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THREE-DIMENSIONAL ASPECTS OF DROPLET NUCLEATION

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1. INTRODUCTION

Correctly representing cloud droplet concentration in numerical models is critical in evaluating aerosol indirect effects and requires a realistic treatment of droplet nucleation. One method of parameterizing the nucleation process is to rely on simple diagnostic relations between cloud condensation nuclei (CCN) and droplet number (e.g. Jones et al. 1994; Lieput and Lohmann 2001; Mechem and Kogan 2003). This method of activation results in a full range of cloud droplet concentrations, but since nucleation process cannot respond directly to model dynamics, the relationship between droplet and CCN concentrations is constrained more rigidly than in nature. Recent investigations of droplet nucleation tend toward a more detailed specification of aerosol parameters in a simplified dynamical framework, typically that of non-entraining adiabatic ascent (Abdul-Razzak et al. 1998; Snider et al. 2003).

Classical theory predicts that CCN activation occurs at or just above cloud base in buoyant updrafts, where supersaturation is maximum. As a parcel ascends adiabatically and becomes slightly supersaturated, the larger nuclei activate first. Once CCN are activated, the rapid flux of vapor to the droplet constitutes a sink to the ambient supersaturation field. Assuming the increase in supersaturation from adiabatic ascent remains larger than the vapor uptake from the activated CCN, supersaturation continues to increase, enabling smaller CCN to nucleate droplets, which then grow by condensation. Eventually, the rate of vapor uptake by the droplets will be greater than the rate of supersaturation increase from adiabatic ascent, reducing the parcel supersaturation to zero. This complicated process occurs just above cloud base.

We present preliminary results from a three-dimensional large eddy simulation (LES) employing size-resolved microphysics that suggest aspects of aerosol activation in marine stratocumulus not captured by nucleation schemes based on simple empirical relations or parcel theory. In contrast, we argue that all regions of positive supersaturation, not merely at cloud base, more completely represent the droplet nucleation zone. Furthermore, since nucleation does not occur uniformly but rather only in supersaturated updraft regions, the number of nucleated droplets should be considered an upper bound on grid-mean droplet concentration.

2. 3D STRUCTURE OF NUCLEATION

Simulations employ the CIMMS large eddy simulation (LES) model with size-resolving microphysics, described in detail in Kogan (1991), Kogan et al. (1995), and Khairoutdinov and Kogan (1999). The CIMMS LES is based on 3D Boussinesq dynamics and explicitly represents the turbulent boundary layer eddies. The grid spacing is ideally chosen to be in the inertial subrange, so that the subgrid-scale contribution to the turbulent kinetic energy is small, except near flow interfaces such as the surface and the inversion.

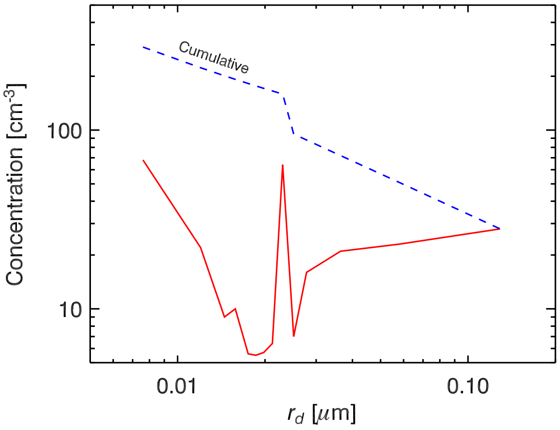

Figure 1. Cumulative CCN spectrum (blue) used to initialize the model. The red line indicates the number of CCN in each bin interval.

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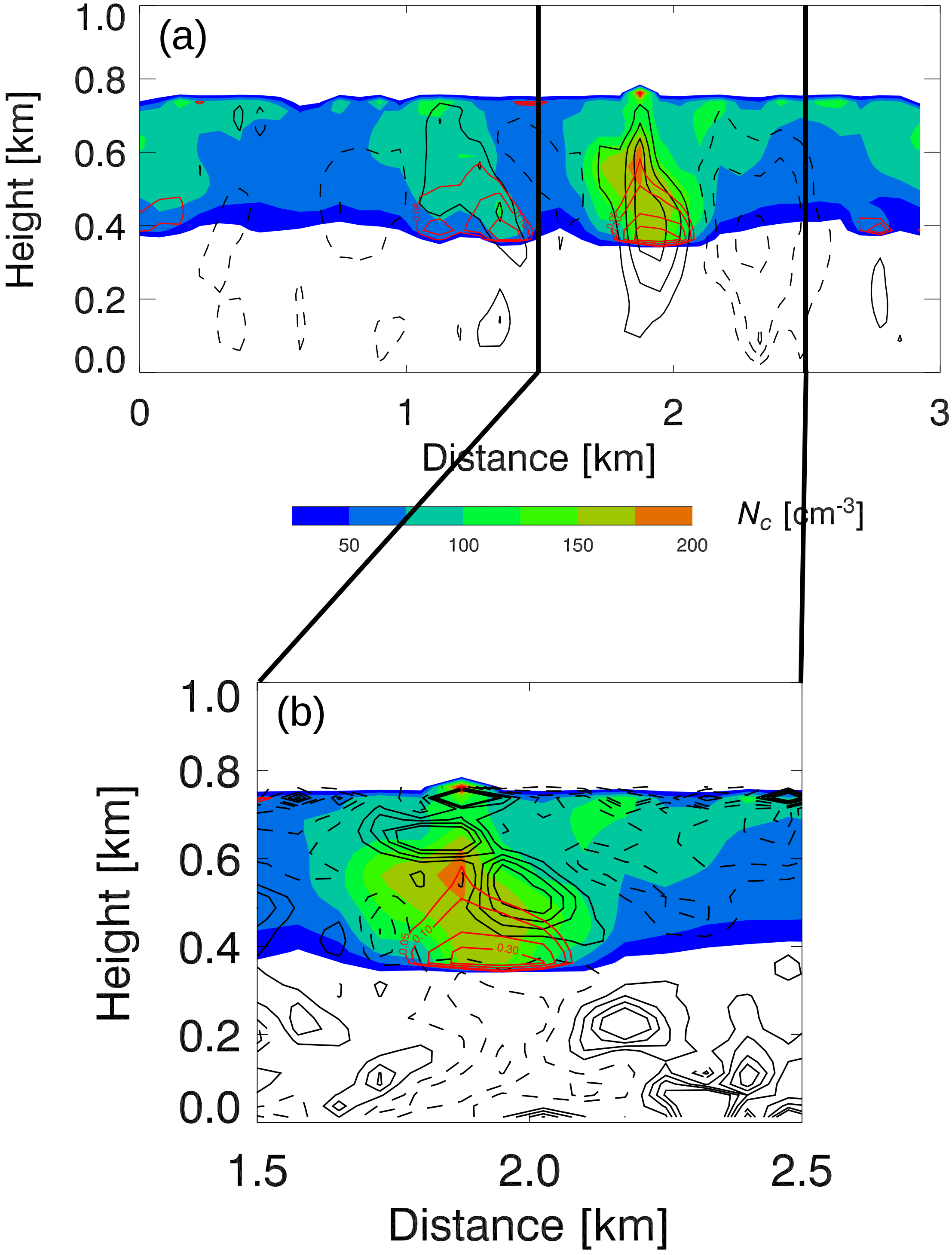

Figure 2. Vertical cross sections of LES results. (a) droplet concentration (color-filled contours), supersaturation (red contours; values of 0.5, 0.1, 0.2, and 0.3%), and vertical velocity (black contours with an interval of 0.5 m s $^{-1}$; negative values dashed); (b) droplet concentration and horizontal convergence (black contours with an interval of 2.5×10^{-3} s^{-1}; negative values dashed).

Size-resolved cloud physics processes are formulated based on prognostic equations for 19 CCN and 25 cloud/drizzle droplet bins. The model has been extensively verified against aircraft observations (Khairoutdinov and Kogan 1999; Liu et al. 2000), and indirect tests of a bulk drizzle parameterization derived from model drop size distributions showed good agreement with in-situ aircraft data (Wood 2005).

The LES is configured based on a cloud-topped boundary layer observed during the Atlantic Stratocumulus Transition Experiment (ASTEX) (A209 case sounding; Duynkerke et al. 1995). LES domain size is 3×3×1.25 km, with grid spacings of 75 m in the horizontal and 25 m in the vertical. Total CCN concentration is 291 cm^{-3}, with a spectral shape (Fig. 1) such that the cloud-mean concentration becomes approximately 75 cm^{-3} at supersaturations typical of marine stratocumulus.

Classical parcel theory predicts that all of the activation occurs at cloud base, where supersaturation is a maximum. A vertical cross section through the LES domain, on the other hand, shows that for some updrafts (e.g. near X=1.9 km in Fig. 2a), the maximum in N_c is well above cloud base, indicating that additional droplet nucleation is occurring in a continuous fashion from cloud base up to the level of maximum N_c. This nucleation is nevertheless restricted to regions of supersaturation, while regions of in-cloud convergence adjacent to supersaturated regions (e.g. left side of the updraft in Fig. 2) strongly suggest that *previously non-activated* aerosol are being supplied to the supersaturated updraft via lateral entrainment. Parcel models do not capture this continuous aspect of nucleation.

The neglect of this additional nucleation of droplets could conceivably lead to an underestimate of N_c. Droplet number at cloud base is well correlated with vertical velocity, especially over supersaturated regions at cloud base (Fig. 3), a dependence that can be represented as a simple power law. This relationship is reflected in frequently employed Cs^k relations and the fact that vertical velocity is a primary factor in determining supersaturation. The relationship between nucleated droplet concentration and updraft is more complicated when the analysis is expanded to all supersaturated regions (the gray “+” marks in Fig. 3), since the additional activation occurring in updrafts above cloud base more completely represents the aerosol activation zone.

Probability distribution functions (PDFs) in Fig. 4 indicate that mean droplet concentration is indeed higher when all supersaturated regions are considered, relative to N_c over supersaturated cloud base regions only (82 vs. 72 cm^{-3}), the difference likely being the direct result of the continuous activation process occurring in the rising parcel. Cloud mean values of N_c for this particular case are 69-71 cm^{-3}, depending on the sampling method employed. Cloud mean values are never greater than the droplet concentrations over the supersaturated nucleation regions.

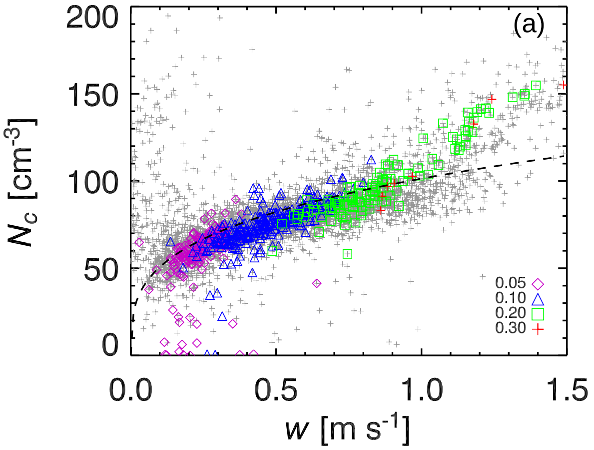

Figure 3. Scatterplot of cloud base droplet concentration as a function of updraft magnitude, stratified by supersaturation [%] according to color and symbol. Gray marks represent all regions of supersaturation. Dashed curve represents a power law fit of $109.9w^{0.459}$.

3. CONCLUSIONS

Results from large eddy simulation suggest that all regions of supersaturation, including those well above cloud base, represent the droplet nucleation zone more completely than simply regions of peak supersaturation at cloud base. Since nucleation does not occur uniformly but rather only in supersaturated updrafts, the number of CCN activated should be considered an upper bound on grid-mean droplet concentration

The pattern of droplet concentration increasing with height in updraft regions suggests a source of unactivated CCN deriving from lateral entrainment. Our future work will employ trajectory calculations to explore the origin of these CCN.

Clearly a non-entraining adiabatic model cannot capture this continuous activation. Including an entrainment term in a 1D model, however, may be able to represent this process for some cases. The concept of parcel and environment is straightforward for a case of low cloud fraction shallow cumulus, but for a solid stratocumulus deck what should be the concept of the "environmental" air that is mixed with an updraft parcel? For this reason, the full three-dimensional aspect of CCN activation may play an important role in dictating cloud layer droplet concentration, and representing this process with a 1D parcel model may prove nontrivial.

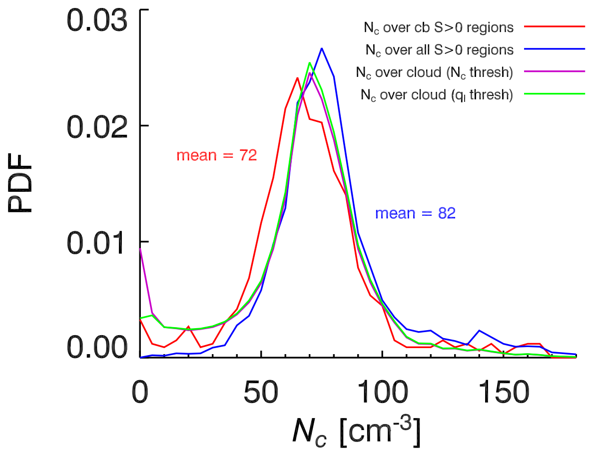

Figure 4. Probability distribution functions of droplet concentration throughout the cloud (purple and green, each representing a different sampling method) and conditionally sampled over supersaturated cloud base (red) and all supersaturated regions (blue).

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REFERENCES

Abdul-Razzak, H., S. J. Ghan, and C. Rivera-Carpio, 1998: A parameterization of aerosol activation, 1. Single aerosol type. *J. Geophys. Res.*, **103**, 6123-6131.

Duynkerke, P. G., H. Zhang, and P. J. Jonker, 1995: Microphysical and turbulent structure of nocturnal stratocumulus as observed during ASTEX. *J. Atmos. Sci.*, **52**, 2763-2777.

Jones, A., D. L. Roberts, and A. Slingo, 1994: A climate model study of indirect radiative forcing by anthropogenic sulphate aerosols. *Nature*, **370**, 450-453.

Khairoutdinov, M. P., and Y. L. Kogan, 1999: A large eddy simulation model with explicit microphysics: Validation against aircraft observations of a stratocumulus-topped boundary layer. *J. Atmos. Sci.*, **56**, 2115-2131.

Khairoutdinov, M., and Y. Kogan, 2000: A new cloud physics parameterization in a Large-Eddy Simulation model of marine stratocumulus. *Mon. Wea. Rev.*, **128**, 229-243.

Kogan, Y. L., 1991: The simulation of a convective cloud in a 3-D model with explicit microphysics. Part I: Model description and sensitivity experiments. *J. Atmos. Sci.*, **48**, 1160-1188.

Kogan, Y. L., M. P. Khairoutdinov, D. K. Lilly, Z. N. Kogan, and Q. Liu, 1995: Modeling of stratocumulus cloud layers in a large eddy simulation model with explicit microphysics. *J. Atmos. Sci.*, **52**, 2923-2940.

Lieput, B. G., and U. Lohmann, 2001: A comparison of sulfate observations and ECHAM4-GCM experiments and its relevance to the indirect aerosol effect. *J. Clim.*, **14**, 1078-1091.

Liu, Q., Y. L. Kogan, D. K. Lilly, D. W. Johnson, G. E. Innis, P. A. Durkee, and K. Nielson, 2000: LES modeling of ship track formation and its sensitivity to boundary layer structure. *J. Atmos. Sci.*, **57**, 2779-2791.

Mechem, D. B., and Y. L. Kogan, 2003: Simulating the transition from drizzling marine stratocumulus to boundary layer cumulus with a mesoscale model. *Mon. Wea. Rev.*, **131**, 2342-2360.

Snider, J. R., S. Guibert, and J.-L. Brenguier, 2003: Aerosol activation in marine stratocumulus clouds: 2. Köhler and parcel theory closure studies. *J. Geophys. Res.*, **108**, 8629, doi: 10.1029/2002JD002692.

Wood, R., 2005: Drizzle in stratiform boundary layer clouds. Part II: Microphysical aspects. *J. Atmos. Sci.*, **62**, 3034-3050.