Supporting online material

Materials and methods

Model description

We use a 48-km square horizontal by 24-km vertical domain with uniform 500-m horizontal by 375-m vertical grid spacing. A dynamics model (*S1*) integrates the anelastic equations for deep convection (*S2*) in conservative form using a 5-s time step, a third-order advection scheme, a standard Smagorinsky subgrid parameterization, and a sponge layer above 20 km. Boundary conditions are rigid at the top and bottom and open at the horizontal faces, where a cosine-squared forcing function is used to nudge values to the initial conditions in a 4.5-km swath along each face. In lieu of a soil model, we assimilate surface heat and moisture fluxes from forecasts of NASA Langley's Advanced Regional Prediction System (ARPS) mesoscale model (*S3*) initialized at 12:00 UTC on July 18. Starting at 16:00 UTC in the domain of the observed updraft, we spatially and temporally interpolate ARPS output from 15-km and 30-min resolution to our 500-m and 5-s resolution.

An embedded microphysics model (*S4, S5*) discretizes the size distributions of aerosols, drops, and ice particles into 16 mass bins each, spanning diameters of 5 nm–1 μm, 2 μm–1 cm, and 2 μm–>3 cm, respectively. Drop density is set to 1.0 g cm^{-3} and ice density is set to 0.9 g cm^{-3} below 300 μm in diameter, above which it decreases according to observations for lump graupel (*S6*). Height-dependent fall velocities are calculated as a function of particle mass and size according to Reynolds number regime, assuming spherical particles of deformable liquid or rigid, rough ice (*S7*). Fall velocities are combined

with radius-dependent collision efficiencies (*S8*) to derive a height-dependent gravitational collection kernel that is integrated semi-implicitly (*S9*). Drops coagulating with ice are assumed to freeze instantaneously. Additional kernels for spontaneous and collision-induced drop breakup (*S8, S10*) are integrated explicitly, but the effect of turbulence on coagulation is currently neglected. Gravitational collection and drop breakup are the only microphysical processes always advanced with the 5-s dynamical time step, whereas all other microphysical processes are advanced with a time step that varies to a minimum value of 0.2 s.

We have extended previous modeling studies of continental deep convection (*S11, S12*) by using aerosol size distributions based on measurements throughout the boundary layer and free troposphere, calculating supersaturations required for activation (*S7*), removing aerosols from the size distribution as they are activated, and allowing for three-dimensional transport. By contrast, the previous studies use a power-law formulation to estimate the number of aerosols activated, $N = Cs^k$, where s is the supersaturation in percent and C and k are positive constants that do not vary. While numerically efficient, the power-law formulation provides an infinite source of aerosols with increasing supersaturation and does not distinguish between the properties of boundary layer and free tropospheric aerosols. These factors become important in our simulated updrafts because high supersaturations activate all of the initial boundary layer aerosols, leaving negligibly few except for those entrained from the free troposphere, which exhibit markedly different number size distributions. Our treatment does require a knowledge of aerosol chemical composition, which we assume to be ammonium bisulfate. Chemical composition is undoubtedly more complex, but an aerosol/cloud condensation nuclei closure study has shown that assuming ammonium bisulfate nonetheless can provide a remarkably accurate representation of activation during CRYSTAL-FACE (*S13*).

Homogeneous freezing of aerosols and drops is treated classically here (*S7*) and results

are insensitive to a newer parameterization (*S14*). Results are also insensitive to inclusion of heterogeneous freezing by low concentrations of ice nuclei (IN), which we relate to temperature, ice supersaturation, and the numbers of particles present as follows. IN are assumed to be present at a background concentration of 0.06 cm^{-3}, consistent with observations (*S15*) on July 18, and are assumed to be uniformly distributed among the total aerosols, drops, and ice particles in each grid cell. IN in drops are assumed to activate linearly with decreasing temperature over the range -16 to $-28°C$ (*S7*), whereas IN in aerosols are assumed to activate nonlinearly with increasing ice supersaturation (*S16*) over the range 4–32% at temperatures below $-5°C$. Based on the simplifying assumption that temperature decreases and supersaturation increases monotonically with increasing elevation in updrafts, the IN incrementally freeze aerosols and drops in a given grid cell if updraft velocity exceeds 5 m/s and an incremental temperature or supersaturation threshold is passed during traversal from the grid cell below. Overall, homogeneous freezing of drops contributes the majority of ice crystals in our simulations, although all other freezing mechanisms also contribute small numbers. This result appears reasonable in light of the low ratio of IN to peak anvil ice crystal numbers and the general agreement between simulated and observed particle numbers and size distributions.

All cloud-resolving model results are dependent upon parameterizations, such as the ones just described for ice formation, as well as spatial resolution. Our spatial resolution is determined by the requirement of sufficient domain size to contain growth of a realistic cluster of updrafts, on the one hand, and the requirement of sufficient computational memory, on the other hand. Previous modeling of shallow cumulus with bulk microphysics has demonstrated that some aspects of cloud simulations can be extremely difficult to converge with respect to increasing spatial resolution (*S1*). Using limited available time on 512 processors of the Department of Energy's National Energy Research Scientific Computing Center IBM SP RS/6000, we find that no aspect of our analysis here is affected by

a doubling of either spatial resolution or bin number. These sensitivity tests reach the bin resolution of recent two-dimensional simulations of deep convection that focus on tracking more particle types (*S11*) and exceed the spatial resolution of recent three-dimensional simulations of deep convection with bulk microphysics (*S17*). However, some aspects of our simulations, such as the width and complexity of updraft features, could become more realistic with increasing resolution (Fig. 3, as noted).

Meteorological and aerosol measurements

Using open lateral boundary conditions, all faces of the model domain are nudged to the initial meteorological and aerosol profiles throughout the simulation. We derive a single representative meteorological profile from rawinsonde measurements on July 18 at the Miami airport, located approximately 100 km south of the observed updraft and at a similar distance from the coastline. We use soundings measured at 14:35 UTC and 17:36 UTC with a Vaisala 80H sensor, corrected for known sensor time-lag and bias errors. Thermodynamic data from the later sounding, taken 13 minutes prior to the updraft penetration, were fouled by sensor icing from supercooled liquid water. However, wind data from the earlier sounding were poorly sampled over extreme speed changes at crucial mid-tropospheric elevations. We thus combine thermodynamic variables from the earlier sounding with winds from the later sounding.

We derive initial representative aerosol profiles for the baseline simulation and sensitivity tests (Table 1) from measurements made on three aircraft. From the Twin Otter aircraft data (below 4 km), we estimate aerosol number $>$ 10 nm from sequential condensation particle counter (CPC) measurements of aerosols $>$ 7 and 12 nm in diameter and we estimate aerosol size distribution from simultaneous differential mobility analyzer measurements reported in 100 size bins spanning 10 nm–1 μm in diameter. From the Citation aircraft data (5–12 km), we estimate aerosol number from CPC measurements of aerosols $>$ 10 nm in diameter (aerosol size distribution was not measured on the Citation). From the WB-57

aircraft data (12–16 km), we estimate both aerosol number > 10 nm and size distribution from sequential nucleation mode aerosol size spectrometer (NMASS-II) measurements of aerosols > 4, 8, 15, 30, and 64 nm in diameter. Since no WB-57 measurements are available from July 18, we use measurements from July 19 to estimate the fall-off of aerosol number with elevation above 10 km and the size distribution of aerosols above 5 km.

While we benefit from a broad analysis of aerosol number and size distribution throughout the CRYSTAL-FACE experiment, precise aerosol conditions in and around the July 18 updraft remain uncertain since aerosol size distribution was not measured by the Citation, the WB-57 made no such measurements on July 18, and some of the Citation's in-cloud aerosol number measurements appear suspect. Aerosol number > 10 nm measured out-of-cloud is approximately 3000 and 4500 cm^{-3} upon entry into and exit from the case study cloud, respectively. Aerosol number measured in-cloud also remains persistently high, never dropping below 1800 cm^{-3} throughout the strong updraft. However, the extreme variability of numbers measured in the updraft is indicative of known CPC instrument errors in the presence of liquid water (*S18*), so we do not consider that data to be reliable. Thus, we focus here on the out-of-cloud aerosol measurements, with acknowledged uncertainty in their size distribution. For simplicity, we use typical monomodal parameters derived from size distributions measured by the WB-57 on July 19 in the absence of nucleation-mode particles and we arbitrarily use the entry value in our baseline profile at 10 km (Table 1). Our results are surprisingly insensitive to an increase of aerosol number up to the exit value using the monomodal parameters or using an additional mid-tropospheric nucleation mode, which can also explain some numbers remaining unactivated within the core.

Cloud measurements

Cloud particle size distributions measured on the Citation aircraft with a forward spectrometer scattering probe (FSSP-100) are reported as a function of maximum particle diameter in 15 bins spanning 4–62 μm. While FSSP measurements may be unreliable in the

presence of large ice crystals (*S19*), we find that integrations of the measured small particle size distributions match the volume of liquid water measured by both King and Rosemount icing probes. Cloud particle size distributions measured on the WB-57 aircraft with a cloud, aerosol, and precipitation spectrometer (CAPS) and an FSSP scattering spectrometer probe (SPP-100) are composited and reported as a function of maximum particle diameter in 25 bins spanning 0.35 μm–1.6 cm. Owing to the persistent presence of secondary peaks in particle concentration at diameters smaller than 6 μm and unexpected flattening of particle size distributions at diameters larger than 0.5 cm, we focus here on the central range of particle sizes between those limits (Fig. 4).

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