1 Quantifying the effect of aerosol on vertical velocity and effective terminal

2 velocity in warm convective clouds

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10 Abstract

11 Better representation of cloud-aerosol interactions is crucial for an improved 12 understanding of natural and anthropogenic effects on climate. Recent studies have 13 shown that the overall aerosol effect on warm convective clouds is non-monotonic. 14 Here, we reduce the system's dimensions to its center of gravity (COG), enabling 15 distillation and simplification of the overall trend and its temporal evolution. Within 16 the COG framework, we show that the aerosol effects are nicely reflected by the 17 interplay of the system's characteristic vertical velocities, namely the updraft (w) and 18 the effective terminal velocity (η). The system's vertical velocities can be regarded as 19 a sensitive measure for the evolution of the overall trends with time. Using bin-20 microphysics cloud-scale model, we analyze and follow the trends of the aerosol 21 effect on the magnitude and timing of w and η , and therefore the overall vertical COG 22 velocity. Large eddy simulation model runs are used to upscale the analyzed trends to 23 the cloud-field scale and study how the aerosol effects on temporal evolution of the 24 field's thermodynamic properties are reflected by the interplay between the two 25 velocities. Our results suggest that aerosol effects on air vertical motion and droplet 26 mobility imply an effect on the way in which water is distributed along the 27 atmospheric column. Moreover, the interplay between w and η predicts the overall 28 trend of the field's thermodynamic instability. These factors have an important effect 29 on the local energy balance.

31 1. Introduction

Clouds are key players in the Earth's climate system via their influence on the energy 32 33 balance (Baker and Peter, 2008; Trenberth et al., 2009) and hydrological cycle. Of all 34 of the anthropogenic effects on climate, aerosol's effect on clouds remains one of the 35 most uncertain (Boucher et al., 2013). In warm clouds, aerosol act as cloud 36 condensation nuclei (CCN) around which droplets can form, and therefore aerosol 37 amount and properties determine the initial number of droplets and their size 38 distribution (Squires, 1958; Rosenfeld and Lensky, 1998; Andreae et al., 2004; Koren et 39 al., 2005). The initial droplet concentration affects cloud dynamics via microphysical 40 and dynamical feedback throughout their lifetime. For example, the onset of 41 significant collision events between droplets in polluted clouds (which are initially 42 smaller and more numerous than in clean clouds (Squires, 1958)) is delayed (Gunn 43 and Phillips, 1957; Rosenfeld, 1999, 2000; Squires, 1958; Warner, 1968). This delay 44 can have opposing effects on cloud development by increasing both the water loading 45 (which reduces cloud buoyancy and vertical development) and the latent heat release 46 resulting from the longer and more efficient condensation (increasing cloud buoyancy 47 and vertical development) (Dagan et al., 2015a;Dagan et al., 2015b;Pinsky et al., 48 2013;Koren et al., 2014). We note that often, these opposing effects act at different 49 stages of the cloud's lifetime, further complicating the prediction of overall trends.

50 Vertical velocities (w) are among the key processes driving convective clouds. The 51 intensity, duration and characteristic size of the updrafts determine the convective 52 clouds' properties. In addition, the clouds' vertical velocity affects the distribution of 53 water along the atmospheric column, thereby having a strong effect on radiation 54 (Koren et al., 2010) and heat balance (Khain et al., 2005). Although previous studies 55 have focused on deep convective clouds, these effects are expected to be significant in 56 warm convective clouds as well. Moreover, warm processes serve as the initial and 57 boundary conditions for mixed-phase processes in deep convective clouds, and therefore gaining a better process understanding of the warm phase is essential for 58 59 understanding the deeper systems (Chen et al., 2017).

60 The system has another characteristic velocity that measures droplet mobility. This 61 velocity, defined as the effective terminal velocity (η), measures the weighted-by-62 mass terminal velocity of all hydrometeors within a given volume and therefore defines the falling velocity of the volume's center of gravity (COG) (Koren et al., 2009;Koren et al., 2015) compared to the air vertical velocity. Smaller droplets imply smaller $|\eta|$ (higher mobility) and therefore less deviation from the surrounding air movement. Since η is always negative, smaller $|\eta|$ implies that per a given air updraft, the collective liquid water mass will be carried up higher in the atmosphere. The movement of the COG compared to the surface, defined as V_{COG}, is the vector sum of the two velocities: V_{COG} = $w - |\eta|$.

70 V_{COG} has recently been shown to be a good measure for the temporal evolution of 71 thermodynamic instability in cloud fields (Dagan et al., 2016). V_{COG} represents the 72 vertical movement of liquid water, which is downward gradient of the net 73 condensation-less-evaporation profile. A negative V_{COG} implies net transport of the 74 liquid water from the cloudy layer to the sub-cloud layer. This holds true for clean 75 (low aerosol concentration) precipitating cases (Dagan et al., 2016), in which the 76 water that condenses in the cloudy layer sediments down to the sub-cloud layer where 77 it partially evaporates. The net condensation in the cloudy layer and the net 78 evaporation in the sub-cloud layer produce a decrease in the thermodynamic 79 instability with time. On the other hand, for the polluted non-precipitating cases, V_{COG} 80 is positive; indicating that the net liquid water movement is upward. The water that is 81 being condensed in the lower part of the cloudy layer is transported upward and 82 evaporates in the upper cloudy and inversion layers (Dagan et al., 2016). The end 83 result of this vertical condensation-evaporation profile is an increase in 84 thermodynamic instability with time.

85 Khain et al. (2005) used a two-dimensional cloud model with spectral (bin) microphysics to study the aerosol effect on deep convective cloud dynamics. They 86 87 concluded that one of the reasons for comparatively low w in clean maritime 88 convective clouds compared to polluted continental ones is the rapid creation of 89 raindrops. This increases the liquid water loading in the lower part of the cloud, 90 thereby reducing buoyancy. They also claimed that the delayed raindrop production in 91 the continental cloud increases the duration of the diffusion droplet growth stage, 92 which in turn, increases the latent heat release by condensation.

93 Seigel (2014) showed an increase in *w* with increasing aerosol loading in the cloud 94 core in numerical simulations of a warm convective cloud field. He also showed a decrease in cloud size under polluted conditions due to increased mixing between the
clouds and their dry environment driven by stronger evaporation of smaller droplets in
polluted cases.

98 It has been recently shown (Dagan et al., 2015a;Dagan et al., 2015b;Dagan et al., 99 2017) that under given environmental conditions, warm convective clouds have an 100 optimal aerosol concentration (N_{op}) with respect to their macrophysical properties 101 (such as total mass and cloud top height) and total surface rain yield. For 102 concentrations smaller than N_{op} , the cloud can be considered as aerosol-limited 103 (Koren et al., 2014;Reutter et al., 2009), and an increase in the mean cloud properties 104 with aerosol loading can be expected due to an increase in the condensation efficiency 105 and droplet mobility (Koren et al., 2015; Dagan et al., 2015a; Dagan et al., 2017). 106 Suppressive processes such as enhanced entrainment and water loading take over 107 when the concentrations are higher than N_{op} and reverse the trend. It has also been shown that the value of $N_{_op}$ depends heavily on the environmental conditions 108 109 (thermodynamic conditions that support deeper clouds would have a larger N_{op}).

110 In this work, a bin-microphysics cloud model and large eddy simulation (LES) of a 111 cloud field were used to explore how changes in aerosol concentration affect w and η , 112 the interplay between them and, as a result, the height of the COG in warm convective 113 clouds (Koren et al., 2009).

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115 **2. Methodology**

116 2.1 Single-cloud model

The Tel Aviv University axisymmetric nonhydrostatic cloud model (TAU-CM) with detailed treatment of cloud microphysics (Reisin et al., 1996;Tzivion et al., 1994) was used. The included warm microphysical processes were nucleation of droplets, condensation and evaporation, collision–coalescence, breakup, and sedimentation. The microphysical processes were formulated and solved using the method of moments (Tzivion et al., 1987).

123 The background aerosol size distribution used here represents a clean maritime 124 environment (Jaenicke, 1988). The aerosols are assumed to be composed of NaCl.

The different aerosol concentrations (25, 500 and 10,000 cm⁻³, denoted hereafter as 125 25CCN, 500CCN and 10000CCN, respectively) and size distributions are identical to 126 127 those used in Dagan et al. (2015a). To study the involved processes, we used a wide 128 range of aerosol loading conditions, from extremely pristine to extremely polluted. 129 These specified three aerosol concentrations represent conditions which are below, 130 around and above the optimal aerosol concentration (N_{op}) . To avoid giant CCN 131 effects, the aerosol size distribution was cut at 1 µm (Feingold et al., 1999; Yin et al., 132 2000;Dagan et al., 2015b).

133 The model resolution was set to 50 m, in both the vertical and horizontal directions, 134 and the time step to 1 s. The initial conditions were based on theoretical atmospheric 135 profiles that describe a tropical environment (Malkus, 1958) (see profile T1RH2 in 136 Fig. 1 in Dagan et al., 2015a). They consisted of a well-mixed sub-cloud layer 137 between 0 and 1000 m, a conditionally unstable cloudy layer (6.5°C/km) between 138 1000 and 4000 m, and an overlying inversion layer (temperature gradient of 2°C over 139 50 m). The relative humidity (RH) in the cloudy (inversion) layer was 90% (30%). 140 The results presented here were examined for a few different sets of initial conditions 141 (different inversion-base heights and cloudy layer RH). Although for different initial atmospheric conditions the transition between aerosol invigoration to suppression 142 143 occurs at different aerosol concentration (Dagan et al., 2015a), the conclusions were 144 found to be general for different sets of initial conditions.

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To examine the effect of aerosols on the entire cloud, the properties presented in this
work are cloud mean values weighted by the liquid water mass in each grid cell.
Cloudy grid cells were defined as cells with liquid water content larger than 0.01 g/kg.
The cloud's COG (Koren et al., 2009) was calculated as:

$$COG = \frac{\Sigma m_i z_i}{\Sigma m_i} \tag{1}$$

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151 where m_i and z_i are the mass [kg] and height [m] of voxel *i*, respectively.

152 The η (effective terminal velocity) was calculated according to Koren et al. (2015):

$$\eta = \frac{\Sigma V t_j m_j n_j}{\Sigma m_j n_j}$$
(2)

where V_{t_j} , m_j and n_j are the terminal velocity [m/s], mass [kg] and concentration [cm⁻³] of droplets in bin *j*, respectively. This was calculated for all cloudy grid cells.

156 For being consistent with the COG point of view, the mean air vertical (*w*) was157 calculated as a mean weighted by the liquid mass:

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$$w = \frac{\sum m_i w_i}{\sum m_i}$$
(3)

The axisymmetric model uses a geometry that is only a simplification and idealization of a full 3D flow and does not account, for example, for wind shear and processes acting on larger scales like clouds effect on the environmental conditions with time. For accounting for these processes, we used 3D cloud field scale simulations as well (see Sec. 2.2 below).

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165 **2.2 Cloud field simulations**

166 We used the System for Atmospheric Modeling (SAM) LES model (Khairoutdinov 167 and Randall, 2003) with a bin-microphysics scheme (Khain and Pokrovsky, 2004) to 168 simulate the BOMEX (Barbados Oceanographic and Meteorological EXperiment) 169 warm cumulus case study (Holland and Rasmusson, 1973;Siebesma et al., 2003). The horizontal resolution was set to 100 m, the vertical resolution to 40 m. The domain 170 size was 12.8 x 12.8 x 4.0 km³ and the time step was 1 s. We ran the model for 16 h, 171 but the statistical analysis included only the last 14 h of the simulation. We used 8 172 different aerosol concentrations: 5, 25, 50, 100, 250, 500, 2000 and 5000 cm⁻³. Again, 173 we used a marine background aerosol size distribution (Jaenicke, 1988). Further 174 175 details about the simulations can be found in Dagan et al. (2017).

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180 **<u>3. Results and discussion</u>**

181 <u>3.1 Single cloud: vertical velocity and effective terminal velocity</u>

182 Starting from the single-cloud scale, we first followed the entire cloud mean w (eq. 3), mean η (eq. 2), mean V_{COG}, and COG height (eq. 1) as a function of time for the three 183 different levels of aerosol loading (25, 500, and 10,000 cm⁻³). From an early stage of 184 the cloud's evolution, the cleanest cloud (25CCN) had the lowest COG. This was a 185 186 result of the lower w (Fig. 1a) and larger absolute value of the negative η (caused by 187 the initially larger droplets – Fig. 1b), which together cause a lower V_{COG} (Fig. 1c). At 188 the early stages of the polluted clouds, the 500CCN and 10000CCN COG moved 189 upward at the same rate. After about 60 min of simulation, the 500CCN's COG started to decrease while the 10000CCN's COG remained relatively high. This trend could 190 191 not be explained by the cloud's mean w (Fig. 1a). The 500CCN's w was higher than 192 that of the 10000CCN during the period between 50 and 63 min of simulation. 193 Without considering the effect of η on the COG, one would expect that the 500CCN's 194 COG would be higher than that of the 10000CCN. The 500CCN had lower (more 195 negative) values of η than the 10000CCN, which decreased the height of its COG 196 compared to the 10000CCN. These larger negative values of η in the 500CCN were 197 due to the rain that developed from this cloud (the rain from the 10000CCN is negligible), which led to lower mobility (lower ability to move with the ambient air 198 199 (Koren et al., 2015)).

We note that the vertical change in the COG height is determined by changes in the vertical distribution of water mass due to microphysical processes like condensation, evaporation and removal of mass by rain (in addition to movement according to V_{COG}). Hence, in some parts of the simulations the V_{COG} was not a perfect predictor of the COG evolution. This is especially true when rain and evaporation are strong i.e. toward the end of the clouds' lifetime.

Figure 1 demonstrates the importance of the aerosol effect on both w and η in determining the COG height. Figure 2 presents the evolution of the clouds on the phase space span by w vs. V_{COG}. All clouds began their evolution on the 1:1 line. This means that at the early stages of the cloud's evolution, $\eta \sim 0$ and hence V_{COG} $\sim w$. After about 40 min of simulation, the cleanest cloud's (25CCN) trajectory began to deviate from the 1:1 line to the left, demonstrating an increase in $/\eta$ / and hence lower 212 droplet mobility. The deviation from the 1:1 line occurred later (at about t = 55 min of 213 simulation) in the more polluted simulation (500CCN), whereas for the most polluted 214 clouds (10000CCN), the lack of significant collision–coalescence and rain production 215 resulted in evolution on the 1:1 line throughout the cloud's lifetime. This delay in the 216 deviation from the 1:1 line (increasing the time for which $\eta \sim 0$) demonstrates the 217 increase in droplet mobility with aerosol loading. The longer period for which $\eta \sim 0$ in 218 the polluted cases enables the water mass to be pushed higher into the atmosphere and 219 hence (together with the increase in the air vertical velocity - Fig. 1a) to produce 220 cloud invigoration by the aerosol (Koren et al., 2015).

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3.2 LES results: aerosol effect on the vertical velocity and effective terminal velocity in cloud fields

224 Shifting our view from the single-cloud scale to the cloud-field scale adds another 225 layer of complexity as clouds affect the way in which the whole field's 226 thermodynamics evolve with time. Moreover, 3D simulations account for the effect of 227 wind shear. Aerosol concentration has recently been shown to determine the trend of 228 this evolution (Dagan et al., 2016;Dagan et al., 2017). Clean precipitating clouds act 229 to consume the initial instability that created them by warming the cloudy layer (in 230 which there is net condensation) and cooling the sub-cloud layer (by rain 231 evaporation). On the other hand, polluted non-precipitating clouds act to increase the 232 field's instability by cooling and moistening the upper cloudy and inversion layers.

233 Figure 3 presents the domain's mean w (in both space and time, weighted by the liquid 234 water mass to be consistent with the COG view - see eq. 3 above) vs. the domain 235 mean η . The color-coding in Fig. 3 denotes the different aerosol concentrations. In 236 agreement with previous studies (Saleeby et al., 2015;Seigel, 2014), an increase in 237 aerosol loading yielded an increase in w. In our simulations, this increase is driven by 238 larger latent heat contribution to the cloud's buoyancy due to the increased 239 condensation efficiency (Dagan et al., 2015a;Dagan et al., 2017;Koren et al., 240 2014; Pinsky et al., 2013; Seiki and Nakajima, 2014) and thermodynamic instability 241 (Dagan et al., 2016;Dagan et al., 2017). In parallel, aerosol shifts to smaller droplets 242 (Squires, 1958) and reduces the magnitude of η , indicating better mobility of the 243 smaller droplets (Koren et al., 2015). The outcome of these two effects (that work

together to push the water mass higher in the atmosphere) is an increase in COG
height with aerosol loading (Heiblum et al., 2016b;Dagan et al., 2017).

246 In the single-cloud-scale analysis (section 3.1), we showed how the timing of the 247 evolution of the two velocities dictates the aerosol effect. Here, having many clouds in 248 the field in different stages of their lifetimes, we first analyzed the bulk properties of 249 the two velocities. With the intention of quantifying the relative contribution of the aerosol effect on the mean COG height by modulating w and η , we plotted them one 250 251 against the other for all of the simulations that differed in aerosol loading and for all 252 clouds in the domain (Fig. 3a). For the entire simulation period, the η vs. w scatter plot resulted in an almost a straight line ($R^2 = 0.96$) which was sorted by aerosol 253 254 concentration with a slope of 0.69. This means that an increase in aerosol 255 concentration that will result in a 1 m/s increase in mean w will drive a decrease in the 256 magnitude of $|\eta|$ by 0.69 m/s. In other words, the relative contribution to the changes 257 in the mean COG height in the domain caused by the increase in aerosol loading 258 (Heiblum et al., 2016b;Dagan et al., 2017) during the entire simulation is ~60% due to 259 changes in w and ~40% due to changes in η .

260 To include the aerosol effect on the cloud-field thermodynamic properties, we divided the simulation period into three equal thirds (excluding the first 2 h, each third of a 261 262 period covered 4 h and 40 min). The x and * markers in Fig. 3a represent the first 263 third (2 h to 6 h 40 min into the simulation) and last third (11 h 20 min to 16 h into the simulation), respectively. During the first third, the slope of w vs. η was steeper than 264 the mean over the entire simulation (slope of 0.92 with $R^2 = 0.96$); during the last 265 third, it was more gradual (slope of 0.47 with $R^2 = 0.87$). The almost 1:1 relation 266 between w and η in the first third of the simulation period suggests a comparable 267 268 contribution in determining the aerosol effect on mean COG height. However, the 269 relative contribution of η decreases as the simulation progresses, to about 1/3 during 270 the last third of the simulation period (compared with 2/3 of *w*).

The decrease in the w vs. η slopes toward the end of the simulations is driven by the changes in the thermodynamic instability. The increase in instability under polluted conditions produces an increase in mean w (Dagan et al., 2016). Nevertheless, increased instability and deepening of the cloud layer are not sufficient to produce a significant amount of rain under the most polluted simulations and hence, there is no increase in the magnitude of η . An increase in w with no change in η is manifested as a horizontal shift to the right on the η vs. w phase space (red arrow in Fig. 3a). On the other hand, the decreased instability under clean conditions produces a decrease in both mean w and the rain amount (Dagan et al., 2017), and therefore in $|\eta|$ (blue arrow in Fig. 3a). The end result of the different changes in w and η under clean and polluted conditions is a decrease in the slope of η vs. w and therefore, a decrease in the relative contribution of η to the aerosol effect on the mean COG.

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284 In Fig. 3a, the presented quantities are domain and time averages. Figure 1 showed 285 that the relative contribution of w and η to the aerosol effect on COG height strongly 286 depends on the stage of the cloud's evolution. The averaging in Fig. 3a mixes many 287 clouds at different stages in their evolution and represents the effect on the mean COG 288 in the domain. To further explore the relative contribution of the aerosol effect on w289 and η as a function of cloud-evolution stage, we used a cloud-tracking algorithm 290 (Heiblum et al., 2016a). We identified the growing stage of the clouds as the stage for 291 which the cloud top ascends. Figure 3b presents the η vs. w phase space only for 292 clouds in their growing stage. Table 1 presents the slopes of the linear regression lines 293 for the entire simulation time and for the different thirds of the simulation period. The 294 decrease with time in the relative contribution of η compared to w to the aerosol effect 295 on COG height was also seen for the growing clouds (see the decrease in the slope 296 with time). This, again, was due to the changes in thermodynamic conditions.

As shown for the cloud scale, one of the most notable aerosol effects can be viewed as delaying the onset of significant collection processes in the polluted clouds (Koren et al., 2015), and therefore delaying the increase in $|\eta|$ values early in the cloud's lifetime. Therefore, during the growing stage, the relative contribution of η was higher (Fig. 3b) as compared to "all clouds" (Fig. 3a). This was demonstrated by the increasing slope of the η vs. *w* phase space during the growing stage (Table 1).

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To quantify the evolution of the thermodynamic instability with time as a function of aerosol loading (on a cloud field scale), we looked at the time trends in the η vs. w phase space. We defined the angle 'A' as the angle between the time trend points on 307 the η vs. w phase space per given aerosol loading (the line that connects the first and 308 last thirds of the simulation and the x-axis on the η vs. w phase space —see schematic 309 definition of A in Fig. 3b). We note that A rotates counter-clockwise with increasing aerosol loading (Fig. 3a). It starts as ~100° for the cleanest simulation and 310 311 monotonically increases with aerosol loading to ~360° for the most polluted 312 simulations (Fig. 4b). A between 90° and 180° (as shown for clean cases—Fig. 4b) 313 represents a decrease in both w and $|\eta|$ and hence a decrease in the thermodynamic 314 instability with time. A between 270° and 360°, on the other hand (as shown for the 315 most polluted cases—Fig. 4b), represents an increase in both w and $|\eta|$ and hence an 316 increase in the thermodynamic instability with time.

317 The sign of V_{COG} has been shown to predict the evolution of thermodynamic 318 instability (Dagan et al., 2016). Thus, correlations between A and V_{COG} are expected. 319 Figure 4 presents V_{COG} (Fig. 4a) and A (Fig. 4b) as a function of the aerosol loading, 320 and V_{COG} vs. A (Fig. 4c). Figure 4a and b demonstrates that both the V_{COG} and A 321 increase monotonically with aerosol loading following a similar trend. V_{COG} and A 322 cross the 0 and 180° lines, respectively, at similar aerosol concentrations, representing 323 the transition between consumption and production of the thermodynamic instability 324 (Dagan et al., 2016). Figure 4c further demonstrates an almost perfect linear correlation ($R^2 = 0.99$) between V_{COG} and A sorted by aerosol concentration. 325

326

327 3.3 Summary

328 Clouds form a complex system in which microphysical and dynamical processes are 329 tightly linked and modulated by the thermodynamic properties of the environment. In 330 turn, on the cloud-field scale, clouds affect the field's thermodynamic conditions. The 331 aerosol effect on the droplet size distribution therefore affects all of the above. Better 332 process-level understanding of aerosol effect on cloud and rain properties in the case 333 of warm convective clouds is essential for improving our understanding of the climate 334 system. In this study, our aim was to better understand and quantify the aerosol effect 335 on the air vertical velocity and droplet terminal velocity. Both characteristic vertical 336 velocities quantities modulate the distribution of water along the atmospheric column, 337 and hence affect the radiation (Koren et al., 2010) and heat balance (Khain et al., 338 2005). The findings presented here for the single cloud and cloud field scales could be 339 used in future works to better represent cloud-aerosol interactions in coarser 340 resolution models (like climate models) as it provides a compact way to represent 341 aerosol effect on the liquid water vertical mass flux and clouds effect on the 342 thermodynamic conditions.

343 Analyzing the two characteristic velocities on the cloud scale allows separation, as a 344 first approximation, between the aerosol effects on condensation/evaporation 345 efficiencies (reflected by the magnitude of w) and those on droplet mobility (reflected by the inverse magnitude of η). The magnitudes of w and η act in opposite ways, i.e., 346 347 stronger w and smaller $|\eta|$ imply more efficient transport of liquid water to the upper 348 atmosphere. We use their sum, defined as V_{COG} , to estimate the overall effect on the 349 COG's vertical movement. Single-cloud analysis showed the timing of this interplay 350 and how each velocity affects the COG elevation. It showed that the invigorating 351 aerosol effect can be viewed mostly at the early stages of cloud development, when an 352 increase in aerosol loading enhances the condensation efficiency (reflected as higher 353 *w* levels) and delays the onset of significant collection processes (reflected as a delay 354 in the sharp increase in η). Both act to transfer liquid water higher into the atmosphere 355 (Koren et al., 2015). Later, as the cloud dissipates, the "payment" is viewed as 356 enhanced evaporation, and if the cloud manages to reach the significant collection-357 process stage, then the surface rain is stronger (expressed as a sharp increase in $|\eta|$).

358 Similar to the single-cloud case, the cloud field (LES) results (that unlink the single 359 clouds simulations, account for 3D processes such as wind shear) demonstrated an 360 increase in w and decrease in the magnitude of η (less negative η) with aerosol 361 loading, both yielding a higher COG. We analyzed the bulk properties of the two 362 velocities for the entire simulation time (14 h) and for all clouds in the domain and showed that the relative contribution of the aerosol effect on w and η in determining 363 COG evolution is comparable (60% and 40%, respectively). However, at the 364 365 beginning of the simulation, this ratio was almost 1:1, and the relative contribution of η decreased with time. Such temporal changes in the w vs. η slope indicate changes in 366 367 the thermodynamic properties of the field (Dagan et al., 2016). Increasing 368 thermodynamic instability under polluted conditions results in an increase in mean w, 369 while the decreasing instability under clean condition results in a decrease in rain 370 amount and hence, in η . Both trends act to reduce the slope. We have defined the 371 angle A, which represents the evolution of the thermodynamic conditions with time. A

- 372 can serve as a compact measure of the thermodynamic instability evolution in future 373 observational or numerical studies that quantify *w* and η .
- Using a cloud-tracking algorithm, we identified the growing stage of the clouds and examined the relative contribution of the aerosol effect on COG height by modulating w and η during this stage. We showed that the relative contribution of the aerosol effect on η is larger during the growing stage (for which aerosol loading acts to maintain lower $|\eta|$ for a longer time) compared to the mature and dissipating stages, thereby strengthening the argument that most of the aerosol invigoration effect occurs
- arly in the cloud's evolution (Koren et al., 2015).
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382 *Data availability*. Information about the model and initialization files are available 383 upon request to the contact author.

- 384
- 385 *Competing interests.* The authors declare that they have no conflict of interest.

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509 Figure 1. (a) Mean vertical velocity (w), (b) mean effective terminal velocity (η),

510 (c) mean vertical velocity plus effective terminal velocity, and (d) cloud center of

511 gravity (COG) as a function of time for three different aerosol concentrations.

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Figure 2. Cloud evolution on the phase space span by w vs. V_{COG}. The arrows mark the direction of the trajectories and the thin black line is the 1:1 line. Stars and diamonds denote t = 40 min and 55 min of the simulation, respectively.



519 Figure 3. Temporal and spatial averages of the ambient air vertical velocity (w) 520 vs. effective terminal velocity (η) . Color-coding denotes the different aerosol 521 concentrations. Dots represent averages of the entire simulation data (excluding the first 2 h spin-up time). The x and * markers represent the first third (2 h to 6 522 523 h 40 min) and last third (11 h 20 min to 16 h) of the simulation period, 524 respectively. (a) All clouds in the domain. (b) Only clouds in the growing stage. 525 The black line in (a) is the zero-sum line for which $V_{COG} = 0$ (below the line V_{COG} 526 < 0 and above it $V_{COG} > 0$). The angle A that measures the η vs. w time trend per 527 aerosol level is illustrated in the inset in panel b.

- 529 Table 1. Linear regression slope on the η vs. w phase space for the different
- 530 periods of the simulations for all clouds and growing-stage clouds in the domain.
- 531 \mathbf{R}^2 of the regression lines is presented in parentheses.

	All clouds	Growing clouds
Total simulation period (2–14 h)	0.69 (0.96)	0.79 (0.98)
First period of simulation (2–6:40 h)	0.92 (0.96)	0.99 (0.93)
Last period of simulation (11:20–16 h)	0.47 (0.87)	059 (0.98)

533





Figure 4. (a) The cloud field's mean value of V_{COG} and (b) the angle A between the line that connect the first and last thirds of the simulation period and the xaxis on the η vs. w phase space for all clouds in the domain (Fig. 3a) as a function of aerosol loading. (c) V_{COG} vs. A. Color-coding denotes the different aerosol concentrations.