Supplement of

Lagrangian Particle–Based Simulation of Aerosol-Dependent Vertical Variation of Cloud Microphysics in a Laboratory Convection Cloud Chamber

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Analysis of Microphysical Equilibration Timescales and Their Dependence on Aerosol Concentration

Figure 2 in the text shows that each microphysical variable—cloud droplet number concentration (N_c), mean droplet radius (r_m), and cloud water mixing ratio (q_c)—exhibits distinct timescales to reach quasi-equilibrium conditions, with these timescales varying systematically according to aerosol concentration. Figures S1 and S2 further elucidate how aerosol-induced changes in supersaturation impact these equilibration timescales. Figure S1 presents the equilibration times, defined as the time to reach 95% of their 15-30 minute average values, for q_c, N_c, r_m, and volume-mean droplet radius (r_v) across a range of aerosol concentrations. These equilibration times differ systematically: N_c equilibrates fastest because most aerosol particles rapidly exceed the cloud droplet threshold radius of 1 μm used in this study. Droplet radius (r_m, r_v) equilibrate next, whereas q_c reaches equilibrium slowest, particularly under high aerosol loading, due to the continued slow growth of larger droplets. Figure S2 explicitly compares the equilibration times of q_c and r_v, highlighting how their difference increases with aerosol concentration. This result indicates that q_c continues to evolve significantly after droplet sizes have equilibrated. Physically, this might occur because q_c, being weighted more heavily toward larger droplets (via r^3), continues to increase as those large droplets slowly grow. Under high aerosol concentrations, reduced supersaturation prolongs the growth of these larger droplets, further delaying the equilibration of q_c relative to r_v. Consequently, the gap between the two equilibration timescales becomes increasingly pronounced as aerosol loading might increase.

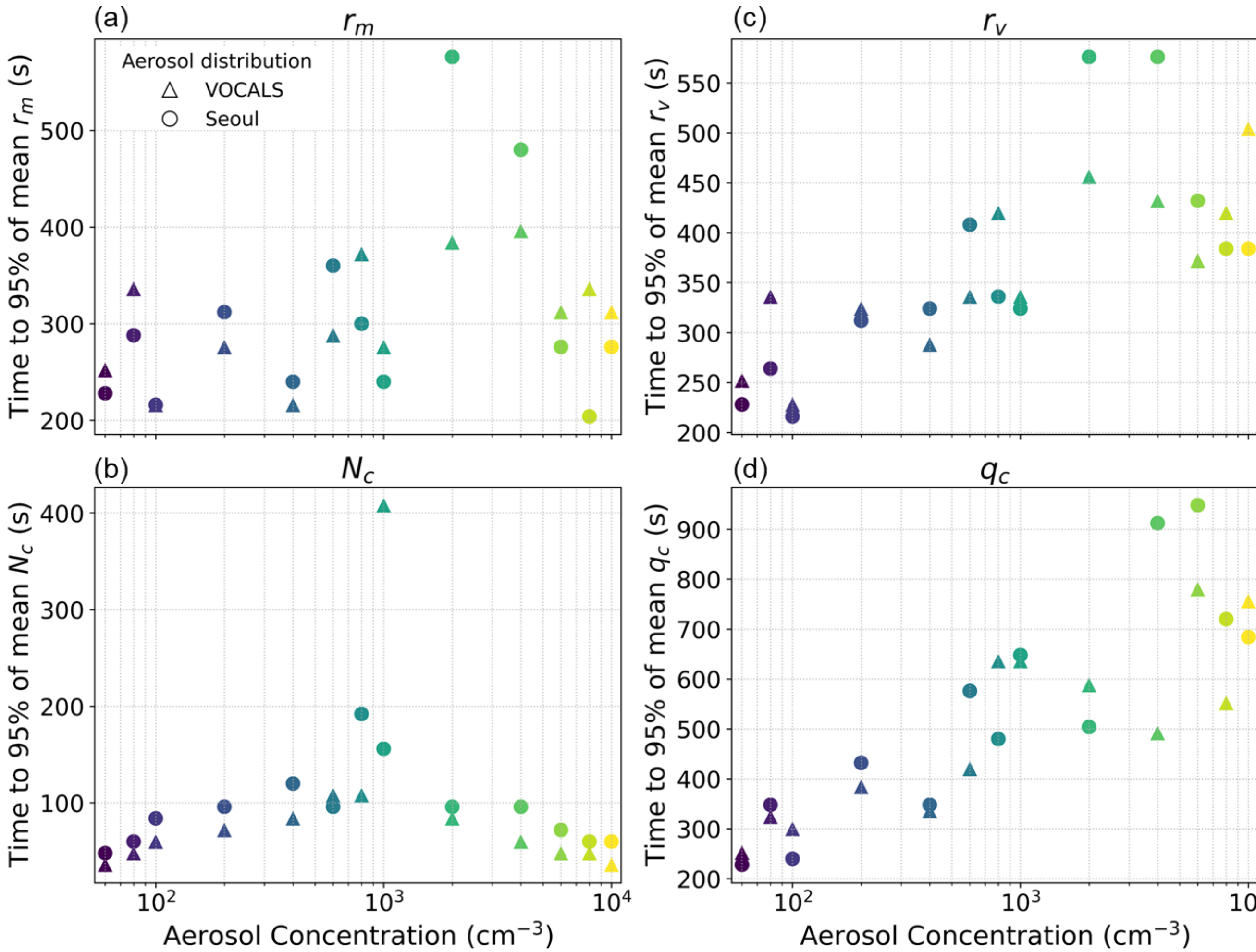

Figure S1. Time required for each microphysical variable to reach 95% of its time-averaged value over the quasi-equilibrium period (15–30 minutes), as a function of aerosol concentration, is shown for VOCALS (triangles) and Seoul (circles) cases. Panels show the timescales associated with the following variables: (a) mean droplet radius (r_m), (b) cloud droplet number concentration (N_c), (c) volume-mean droplet radius (r_v), and (d) cloud liquid water mixing ratio (q_c). The plots highlight distinct timescales for each variable to approach steady-state behavior.

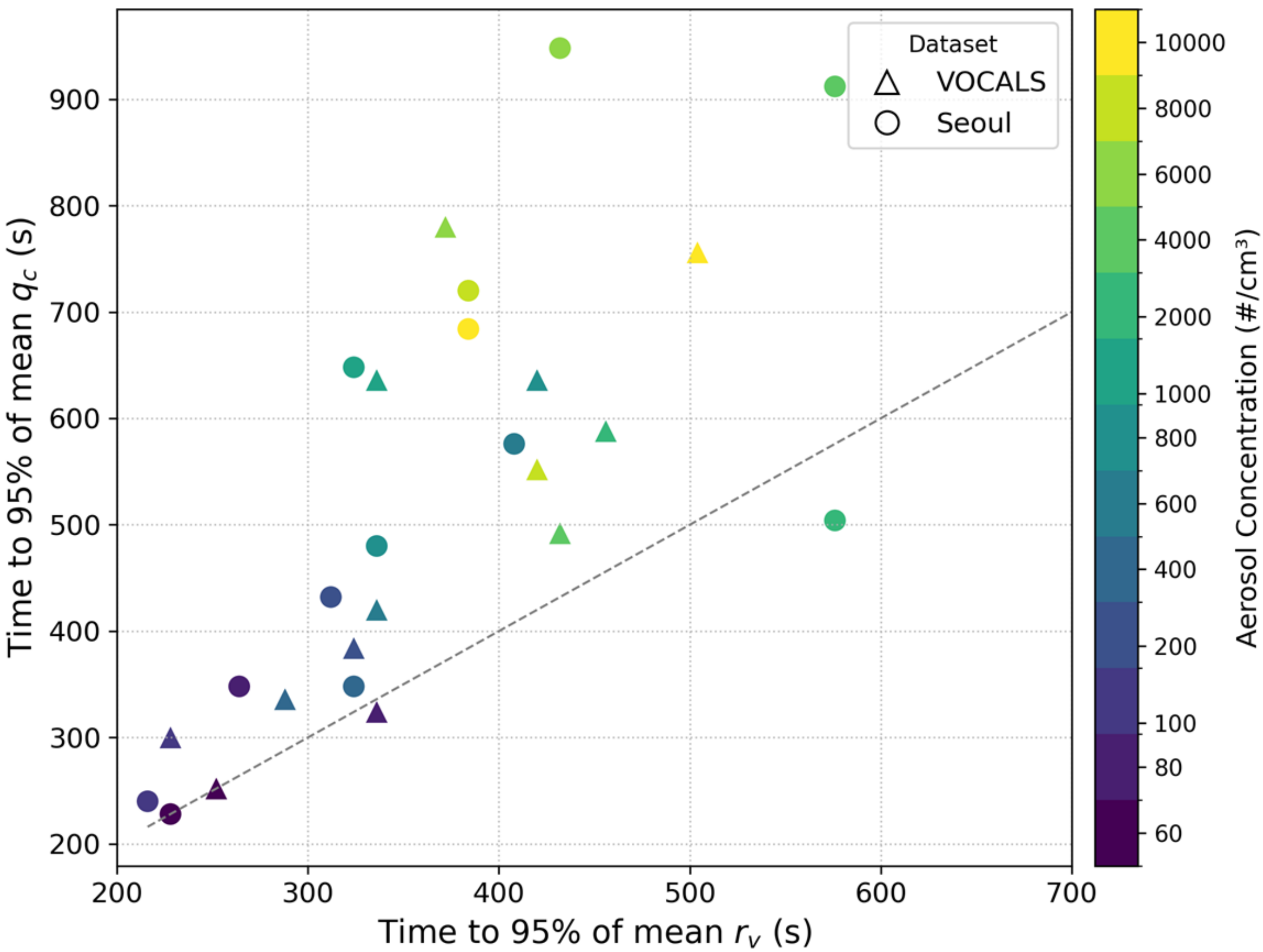

Figure S2. Relationship between the time to reach 95% of the mean quasi-equilibrium (15–30 min) volume-mean droplet radius (r_v) and the mean quasi-equilibrium cloud water mixing ratio (q_c), for VOCALS (triangles) and Seoul (circles) cases. Marker color represents aerosol concentration. The dashed grey 1:1 line highlights that q_c generally equilibrates more slowly than r_v, especially under high aerosol loading.

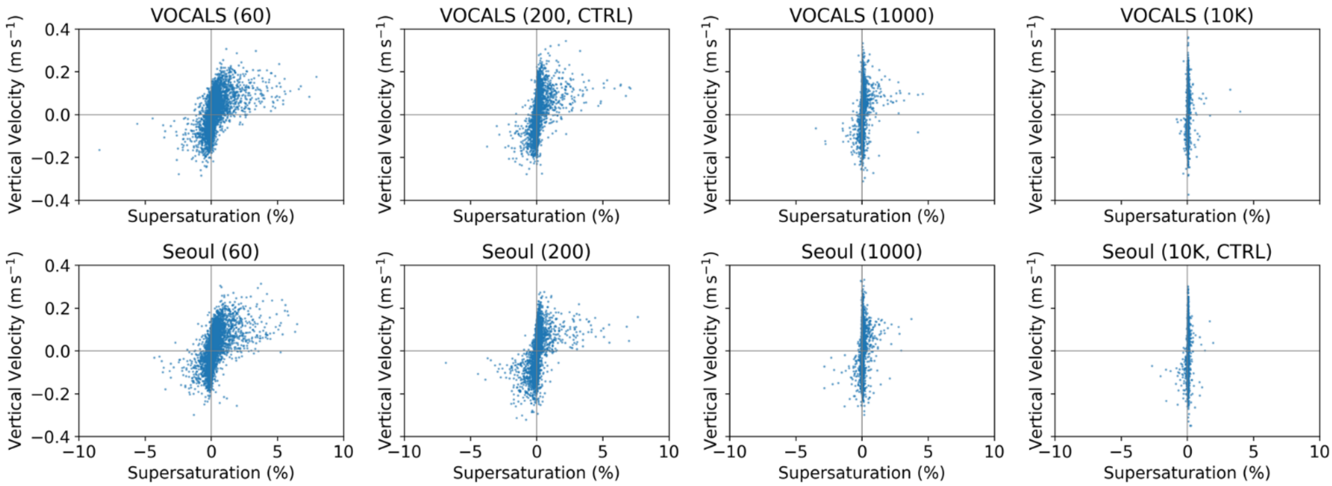

Figure S3. Scatterplot of the vertical velocity as a function of the supersaturation for the last 15 min of the simulations. Only 0.1% of randomly selected grid points away from chamber boundaries are included in the plot.