Untangling Microphysical Impacts on Deep Convection Applying a Novel Modeling Methodology. Part II: Double-Moment Microphysics

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ABSTRACT

The suggested impact of pollution on deep convection dynamics, referred to as the convective invigoration, is investigated in simulations applying microphysical piggybacking and a comprehensive double-moment bulk microphysics scheme. The setup follows the case of daytime convective development over land based on observations during the Large-Scale Biosphere–Atmosphere (LBA) experiment in Amazonia. In contrast to previous simulations with single-moment microphysics schemes and in agreement with results from bin microphysics simulations by others, the impact of pollution simulated by the double-moment scheme is large for the upper-tropospheric convective anvils that feature higher cloud fractions in polluted conditions. The increase comes from purely microphysical considerations: namely, the increased cloud droplet concentrations in polluted conditions leading to the increased ice crystal concentrations and, consequently, smaller fall velocities and longer residence times. There is no impact on convective dynamics above the freezing level and thus no convective invigoration. Polluted deep convective clouds precipitate about 10% more than their pristine counterparts. The small enhancement comes from smaller supersaturations below the freezing level and higher buoyancies inside polluted convective updrafts with velocities between 5 and 10 m s\(^{-1}\). The simulated supersaturations are large, up to several percent in both pristine and polluted conditions, and they call into question results from deep convection simulations applying microphysical schemes with saturation adjustment. Sensitivity simulations show that the maximum supersaturations and the upper-tropospheric anvil cloud fractions strongly depend on the details of small cloud condensation nuclei (CCN) that can be activated in strong updrafts above the cloud base.

1. Introduction

The impact of atmospheric aerosols on the microphysics and dynamics of deep convection continues to be a subject of considerable interest in modeling and observations. A recent review by Tao et al. (2012) includes around 300 relevant references, while chapter 7, "Clouds and Aerosols," of the Working Group 1 contribution to the IPCC Fifth Assessment Report (IPCC 2013; see http://www.ipcc.ch/report/ar5/s) provides a broader context for the cloud–aerosol interactions and their role in the climate and climate change problem.

The study reported here focuses on a very specific aspect: namely, the possible invigoration of deep convective clouds in polluted environments (e.g., Andreae et al. 2004; Rosenfeld et al. 2008). The invigoration has been argued to originate from the enhanced latent heating when large amounts of liquid water freeze after being transported above the 0°C level by convective updrafts, followed by off-loading of the frozen condensate through precipitation processes.\(^1\) This can occur when collision–coalescence is suppressed in the lower portions of polluted deep convective clouds as a result of high droplet concentrations and reduced droplet sizes. However, the presence of deeper clouds and/or clouds

\(^{1}\) It is emphasized that both latent heating due to freezing and off-loading are needed for invigoration to occur. Latent heat of freezing alone merely balances the loading of the liquid condensate carried aloft. Off-loading is required to increase cloud buoyancy after freezing of the condensate.
with a larger upper-tropospheric cloud cover [cf. Fig. 2 in Rosenfeld et al. (2008)] does not have to imply more vigorous convection. Changes in cloud microphysics, for instance, leading to changes in the partitioning between cloud condensate carried by cloud updrafts and precipitation falling out of them can also have similar effects without any changes in the cloud dynamics (e.g., Morrison and Grabowski 2011).

Cloud modeling is especially useful for studying aerosol effects on deep convective clouds because one can separate meteorological and aerosol conditions and quantify updraft and cloud buoyancy statistics. However, because deep convection involves nonlinear fluid dynamics, separating statistically significant physical effects from the effects of different flow realizations is difficult. One possibility is to consider high-resolution convection-permitting numerical weather prediction (NWP)-type simulations driven by observed meteorology. If the simulation domain is large enough, such an approach includes many clouds and thus incorporates feedbacks between cloud processes and the large-scale environment. If the simulation period is long, say, many days, such simulations sample realistic meteorological conditions (forcings by synoptic variability, topography, etc.), and they include realistic impacts on the surface energy budget. Seifert et al. (2012) were the first to report such simulations applying a convection-permitting limited-area NWP model with a double-moment cloud microphysics scheme [i.e., predicting both mass and number mixing ratios and allowing explicit responses to changes in cloud condensation nuclei (CCN) and ice nuclei (IN)] for three summer seasons’ convective precipitation over Germany. They concluded that (see the abstract) “the CCN and IN assumptions have a strong effect on cloud properties, like condensate amounts of cloud water, snow and rain as well as on the glaciation of the clouds, but the effects on surface precipitation are—when averaged over space and time—small” [Seifert et al. (2012, p. 709); see Fig. 9 therein]. Fan et al. (2013) performed month-long NWP-type simulations over three regions—the tropical western Pacific (TWP), summertime southeastern China (SEC), and summertime southern Great Plains (SGP)—using the Weather Research and Forecasting Model with bin microphysics and contrasted simulations specifying pristine and polluted conditions. The impact on the surface rain accumulation was relatively small: the accumulation was a few percent larger in the polluted case for the TWP and SEC simulations and a few percent smaller (larger) for the polluted case for the initial 20 days (final 10 days) for the SGP site. However, the polluted minus pristine difference was smaller than differences between the simulated and observationally estimated surface rainfall.

The simulated impact on bulk cloud parameters (such as the cloud fraction, cloud-top height, and cloud thickness) was large, in agreement with results discussed in Seifert et al. (2012).

A novel modeling methodology has recently been developed to allow the separation of purely microphysical effects from cloud dynamical impacts (Grabowski 2014, 2015, hereinafter G15). Grabowski (2014) applied the methodology to simulations of shallow convection, whereas deep convection was considered in G15. The methodology is referred to as “microphysical piggybacking.” The main idea is to apply two sets of thermodynamic variables (the potential temperature, water vapor mixing ratio, and all variables describing aerosol, cloud, and precipitation particles) in a single cloud field simulation. The first set is coupled to the dynamics and drives the simulation (set D, as in “driving”), and the second set piggybacks the simulated flow and does not affect it (set P, as in “piggybacking”). The methodology allows for assessing the impacts with an unprecedented accuracy, and it is capable of detecting even minuscule impacts on bulk cloud properties, such as the cloud cover, liquid and ice water path, and surface precipitation. It also allows for comparing local cloud buoyancies between driving and piggybacking sets of thermodynamic variables and thus exploring possible impacts on cloud dynamics. The impact on the dynamics is assessed by performing a second simulation with microphysical sets swapped so the D set becomes the P set, and vice versa. If the differences between results from the D and P sets in the two simulations (i.e., in the original simulation and in the one with sets swapped) are the same except for the sign, then the cloud dynamics is arguably insignificantly affected by the cloud microphysics. This was the case in shallow convection simulations discussed in Grabowski (2014). If the D-minus-P differences have different magnitudes, then an impact on the dynamics is likely.

For the case of unorganized (scattered) deep convection, G15 applied microphysical piggybacking to contrast surface precipitation and updraft–downdraft properties simulated with two single-moment bulk microphysics schemes assuming cloud droplet concentrations corresponding to either pristine or polluted conditions. The two bulk schemes, referred to as simple ice (SIM; Grabowski 1998) and ice A and B (IAB; Grabowski 1999), apply saturation adjustment (i.e., no supersaturation is allowed), and they differ significantly in the representation of ice processes. Applying different schemes in the piggybacking simulations resulted in large differences in the accumulated surface precipitation (see Table 1 in G15). The magnitude of the difference between the D and P sets was about 50% larger when SIM
was driving and IAB was piggybacking compared to when IAB was driving and SIM was piggybacking. This suggests significant differences in the cloud dynamics, an aspect supported by the difference in the cloud updraft statistics (cf. Fig. 7 in G15). However, applying either set (i.e., SIM or IAB) and changing assumed droplet concentrations between the D and P sets resulted in similar magnitudes of the D-minus-P difference (see Table 2 in G15), suggesting a small impact on cloud dynamics due to aerosols. The latter was shown to agree with small differences in the cloud updraft statistics (Fig. 7 in G15). Overall, these results, as well as results discussed in Grabowski (2014) and in Grabowski and Jarecka (2015), document the utility of the piggybacking methodology.

This paper reports piggybacking simulations following G15 but applying a double-moment microphysics scheme. The next section provides a short description of the models applied in this study and the modeling setup. Section 3 discusses simulation results, focusing on aspects discussed in G15 and separating purely microphysical effects from effects on convective dynamics. Because the analysis shows a strong microphysical effect on upper-tropospheric ice clouds, we also apply a radiative transfer model offline to further quantify the impacts. Section 4 provides a brief discussion of sensitivity simulations prompted by results discussed in section 3. A discussion in section 5 concludes the paper.

2. The models

a. Cloud model and modeling setup

The cloud model used in this study, the same as in Grabowski (2014), G15, and Grabowski and Jarecka (2015), is a simplified serial version of the 3D non-hydrostatic anelastic Eulerian–semi-Lagrangian (EULAG) model (http://www.mmm.ucar.edu/eulag/), referred to as the babyEULAG. Herein, the same modeling setup as in G15 is used with the double-moment scheme of Morrison and Grabowski (2007, 2008a,b).

The modeled case mimics daytime convective development over warm-season continents and features a transition from shallow to deep convection due to strongly increasing surface latent and sensible fluxes. The 12-h simulations (i.e., from 0730 to 1930 local time) apply evolving sensible and latent surface fluxes, as in Grabowski et al. (2006) [see Fig. 1 in G15 and the appendix in Grabowski et al. (2006)]. The model setup is exactly the same as in G15. A horizontal domain of 50 km × 50 km is covered by a uniform 400-m grid. In the vertical, the domain extends up to 24 km, applying 81 levels with a stretched grid. The vertical grid length is around 100 m near the surface, with about a dozen levels below 1.5 km. The grid length increases to about 300 and 400 m at 5 and 15 km, respectively. The model time step is 4 s.

The main difference between the current simulations and G15 is that a double-moment bulk cloud microphysics scheme is used here. The warm-rain scheme is that of Morrison and Grabowski (2007, 2008a). It predicts number and mass mixing ratios for cloud water and drizzle/rain (i.e., four prognostic variables). The Khairoutdinov and Kogan (2000) representation of autoconversion and accretion is used [see Morrison and Grabowski (2007) for details]. Ice processes are represented using the double-moment three-variable scheme of Morrison and Grabowski (2008b). In this approach, the ice-particle mass–dimension and projected-area–dimension relationships vary as a function of particle size and rimed mass fraction. The rimed mass fraction is derived locally by separately predicting two ice mass mixing ratios: one including the ice mass grown by vapor deposition and the other including that grown by riming. The third prognostic ice variable is the number mixing ratio of ice particles. This approach allows for representing the gradual transition from small to large ice particles due to growth by water vapor deposition and aggregation, and from unrimed crystals to rimed ice particles and eventually to graupel due to riming. The scheme was used in Slawinska et al. (2009) in kinematic model simulations of deep organized convection, in Grabowski and Morrison (2011) in idealized simulations of convective–radiative quasi equilibrium, and in Morrison and Grabowski (2011, 2013) for simulations of tropical deep convection from the Tropical Warm Pool–International Cloud Experiment (TWP-ICE).

The scheme applied here predicts the evolution of the supersaturation field (Morrison and Grabowski 2008a), instead of using saturation adjustment as bulk microphysics schemes typically do. This is an important feature of the scheme, as illustrated by simulation results discussed below. Predicting supersaturation allows an explicit treatment of the cloud droplet activation. As discussed in Morrison and Grabowski (2008a), an additional prognostic model variable, the number mixing ratio of activated CCN, is used to keep track of aerosol that may still be available to activate cloud droplets above the cloud base, either because of entrainment or an increasing updraft velocity. The number mixing ratio of activated CCN is predicted separately for the two sets of thermodynamic variables (i.e., the one driving the simulation and the one piggybacking it).

The cloud droplet activation scheme is the same as in Morrison and Grabowski (2007, 2008a). It assumes a lognormal CCN single-mode size distribution with the
prescribed total CCN number mixing ratio, mean dry radius, geometric standard deviation of the distribution, and the soluble fraction [cf. Eqs. (9)–(12) in Morrison and Grabowski (2007)]. Following Morrison and Grabowski (2007, 2008a), the latter three parameters are selected as 0.05 μm, 2.0, and 0.7, respectively, with the soluble fraction consisting of ammonium sulfate. The increase in droplet number from activation is calculated from the predicted supersaturation field and specified CCN characteristics using Kohler theory, taking into account the prognosed number of CCN already activated. We contrast convection developing in pristine and polluted environments by assuming a total CCN number mixing ratio of either 100 mg$^{-1}$ (i.e., per cubic centimeter for the air density of 1 kg m$^{-3}$) or 1000 mg$^{-1}$. These will be referred to as pristine (PRI) and polluted (POL), respectively. Sensitivity simulations with an additional small CCN mode will also be discussed later in the paper.

Primary ice initiation in the scheme occurs through several mechanisms. Deposition/condensation-freezing nucleation is parameterized following the supersaturation-dependent formulation of Meyers et al. (1992), applied at temperatures below −8°C and ice supersaturation greater than 5%, and limited to a maximum of 100 L$^{-1}$. Cloud droplet and raindrop heterogeneous freezing follows from the volume-dependent formulation from Bigg (1953). Contact freezing of cloud droplets follows from the approach in Morrison and Pinto (2005). Homogeneous freezing of all cloud and rainwater occurs at −40°C. Note that, for simplicity and to focus on the effects of changes in CCN, IN concentrations acting in contact and deposition/condensation-freezing modes are the same in polluted and pristine environments. However, differences in the cloud and raindrop concentrations in the two environments impact the initiation of ice by heterogeneous and homogeneous droplet/drop freezing.

As in the simulations discussed in Grabowski (2014) and G15, we apply a five-member miniensemble for the piggybacking simulations, with individual members referred to as D-PRI/P-POL (for PRI driving and POL piggybacking) and D-POL/P-PRI (for POL driving and PRI piggybacking). The ensemble members are generated by applying different sets of random numbers in the model initialization and during the simulation (cf. Grabowski et al. 2006). This miniensemble methodology is useful because of the highly nonlinear nature of moist convection. It also improves robustness of the piggybacking results and alleviates, to some extent, the impact of a relatively small horizontal domain applied here, as only a few deep convective clouds can coexist in the period of the strongest forcing (see Fig. 2 in G15 and Fig. 2 herein).

Simulation results are saved as snapshots of model fields every 6 min of the simulation time (i.e., 10 h$^{-1}$). Because of high spatial and temporal variability, surface precipitation is saved every 3 min as the average over all time steps from the preceding 3-min period. These data are used in the analysis presented in section 3.

b. Radiation transfer model

Significantly different mean cloud fields in POL and PRI simulations, as documented in the next section, imply a strong impact of clouds on radiative processes and surface energy budget (Grabowski 2006; Grabowski and Morrison 2011; Fan et al. 2013). To further quantify this aspect, we apply a radiation transfer model to snapshots of model fields. The independent-column approximation model, the same as in Grabowski (2006), comes from the National Center for Atmospheric Research (NCAR)’s Community Climate System Model (Kiehl et al. 1994). The solar radiation geometry is set to fit the geographical location and the time of the year of the field project on which the modeling case is based (Grabowski et al. 2006). The same applies to the time of day that the simulations cover, from 0730 to 1930 local time. The effective radii of water droplets and ice crystals for the radiation transfer model are diagnosed from assumed spectral characteristics and ice particle mass–dimension and projected area–dimension relationships [see Morrison and Grabowski (2008a,b) for details]. The effective radii predicted by the double-moment warm-rain and ice microphysics schemes are additionally limited to be between 4 and 16 μm for the water drops and between 13 and 130 μm for the ice crystals. Such limits are required to avoid unphysical values predicted by the double-moment scheme at grid volumes with extremely low water or ice mixing ratios that causes problems when passed to the radiation transfer code.

3. Results

a. Bulk cloud properties

Figure 1, in a format similar to Figs. 3 and 4 in G15, shows evolution of the cloud fraction profiles (derived from cloud variables in both the driving and the piggybacking sets of thermodynamic variables) for all simulations. The cloud fraction at each level is defined as the fraction of grid points with a sum of cloud water and total ice mixing ratio larger than 0.01 g kg$^{-1}$. The evolution of the cloud field agrees with the results discussed in Grabowski et al. (2006) and in G15. Shallow convective clouds develop in the second hour of the simulation, with the cloud fraction profiles typical for shallow
convection (e.g., Fig. 6 in Siebesma et al. 2003; Fig. 4 in vanZanten et al. 2011). The boundary layer and cloud field deepen as surface fluxes continue to increase, with the transition from shallow to deep convection taking place between hours 4 and 5. Deep convection reaches maximum strength around hour 6, shortly after the maximum surface heat flux. Deep convective clouds dissipate in the afternoon as the surface fluxes decrease, with only upper-tropospheric anvils, remnants of the earlier deep convective clouds, present at the end of the simulation.

Differences between the simulations are relatively minor until significant anvils develop after hour 6. In contrast to results from single-moment simulations discussed in G15 (cf. Figs. 3 and 4 therein), there are large differences in the anvils between PRI and POL, with POL featuring high upper-tropospheric cloud fractions regardless of whether POL drives or piggybacks the simulation. At the end of the simulations, POL sets still have high upper-tropospheric cloud fractions, typically exceeding 0.6, whereas cloud fractions for PRI are typically below 0.2. This is because of contrasting cloud droplet concentrations in POL and PRI that translate into contrasting ice concentrations (i.e., significantly higher in POL) and thus smaller sizes and fall velocities (see below). One can thus expect a strong radiative effect due to these differences, as documented later in the paper. Cloud fractions in D-POL and P-POL are similar, and so are cloud fractions in D-PRI and P-PRI, suggesting rather small impacts on the cloud dynamics. Overall, the results are consistent with those discussed in more detail in bin microphysics simulations of Fan et al. (2013; see Figs. 2–4 therein), although the differences between PRI and POL in simulations discussed here are larger than in the Fan et al. (2013) study. Note that the inclusion of a small-mode CCN substantially reduces the magnitude of these differences in anvil cloud fraction between PRI and POL, illustrated by the sensitivity simulations discussed in section 4. Figure 1 also illustrates the need for an ensemble approach because differences between ensemble members are large in the second half of the simulations.

FIG. 1. Evolution of the cloud fraction profiles for five members of (top) D-POL/P-PRI and (bottom) D-PRI/P-POL piggybacking simulations for (left)–(right) hours 2, 4, 6, 8, 10, and 12. Solid (dashed) lines are for thermodynamic sets driving (piggybacking) the simulated flow.
For illustration, Fig. 2 shows snapshots of the top of the atmosphere (TOA) albedo for randomly selected D-PRI (left column) and D-POL (right column) simulations for hours 2, 6, and 10. In agreement with the cloud fraction profiles in Fig. 1 (and maps of the liquid plus ice water path in Fig. 2 of G15), only shallow and optically thin cumuli are present at hour 2. Because the two simulations represent corresponding ensemble members (i.e., with the same random numbers applied in either pristine or polluted simulations), the cloud fields at hour 2 are close to each other. The mean albedo of the entire scene at hour 2 is around 0.16, only slightly different between PRI and POL. However, the difference between the average cloudy column albedo is about 10% (0.22 for PRI and 0.24 for POL). Deep convection and some anvils, with different cloud realizations, dominate the scene at hour 6. The mean albedo is around 0.30 (around 0.02 larger for POL). At hour 10, there are only a few deep highly reflective convective cores and significant upper-tropospheric anvils. The mean albedo is larger for POL (0.51 vs 0.37), consistent with the higher cloud cover indicated by the cloud fraction profiles in Fig. 1.

Figures 3–5 show profiles of cloud microphysical fields at hour 3, 6, and 11, respectively, for randomly selected members of the D-PRI/P-POL and D-POL/P-PRI ensembles (results from other members are similar and thus are not shown). Variables from both driving and piggybacking sets are shown using solid and dashed lines, respectively. The conditional sampling of microphysical fields is done in the same way as in Grabowski and Morrison (2011): that is, cloud water fields (mass and number mixing ratios $q_c$ and $N_c$) are averaged over points with $q_c$ larger than 0.01 g kg$^{-1}$, rainwater fields ($q_r$ and $N_r$) are averaged over points with $q_r$ larger than 0.001 g kg$^{-1}$, and ice fields (mixing ratios acquired by water vapor diffusion and by riming, $q_{id}$ and $q_{ir}$, respectively, and ice number mixing ratio $N_i$) are averaged over points with the total ice mixing ratio ($q_{id}$ plus $q_{ir}$) larger than 0.001 g kg$^{-1}$. The sampling for a given hour includes snapshots for the hour and four time levels around the hour (i.e., 6 and 12 min before and after) to increase the dataset size. The figures illustrate the key differences between pristine and polluted cloud fields, with the piggybacking approach allowing confident assessment despite the fact that the two D-PRI/P-POL and D-POL/P-PRI members feature different realizations of the cloud field.

While considering the data shown in Figs. 3–5, one needs to be aware that conditional averaging does not allow for assessing domain-averaged properties, such as the surface precipitation rate or cloud fraction. For these, one needs to know the number of data points that are involved in the conditional sampling. For instance, for the surface precipitation, the conditionally sampled surface rain mixing ratios are the same in the left and right panels in Fig. 3, but the higher precipitation rate in the pristine case for the shallow convection phase comes from about twice as many data points used in the conditional sampling of the rain field in PRI when compared to POL (regardless of which set is driving). The same applies to the surface rain field at hour 6 (deep convection phase; see Fig. 4), where there are about 10% more data points for POL regardless of which set is driving. Differences in the in-cloud mean (conditionally sampled) upper-tropospheric anvil clouds are relatively large (Fig. 5), and the number of cloudy grid points is much larger in POL set than in PRI. These lead together to large differences seen in the upper-tropospheric cloud fractions (see Fig. 1).

Only shallow convective clouds are present at hour 3 (Fig. 3). As expected, PRI clouds have lower cloud droplet number mixing ratios (around 20 mg$^{-1}$ for PRI vs around 100 mg$^{-1}$ for POL) and thus more rain,
especially in upper parts of the cloud field. Averaged accumulated surface rain is small at this stage (see Fig. 6 to be discussed later), but more rain in the PRI case leads to less cloud water, as shown by the small but systematic differences in $q_c$ regardless of which thermodynamic set drives the flow.

Deep convection is at full strength at hour 6. Figure 4 shows conditionally sampled cloud water, rain, and ice profiles at hour 6, with the horizontal lines in all panels showing approximate heights of $0^\circ$ and $-40^\circ$C based on the initial sounding. Cloud and rainwater fields below the freezing level show differences consistent with those for shallow convection (i.e., lower droplet concentration, more rain, and less cloud water in the PRI case). There are small but systematic differences in all microphysical species between $0^\circ$ and $-40^\circ$C (i.e., between the horizontal lines in all panels). For the liquid water, the differences are similar to the differences above $0^\circ$C, with smaller cloud droplet number mixing ratios, higher rain number, and less cloud water mass for PRI. For the ice, there are higher ice particle concentrations, and more ice is grown by diffusion in the POL case, arguably because of higher cloud droplet concentrations in this case. There is also more ice mass grown by riming in the POL case in the upper half of the layer between $0^\circ$ and $-40^\circ$C (the differences close to the melting level may depend on the particular flow realization, as the differences between D-PRI/P-POL and D-POL/P-PRI do not seem systematic). Differences in the ice field for temperatures below $-40^\circ$C are significant and systematic, especially for the ice number mixing ratios, over an order of magnitude higher for the POL case. The difference in ice concentrations below and above the $-40^\circ$C level (note the logarithmic scale on the horizontal axis) documents an important role of homogeneous freezing in deep convection. For the ice mass mixing ratios, the differences between D-PRI/P-POL and D-POL/P-PRI simulations reflect different cloud field realizations, but the differences between POL and PRI are similar: more $q_{id}$ for POL around 14 km and more $q_{id}$ for PRI around 10 km. These differences likely come not only from differences in the ice initiation but also from the subsequent growth and fallout.

Only anvil clouds are present late during the day, remnants of earlier deep convection. This is illustrated in Fig. 5, showing ice profiles at hour 11. Ice number mixing ratios in the upper troposphere are about an order of magnitude higher in the POL case regardless of which set drives the simulation. The maximum of the diffusively grown mass mixing ratio around 12 km for the POL set arguably comes from the low sedimentation rate of small ice particles. As one might expect, the mass
mixing ratio grown by riming is practically zero. Differences for heights between 0° and −40°C are small, and it is unclear if they are statistically significant.

The left panels in Fig. 6 show evolution of the cloud cover and accumulated rain in all members of D-POL/ P-PRI and D-PRI/P-POL piggybacking simulations. Evolution of the difference between POL and PRI is shown in the right panels. As in Grabowski et al. (2006) and in G15, the cloud cover is defined as the fraction of model columns with cloud condensate path larger than 0.01 kg m⁻², with the cloud condensate defined as in the cloud fraction profiles (i.e., the sum of cloud water and total ice mass mixing ratios). Table 1 shows the time-averaged cloud cover and total rain accumulation for all ensemble members, together with POL and PRI differences in the piggybacking simulations. Figure 6 shows large differences between cloud cover in POL and PRI starting around hour 6, in agreement with data shown in Fig. 1. At the end of the simulations, PRI sets are practically cloudless regardless of whether PRI is driving or piggybacking, whereas POL sets still have 100% cloud fractions. PRI has about 10% lower rain accumulation at the end of simulation. However, PRI has higher mean precipitation rate during the shallow convection period, but it insignificantly contributes to the total accumulation that is dominated by deep convection. The right panel shows that the difference in rain accumulation between POL and PRI is about 50% larger when POL is driving (around 0.3 mm when POL is driving vs 0.2 mm when PRI is driving). As argued in
G15, this difference may come from a small but systematic difference in the cloud dynamics, an aspect quantified later in the paper. The figure and table show that the total rain accumulations are close to 3 mm. This agrees with the accumulations from G15’s comprehensive single-moment ice scheme referred to as IAB. The differences in mean cloud cover between driving and piggybacking sets do not seem as systematic as in the rain accumulations, perhaps because of a significant spread in the upper-tropospheric cloudiness among ensemble members, as shown in Fig. 1.

The presence (absence) of statistical significance of the precipitation accumulation (cloud cover) difference between the two ensembles is supported by the Student’s $t$ test statistic. The statistic is calculated as $|a_1 - a_2|/\left[\sum (s_1^2 + s_2^2)/n\right]^{1/2}$, where $a_1$ and $s_1$ ($a_2$ and $s_2$) are the absolute value of the mean and the standard deviation of the D $-$ P (P $-$ D) differences shown in Table 1, $t$ is the confidence interval (taken as 2.3 for the 95% confidence level and $2n - 2 = 8$ degrees of freedom of the two ensembles), and $n = 5$ is the single ensemble size. The values of the statistic larger/smaller than 1.0 imply that the difference is statistically significant or insignificant. The statistic is equal to around 6 for the total rain accumulations and around 0.6 for the cloud cover. This shows that the difference between D-PRI minus P-POL and D-POL minus P-PRI in the rain accumulation is statistically significant, whereas it is not for the cloud cover.

**b. Impact on radiative transfer**

Figure 7 shows evolution of the horizontally averaged TOA outgoing longwave radiation (OLR), TOA albedo, and the surface net radiative flux for all simulations. The 12-h-averaged values are shown in Table 2. As Fig. 7 shows, differences between PRI and POL ensembles emerge around hour 3 in the surface energy and TOA albedo regardless of whether PRI or POL drives the simulation. Because only shallow convection is present at that time, the differences are dominated by the Twomey effect (Twomey 1974): that is, more solar radiation reflected back to space by polluted clouds with smaller cloud droplets. As one might expect, OLR is only weakly affected by the presence of these shallow clouds. OLR begins to decrease around hour 4, when the cloud field deepens. The TOA albedo steadily increases, faster in the POL cases. The OLR differences between PRI and POL become apparent around hour 6, and they continue to increase as a result of increasing differences in the upper-tropospheric anvils, reaching values up to 100 W m$^{-2}$ around hour 9. The anvils also lead to large differences in the TOA albedo, between 0.2 and 0.3 around hour 9. Because the upper-tropospheric anvils begin to disappear in the last couple hours of the simulations in PRI cases (regardless of whether PRI is driving or piggybacking), OLR begins to recover and reaches values close to those at the start of the simulation. POL simulations, however, even at hour 12 have OLR around 200 W m$^{-2}$ smaller than the initial value. The OLR differences between PRI and POL in the last 2 h of the simulations mostly affect the atmosphere because the differences in the net surface radiation are only 10–20 W m$^{-2}$. This is confirmed by the mean profiles of the atmospheric radiative heating rates that show large-amplitude (up to 10 K day$^{-1}$) heating–cooling upper-tropospheric dipoles (i.e., heating at the cloud base and cooling at the cloud top) because of the presence of the anvils.

Table 2 shows 12-h averages of the net surface radiation, OLR, and TOA albedo (the latter defined as the...
12-h mean reflected solar flux at TOA divided by the 12-h-mean TOA incoming solar flux). As the table documents, the differences are large: 20–30 W m\(^{-2}\) for the net surface radiation and OLR and up to 0.05 for the TOA albedo. The differences between simulations driven by PRI and POL are not statistically significant. For instance, the mean D-PRI minus P-POL for the net surface radiative flux is around 33 W m\(^{-2}\) with a standard deviation of about 4 W m\(^{-2}\) for the five-member ensemble. The mean D-POL minus P-PRI value is around −29 W m\(^{-2}\) with a similar standard deviation. Thus, the difference in the mean, around 4 W m\(^{-2}\), is smaller than the sum of the standard deviations (8 W m\(^{-2}\)). The statistic for the Student’s

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<th>Table 1. Rain accumulation and 12-h-mean cloud cover in all piggybacking simulations.</th>
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<td>Rain accumulation (mm)</td>
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<td>D-PRI</td>
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test is around 0.7, similar to the cloud cover statistic at the end of section 3a.

The above results document a large impact on the surface mean energy budget. If the simulations were run for many days with interactive radiation and land surface models (e.g., as in Fan et al. 2013), such a large difference between PRI and POL would arguably lead to a significant feedback on clouds and precipitation. Such an argument may explain why the impact shown in Table 2 is much larger than the one shown in Fan et al.

### Table 2. The 12-h-mean net surface radiative flux, OLR, and TOA albedo in all piggybacking simulations.

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<th></th>
<th>Net surface radiation (W m⁻²)</th>
<th>OLR (W m⁻²)</th>
<th>TOA albedo</th>
</tr>
</thead>
<tbody>
<tr>
<td>D-PRI</td>
<td>396 403 398 397 403</td>
<td>242 247 244 240 245</td>
<td>0.257 0.248 0.255 0.257 0.249</td>
</tr>
<tr>
<td>P-POL</td>
<td>357 373 364 361 375</td>
<td>196 209 203 198 217</td>
<td>0.310 0.290 0.301 0.304 0.286</td>
</tr>
<tr>
<td>Difference</td>
<td>39 30 34 36 28</td>
<td>46 38 41 42 28</td>
<td>-0.053 -0.042 -0.046 -0.047 -0.037</td>
</tr>
<tr>
<td>D-POL</td>
<td>377 371 373 364 381</td>
<td>211 200 213 197 220</td>
<td>0.284 0.292 0.289 0.301 0.279</td>
</tr>
<tr>
<td>P-PRI</td>
<td>405 401 405 397 405</td>
<td>250 239 251 242 250</td>
<td>0.245 0.251 0.243 0.255 0.245</td>
</tr>
<tr>
<td>Difference</td>
<td>-28 -30 -32 -33 -24</td>
<td>-39 -39 -38 -45 -30</td>
<td>0.039 0.041 0.046 0.046 0.034</td>
</tr>
</tbody>
</table>
However, sensitivity simulations presented in section 4 also show that the difference between PRI and POL becomes significantly smaller if an additional mode of small aerosols is included in the simulations (cf. Fig. 12). It is also worth pointing out that such a feedback cannot develop in simulations discussed in Seifert et al. (2012) because of their modeling methodology that features 48-h hindcasts and not extended-range free-running simulations.

c. Cloud buoyancy and dynamics

To search for possible dynamical differences between PRI and POL simulations, we consider buoyancy and vertical velocity statistics between various simulations following G15. Buoyancy is defined as in G15 (see p. 2459) and it includes temperature, humidity, cloud condensate, and precipitation perturbations. Figure 8 (in a format similar to Figs. 11, 12 in G15) shows scatterplots of the difference between the local buoyancy calculated applying D and P sets of thermodynamic variables as a function of the D set buoyancy at 3- and 9-km height for hour 6 of the simulation. The data come from single ensemble members of D-POL/P-PRI and D-PRI/P-POL simulations, including only points with the sum of all cloud and precipitation mixing ratios larger than 0.1 g kg$^{-1}$ and vertical velocity larger than 1 m s$^{-1}$ (the figures for other members show similar patterns). The heights shown in Fig. 8 are selected because one expects only warm-rain processes within cloud updrafts at 3 km (ambient temperature around 9°C) and ice processes dominant within updrafts at 9 km (ambient temperature around −27°C). At 9 km (top panels in

![Figure 8: Scatterplots of the difference between buoyancy calculated applying D and P sets of thermodynamic variables as a function of the buoyancy for cloudy updraft points calculated applying the D set. Data for grid points at (bottom) 3 and (top) 9 km for hour 6 of a randomly selected member for (left) the D-PRI/P-POL and (right) the D-POL/P-PRI ensemble. Note the different scale on vertical axes of top and bottom panels.](image-url)
Fig. 8), the differences between buoyancies from the D and P sets are relatively small (note different scales on the vertical and horizontal axes). Positive D-PRI buoyancies (the top-left panel) typically correspond to (much smaller in the absolute sense) negative differences between P-POL and D-PRI buoyancies; that is, buoyancies in the polluted set are typically slightly smaller than those in the pristine set driving the simulation. Similarly, positive D-POL buoyancies in the top-right panel typically correspond to small positive differences between P-PRI and D-POL buoyancies. These small differences are in the opposite direction to what one expects from the hypothesized upper-tropospheric invigoration of polluted deep convective clouds (e.g., Rosenfeld et al. 2008). Buoyancy plots (not shown) at 7 km (as in G15; ambient temperature around −12°C) as well as at 11 km (ambient temperature of around −42°C; i.e., above the homogeneous freezing level) indicate larger scatter around the zero line for the P-POL/D-PRI and P-PRI/D-POL differences, with no clear tendency for the mean buoyancy difference, similar to the analysis for the IAB scheme in G15 (Fig. 12 therein).

Buoyancies at 3 km (bottom panels in Fig. 8; note a different scale on the vertical axes than in the top panels) clearly show that the P-POL/D-PRI and P-PRI/D-POL differences are much larger than at 9 km, up to as much as half of the driving buoyancy or even larger. For positive buoyancies of the driving set, the buoyancy in the polluted set is typically larger [i.e., positive (negative) differences in left (right) bottom panels]. This is reminiscent of the difference between pristine and polluted shallow warm convective clouds investigated by Grabowski and Jarecka (2015), with polluted clouds experiencing more buoyancy because of smaller supersaturation. Grabowski and Jarecka (2015) showed that the effect is small for shallow convection, with the density temperature lower by 0.1 K for a supersaturation of around 1% when compared to saturated conditions. The buoyancy differences in the bottom panels of Fig. 8 are much larger: a buoyancy difference of 0.02 m s⁻² corresponds to about 0.5-K density temperature difference. Note that the difference is opposite in sign to the effect of cloud water off-loading as a result of drizzle/rain formation and fallout, as in the case of the simulations in G15, but it is consistent with the effect of the finite supersaturation, smaller in the POL case, as discussed in Grabowski and Jarecka (2015). However, if the buoyancy difference comes from the finite supersaturation, then the magnitude of the supersaturation difference between PRI and POL has to be much larger than in a typical shallow convective cloud considered in Grabowski and Jarecka (2015).

Figure 9 shows scatterplots of the supersaturation in the piggybacking set of thermodynamic variables, the quasi-equilibrium supersaturation, and the number mixing ratio of the activated CCN as a function of the supersaturation in the driving set at a height of 3 km for hour 6. Only cloudy points with q_i larger than 1 g kg⁻¹ and updrafts stronger than 1 m s⁻¹ are included in the analysis. The supersaturations are calculated by applying the temperature and water vapor mixing ratio from either the D or P set. The quasi-equilibrium supersaturation is calculated as S_{qe} = c w t, where w is the updraft speed, t is the phase relaxation time scale defined as 1/τ = d(N(r_c + N(r_s)) (r_c and r_s are the mean radii of cloud droplets and drizzle/rain drops, respectively), and c = 5 × 10⁻⁴ m⁻¹ and d = 3 × 10⁻⁴ m² s⁻¹ are numerical coefficients (cf. Grabowski and Wang 2013). Top panels in Fig. 9 show number mixing ratios of activated CCN in the D and P sets of thermodynamic variables. One striking feature of the figure is the magnitude of the supersaturation, up to 10% and even higher. The S_{qe} values show that such high supersaturation is not unrealistic and arguably comes from large vertical velocities even at the 3-km height. Perhaps more importantly, the number mixing ratios of activated CCN show that, for supersaturation larger than 1% or 2%, essentially all CCN are already activated in either the D or P sets. This result motivated sensitivity simulations with increased concentrations of small mode CCN discussed later in the paper. Note that 5% supersaturation gives about 0.5 K of additional density temperature [when compared to the situation with no supersaturation (Grabowski and Jarecka 2015)], which is sufficient to explain the buoyancy differences in Fig. 8.

Figure 10 includes a fraction of the dataset used in Fig. 9 to highlight the relationship between the supersaturation in the driving set and the variables that affect the quasi-equilibrium supersaturation (middle panels in Fig. 9), the updraft velocity, and the number mixing ratios for the cloud water and rain. As one might expect, large supersaturations (several percent) correspond to large vertical velocities (several meters per second). Supersaturations are larger when PRI is driving (i.e., in the left panels). Despite significant scatter, it is apparent that high supersaturations also correspond to reduced cloud droplet concentrations and increased raindrop concentrations in both driving and piggybacking sets, regardless of which set is driving. The slope of the mean relationship between the vertical velocity and supersaturation is different between PRI and POL, arguably reflecting the dependence of the supersaturation on the vertical velocity and mean microphysical characteristics.

Cloud updraft statistics from all simulations at hours 6 and 7 are compiled in Fig. 11. The large panels show the
vertical velocity histograms at 3- and 9-km heights. The number of updrafts for each 1 m s$^{-1}$ bin is calculated separately for each ensemble member, and then the total and the standard deviation (among ensemble members) are derived. The histograms show results from both PRI and POL simulations (i.e., two bars per 1 m s$^{-1}$ bin). Small panels show whether the differences for each bin between the two ensembles are statistically significant. Two populations can be argued to be statistically different at the 95% confidence level (i.e., confidence interval around 2.3 for the 8 degrees of freedom of the two ensembles) if the Student’s $t$ test statistic is larger than approximately 1.0: that is, the difference of the mean is larger than the mean standard deviation (the latter calculated as the square root of the sum of the squared standard deviations from the two five-member ensembles). The stars in small panels show the ratio between the mean standard deviation sum and the difference between the mean values. Thus, if the star is between the dotted lines (showing the 1 and $-1$ values), then the difference between POL and PRI is statistically significant at the 95% confidence level.

As Fig. 11 shows, the differences between POL and PRI updraft statistics are insignificant at 9 km except for
perhaps a couple bins where the difference might be statistically significant. This is in agreement with the small differences in the buoyancy shown in Fig. 8. However, there are statistically significant differences between POL and PRI at 3-km height. For 2 and 3 m s\(^{-1}\) bins, simulations driven by pristine aerosols have a statistically significant larger number of updrafts. This can be argued to come from more efficient conversion of cloud water into drizzle/rain and its subsequent fallout that increases parcel buoyancy in the PRI case. This was argued to lead to larger rain accumulations in the pristine cases in G15 simulations. On the other hand, for updrafts in the range from 5 to 10 m s\(^{-1}\) in the bottom panel of Fig. 11, polluted clouds have a statistically significant larger number of updrafts. This agrees with the data shown in Fig. 10 and comes from the effects of the finite supersaturation (smaller in POL by several percent for large supersaturations, as shown in Figs. 9, 10) on the buoyancy field (cf. bottom panels in Fig. 8). This mechanism was absent in the simulations reported in G15 because of the saturation adjustment applied in SIM and IAB microphysical schemes there. There is also a statistically significant difference in the number of downdrafts in the −3 to −4 m s\(^{-1}\) range, larger in the pristine case.

**FIG. 10.** Scatterplots of (bottom) vertical velocity, (middle) rainwater number mixing ratio, and (top) cloud water number mixing ratio as a function of the supersaturation in the driving set of thermodynamic variables at a height of 3 km and at hour 6 for (left) D-PRI/P-POL and (right) D-POL/P-PRI. Number mixing ratio in the driving (piggybacking) set is shown using crosses (circles). Only 4% of data points are shown.
The differences discussed above likely come from contrasting effects of the cloud droplet concentration between shallow and deep convective clouds. Note that the simulations feature various convective clouds at hour 6, both deep and shallow, as well as clouds at different stages of their development. In shallow clouds with vertical velocities of a few meters per second and thus with a small effect of the finite supersaturation on the cloud buoyancy (Grabowski and Jarecka 2015), reduced droplet concentration increases conversion from cloud water to rain and thus increases cloud buoyancy in the pristine case. This condensate off-loading mechanism was argued to play the key role in the difference in precipitating shallow convection simulations applying either the gravitational collision kernel or the collision kernel that included effects of small-scale cloud turbulence (Wyszogrodzki et al. 2013; Grabowski et al. 2015). In contrast, for deep cumuli featuring strong cloud updrafts at 3 km (up to 10 m s\(^{-1}\); cf. Fig. 10), lower supersaturations in the polluted case lead to increased cloud buoyancy.

4. Sensitivity simulations with increased small CCN concentrations

Because both POL and PRI sets of thermodynamic variables are completely depleted of CCN that could be activated below the freezing level (cf. Fig. 9), three-member-ensemble piggybacking simulations were run with an additional CCN mode characterized by a mean
dry radius of 0.01 μm and number mixing ratio of either 500 mg \(^{-1}\) for PRI and 5000 mg \(^{-1}\) for POL, with other characteristics prescribed to be the same as the larger modes. The most significant impact of the additional small CCN mode is on the upper-tropospheric cloudiness, making the PRI simulations much closer to POL. At the same time, impacts on the buoyancy, dynamics, and resulting surface precipitation are relatively small. This is illustrated in Figs. 12 and 13, which are in a format similar to Figs. 1, 6, and 9. The cloud fraction profiles for PRI and POL ensembles with the additional CCN mode differ less than in the original simulations (cf. top panels in Figs. 1 and 12). This leads to a relatively small difference in the cloud cover (not shown). The surface rain accumulation is still larger in POL (bottom panels in Fig. 12), and the difference between D-POL and P-PRI accumulation is about 40% larger than the difference between D-PRI and P-POL, similar to the original set of simulations (cf. Fig. 6). The buoyancy plot (as in Fig. 8) is also similar, although the difference in buoyancy at 3 km between POL and PRI is smaller (not shown). The latter is consistent with smaller supersaturations at 3 km, as shown in Fig. 13, typically smaller than 5% in PRI and 3% in POL. The concentrations of activated CCN in sensitivity simulations (top panel in Fig. 13) are higher than in simulations discussed previously, but not all available CCN are activated. The latter is equal to 600 and 6000 mg \(^{-1}\) in PRI and POL,
respectively, as marked by dashed lines in the top panels of Fig. 13 in both driving and piggybacking sets of thermodynamic variables.

5. Discussion and conclusions

Following the study of G15, we report on simulations applying the microphysical piggybacking methodology and double-moment bulk microphysics scheme of Morrison and Grabowski (2007, 2008a,b). Application of the double-moment scheme provides a link between warm-rain processes, the simulated cloud droplet concentration in particular, and the development and growth of the ice field. Such a link was absent in simulations applying the single-moment schemes discussed in G15. Moreover, the double-moment scheme used here predicts cloud supersaturation (in contrast to G15’s single-moment SIM and IAB schemes that apply saturation adjustment), allowing simulation of the CCN activation near the cloud base as well as aloft because of increasing updraft speed or entrainment (Morrison and Grabowski 2008a; Slawinska et al. 2012). Contrasting CCN number mixing ratios are specified, either 100 or 1000 mg$^{-1}$, referred to as pristine and polluted, respectively. Homogeneous and heterogeneous ice formation processes are represented in the simulations, with IN concentrations for the latter identical in both pristine and polluted conditions to focus solely on the effects of CCN on the cloud microphysics and dynamics. The simulation setup, the same as in G15, considers...
daytime convective development over land due to increasing surface heat fluxes (Grabowski et al. 2006). There are no clouds at the simulation onset, and shallow convection develops after a couple hours, followed by shallow-to-deep convection transition in midday. Only upper-tropospheric anvil clouds are present in the last few hours of the 12-h simulation, remnants of the earlier deep convection.

Application of the double-moment scheme to the same simulation setup as in G15 results in large differences between pristine and polluted clouds. The most important impact is on the upper-tropospheric anvil clouds in the second half of the day. This agrees with previous studies, such as Morrison and Grabowski (2011), Seifert et al. (2012), and Fan et al. (2013). The impact comes from microphysical effects involving more numerous cloud droplets leading to more numerous, smaller, and slower-falling ice crystals and hence greater anvil cloud fraction. While differences in anvil cloud fraction between pristine and polluted are evident in the simulations that include a small CCN mode, these differences are much smaller than in the simulations without this small mode.

There seems to be a small impact on the upper-tropospheric cloud dynamics in contrast to the convective invigoration hypothesis of Andreae et al. (2004) and Rosenfeld et al. (2008). When averaged over the 12-h simulation period, polluted clouds produce about 10% more surface precipitation. The difference comes not from ice processes, as hypothesized by Andreae et al. (2004) and Rosenfeld et al. (2008), but from the dynamical mechanism for cloud regions with temperatures above freezing. The mechanism involves stronger deep convective updrafts below the freezing level for polluted clouds as a result of smaller supersaturations and thus greater latent heating and cloud buoyancy. Grabowski and Jarecka (2015) showed that 1% supersaturation corresponds to about 0.1-K potential density temperature reduction compared to the situation without supersaturation. Deep convective clouds simulated by the double-moment scheme show not only larger supersaturations below the freezing level [up to 10% (Figs. 9, 10) for the original setup and up to 5% when an additional small CCN mode is included (Fig. 13)], but also large differences between driving and piggybacking sets of thermodynamic variables. Such differences are sufficient to affect cloud buoyancy, larger in the polluted case, as shown in Fig. 8. Figure 8 also shows that there is no systematic difference in the cloud buoyancy above the freezing level. These differences in the cloud buoyancy, or the lack thereof, explain differences in the cloud updraft statistics (Fig. 11).

Large supersaturations simulated by the double-moment bulk scheme and their impact on the cloud buoyancy and updraft strength below the freezing level in deep convective clouds simulated here may be quite important for the cloud dynamics. For instance, there is a negative feedback between the supersaturation and updraft strength, with stronger updrafts leading to larger supersaturations and thus lower cloud buoyancy (Grabowski and Jarecka 2015), which in turn reduces the updraft strength. It is unclear what role in convective dynamics such considerations play. Hall (1980, p. 2502, bottom of the left column, and Fig. 7 therein) noticed supersaturation values higher than 5% in his idealized bin microphysics warm-rain and ice simulations of moist convection initiated by a warm bubble. These high supersaturation values were simulated in the upper part of the cloud (around 6-km height, ambient temperature around −15°C), where updraft velocity exceeded 8 m s$^{-1}$. Hall argued that these high supersaturations originated from removal of cloud water by precipitation processes and inability of the remaining cloud water to absorb water vapor available for diffusional growth in the strong cloud updraft. The same mechanism seems to operate in our simulations, because the high supersaturations are present in cloudy volumes with reduced cloud droplet concentrations and increased raindrop concentrations in both pristine and polluted conditions (Figs. 9, 10). Large supersaturations are also accompanied by a lack of CCN available to activate, as shown in Fig. 9. However, large supersaturations are still present when an additional CCN mode is added in sensitivity simulations, with some CCN remaining unactivated. Lebo et al. (2012, Fig. 13 therein) show similar supersaturations in the case of supercell simulations. Because supercells feature even stronger vertical velocities (up to several tens of meters per second), large values of the supersaturation and activation of all available CCN should not be surprising. As results presented herein show, large supersaturation can also exist within unorganized (scattered) deep convection, like that over the Amazon basin.

The mechanisms discussed here cannot be simulated by models applying saturation adjustment, and thus they were absent in the simulations of G15. It is perhaps worthwhile to stress that most double-moment schemes (or partly double-moment schemes: that is, applying mass and number mixing ratios for some but not all condensate and precipitation species) use saturation adjustment to calculate condensation rate rather than calculating condensation from the predicted supersaturation (e.g., Cohard and Pinty 2000; Thompson et al. 2004, 2008; Morrison et al. 2009). Thus, the negative
feedback between the supersaturation and updraft strength mentioned above cannot be simulated. Note that saturation adjustment leads to the highest buoyancy possible (and thus arguably to the strongest updrafts and downdrafts), with finite supersaturations leading to a reduction of cloud buoyancy (Grabowski and Jarecka 2015). Such effects can be exposed with confidence applying the piggybacking methodology.

Sensitivity simulations discussed in section 4 show that small interstitial aerosols, unable to activate at the cloud base, may strongly impact upper-tropospheric anvil clouds (cf. Figs. 1, 12). These aerosols can get activated above the cloud base because of high supersaturations driven by vertical velocity increasing with height. They subsequently affect the concentration of ice crystals when the cloud droplets freeze after being transported into the upper troposphere. This aspect is quantified in Fig. 14. It shows the variability of the mean upper-tropospheric ice concentration (in an approximately 3-km-deep layer below the cloud top) versus the mean concentration of cloud droplets (in the layer between 2 and 4 km) in deep convective columns, defined as columns with a cloud base below 2 km, cloud top above 11 km (temperature around –42°C), and maximum vertical velocity in the column larger than 5 m s⁻¹. The data come from all time levels between hours 6 and 8: that is, when deep convection is in its maximum strength. Although the scatter of the individual data points is large, a clear pattern is evident: moving from PRI to POL increases both the droplet concentration and the ice concentration, and adding small CCN shifts both PRI and POL distributions toward higher concentrations. This provides strong support for the link between CCN and upper-tropospheric ice concentrations in deep convection. An additional aspect, not considered in our study, is the vertical variability of aerosols. Because some CCN may come from air entrained into a cloud above the cloud base, differences between boundary layer and free-tropospheric aerosols may play a role as well. Assuming that CCN do not have volatile components, one may attempt to investigate the role of small CCN by considering differences in residuals of cloud droplets (e.g., as in Twohy et al. 2013) collected just above the cloud base and those collected at higher levels. If samples collected at higher levels contain a larger fraction of small CCN than just above the cloud base, such measurements can provide indirect support for the significance of in-cloud droplet activation well above the cloud base.

Finally, double-moment bulk microphysics schemes still rely on considerable simplifications for the formation, growth, and fallout of ice particles. We plan to expand simulations discussed here through the application of the bin microphysics, as in Fan et al. (2013). We will report the results of such simulations in the next installment of this series.

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