%, mostly due to overestimation (McFiggans et al., 2006).

In this section, sensitivity tests are conducted to assess changes in DCPM simulations to variations in key inputs and assumptions. Test results are further compared with in-cloud observations from the aircraft to assess the role of individual state variables and processes for the cumulus congestus case-study on 12 June during IPHEx. Selected parameters are perturbed one at a time while other assumptions and input parameters remain as specified in Sect. 4.1. Table 3b presents a summary of the ranges of physiochemical parameters and initial conditions tested in the sensitivity analysis.

4.2.1 Condensation coefficient

Condensation plays a dominant role in the early stages of cloud formation and one key factor in this process is the condensation coefficient (a_c) that governs activation and condensational growth. A laboratory study by Chuang (2003) reported a_c values ranging from 4×10^{-5}–1, and experimental values from field campaigns and from chamber studies of individual droplet growth also differ over a wide range (0.007–0.1) as reviewed in Sect. 1. In this study, a_c was made to vary in the range [0.001, 1.0] on the basis of Fountoukis and Nenes (2005). For the targeted in-cloud region (IC), Fig. 6 shows simulated profiles of updraft velocity, supersaturation, total CDNC, LWC, and their sensitivity to selected a_c values in comparison with the airborne observations (denoted by black crosses). Measurements from the IC region along the lowest cloud transect (highlighted in the blue circle in Fig. 3a) are used to evaluate model performance, since no observations are available in the upper unmixed cloudy areas to assess the entire vertical profiles simulated by the CPM. Only simulations with reasonable agreement with the observations are presented, thus results with a_c from 0.06 to 1.0 are not shown here. Particles above 1 μm in diameter are considered cloud droplets and are included in the integration to calculate LWC. Note that ground elevations under the IC region vary from 928 m to 1,184 m MSL (see Fig. 3b), and the region is on a small hill in the middle of the valley and surrounded by much higher ridges (terrain elevation ~ 1,500 m MSL). Hereafter, aircraft measurements are expressed as AGL to facilitate their comparisons with the model results.

As illustrated in Fig. 6a, large values of a_c (> 0.01) have negligible influence on the vertical velocity profiles. Although it is apparent that a_c has a significant impact on the simulated supersaturation profiles (Fig. 6b). The black crosses indicate the quasi-steady approximation of supersaturation (S_{qs}), calculated according to Eq (A8) (also Eq (3) in Pinsky et al. (2013)). We should keep in mind that large uncertainties are associated with the temperature measurements from the aircraft, which are used to compute the S_{qs}. Low values of a_c strongly inhibit the phase transfer of water vapour molecules onto aerosol particles (aerosol wetting), slowing the depletion of water vapor in the parcel, and thus substantially increasing maximum supersaturation (S_{max}). Consequently, smaller aerosol particles with high concentrations are activated due to a higher S_{max}, resulting in a direct increase in cloud droplet numbers with lower values of a_c (Fig. 6c). Overall, these results are in agreement with earlier studies (Nenes et al., 2002;Simmel et al., 2005) that investigated the dependence of cloud droplet number concentrations on the condensation coefficient. Moreover, Fig. 6c shows that the simulation with a_c = 0.01 (green line) captures well the observed drop concentrations between 1,500 m and 1,600 m AGL (highlighted in yellow shade), whereas a condensation coefficient that is one order of magnitude lower (a_c = 0.002, blue line) yields better results for the observations above 1,600 m. As summarized in Table 4, the simulated CDNC for the region between 1,500 m and 1,600 m AGL along the hillslope (shaded in Fig. 6b, reference sub-region within IC) for α_c = 0.01 attains an average CDNC of 354 cm^{-3}, which is only ~1.3% higher than the observed average between 1,500 m and 1,600 m (349.4 cm^{-3}). The corresponding LWC is also in reasonable agreement with the range of observed values (Fig. 6d). The observed CDNC for the cluster between 1,600 m and 1,750 m (397.5 cm^{-3}) is underestimated by all simulations with different α_c values and the averaged CDNC using a much lower condensation coefficient (0.002) is ~ 8% lower than the averaged observation. Inspection of Fig. 6c suggests that within IC there are two clusters of air parcels at different levels above ground. Interestingly, the cluster at lower elevation (Fig. 3b) is better reproduced using a lower condensation coefficient, whereas a higher condensation coefficient better captures the reference region that includes the maximum updrafts near the hilltop. Good agreement between the model results and airborne observations for the lower cluster provides more confidence in the conclusions about the sensitivity tests. Thus, the lower cluster over the higher terrain (Fig. 3b and 3d) is considered as the reference region within IC and will be the focus of this study.

The sensitivity of predicted spectra at 1,500 m (in solid lines, Fig. 7a) to α_c varying from 0.002 to 0.06 is very high. The observed spectrum (black dotted line) is the average from five individual CDP measurements (dotted lines in Figs. 3c and d, also highlighted in the yellow shaded area in Fig. 3b) between 1,500 m and 1,600 m AGL (see Fig. 6d for their LWC in shade). Generally, spectra simulated with lower values of α_c are broader with higher numbers of small droplets, while simulations with large values of α_c yield narrower spectra shifted to larger droplet sizes. The differences in drop size range and spectra shape can be explained by inspecting the vertical profiles of the parcel supersaturation and S_{eq} for six illustrative aerosol particle diameter (D_{aero}) depicted in Fig. S19. Growth by water vapour condensing on different sizes of cloud droplets is determined by the difference between S and S_{eq} (Eq. 6 in Sect. 2.1). At low S, small particles become interstitial aerosols, and their corresponding S_{eq} remains in equilibrium with the parcel supersaturation ($S - S_{eq} = 0$). At high S, as a result of low α_c values, activation of small aerosols contributes to significant spectra broadening, produces larger CDNC, and shifts the droplet size distribution toward smaller diameters due to slower condensational growth. This is consistent with Warner (1969) who found that low condensation coefficients (< 0.05) were required to capture the observed dispersion of droplet spectra in natural clouds, especially for small sizes (i.e. left-hand side of the spectra). Figure 7b displays the simulated droplet number distributions at different levels for α_c = 0.01 in comparison with the individual droplet spectra measured by the CDP. The simulated spectra are representative of the evolution of cloud droplet distributions in one parcel at different cloud development stages. The observed spectrum at 1,559 m AGL (black dotted line) and its CDNC (357 cm^{-3}) and LWC (0.37 $g\ m^{-3}$) is chosen for the comparison as best agreement is achieved with the DCPM results. The results are also consistent with the parameterizations of CDNC and cloud droplet spectra at different heights given the updraft velocity and the number of CCN that can be activated at moderate supersaturation levels developed by Kuba and Fujiyoshi (2006) using a cloud microphysical model. Simulated spectra at 1,500 m and 1,600 m altitude show very good agreement with the observed number concentration and drop size range. Below 1,600 m, a shift of the unimodal spectra to larger drop sizes suggests that the condensation process currently dominates the growth of cloud droplets. Larger drops above 1,700 m could be produced by coalescence growth, leading to the formation of a second mode at larger sizes in the upper portion of the cloud. For the analyses presented hereafter, we consider a_c = 0.01 together with other initial conditions as prescribed in Sect. 4.1, as the reference simulation (denoted by the grey line in the following figures).

Further examination using data from other cloud and precipitation probes suggests that concentrations of droplets larger than 30 μm in diameter are negligible during the first horizontal flight leg. Considering that droplets with diameters larger than 30–32 μm are required to trigger effective droplet collisions (Pinsky and Khain, 2002), we conclude that the collision-coalescence process is not important in the sampled IC region, and it is unlikely that it contributes to the wide bimodal spectra observed at early stages of cloud growth. It is noteworthy that small drops are absent in the simulated spectra, in contrast to the observed spectrum that exhibits a broad drop size range and two distinct modes (see Fig. 7b). One possible explanation is that the moving bin grid determined by the condensation process tends to widen the spectral gap between the growing droplets and non-activated aerosol particles in the ascending parcel. Thus, a geometric size distribution with 1,000 bins is utilized herein to further refine the discretization for small particle sizes. Another explanation relates to the uncertainties of the input sounding extracted from the WRF simulation. Even though ambient aerosols are entrained continuously through lateral boundaries, most of them remain as interstitial aerosol particles because the low supersaturation in the parcel is insufficient to enable activation (see Fig. 6b). The WRF sounding in Fig. 5b exhibits a lapse rate of -4.1 °C km^{-1} from 1,270 m (CBH) to 2,200 m, corresponding to stable atmospheric conditions unfavourable for cloud development. To assess the impact of the environmental conditions on cloud growth, an additional model simulation was performed by altering lapse rate at lower levels (see Appendix B1). The results point out that uncertainties of the assumed environmental thermodynamic conditions (e.g., temperature) impose significant constraints in the vertical development of clouds, thus posing as a significant challenge in cloud modelling studies.

4.2.2 Entrainment strength

To access the influence of entrainment on cloud drop concentrations and LWC, different strengths of lateral entrainment are examined by altering the initial cloud parcel size R at the cloud base. Figure 8 displays the vertical profiles of total CDNC and LWC, and cloud droplet spectra formed at three altitudinal levels (1,500 m: solid line, 1,600 m: dotted line, and 1,700 m: dashed line) for simulations using different initial parcel radii as compared to the CDP observations in the IC region (denoted by black crosses in Figs. 8a and b and the black dotted line in Fig. 8c). Entrainment appears to have a dominant influence on the cloud vertical structure as small rising parcels associated with higher entrainment dissipate faster by intensive mixing of dry ambient air through lateral cloud boundaries. Stronger entrainment strength results in a direct decrease in drop concentrations and LWC, while it has little influence on the droplet size range. The best agreement on droplet numbers is between the reference simulation (R = 500 m, a_c = 0.01; grey line in Fig. 8a) and the reference sub-region within IC (between 1,500 m and 1,600 m AGL), whereas results for R = 1,500 m better captures the higher cluster of cloudy samples (above 1,600 m AGL). Recall that previously, when R was held constant the higher cluster is better reproduced using a_c values one order of magnitude smaller than the reference value. Thus, the sensitivity analysis does suggest there is a trade-off with weaker entrainment for a higher condensation coefficient (R = 1500 m and a_c = 0.01; orange line in Fig. 8a) when other parameters in the reference simulation remain the same.

Given R = 500 m, an additional test was conducted using the jet model parametrization of lateral entrainment (Eq. 5 in Sect. 2.1). The comparison of two entrainment parameterizations indicates that the bubble model (the grey line) has stronger entrainment strength than the jet model (red line) given the same initial parcel size (500 m). Nevertheless, continuous increases in simulated LWC in the upper portion of the cloud (see Fig. 8b) for both parameterizations are unrealistic in real clouds (Paluch, 1979). This problem can be likely ascribed, at least in part, to the uncertainties in the environmental conditions associated with the WRF sounding. As noted in Fig. B1, decreases in LWC are manifest at the upper portion of the cloud, as indicated in the simulations with modified sounding inputs. The lack of sufficient mixing with dry ambient air near cloud top is an inherent deficiency in the simple parameterization of lateral homogenous entrainment, assuming decreasing entrainment strength with height, but this assumption does not significantly affect our conclusions for in-cloud regions below cloud top.

4.2.3 Initial aerosol concentration

The initial aerosol concentration at cloud base can also have significant effects on cloud development. Because aerosol size distributions were not sampled by the aircraft during IPHEx, they are estimated by extrapolating surface aerosol number concentrations according to an exponential decay with a given scale height (H_S). To probe and characterize the dependence of droplet formation on aerosol concentrations available at cloud base, sensitivity to H_S was explored by varying its values from 800 m to 1,200 m, which correspond to the range of LCL at valley locations along the flight (Supplement Sections S3 and S4). Figure 9 shows the simulated profiles of the total CDNC and LWC, and cloud droplet spectra formed at three altitudinal levels (1,500 m: solid line, 1,600 m: dotted line, and 1,700 m: dashed line). It is not surprising that aerosol concentrations at cloud base have a substantial influence on the resulting droplet concentrations. Higher aerosol concentrations, inferred from larger H_S lead to larger drop numbers with smaller average droplet sizes, which is known as the first indirect effect of aerosols (Twomey, 1977). Yet, here, LWC appears insensitive to the initial aerosol concentration as it is limited by moisture content available in the parcel. The reference value of H_S = 1,000 m yields the best agreement in CDNC between the DCPM simulations and the average droplet spectra observed by the CDP (black dotted line in Fig. 9c, see reference sub-region within IC shaded in Fig. 3b), which lies within the typical H_S range (550–1,100 m) of aerosol number concentration measurements for remote continental type (Jaenicke, 1993).

Because of the uncertainty in the characterization of environmental conditions due to the lack of sounding observations and the complexity of 3D circulations in the inner mountain region, an additional set of CPM simulations were conducted assuming a well-developed and well-mixed CBL, and thus uniform distribution of dry aerosol concentrations below CBH. This enables contrasting the results using the well-mixed CBL and the vertical venting mechanism to pump low level aerosol to the atmosphere above the mountain ridges. The modelling results are present and discussed in Sect. S4. The surface aerosol concentration at MV (see Fig. 4) is used as model input at cloud base, and other input parameters remain as specified in Sect. 4.1. Although there is good agreement in CDNC between simulations with surface aerosols at cloud base and the airborne observations as expected using a conservative CPM, there are large discrepancies between the observed and simulated cloud droplet spectra with respect to spectral width, peak diameter and peak concentration number above CBH. Nevertheless, aerosols in the atmosphere exhibit a significant space-time variability especially in regions of complex terrain with heterogeneous mixing by different ventilation processes in addition to the possibility of remote transport (e.g., see for example Figs. 3, 5, and 11 in De Wekker and Kossmann, 2015), all of which can contribute to the diverse cloud droplet spectra observed across the cloud transect (see Figs. S15a-d). This cannot be captured by the current model simulations that assume a homogenous aerosol distribution at cloud base and 1D vertical flow (updraft) with parameterized lateral entrainment. .

4.2.4 Hygroscopicity

Another key element in the condensation process is the hygroscopic property that governs the influence of aerosol chemical composition on CCN activity. To account for its temporal variability observed during IPHEx, a κ value varying from 0.1–0.4 (within the typical range measured at the surface site, see Figs. S8a and S9c) is applied uniformly for all particle sizes. As noted from Fig. 10, simulated profiles of total CDNC and LWC exhibit a weak dependence on the hygroscopicity with only a slightly increase in total CDNC with more hygroscopic aerosols. Predicted droplet spectra at three altitudinal levels (1,500 m: solid line, 1,600 m: dotted line, and 1,700 m: dashed line) also show little sensitivity to the variations in κ. As discussed in Sect. S2, hygroscopic properties of aerosols have been found to vary with particle sizes. Potential uncertainties might remain by assuming a constant κ, but its variation with droplet sizes is not addressed in the current study. We should also note the hygroscopicity derived from surface measurements may not be representative for aerosols beneath the cloud (Pringle et al., 2010). However, the vertical variability of aerosol hygroscopicity is not taken into account in this study.

4.2.5 Summary of sensitivity analysis

Under realistic assumptions, the total number concentration and size distributions from the airborne observations are captured well by the reference simulation. Sensitivity tests by changing a_c in the range of 0.001–1.0 suggest that the predicted CDNC, LWC, and thermodynamic conditions are highly dependent on the condensation coefficient. At early stages of cloud development, the condensation coefficient plays a key role in the simulated spectra width and shape that increases in a_c lead to a shift towards larger droplet sizes and narrower spectra widths. Entrainment has a substantial impact on the cloud depth, droplet numbers, and LWC, whereas initial aerosol concentrations have a strong effect on number concentrations and size distributions of cloud droplets, but induce little effects on LWC. Hygroscopicity has negligible influence on simulated total CDNC and LWC. Additional tests regarding the sounding inputs and initial updraft velocity were conducted and discussed in Appendix B.

Due to the limited dataset from the campaign, a specific set of initial conditions are inferred from surface and airborne observations and reasonable assumptions are made based on the literature and WRF model results. It is important to keep in mind the uncertainties associated with the determination of CBH, which is estimated from the WRF model simulations as concurrent soundings are not available during IPHEx. If the CBH is lifted by 100 m, simulations using different a_c values (0.002 – 0.06) are in better agreement with the airborne measurements of LWC. The CDNC in the reference region (yellow shade, Fig. 6c) is captured better with a higher a_c value (0.015) but narrower spectra results are associated with increasing a_c values, inconsistent with the observed spectra (not shown here). These caveats highlight the need for comprehensive concerted observations of end-to-end processes in future field campaigns.

Based on the sensitivity tests, model simulations using a relatively low value of a_c (0.01) exhibit CDNC and spectra consistent with the cloud spectra observed in the inner region of the SAM for early development of cumulus congestus on 12 June. Assuming adiabatic conditions (i.e. in the absence of entrainment) and for aircraft observations near cloud base (discussed in Sect. 1), higher values of the condensation coefficient have been specified in previous studies such as $a_c = 0.06$ for warm cumulus during CRYSTAL-FACE (Conant et al., 2004), a_c = 0.042 for stratocumulus during Coastal Stratocumulus Imposed Pertubation Experiment (CSTRIPE, Meskhidze et al., 2005), and a_c = 0.06 for cumuliform and stratiform clouds during ICARTT (Fountoukis et al., 2007), consistent with the modal values in Shaw and Lamb (1999). In this work, entrainment is included and comparisons against the observations are performed several hundred meters above cloud base. Exploratory simulations assuming a higher aerosol number concentration at cloud base (H_S = 1,200 m, Fig. B3b) show a highly nonlinear response to changes in a_c and R with the best agreement in CDNC being achieved with higher a_c values (0.03 and 0.06) and weak entrainment (R = 1500 m) consistent with the nonlinear trade-offs between entrainment (stronger, R = 500 m) and condensation (lower, $a_c = 0.01$) for the reference simulation as discussed in Sect. 4.2.2. Nevertheless, the corresponding spectra simulated with higher a_c values show larger discrepancies in spectral width and shape against the observations within IC (not shown here), and thus predictions of inferior skill with regard to cloud vertical development.

Finally, the lower value of a_c that is in good agreement with aircraft observations above cloud base in the present study is consistent with diffusion-kinetic theory that accounts for the feedbacks between latent heat and temperature in the boundary-layer of growing droplets (Fukuta and Myers, 2007). Further, results from laboratory experiments of direct contact condensation on aerosols in cloud chambers with horizontal or vertical moist flows, point to a_c values around 0.01 (Garnier et al., 1987;Hagen et al., 1989) in contrast with the most probable value (0.06) found in the levitation cell by Shaw and Lamb (1999).

5 Discussion and Conclusions

The vertical microphysical structure of clouds in regions of complex terrain is tightly linked to increases in rainfall intensity via seeder-feeder interactions (e.g. Barros and Lettenmaier, 1994). In this study, a new entraining cloud parcel model

(DCPM) with explicit bin microphyscis is presented and applied to explore the vertical structure of cloud development, and to investigate dominant factors in determining the microphysical development of clouds in the complex terrain of the inner Southern Appalachian Mountains using observations collected during the Integrated Precipitation and Hydrology Experiment in 2014 (IPHEx) as part of the Global Precipitation Measurement (GPM) mission ground-validation (Barros, et al. 2014) . The DCPM was evaluated against standard parcel models for a range of input parameters and the implementation correctly captures the known microphysics encoded in the supersaturation balance equation. The model was applied to a case study of the development of a mid-day cumulus congestus case on 12 June 2014 when aircraft measurements are available during the IPHEx campaign. Although this flight sampled three distinct cloud regions along the lowest aircraft transect, the targeted in-cloud region and the Maggie Valley supersite located nearby in the inner valley region of the mountains, and thus a detailed modelling study could be conducted leveraging ground-based aerosol measurements and W-band radar profiles available at Maggie Valley to inform model initialization. Because measurements of key input parameters are not available from IPHEx, or could not be resolved by current sampling techniques, a specific set of initial conditions and parameters was inferred from Maggie Valley observations and the literature. Sensitivity experiments were conducted to examine the model dependence on key inputs and assumptions within their possible ranges. Albeit a large variability in cloud microphysical properties was observed at sub-km scale (~ 90 m is the spatial averaging resolution of the measurements along the flight track) over the complex terrain of the inner Southern Appalachian Mountains and even within the targeted in-cloud region, modelling results for the reference simulation achieved good agreement with the observed cloud droplet number concentration, liquid water content, and droplet size spectra collected roughly 300–500 m above cloud base in the centre of the cloud.

In the framework of the physically based cloud parcel model, sensitivity of the simulated cloud microphysical characteristics to variations in key parameters was investigated within the context of in situ measurements. Results from sensitivity tests show that condensation coefficient exerts a profound influence on the droplet concentration, size distribution, liquid water content, and thermodynamic conditions inside the parcel. A decrease in a_c leads to an increase in cloud droplet number, a broader droplet spectrum, and a higher maximum supersaturation near cloud base. The case-study during IPHEx reveals that the observed cloud features are better captured by a low value of a_c (0.01) and strong entrainment strength corresponding to R = 500m using the bubble parameterization of entrainment processes. As expected, entrainment is found to be a major process controlling the vertical structure, cloud droplet number concentration, and liquid water content of the cloud. Further, it was shown that with other input parameters remain the same as reference simulation conditions, there is a trade-off between the cloud droplet number concentration sensitivity to entrainment strength and the condensation coefficient: strong entrainment (meaning the characteristic scale R in the bubble parameterization is small) is compensated by lower a_c values, and vice-versa. This competitive interference explains higher values found in previous aerosol-cloud droplet closure studies using adiabatic parcel modeles that neither include entrainment nor collision-coalescence. Initial aerosol concentrations at cloud base also have a large impact on droplet numbers but negligible influence on liquid water content. Hygroscopicity also has little effect on the simulated profiles of droplet number concentration and liquid water content. Nevertheless, analysis of the effect of the interdependence of initial aerosol concentration, condensation coefficient and entrainment strength on the droplet number concentration revealed ambiguous behavior that could only be resolved by assessing the properties of the simulated droplet spectra (shape, range) against the aircraft measurements at different altitudes throughout the clouds (i.e., well above cloud base). Overall, these findings provide insight into key parameters of aerosol cloud interaction processes in this region.

Finally, data and model limitations should be acknowledged. Regarding data limitations to constrain and force the CPM, reasonable assumptions were made based on the literature, surface and airborne observations from IPHEx, and WRF model simulations due to the lack of near cloud-base measurements and sounding, as pointed out earlier. Next, it is important to recognize the limitations of the lateral homogeneous entrainment employed in the model. Its concept is based on a simple assumption that entrained aerosols are mixed instantly across the parcel, which neglects the inhomogeneous supersaturation and microphysical structure inside the cloud associated with discrete entrainment events on different spatial scales (Baker et al., 1980; Khain et al., 2000). Realistic entrainment and mixing with cloud surroundings have been found to contribute significantly to droplet spectrum broadening. Turbulent mixing (Krueger et al., 1997) can break down entrained blobs of air into smaller scales and subsequently form small adjacent regions with uniform properties on account of molecular diffusion, thus leading to considerable spectrum broadening. In addition, the parameterization of entrainment through lateral boundaries neglects entrainment with dry air at cloud top that is expected to be an important element to cloud vertical development (Telford et al., 1984). Downdrafts induced by the penetration of dry air at cloud top can sink and mix with updrafts, effectively diluting number concentrations and broadening droplet spectra in clouds (Telford and Chai, 1980). Another limitation in the current approach is the assumption of uniform hygroscopic properties for all particle sizes. In reality, the aerosol distribution is an aggregate of particles with different physicochemical properties, including different shapes, solubility, and chemical species (Kreidenweis et al., 2003;Nenes et al., 2002). Even if specified initial aerosol characteristics were to capture the variation of κ with size, how to track the evolution of κ as particles among different bins undergo coalescence and breakup remains a challenge. Nevertheless, the sensitivity analysis indicates that the cloud droplet growth is generally insensitive to hygroscopicity (Sect. 4.2.4), thus the constant κ value used in this study does not significantly affect our modelling results.

For unstable cloud layers, complexity of in-cloud vertical velocity fields with localized areas of much stronger updrafts has been found to support the formation of wide bimodal spectra in cumulus clouds due to in-cloud nucleation of new droplets from interstitial aerosols when the parcel supersaturation higher up in the cloud exceeds the cloud base maximum (Pinsky and Khain, 2002). As a result, this mechanism can lead to the formation of a secondary mode of small droplets in individual spectra, different from our observed spectra with a second mode centred at a larger droplet size (Figs. 3 and 7). In this study, however, supersaturation does not increase above the cloud base maximum under the conditions of the original and modified model environments, likely attributed to the ambiguities in the sounding input from WRF, even if direct aircraft measurements albeit highly uncertain suggest otherwise. Because the collision-coalescence efficiency kernels are dependent on particle size, nonlinear stochastic behaviour can also lead to the development of a second mode of larger drops especially because collision-break-up mechanisms are not active (Prat et al. 2012).

In the present study, the local sensitivity of selected model parameters are assessed individually over certain ranges based on IPHEx data and the literature. Overall, thirty different simulations correspondingto 30 different parameter combinations were conducted, and the results suggest that the ranges of parameters that lead to physically-meaningful results consistent with observations are small. Nevertheless, the present study underlines the importance of the relationhsip between entrainment processes that determine the local- (microscale) and cloud-scale thermodynamic environment around individual particles, and the aerosol condensation coefficient that measures the effectiveness of condensation processes in the same thermodynamic environment. Given the multiscale thermodynamic structure of clouds, these interactions suggest that realistically the condensation coefficients in the natural environment are transient and spatially variable. Therefore, further research is necessary to map the joint nonlinear dynamics of aerosol-cloud-interaction parameters to identify physically meaningful regimes in parameter-space and to link these regimes to environmental conditions and aerosol properties toward reducing aerosol-cloud interaction related uncertainties in the estimation of the aerosol indirect effect. Ongoing work is focusing on exploring the sensitivity of the DCPM in a multi-dimentional parameter space to quantify multiple parameter interactions (Gebremichael and Barros, 2006;Yildiz and Barros, 2007) on aerosol-cloud processes using the factorial design method (Box et al., 1978).

Appendix A

Glossary of Symbols

a_c	condensation coefficient
a_T	thermal accommodation coefficient
c_p	specific heat of dry air
D_v, D_v'	diffusivity of water vapor in air, modified diffusivity of water vapor in air
e_s	saturation vapor pressure
g	gravitational constant
G	growth coefficient
H_S	scale height
k_a, k_a'	thermal conductivity of air, modified thermal conductivity of air
L	latent heat of evaporation
M_a, M_w	molecular weight of dry air, of water
N, N'	number concentration of cloud droplets, of ambient aerosol particles
p	pressure
r, r_c	radius of cloud droplet, of dry aerosol particle

R universal gas constant

R_a specific gas constant for moist air

R_v Specific gas constant for water vapor

R_B, R_J radius of air bubble, of convective jet

S supersaturation

S_{eq} droplet equilibrium supersaturation

T (T') temperature of air parcel (ambient air)

V parcel updraft velocity

v, v' droplet volumes

w_L mixing ratio of liquid water in parcel

w_v (w_v') mixing ratio of water vapor in parcel (in environment)

κ hygroscopicity parameter

μ entrainment rate

ρ_a, ρ_w density of dry air, of water

σ_w droplet surface tension

<u>*Parameter Formulae*</u>

$$G = \left[\frac{\rho_w RT}{e_s D_v' M_w} + \frac{L\rho_w}{k_a' T}\left(\frac{LM_w}{RT} - 1\right)\right]^{-1} \tag{A1}$$

where the modified diffusivity (D_v') and thermal conductivity (k_a') of water vapor in air account for non-continuum effects (Seinfeld and Pandis, 2006) and are described as follows

$$D_v' = \frac{D_v}{1 + \frac{D_v}{a_c r}\sqrt{\frac{2\pi M_w}{RT}}} \tag{A2}$$

$$k_a' = \frac{k_a}{1 + \frac{k_a}{a_T r \rho_a c_p}\sqrt{\frac{2\pi M_a}{RT}}} \tag{A3}$$

where the thermal accommodation coefficient (a_T) is taken as 0.96 (Nenes et al., 2001). Additional sensitivity tests of CDNC to a_T, ranging from 0.1 to 1 (Shaw and Lamb, 1999), were conducted and the resulting droplet concentrations indicate little sensitivity to this input parameter (not shown here).

The hygroscopicity parameter (κ) is adopted to characterize aerosol chemical composition on CCN activity according to κ-Köhler theory (Petters and Kreidenweis, 2007). $S_{eq,i}$ for droplets in the i^{th} bin ($i = 1, 2, \ldots, nbin$) can be written as

$$S_{eq,i} = \frac{r_i^3 - r_{c,i}^3}{r_i^3 - r_{c,i}^3(1-\kappa_i)} exp\left(\frac{2M_w\sigma_w}{RT\rho_w r_i}\right) - 1 \tag{A4}$$

where $r_{c,i}$ and r_i are the radius of the dry aerosol particle and the corresponding growing droplet, respectively. Droplet surface tension (σ_w) is a function of the parcel temperature (Pruppacher and Klett, 1997).

$$\alpha = \frac{gM_wL}{c_pRT^2} - \frac{gM_a}{RT} \tag{A5}$$

$$\gamma = \frac{pM_a}{e_sM_w} + \frac{M_wL^2}{c_pRT^2} \tag{A6}$$

Liquid water content (g m^{-3}):

$$LWC = \frac{4\pi}{3}\rho_w \sum_{i=1}^{bins} N_i r_i^3 \tag{A7}$$

Quasi-steady approximation of supersaturation S_{qs} (Pinsky et al., 2013):

$$S_{qs} \approx \frac{A_1V}{4\pi D_v N\bar{r}} \tag{A8}$$

where $\bar{r}$the average droplet radius, and N is the total droplet number concentration.

$$A_1 = \frac{g}{R_aT}\left(\frac{LR_a}{c_pR_vT} - 1\right) \tag{A9}$$

Appendix B

1. Sensitivity to environmental conditions

To account for the uncertainties associated with the environmental condition from WRF and examine its impact on cloud formation, one additional simulations was conducted with modified temperature profiles at the lowest 2 km above CBH (1,270 m), as displayed in Fig. B1. Here, we adjusted the original lapse rate (-4.1 °C km^{-1} from the WRF sounding, Fig. 10b) to -7 °C km^{-1} (Γ_1) for 1,270–2,200 m. The lapse rate for 2,200–3,200 m was changed to -4 °C km^{-1} to keep the ambient temperature below CBH and above 3,200 m unchanged. As expected, deeper clouds are formed in the modified environment, representing conditionally unstable atmosphere. As expected, LWC is significantly enhanced and droplet growth is faster under fast cooling condition in the atmosphere.

2. Sensitivity to initial updraft velocity

Cloud dynamics also play a crucial role in the microphysical evolution of cumulus clouds. One major parameter in the cloud dynamical field is the updraft velocity. In accordance with the observed vertical velocities by the aircraft and the W-band radar (see Fig. S12b), a reasonable variability in the initial updraft velocity at cloud base is introduced to assess its effects on the parcel supersaturation and cloud droplet concentrations, as shown in Fig. B2. By varying the initial updraft in a range of 0.1–1.5 m s^{-1}, simulated results display similar vertical velocities at the observation levels, which are still higher than the measured range (not shown here). As expected, slight increases in maximum supersaturation are resulted from larger initial updraft velocities, thus leading to slight enhancement of total droplet numbers. The simulated spectra show a slightly shift towards larger drop sizes due to weaker updrafts, which allow more time for cloud droplets to grow in a rising parcel.

Data availability: The IPHEx data are accessible at Global Hydrology Resource Center (GHRC) Distributed Active Archive Center (https://ghrc.nsstc.nasa.gov/home/field-campaigns/iphex).

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Table 1: Cloud parcel models with detailed microphysics from the literature and in this study (Duke CPM). NA denotes information is not described in the reference paper.

Parcel model	Binning	Condensation	Coalescence	Entrainment	Numerics
Abdul-Razzak et al. (1998)	Discrete	Leaitch et al. (1986)	Not included	No included	LSODE solver (Hindmarsh, 1983)
Cooper et al. (1997)	Moving discrete	Fukuta and Walter (1970)	Modified Kovetz and Olund (1969)	Not included	Fifth-order Runge-Kutta (adaptive-size)
Flossmann et al. (1985)	Discrete	Pruppacher and Klett (1978)	Berry and Reinhardt (1974)	Lateral homogeneous bubble model	NA
Jacobson and Turco (1995)	Hybrid discrete	Jacobson and Turco (1995)	Jacobson et al. (1994)	Not included	SMVGEAR (Jacobson and Turco, 1994)
Kerkweg et al., (2003)	Discrete	Pruppacher and Klett (1997)	Bott (2000)	Lateral homogeneous bubble model	NA
Nenes et al. (2001; 2002)	Moving discrete	Pruppacher and Klett (1997); Seinfeld and Pandis (1998)	Not included	Not included	LSODE solver (Hindmarsh, 1983)
Pinsky and Khain (2002)	Moving discrete	Pruppacher and Klett (1997)	Bott (1998); turbulent effect on drop collision	Not included	NA
Snider et al. (2003)	Discrete	Zou and Fukuta (1999)	Not included	Not included	NA
Duke CPM	Moving discrete	Pruppacher and Klett (1997); Seinfeld and Pandis (2006)	Bott (1998); turbulent effect on drop collision	Lateral homogeneous bubble/jet model	Fifth-order Runge-Kutta (adaptive-size)

Table 2. Lognormal fit parameters characterizing the aerosol number distribution of four modes. Note N = total number of aerosol particles per cm^3; Dg = geometric mean diameter (µm); σ_g = geometric standard deviation for each mode. N_{surf} and N_{CBH} represent total aerosol number concentrations at the surface and cloud base height (CBH: 1,270 m), respectively.

Mode #	N_{surf} (cm^{-3})	N_{CBH} (cm^{-3})	D_g (µm)	σ_g
1	1401.9	393.7	0.076	1.63
2	415.7	116.8	0.195	1.35
3	0.300	0.084	0.750	1.30
4	0.300	0.084	2.200	1.40

Table 3. a) Summary of model numerical configuration parameters used in simulations presented here.

Bin Discretization	Volume Ratio	Number of bins	Time-step (condensational growth)	Time-step (advection) (collision-coalescence)
Geometric	1.026	Up to 1,000 for initial CN distribution	5×10^{-4}s	
Geometric, Time-Varying	1.026	Bins for larger diameters added as determined by condensational growth	10^{-3}s	0.2s

Table 3. b) Summary of physiochemical parameter ranges used in sensitivity simulations (reference value in bold).

Parameter	Value and Range
Vo [m/s]	0.1,0.3,**0.5**,1.0,1.5
κ	0.1, **0.14**, 0.2, 0.3, 0.4,
a_T	**0.96**
a_c	0.002,0.005, **0.01**, 0.015,0.03,0.06
Entrainment Radius R_B (bubble model) [m]	300, 400, **500**, 1000,1500, 2000
Entrainment Radius R_J (jet Model) [m]	**500**
Scale Height [m]	800,900,**1000**,1100, 1200

Table 4. Evaluation of the predicted CDNC from simulations using various condensation coefficients against the averaged observation from the CDP.

Condensation coefficient	Prediction[a] (cm^{-3}) (1,500 – 1,600 m AGL)	Difference[b] (%)	Prediction[a] (cm^{-3}) (1,600 – 1,750 m AGL)	Difference[b] (%)
0.002	402.7	15.3	365.9	-7.90
0.005	385.8	10.4	350.5	-11.8
0.010	354.0	1.30	321.6	-19.0
0.015	328.5	-6.00	298.5	-24.9
0.030	281.0	-19.6	255.3	-35.7
0.060	242.1	-30.7	219.9	-44.6

[a]The averaged CDNC in the predictions for the indicated altitudes. The DCPM uses Above Ground Level (AGL) as the altitude coordinate.
[b]Difference (%) = 100×(Prediction - Observation)/Observation. Note that observations between 1,500 m and 1,600 m AGL (349.4 cm^{-3}) over the higher terrain (Fig. 3d) and between 1,600 m and 1,750 m AGL (397.5 cm^{-3}) over the lower terrain (Fig. 3c) are calculated by averaging the cluster of five consecutive CDNC measurements. A shown in Fig. 3b, the two altitudes are approximately the same with respect to Mean Sea Level (MSL).

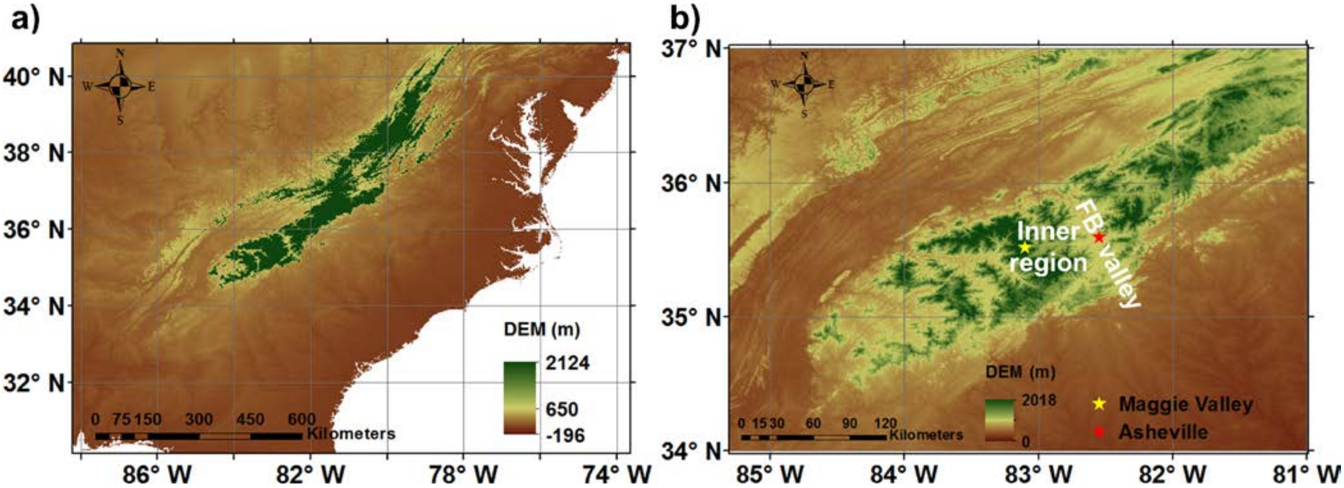

Figure 1: a) Study region of the IPHEx campaign in the SAM (highlighted in the black box), as shown in context of a large scale map of the south-eastern United States. (b) Topographic map of the SAM including the two ground-based IPHEx observation sites referred to in this study. FB valley denotes French Broad valley.

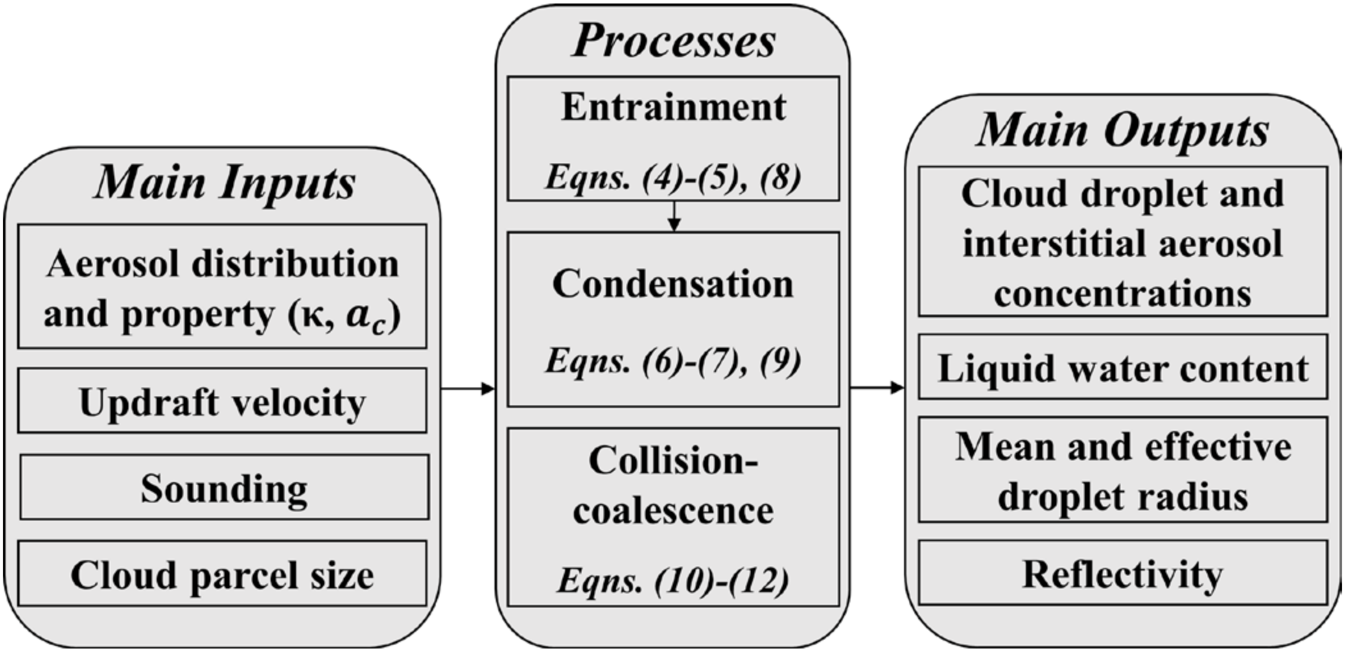

Figure 2: Flowchart of the main inputs, microphysical processes, and main outputs of the DCPM. Equation numbers refer to formulae in Sect. 2.

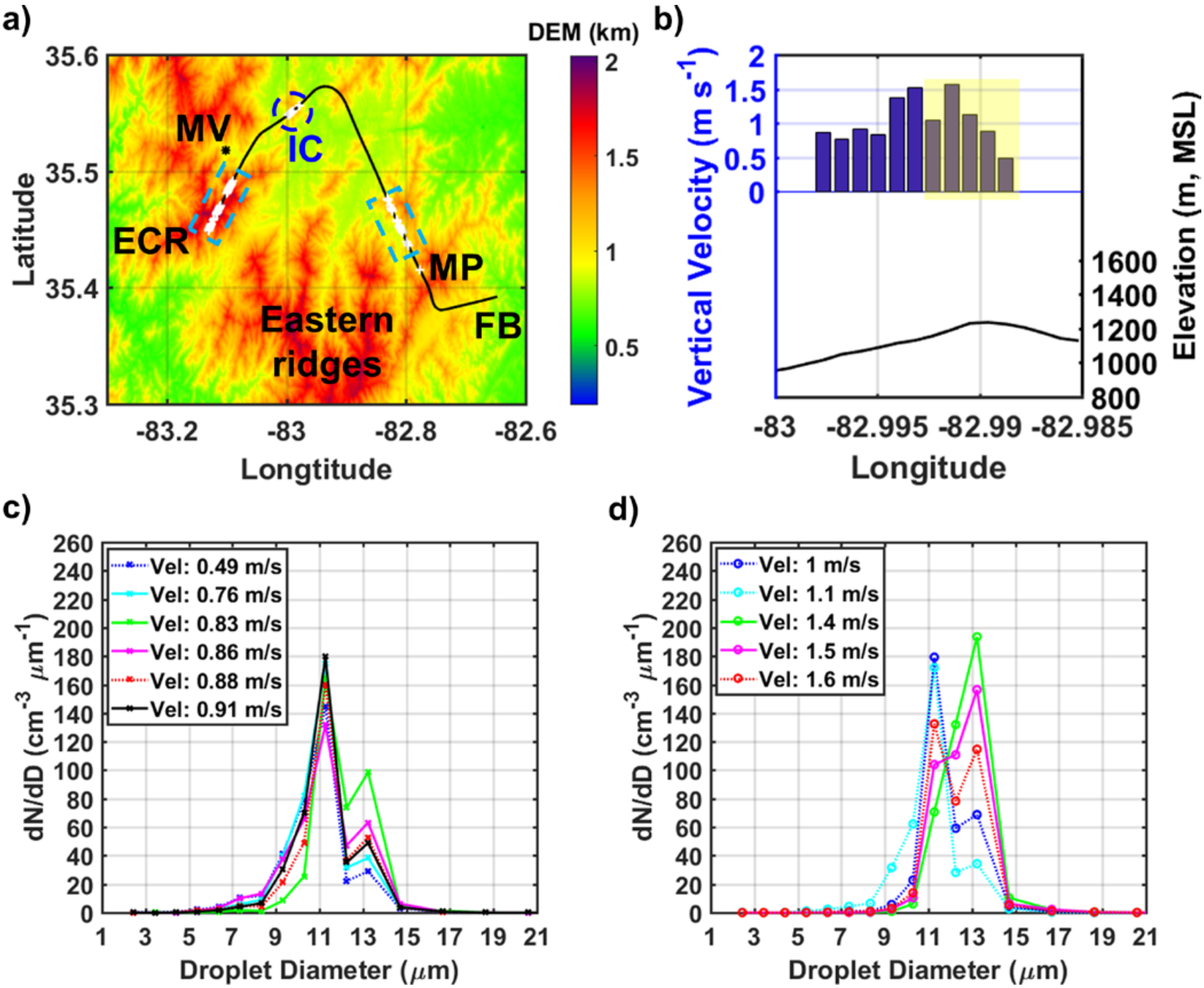

Figure 3: a) Lowest cloud transect of the UND Citation flight track on 12 June 2014. The in-cloud observations are identified as white plus signs, and MV is marked by the black asterisk. For left to right in the map, ECR denotes Eastern Cherokee reservation, MP denotes Mount Pisgah, and FB denotes French Board valley. b) Updraft velocity variations of the targeted in-cloud region, denoted by IC in (a). The in-cloud samples were collected at 1-Hz (~ 90 m in flight distance) resolution. Cloud droplet concentrations of the in-cloud samples in IC (b) with low (0–1 m s^{-1}) and high (1–2 m s^{-1}) updrafts are shown in (c) and (d), respectively. The updraft velocity of each sample is indicated in the legend. Dotted lines represent the droplet spectra in the reference sub-region within IC, within yellow shade region in (b).

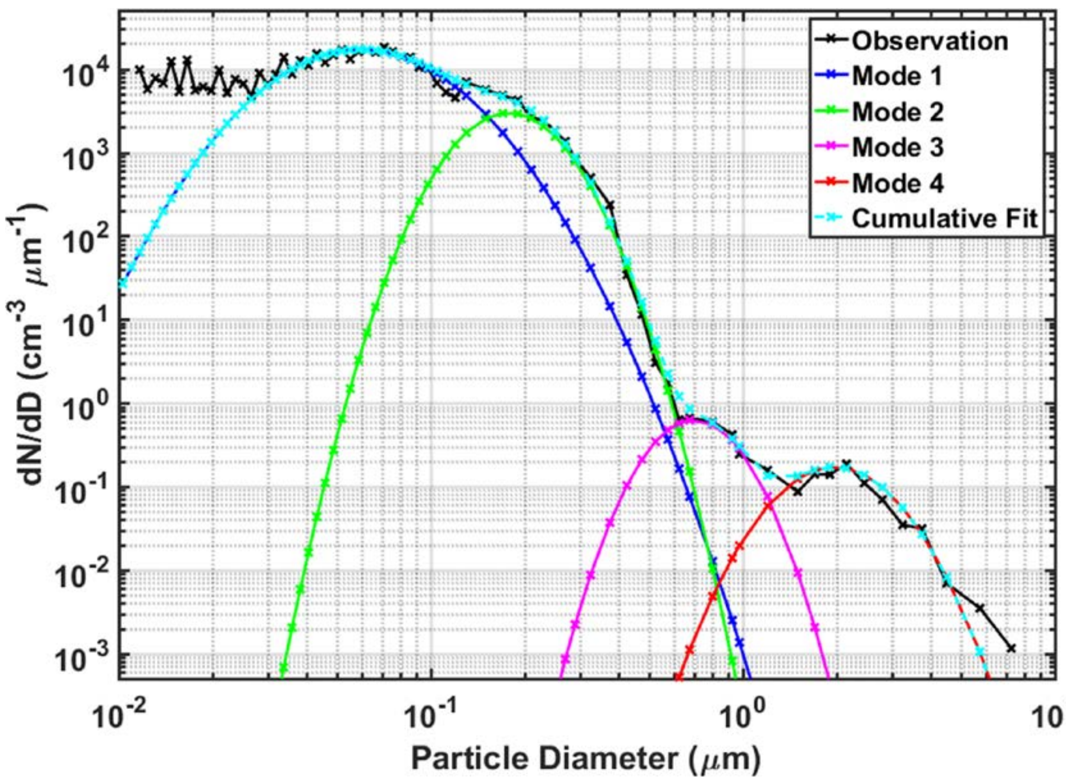

Figure 4: Mean surface aerosol size distribution fitted by four lognormal functions. Observations are merged from the SMPS and PCASP, and are averaged during the first 10 mins (12:14 LT – 12:24 LT) of the 12 June flight. Fitted parameters (total number concentration, geometric mean diameter, and geometric standard deviation) for each mode are summarized in Table 2.

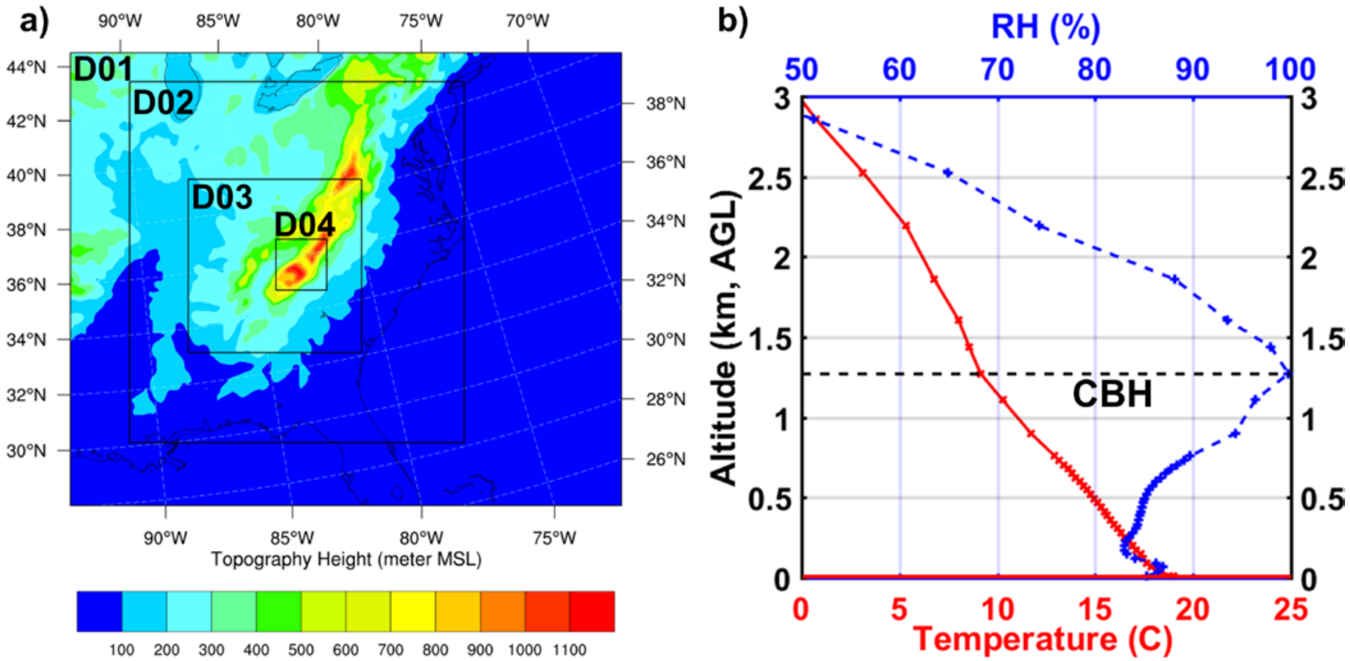

Figure 5: a) WRF model configuration of four one-way nested domains at 15-, 5-, 1.25-, 0.25-km grid resolution, respectively. b) Vertical profile of temperature (red solid line) and relative humidity (dashed blue line) from the spatially-averaged WRF sounding columns at IC (see its location in Fig. 3a). The horizontal dashed line depicts CBH = 1,270 m AGL.

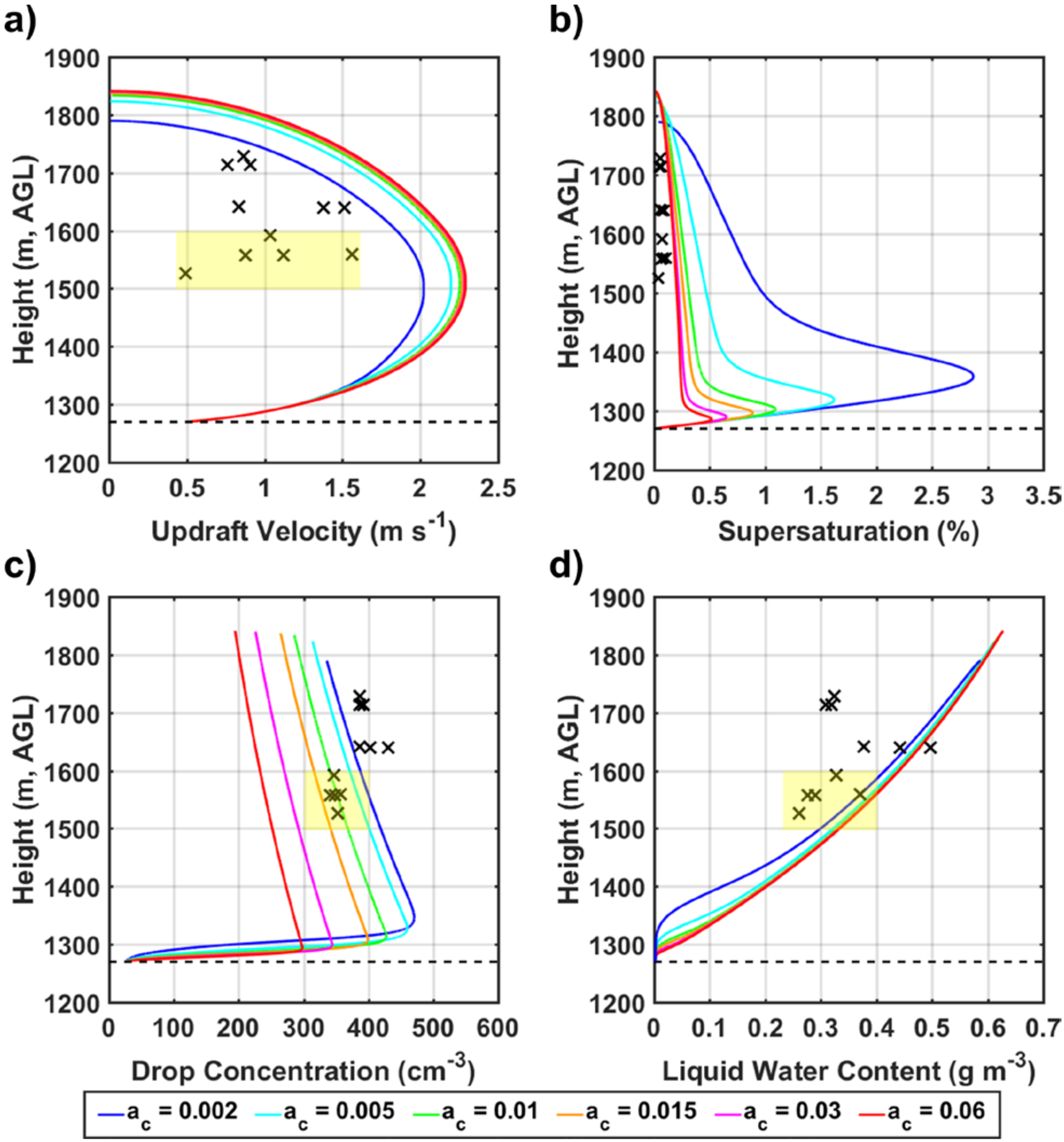

Figure 6: Sensitivity of the updraft velocity (a), supersaturation (b), total drop concentration (c), and LWC (d) to the variations in the condensation coefficient (a_c) as compared to the airborne observations (marked by black crosses). The horizontal dashed line depicts CBH. In (b), the quasi-steady approximation of supersaturation is calculated based on observed temperature. It should be kept in mind that airborne measurements of temperature in clouds are subject to large uncertainties, thus rendering the derivation of supersaturation unreliable.

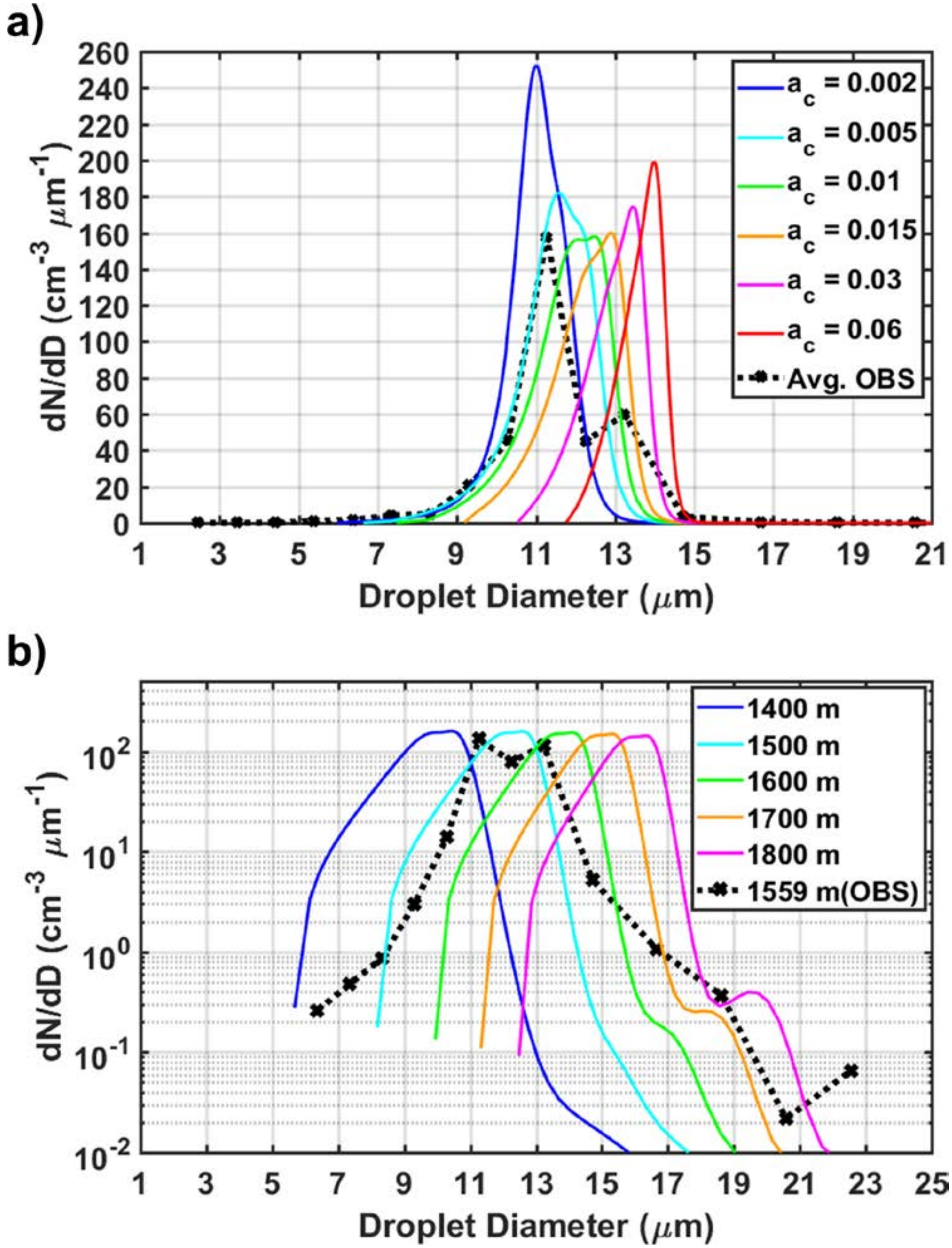

Figure 7: a) Sensitivity of simulated droplet spectra at 1,500 m (solid lines) to the variations in a_c. The black dotted line reflects the average of five droplet spectra observed by the CDP (dotted lines in Figs. 3c and d) between 1,500 m and 1,600 m AGL. b) Simulated evolution of cloud droplet spectra at 1,400 m, 1,500 m, 1,600 m, 1,700 m, and 1,800 m altitude assuming a_c= 0.01. The black dotted line denotes the observed droplet spectrum at 1,559 m that has similar total CDNC and LWC as the simulation with a_c = 0.01 at the same altitude.

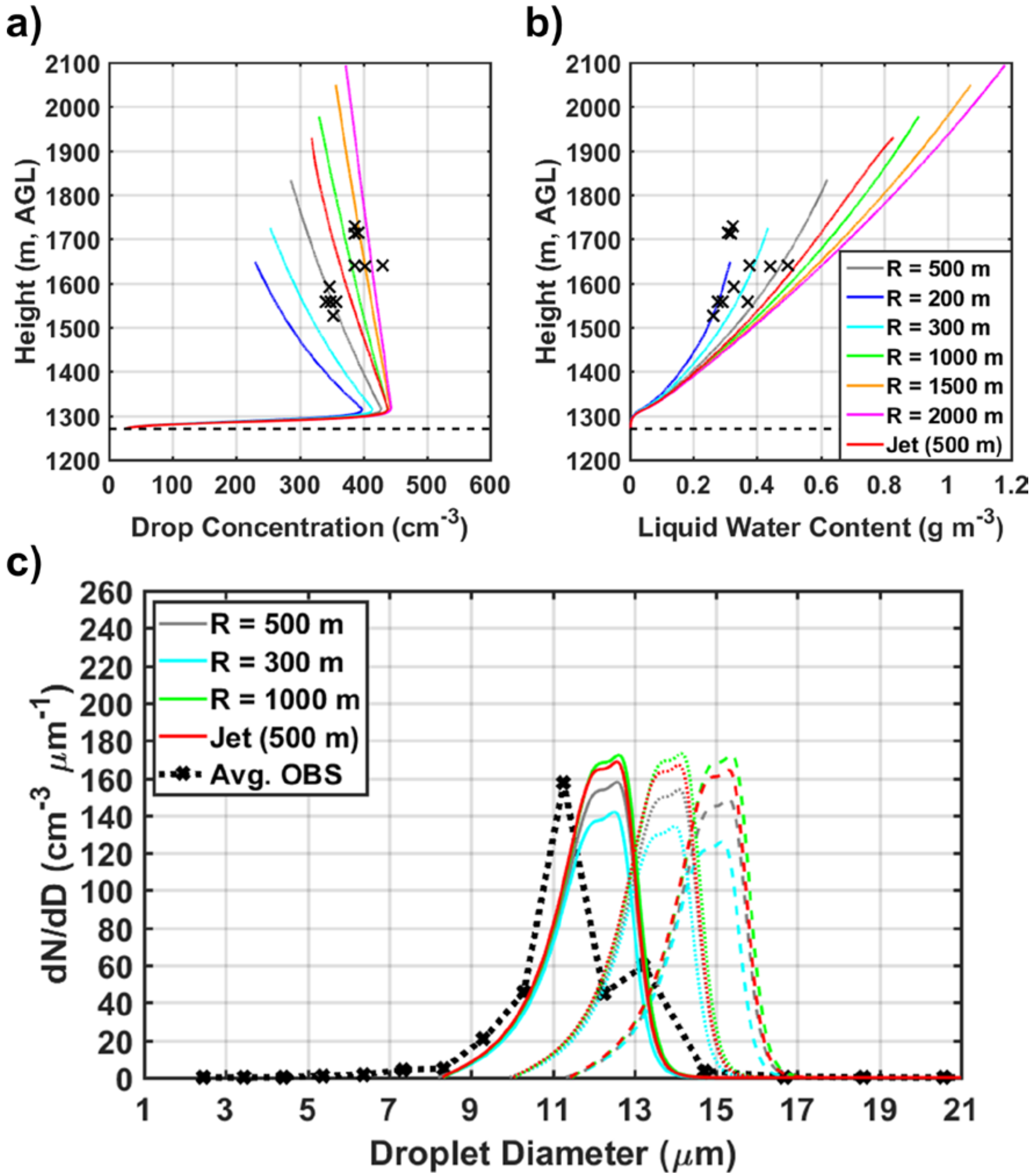

Figure 8: Sensitivity of the total drop concentration (a) and LWC (b) to the variations in the initial parcel radius (R) considering lateral entrainment as a bubble model and a jet model. In (a) and (b), the airborne observations are marked by black crosses, and the horizontal dashed line depicts CBH. c) Predicted droplet spectra at three altitudinal levels (1,500 m: solid line, 1,600 m: dotted line, and 1,700 m: dashed line) using two parameterization schemes for lateral entrainment: the bubble model with R = 500 m (base case, grey lines), R = 300 m (cyan lines), and R = 1,000 m (green lines); the jet model with R = 500 m (red lines). The black dotted line reflects the average of five droplet spectra observed by the CDP (dotted lines in Figs. 3c and d) between 1,500 m and 1,600 m AGL.

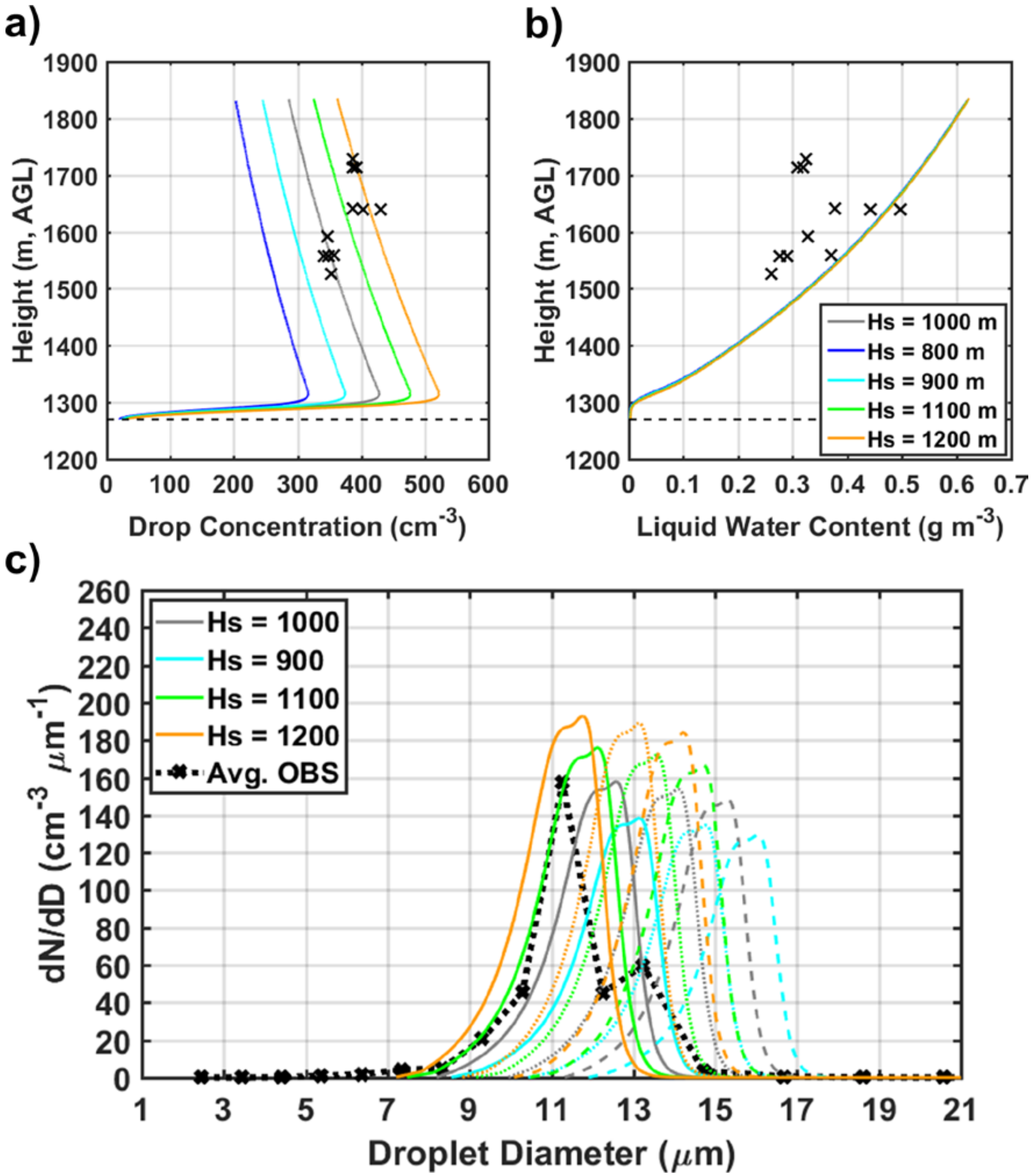

Figure 9: Sensitivity of the total drop concentration (a), LWC (b), and droplet spectra (c) at three altitudinal levels (1,500 m: solid line, 1,600 m: dotted line, and 1,700 m: dashed line) to the variations in initial aerosol concentrations at cloud base, as represented by different values of the scale height (H_S). In (a) and (b), the airborne observations are marked by black crosses, and the horizontal dashed line depicts CBH. The black dotted line in (c) reflects the average of five droplet spectra observed by the CDP (dotted lines in Figs. 6c and d) between 1,500 m and 1,600 m AGL.

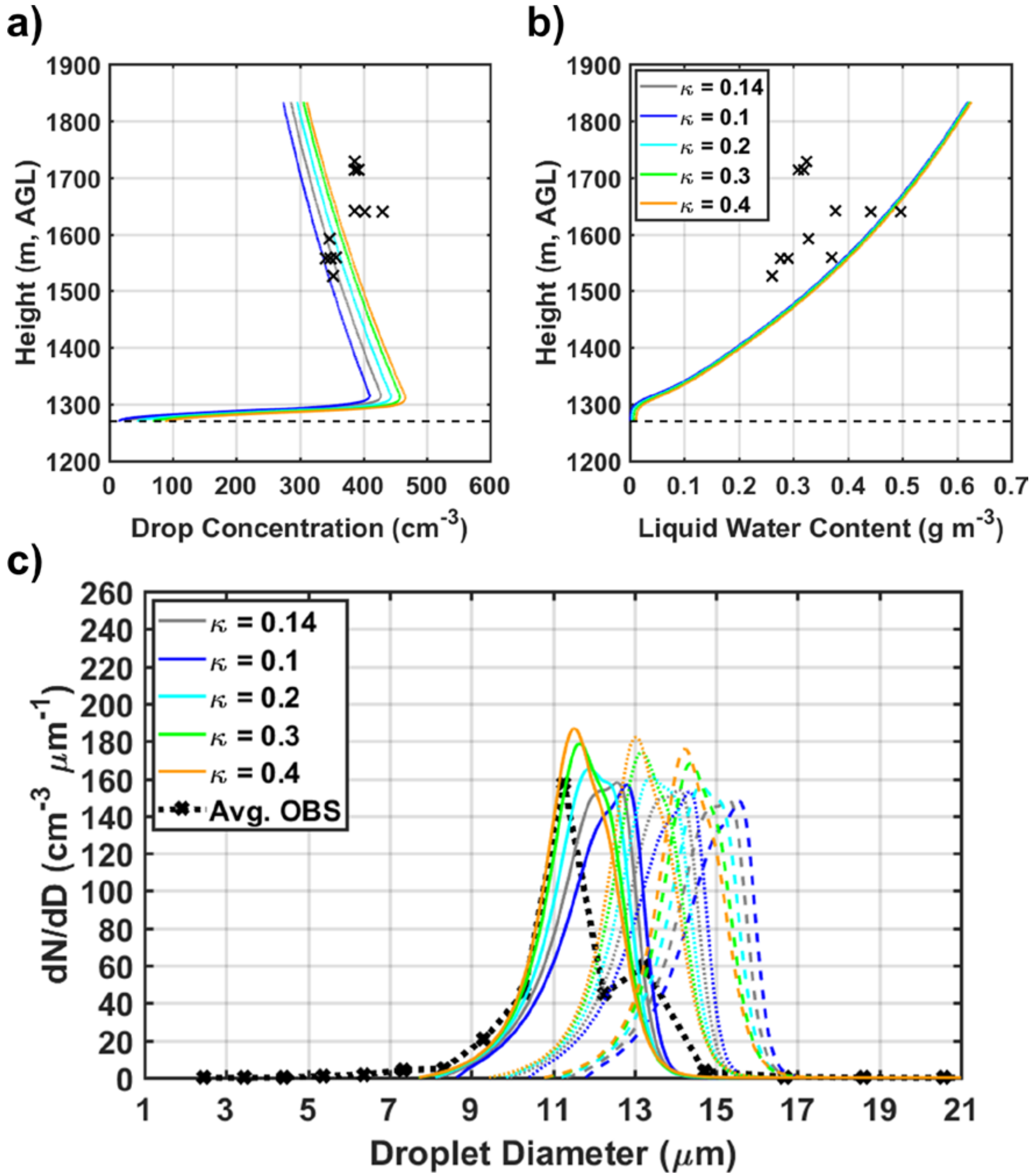

Figure 10: Sensitivity of the total drop concentration (a), LWC (b), and droplet spectra (c) at three altitudinal levels (1,500 m: solid line, 1,600 m: dotted line, and 1,700 m: dashed line) to variations in hygroscopicity parameter (κ). In (a) and (b), the airborne observations are marked by black crosses, and the horizontal dashed line depicts CBH. The black dotted line in (c) reflects the average of five droplet spectra observed by the CDP (dotted lines in Figs. 3c and d) between 1,500 m and 1,600 m AGL.

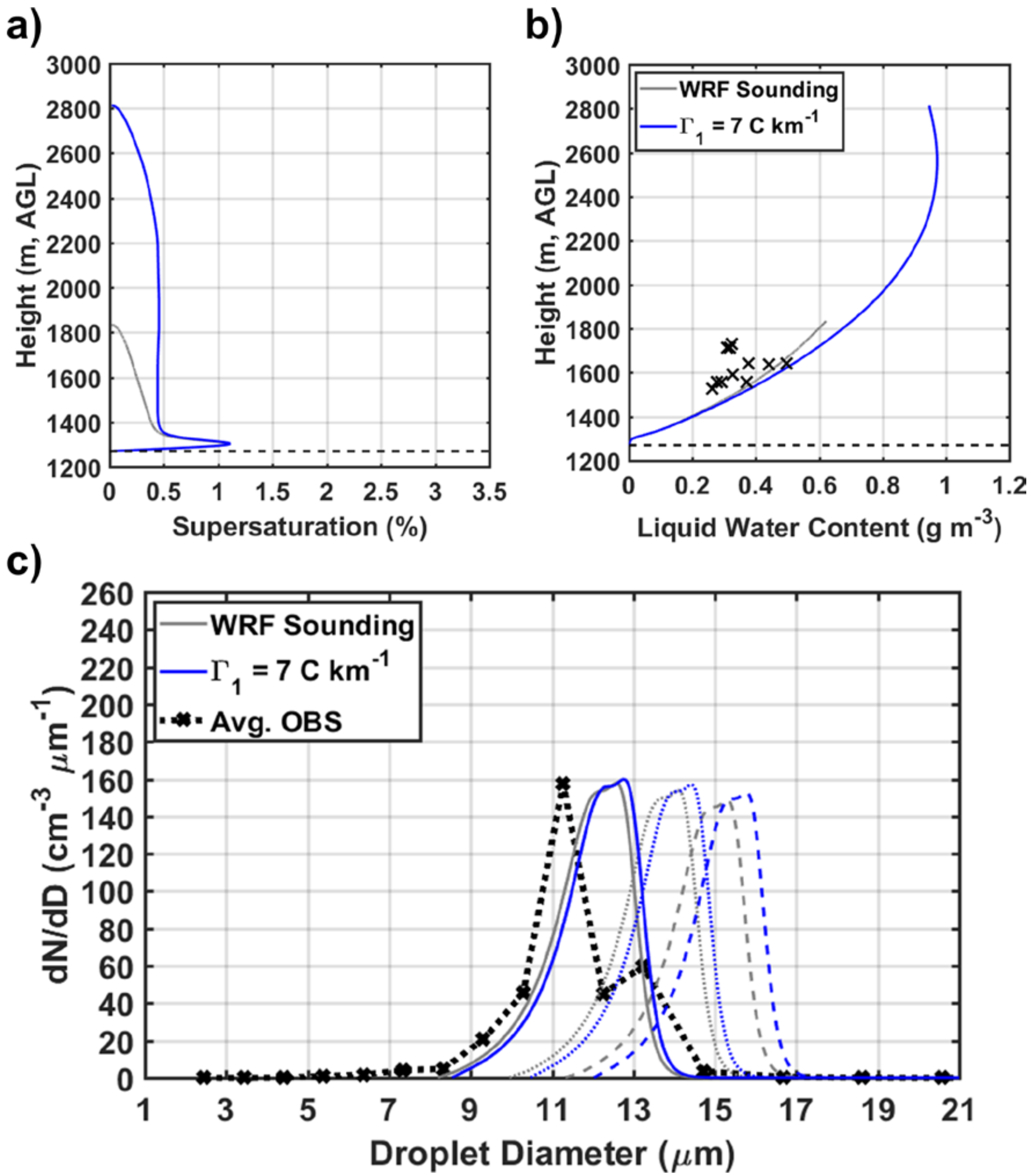

Figure B1: Vertical profiles of the supersaturation (a) and LWC (b) for simulations with the original WRF sounding (grey lines) and modified ambient temperature (blue lines). In (b), the airborne observations are marked by black crosses, and the horizontal dashed line depicts CBH. c) Predicted droplet spectra at three altitudinal levels (1,500 m: solid line, 1,600 m: dotted line, and 1,700 m: dashed line) to the variations in the environmental conditions. The black dotted line reflects the average of five droplet spectra observed by the CDP (dotted lines in Figs. 3c and d) between 1,500 m and 1,600 m AGL.

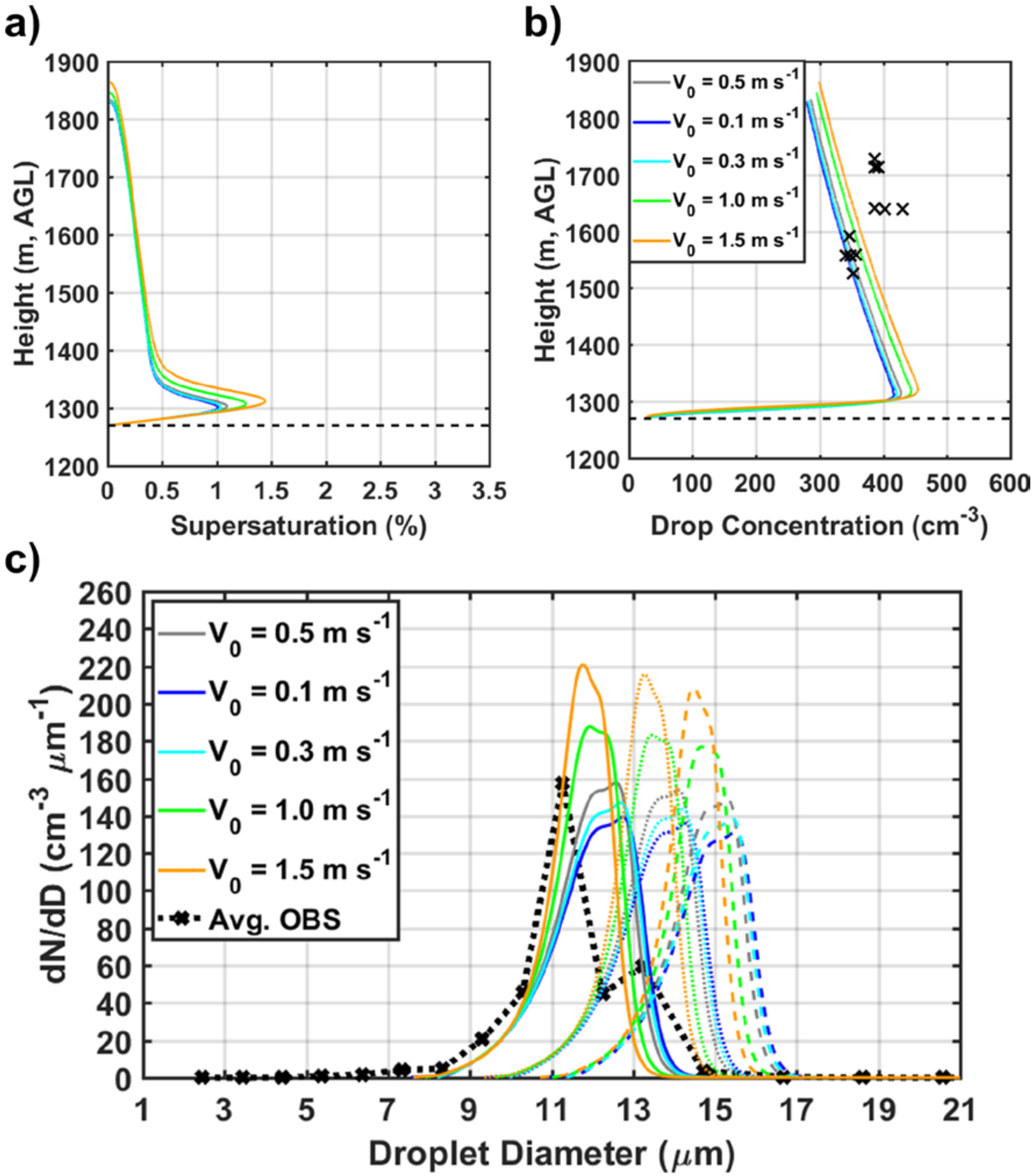

Figure B2: Sensitivity of the supersaturation (a), total drop concentration (b), and droplet spectra (c) at three altitudinal levels (1,500 m: solid line, 1,600 m: dotted line, and 1,700 m: dashed line) to the variations in the initial updraft velocity (V_0) at cloud base. In (b), the airborne observations are marked by black crosses, and the horizontal dashed line depicts CBH. The black dotted line in (c) reflects the average of five droplet spectra observed by the CDP (dotted lines in Figs. 3c and d) between 1,500 m and 1,600 m AGL.

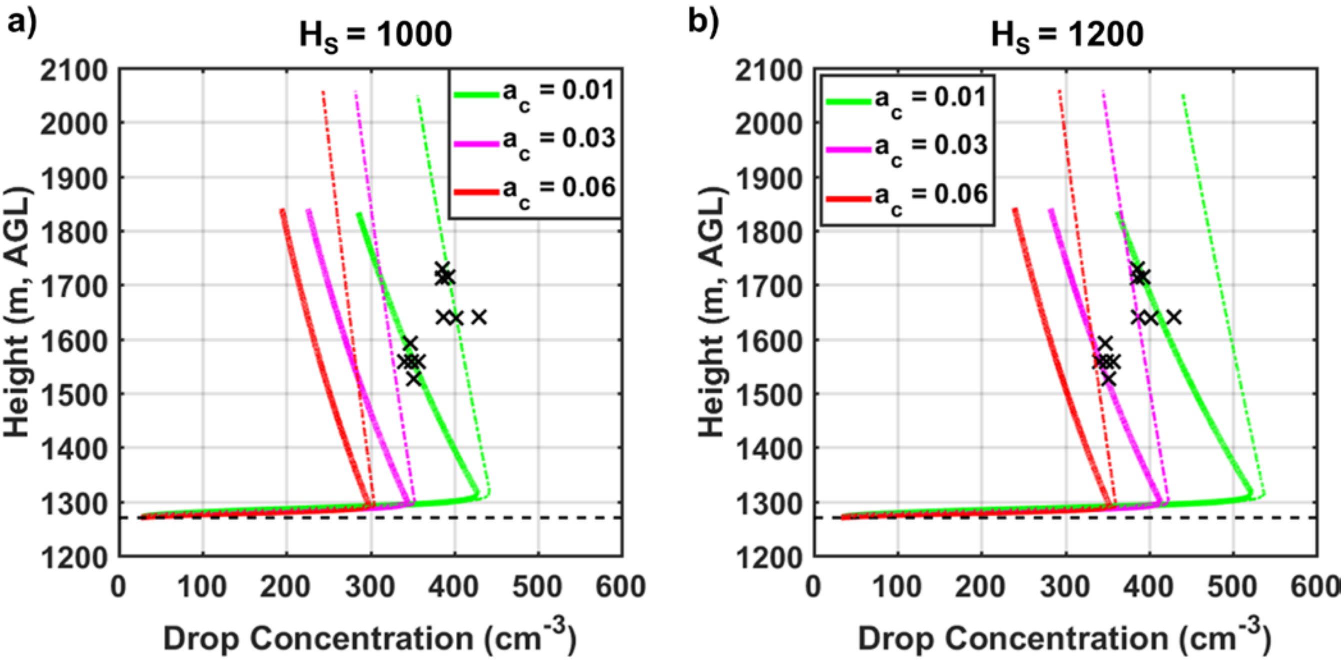

Figure B3: Sensitivity of the total cloud drop concentration to the variations in condensation coefficient and entrainment strength (strong: R = 500 m, solid thick lines; weak: R = 1,500 m, dash-dot thin lines) assuming different initial aerosol concentrations at cloud base (a: H_S = 1,000 m; b: H_S = 1,200 m). The airborne observations are marked by black crosses, and the horizontal dashed line depicts CBH.