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1. INTRODUCTION

Previous studies have suggested that precipitation by stratocumulus in the marine boundary layer (MBL) can significantly affect boundary layer thermodynamics and energetics, leading to profound changes in cloud amount and internal cloud properties (Paluch and Lenschow 1991; Stevens et al. 1998). Accurately representing the sensitivity of cloud and radiative properties to microphysical processes is important for assessing aerosol indirect effects in large scale models and for correctly forecasting MBL cloud systems on shorter timescales.

The classical model of precipitation formation and growth proceeds through the nucleation of cloud droplets on cloud condensation nuclei (CCN), growth by condensation, and growth by coalescence (Beard and Ochs 1993). The long timescale required for condensational growth of droplets large enough for collection efficiencies to become significant has been an issue of ongoing investigation for many years. In short, this simple model of nucleation, condensation, and coalescence (the warm rain process) is insufficient to explain the rapid growth of precipitation-sized droplets in observations. The bottleneck in the classic theory is the development of sufficient droplets in the ~20-25 μm range.

Giant CCN (GCCN; $1 < r < 10 \mu\text{m}$) have been suggested as a mechanism to develop drizzle nuclei in this size range, which may initiate the development of precipitation (Johnson 1982). Studies have shown that sea salt nuclei are often present in concentrations similar to those of drizzle drops, implying they may play an important role in the production of drizzle (O'Dowd et al. 1997). Furthermore, although high CCN concentrations are typically associated with the suppression of precipitation (Albrecht 1989), the presence of GCCN may

enable the precipitation process in such an environment (Johnson 1982).

Because of the high solubility and the large size of sea salt GCCN, they deliquesce into droplets many times their dry size at subsaturated relative humidity (RH). However, only the smallest of the deliquesced nuclei reach their equilibrium (Köhler) size at 100% RH in a short time (order seconds). Because of the slowness of condensational growth, upon entering cloud base the size of the larger particles is typically less than equilibrium size corresponding to 100% RH. Thus, these deliquesced GCCN are not "activated" in a Köhler sense yet are sufficiently large to serve as drizzle nuclei. Mordy (1959) assumed that particles smaller than 0.12 μm at cloud base deliquesced to equilibrium radii at 100% RH, while particles larger than 1.2 μm deliquesced to a size corresponding to equilibrium at 90% RH. A 3.5 μm sodium chloride (NaCl) nuclei at an equilibrium RH of 90% deliquesces to a "wet" nuclei of 20.2 μm (Table 1, Kogan 1991), in the range of drop sizes that can initiate coalescence. Here we present a new parameterization of giant CCN suitable for use in bulk microphysical models.

2. GCCN PARAMETERIZATION

This GCCN parameterization based on first principles uses precise representation of the condensational growth of aerosol particles (CCN wetting or deliquescence) in the subcloud layer. Total concentration (N_g) and the exponent (α) of a Junge power law distribution constitute specification of the GCCN properties. The parameterization includes a prognostic equation for N_g . Based on detailed calculations of nucleation in the subcloud layer performed by Ivanova et al. (1977), the size of a deliquesced CCN exceeds the size of the dry particle by a factor k :

$$k(r_d) = 5.8w^{-0.12}r_d^{-0.214}, \quad (1)$$

where w is vertical velocity in m s^{-1} and r_d is the dry radius in μm . For a characteristic stratocumulus vertical velocity of 0.6 m s^{-1} and GCCN radii from 1

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to 10 μm , the factor k ranges in value from 6.2 to 3.8. Incorporating this expression for k into the third moment of the (Junge) size distribution results in a source term, which can be incorporated into the prognostic rainwater equation of bulk microphysical models:

$$\frac{dq_g}{dt} = 9.82 \times 10^{-7} \frac{\alpha}{(\alpha - 2.358)} \frac{dN_g}{dt}, \quad (2)$$

where the shape parameter α is assumed to be 4, q_g is the rain water source term due to GCCN [$\text{g g}^{-1} \text{s}^{-1}$], and N_g is the deliquesced GCCN concentration [cm^{-3}]. Upon saturation, all deliquesced GCCN are assumed to become drizzle nuclei.

3. GCCN PARAMETERIZATION TESTS

Preliminary tests of the GCCN parameterization employ the CIMMS large eddy simulation (LES) framework under clean and polluted conditions (ASTEX A209 case). We perform three dimensional ($40 \times 40 \times 51$) simulations with horizontal and vertical grid spacings of 100 and 25 m, respectively, using the CIMMS LES (Kogan et al. 1995; Khairoutdinov and Kogan 1999). The LES is equipped with the bulk microphysical parameterization of Khairoutdinov and Kogan (2000) and the GCCN parameterization.

The LES is first run for a spin-up time of 40 minutes in order to establish MBL structure. During this time, cloud properties are diagnosed using simple saturation adjustment. After the spin-up, droplet nucleation proceeds according to Cs^k ,

where the ambient large mode CCN concentration C equals 82 and 628 cm^{-3} for the clean and polluted cases, respectively. Given 0.5 for k , a supersaturation of 0.2% equates to droplet concentrations of 285 in the polluted case. A dynamically integrated nucleation parameterization that predicts cloud droplet formation based on concentrations of CCN and GCCN, and vertical velocity, is under development for use in the bulk model. This model will respond to changes in CCN concentration and will represent the reduction in nucleated droplets arising from the presence of GCCN suppressing the ambient supersaturation field (Ghan et al. 1998).

Sensitivity to giant CCN is explored through two experiment configurations. First, GCCN are specified as an initial value problem with domain-uniform concentrations of 30, 300, and 1360 L^{-1} . The last value corresponds to that of the O'Dowd et al. (1997) sea spray "jet" mode at 10 m s^{-1} . These GCCN are added to the polluted and clean background CCN populations of 628 and 82 cm^{-3} . These simulations are compared to control cases with no GCCN. As expected, the polluted simulations show the greatest sensitivity to GCCN. Relative to the control case, increasing amounts of GCCN produce the expected response of depleted droplet concentrations, reduced liquid water, increased drizzle, and the tendency of the PBL circulation to decouple (Fig. 1). Adding GCCN to the clean case, which is already precipitating, affects the simulation very little (not shown).

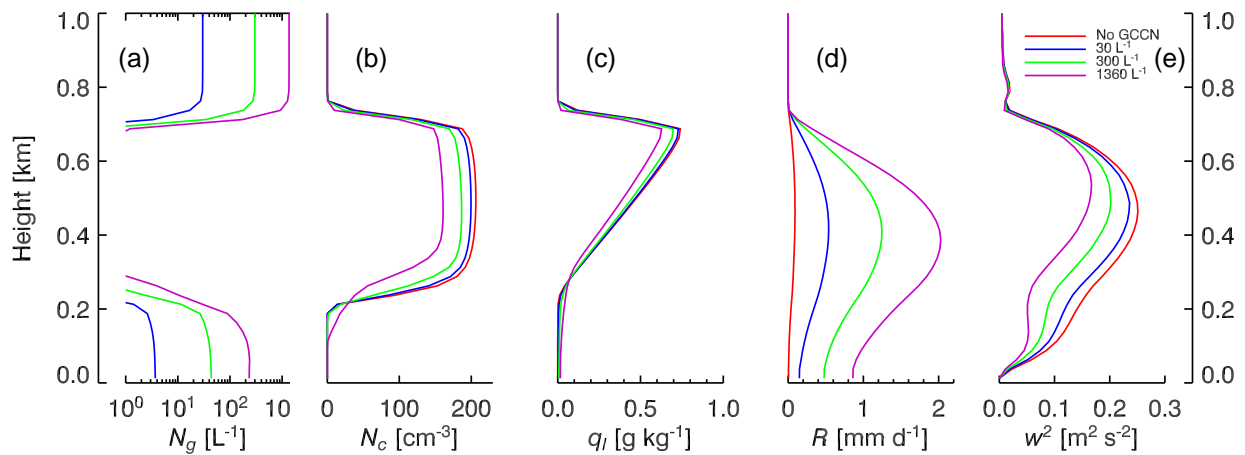


Figure 1. Hourly mean profiles (1-2 h) of LES quantities for the polluted simulation series. Each profile corresponds to a GCCN concentration specified in the initial value problem. (a) GCCN concentration; (b) cloud droplet concentration; (c) liquid water mixing ratio; (d) drizzle rate; (e) vertical velocity variance.

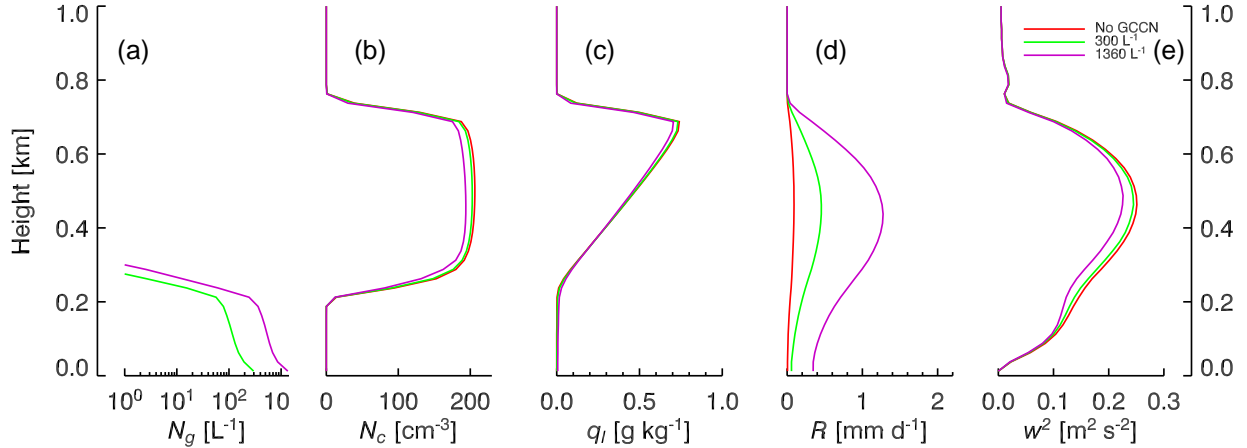


Figure 2. Hourly mean profiles (1-2 h) of LES quantities for the polluted simulation series. Each profile corresponds to a surface GCCN concentration that specifies the boundary value problem. Panels are as in Fig. 1.

Removal of GCCN by cloud processing occurs rapidly in the initial value problem, as indicated by the difference between the free troposphere and subcloud layer GCCN concentrations in Fig. 1a. The e-folding decay time for GCCN in all three simulations is 20 min, and GCCN concentration falls to 1% of the initial value after 114, 149, and 150 minutes (corresponding to the 30, 300, and 1360 L^{-1} simulations, respectively). Because after only 20 min the response to different amounts of drizzle in the GCCN simulations is limited, we infer that the similar e-folding timescale in all three simulations is dictated simply by the eddy turnover timescale. The large variation between the 1% values, on the other hand, implies a feedback of the GCCN-induced drizzle on the MBL dynamics and GCCN transport. A small background source via entrainment from the free troposphere is present but of much smaller magnitude than the cloud processing sink of GCCN.

The initial value series of simulations represent transient responses to environmental conditions. Since the most likely source for GCCN in the MBL is from the sea surface, we perform a series of experiments configured as a boundary value problem, with GCCN initially zero everywhere, except for different GCCN concentrations fixed at the surface. The cloud layer is then “seeded” from below via upward transport of GCCN from the surface interface. These simulations are summarized in Fig. 2 (The effect of 30 L^{-1} is so small that it is not shown). The effects of the GCCN are qualitatively similar though more subtle relative to the initial value problem. GCCN source and sink rates are

such that the profiles of subcloud GCCN in Fig. 2 become nearly invariant over time.

4. INFLUENCE OF GCCN ON CLOUD RADIATIVE PROPERTIES

Our analysis of the influence of GCCN on cloud system radiative properties follows Feingold et al. (1999). Cloud system albedo is a function of optical depth τ ,

$$A = \frac{(1-g)\tau}{2 + (1-g)\tau}, \quad (3)$$

where g is the asymmetry parameter (≈ 0.84). Aerosol indirect effects are frequently formulated as a sensitivity, called “susceptibility,” of albedo to a change in droplet number, which has units of $[cm^3]$ and may be approximated as

$$S = \frac{A(1-A)}{3N}. \quad (4)$$

The effect on radiative properties of adding various concentrations of GCCN to background clean and polluted cases CCN is summarized in Fig. 3. GCCN has little effect on the optical properties for the clean cases, largely because they are already drizzling. Adding GCCN to the polluted background CCN, on the other hand, results in noticeable reductions in optical depth and albedo. The reduction in optical depth and albedo results from a reduction in cloud liquid water content (from drizzle loss), and a decrease in droplet concentration accompanying drizzle production (collection). Less liquid water spread out over fewer drops

decreases the backscatter cross section and the optical depth. Absolute susceptibility varies little over the GCCN concentrations, mainly for the reason that the relative difference in droplet number N between the simulations is quite small. The response of albedo to changes in droplet concentration (S) is smaller in the polluted case. In other words, equivalent changes in N produce more albedo response in the clean case (small N) than in

the polluted case (large N). Yet Figs. 3a and b plainly demonstrate that the polluted case is more sensitive to the addition of GCCN. For this reason, susceptibility relative to the control simulations (Fig. 3d) most aptly illustrates the sensitivity of albedo to change in droplet number. As expected, the relative susceptibility of the polluted case is much greater than that of the clean case, and increases with increasing GCCN.

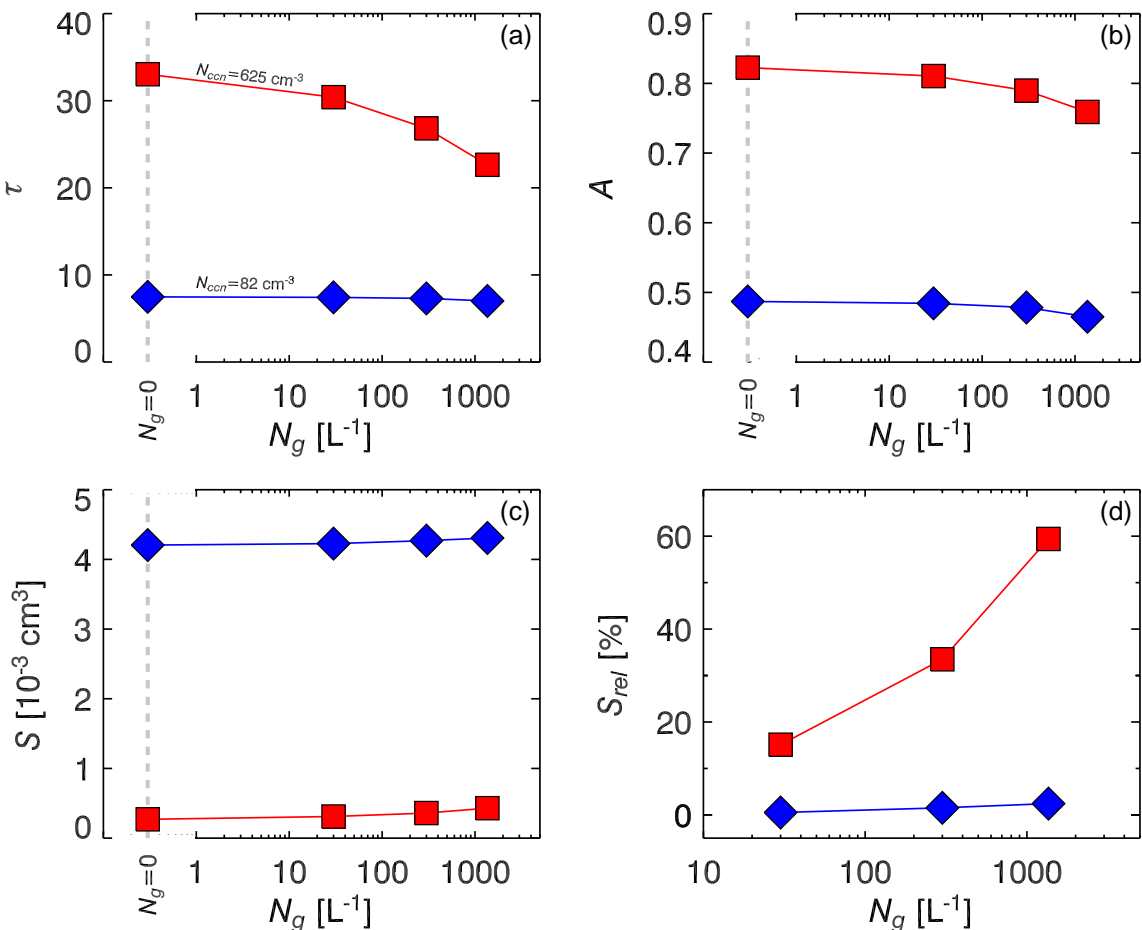


Figure 3. Hourly domain-mean calculations (2-3 h) of radiative quantities for clean (blue) and polluted (red) simulation series. (a) Optical depth; (b) albedo; (c) susceptibility $[A(1-A)/(3N)]$; (d) Susceptibility relative to the control runs without GCCN.

5. SUMMARY AND COMMENTS

The behavior of a new parameterization of giant CCN is tested in a bulk LES framework. Results are consistent with previous simulations using explicit microphysical methods (Feingold et al. 1999). Microphysical and cloud radiative properties exhibit the greatest sensitivity to the addition of GCCN when the background aerosol concentration

is high. Increasing GCCN to the polluted background CCN case results in a decrease of optical depth and albedo, and a significant increase in relative albedo susceptibility.

Rapid removal of GCCN by cloud processing in a few eddy turnover timescales (e-folding time of N_g decrease is ~ 20 min) implies the importance of accurately specifying the flux of GCCN from the ocean surface. Specifying GCCN both as an initial value problem and a boundary value problem both

produce qualitatively reasonable responses; however, the boundary value problem has a more realistic physical basis and is less prone to artificial spikes in drizzle rate early in the simulation. Furthermore, common parameterizations dependent on wind speed may be easily employed to parameterize the source of GCCN from sea spray. In the boundary value problem, however, longer integrations become necessary to “spin-up” the turbulent transport of GCCN from the surface up to the cloud layer. The subcloud GCCN profiles are nearly constant in time, indicating the cloud GCCN uptake and processing are in close balance.

ACKNOWLEDGEMENTS

This research was supported by ONR Grants N00014-05-1-0550 and N00014-03-1-0304, and by the Office of Science (BER), U.S. Department of Energy, Grant No. DE-FG02-05ER64062.

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