

1 **Core and margin in warm convective clouds. Part II: aerosol effects**  
2 **on core properties**

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23 **Abstract:**

24 The effects of aerosol on warm convective cloud cores are evaluated using single cloud  
25 and cloud field simulations. Three core definitions are examined: positive vertical  
26 velocity ( $W_{core}$ ), supersaturation ( $RH_{core}$ ), and positive buoyancy ( $B_{core}$ ). As  
27 presented in Part I, the property  $B_{core} \subseteq RH_{core} \subseteq W_{core}$  is seen during growth of  
28 warm convective clouds. We show that this property is kept irrespective of aerosol  
29 concentration. During dissipation core fractions generally decrease with less overlap  
30 between cores. However, for clouds that develop in low aerosol concentrations capable  
31 of producing precipitation,  $B_{core}$  and subsequently  $W_{core}$  volume fractions may  
32 increase during dissipation (i.e. loss of cloud mass). The  $RH_{core}$  volume fraction  
33 decreases during cloud lifetime and shows minor sensitivity to aerosol concentration.

34 It is shown that a  $B_{core}$  forms due to two processes: i) Convective updrafts –  
35 condensation within supersaturated updrafts and release of latent heat, ii) Dissipative  
36 downdrafts – sub-saturated cloudy downdrafts that warm during descent “undershoot”  
37 the level of neutral buoyancy. The former process occurs during cloud growth for all  
38 aerosol concentrations. The latter process only occurs for low aerosol concentrations  
39 during dissipation and precipitation stages where large mean drop sizes permit slow  
40 evaporation rates and sub-saturation during descent.

41 The aerosol effect on the diffusion efficiencies play a crucial role in the development  
42 of the cloud and its partition to core and margin. Using the  $RH_{core}$  definition, it is  
43 shown that the total cloud mass is mostly dictated by core processes, while the total  
44 cloud volume is mostly dictated by margin processes. Increase in aerosol concentration  
45 increases the core (mass and volume) due to enhanced condensation but also decreases  
46 the margin due to evaporation. In clean clouds larger droplets evaporate much slower,  
47 enabling preservation of cloud size and even increase by detrainment and dilution  
48 (volume increase while losing mass). This explains how despite having smaller cores  
49 and less mass, cleaner clouds may live longer and grow to larger sizes.

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## 53 **1. Introduction**

54 Aerosols remain one of the largest sources of uncertainty in climate predictions, mainly  
55 via their effects on clouds (IPCC, 2013). Here we focus on the aerosol effects on warm  
56 clouds. Aerosols act as cloud condensation nuclei (CCN) during heterogeneous  
57 nucleation of cloud droplets (Köhler, 1936; Mason and Chien, 1962). The number, size,  
58 and composition of aerosol distribution yields differences in the initial cloud droplet  
59 size distribution (DSD). Polluted clouds (i.e. more aerosols) have more, but smaller  
60 droplets, and a narrower DSD compared to clean clouds (Andreae et al., 2004; Twomey,  
61 1977). Changes in the initial DSD drive various effects and feedbacks on the cloud's  
62 evolution and key processes, such as: droplet mobility, condensation/evaporation  
63 budgets, collision-coalescence, and entrainment (Jiang et al., 2006; Koren et al., 2015;  
64 Small et al., 2009; Xue and Feingold, 2006).

65 It is well known that an abundance of small droplets in a cloud (a narrow DSD) reduces  
66 the efficiency of the collision-coalescence process (Squires, 1958; Twomey, 1977;  
67 Warner, 1968), prolongs the diffusional growth time (Khain et al., 2005; Wang, 2005),  
68 and delays or even completely suppresses the initiation of precipitation (Albrecht, 1989;  
69 Hudson and Mishra, 2007; Hudson and Yum, 2001; L'Ecuyer et al., 2009). Moreover,  
70 in-cloud condensational growth is more efficient in consuming supersaturation because  
71 of the larger surface area-to-volume ratio of droplets (Dagan et al., 2015a, 2015b;  
72 Mordy, 1959; Pinsky et al., 2013; Reutter et al., 2009; Seiki and Nakajima, 2014). We  
73 note that throughout this work the word *efficient* will be used to describe both the rate  
74 and the total change of mass attributed to a microphysical process. The processes  
75 described above enable the more polluted cloud to condense more water and intensify  
76 its growth via increased release of latent heat (Kogan and Martin, 1994; Koren et al.,  
77 2014; Saleeby et al., 2015; Sheffield et al., 2015). The smaller droplets are also pushed  
78 higher in the atmosphere due to larger droplet mobility (Koren et al., 2014, 2015).

79 However, the increase in aerosol amount yields suppressing effects as well. An opposite  
80 effect should take place in the sub saturated regions of the cloud, where more numerous  
81 and smaller droplets increase the evaporation rate and loss of cloud mass (Grant and  
82 van den Heever, 2015; Saleeby et al., 2015; Storer and van den Heever, 2013).  
83 Henceforth evaporation will be referred to as a process (i.e. change of mass per unit  
84 time) rather than complete evaporation of a water drop. Increased evaporation can

85 promote entrainment mixing which in turn mixes more sub saturated air into the cloud  
86 and further promotes evaporation (Jiang et al., 2006; Small et al., 2009; Xue and  
87 Feingold, 2006), effectively initiating a positive feedback between evaporation and  
88 mixing with the eventual suppression of cloud growth. This effect may also be  
89 accompanied by a suppressing effect of the larger water loading in polluted clouds  
90 which contain more liquid water mass.

91 The competition between those opposing processes that are driven by enhanced aerosol  
92 loading determines the net aerosol effect on cloud properties such as cloud fraction,  
93 lifetime, albedo, mass, size, and precipitation amount. However, the sign and magnitude  
94 of such effects are non-trivial (Jiang and Feingold, 2006). Previous studies report  
95 opposing findings regarding the total aerosol effects on warm clouds (Altaratz et al.,  
96 2014). Some studies suggest cloud invigoration by aerosols (bigger and deeper clouds)  
97 (Dey et al., 2011; Kaufman et al., 2005; Koren et al., 2014; Yuan et al., 2011) while  
98 some suggest cloud suppression or no effect at all (Jiang and Feingold, 2006; Li et al.,  
99 2011; Savane et al., 2015; Xue et al., 2008). Moreover, other work has shown that the  
100 precipitation susceptibility (i.e. quantifies the sensitivity of precipitation to the aerosol  
101 increase) has a non-monotonic behavior that reaches its maximum at intermediate LWP  
102 values (Sorooshian et al., 2009), implying that the resultant aerosol effects are heavily  
103 dependent on cloud type and environmental conditions (Khain et al., 2008).

104 A different approach to aerosol effects suggests that cloud systems can be buffered to  
105 microphysical effects (Stevens and Feingold, 2009). Several studies have shown that  
106 given enough time for the cloud system to reach steady state, cloud macro-physical  
107 parameters (e.g. cloud fraction, rain yield) show similar results for various aerosol  
108 concentrations (Carrió and Cotton, 2014; Glassmeier and Lohmann, 2018; Seifert et al.,  
109 2015). Based on the idea that clouds can be partitioned to aerosol-limited, updraft-  
110 limited, or aerosol and updraft sensitive regimes (Reutter et al., 2009), a unified theory  
111 for the contradicting results regarding aerosol effects was suggested (Dagan et al.,  
112 2015b). Given an aerosol range that covers all three regimes, the competition between  
113 opposite processes leads to an optimum value of aerosol concentration regarding  
114 various cloud properties like total mass, cloud top, or rain (Dagan et al., 2015b). A cloud  
115 that develops under low aerosol concentration is aerosol limited, as it does not have  
116 enough collective droplet surface area to consume the available water vapor. On the  
117 other side of the trend, a cloud that develops in polluted environment (with more

118 aerosols than the optimum) is influenced significantly by enhanced entrainment and  
119 larger water loading, causing suppression of cloud development. The optimal  
120 concentration is a function of the thermodynamic conditions (temperature and humidity  
121 profiles) and cloud size.

122 Environments that support larger clouds development will have larger cloud cores that  
123 are positively affected by aerosol increase and can be regarded as aerosol limited (i.e.  
124 on the ascending branch of the aerosol trend) up to a higher optimal aerosol  
125 concentration. Environmental conditions that support small clouds are more strongly  
126 affected by cloud suppression processes at the cloud margins (due to higher cloud  
127 surface area to volume ratio) and would have a lower optimal aerosol concentration.  
128 This can explain why studies biased to smaller clouds (mostly numerical modeling  
129 studies) report cloud suppression and studies biased to larger clouds (mostly  
130 observational studies) report cloud invigoration. Similar conclusions were reached for  
131 the cloud field scale as well (Dagan et al., 2017).

132 In addition, it was shown that clouds impact differently the environmental  
133 thermodynamics according to the aerosol level in the field (Dagan et al., 2016; Seifert  
134 and Heus, 2013; Seifert et al., 2015). For example changes in aerosol loading impact  
135 the amount of precipitation reaching the surface and subsequently the evaporative  
136 cooling below cloud base and the organization patterns (Seifert and Heus, 2013; Seigel,  
137 2014; Xue et al., 2008). Moreover, an increase in aerosol loading may increase  
138 evaporation rates around the margins and tops of clouds (Seigel, 2014; Stevens, 2007;  
139 Xue and Feingold, 2006), cooling the upper cloudy layer and increasing the convective  
140 instability. Therefore aerosol effects on phase changes and precipitation result in  
141 vertical redistribution of heat and moisture, which may either stabilize or destabilize  
142 the environment in which subsequent clouds grow (Seifert and Heus, 2013).

143 Irrespective of the definition chosen, the cloud's core and margin are dominated by  
144 different processes (Dagan et al., 2015b). These processes often compete with each  
145 other, with the dominant one changing along the cloud's evolution. For example, at the  
146 initial stage of cloud formation, a cloud is more adiabatic and is controlled by the core's  
147 processes (condensation), and when it dissipates the margin processes are more  
148 dominant (entrainment and evaporation). Aerosols affect each of these processes and  
149 thus each stage in the cloud's lifetime. As a continuation to Part I of this work (hereafter

150 PTI), in this part we analyze aerosols effects on the cloud's partition to core and margin  
151 throughout the lifetime of a cloud. We report the consequences these effects have on  
152 evolution of a cloud, in terms of volume, mass, and lifetime. As opposed to other works  
153 that typically focus on a single cloud core definition, here three different definitions are  
154 used (see Sect. 2), with emphasis put on the sensitivity of each core definition to aerosol  
155 concentration. Moreover, the combination of single cloud with large eddy simulations  
156 enables us to gain process level understanding and test the robustness of our findings.

157

## 158 **2. Methods**

159 The analyses performed here are to the most part identical to those described in PTI of  
160 this work. In this section we shall thus only give a brief review of the methods used.  
161 For single cloud simulations we use the Tel-Aviv University axisymmetric cloud model  
162 (TAU-CM (Reisin et al., 1996)), and for cloud field simulations we use the System for  
163 Atmospheric Modeling (SAM) Model (version 6.10.3, for details see webpage:  
164 <http://rossby.msrc.sunysb.edu/~marat/SAM.html>, (Khairoutdinov and Randall, 2003)).

165 Both models utilize explicit bin microphysics schemes (Khain et al., 2004; Tzivion et  
166 al., 1987), solving nucleation, diffusion (i.e. condensation and evaporation), collisional  
167 coalescence, breakup, and sedimentation microphysical processes. The single cloud  
168 model is initialized using a Hawaiian thermodynamic profile, based on the 91285  
169 PHTO Hilo radiosonde at 00Z, 21 Aug, 2007. The cloud field model is setup based on  
170 the BOMEX case study, including an initialization setup (sounding, surface fluxes, and  
171 surface roughness) and large scale forcing setup (Siebesma et al., 2003). More details  
172 on the model setups and definitions can be found in PTI.

173 To study the effects of aerosols on the cloud cores we run each model setup with three  
174 different aerosol concentrations: clean –  $25 \text{ cm}^{-3}$ , intermediate –  $250 \text{ cm}^{-3}$ , and polluted  
175 –  $2000 \text{ cm}^{-3}$ . The model domain is initialized using an oceanic size distribution  
176 (Altartatz et al., 2008; Jaenicke, 1988), maintaining constant mixing ratio with height.  
177 Aerosol budget includes removal by nucleation and regeneration upon evaporation,  
178 while wet scavenging by precipitation removes aerosols from the domain. Thus, the  
179 aerosol concentration may be depleted by 20%–40% (depending on the precipitation  
180 amount) during the simulation. More on the treatment of aerosols in the cloud field

181 model can be found in previous work (Heiblum et al., 2016a). As defined in PTI, all  
182 pixels with at least  $0.01 \text{ g kg}^{-1}$  of liquid water are considered cloudy. Cloud cores are  
183 defined using three definitions: 1)  $\text{RH}_{\text{core}}$ : relative humidity  $> 100\%$ , 2)  $\text{B}_{\text{core}}$ :  
184 buoyancy  $> 0$ , and 3)  $\text{W}_{\text{core}}$ : vertical velocity  $> 0$ . Relative humidity (RH) and vertical  
185 velocity (W) are standard outputs of the model, while the buoyancy (B) is calculated  
186 based on eq. 1 in PTI, where each cloudy pixel is compared with the mean non-cloudy  
187 thermodynamic reference state per height.

188 In order to reduce the problem's dimensionality and distill signals in a cloud field  
189 system governed by high variance, we use the Gravity vs. Mass (CvM) phase space in  
190 combination with an automated 3D cloud tracking algorithm (Heiblum et al., 2016a).  
191 The CvM phase space enables a compact view of all clouds in the simulation, by  
192 projecting only their Center-of-Gravity (COG) height and mass at each output time step.  
193 Using the cloud tracking, it was shown that the lifetime of a cloud can be described by  
194 a trajectory on this phase space. Hence, the different locations in the CvM space are  
195 associated with different stages in a cloud's lifetime (i.e. growing, precipitating, and  
196 dissipating). For an in-depth explanation of the CvM space, the reader is referred to  
197 Sect. 2.4 in PTI (see schematic illustration - Fig. 1, PTI).

198

### 199 **3. Results – Single cloud simulations**

#### 200 **3.1. Sensitivity of different core types to aerosol concentration**

201 Figure 1 presents time series of single cloud core volume fractions and cores'  
202 properties, for three aerosol concentrations (clean, intermediate, and polluted). Also  
203 included are time series of instantaneous rain-rates [ $\text{mm hr}^{-1}$ ] at the domain surface. For  
204 all aerosol concentrations and during most of the clouds' lifetimes, the volume fraction  
205 of  $\text{W}_{\text{core}}$  tends to be the largest and of  $\text{B}_{\text{core}}$  the smallest. Exceptions to this finding are  
206 seen either at the initial time step for the polluted cloud or the later stages of cloud  
207 lifetime for the lower concentration clouds. In addition, we find that  $\text{RH}_{\text{core}} \subseteq \text{W}_{\text{core}}$   
208 for all stages of cloud lifetime while  $\text{B}_{\text{core}} \subseteq \text{W}_{\text{core}}$ ,  $\text{RH}_{\text{core}}$  for all stages of the  
209 polluted cloud but only applies to the growing stages of lower concentration clouds  
210 before precipitation production. Thus, the main finding from PTI (i.e.  $\text{B}_{\text{core}} \subseteq \text{RH}_{\text{core}}$

211  $\subseteq W_{\text{core}}$ ) generally applies to all aerosol concentrations during the pre-precipitation  
212 stages of the clouds' lifetimes.

213 Lower aerosol concentration simulations produce more rain, and at earlier stages of  
214 cloud lifetime due to efficient collision coalescence. The increase in  $B_{\text{core}}$  volume  
215 fraction at later stages of cloud lifetime in those simulations (clean and intermediate)  
216 coincides with initiation of precipitation production, followed by a consequent increase  
217 in  $W_{\text{core}}$  volume fraction as well (more so for the intermediate concentration). This  
218 dissipating  $W_{\text{core}}$  is mostly contained within the  $B_{\text{core}}$ . The possible mechanism  
219 behind the increase in prevalence of buoyant parcels during precipitation is explored in  
220 Sect. 3.2. The lack of  $RH_{\text{core}}$  pixels at these stages indicates that the  $W_{\text{core}}$  is  
221 composed of pixels with small vertical velocities, insufficient for supersaturation  
222 production. The  $RH_{\text{core}}$  is the only one which is not sensitive to rain and monotonically  
223 decreases during all clouds' lifetimes. Another clear aerosol effect seen in Fig. 1 is an  
224 increase in cloud lifetime with decrease in aerosol concentration. This point will be  
225 further explored in Sect. 3.3.

226

### 227 **3.2. Mechanisms governing positive buoyancy**

228 The theoretical arguments in PTI showed that  $B_{\text{core}}$  should be the smallest of the three.  
229 This was shown for both the adiabatic cloud column case and also the non-adiabatic  
230 case where entrainment mixing and consequent evaporation has a strong net negative  
231 effect has on cloud buoyancy. Despite this fact, results show (see Fig. 1, and Fig. 2 in  
232 PTI) that pockets of positive buoyancy may form independent of the other cores during  
233 dissipation and precipitation stages, even though evaporation is to be expected then.  
234 Since positive buoyancy is the result of either higher temperature or vapor content (or  
235 both) than the surrounding environment, we choose to analyze these two terms during  
236 different stages of the single clouds' lifetimes. The liquid water content buoyancy term  
237 (not shown here) is always negative and typically increases (in absolute value) with  
238 increase in vertical velocity or total buoyancy.

239 Figure 2 shows the values of the temperature ( $B_T$ ) and humidity ( $B_{Q_v}$ ) buoyancy terms  
240 in pixel buoyancy vs. pixel vertical velocity phase space. The scatter plots include all  
241 cloudy pixels during all time steps, for the three different aerosol concentration



242 simulations. The distribution of points for the polluted simulation shows a positive  
243 linear dependence of buoyancy on vertical velocity. Negative vertical velocity is  
244 associated with negative buoyancy and positive vertical velocity shows a transition  
245 from negative to positive buoyancy with increase in magnitude. For this case both  $B_T$   
246 and  $B_{Q_V}$  increase with increase in vertical velocity, as is generally expected in  
247 convective clouds. The sign of pixel buoyancy is mostly dependent on  $B_T$  since all  
248 pixels have positive  $B_{Q_V}$  and a negative water loading term. This behavior is also seen  
249 for lower aerosol concentrations, where the sign of buoyancy is to the most part  
250 determined by  $B_T$ . Hereafter, we refer to positive buoyancy (both  $B_T$  and  $B_{Q_V}$ )  
251 production within updrafts as updraft buoyancy.

252 The clean and intermediate simulations show a similar dependence of buoyancy on  
253 vertical velocity; however, it is apparent that these simulations also include an outlier  
254 scatter region of pixels with positive buoyancy and weak negative vertical velocity  
255 which is absent in the polluted simulation (see white arrows, Fig. 2). Consistent with  
256 the rest of the cloudy pixels, these outlier pixels have positive  $B_T$ , but differ in that they  
257 show neutral  $B_{Q_V}$ . It can also be seen that these pixels are only attributed to the stages  
258 after surface precipitation has commenced (indicated by black dots in markers).  
259 Precipitation is indicative of both downdraft motion and abundance of large drop sizes.

260 Although not usually the focus of studies, the existence of positively buoyant  
261 downdrafts in convective clouds has been reported in both observations (Igau et al.,  
262 1999; Wei et al., 1998) and simulations (Xu and Randall, 2001; Zhao and Austin,  
263 2005a, 2005b). A possible explanation for this can be deduced from previous theoretical  
264 studies predicting mixing induced downdrafts in cumulus clouds (Betts and Silva Dias,  
265 1979; Betts, 1982). It was shown that in some cases cloud - environment mixtures are  
266 negatively buoyant (while still containing liquid water) and the consequent downdrafts  
267 can sometimes descend only part way down to the cloud base before reaching neutral  
268 buoyancy. Similar to convective overshooting, parcels with negative vertical  
269 momentum may then “undershoot” the downdraft equilibrium level and turn positively  
270 buoyant while the downdraft weakens. One can therefore expect the magnitude of  
271 positive buoyancy within the downdraft to reach a maximum when the velocity  
272 approaches zero. Hereafter we refer to positive buoyancy production within downdrafts  
273 as downdraft buoyancy.

274 Downdraft buoyancy production occurs frequently in cumulus fields because the  
275 negatively buoyant downdrafts follow a warming lapse rate which is more unstable than  
276 the environmental one, which is typically between the dry adiabat and moist adiabat (as  
277 is the case for the Hawaiian and BOMEX profiles simulated in this work). On one  
278 extreme, a descending parcel is least buoyant (i.e. coolest) when evaporation (after  
279 mixing) keeps it just barely saturated (Paluch and Breed, 1984, also PTI) so that the  
280 lapse rate of descent tends to moist adiabatic and may remain negatively buoyant. On  
281 the other extreme, if little to no evaporation of liquid water occurs, the descent will  
282 follow the dry adiabat and switch to neutral (and then positive) buoyancy rapidly. Thus,  
283 the ability of a negatively buoyant cloudy downdraft to sustain itself depends on  
284 continuous inflow of liquid water (by mixing) and its consequent evaporation (Knupp  
285 and Cotton, 1985).

286 Indeed, the results in Fig. 2 match the hypothesis explained above, where positively  
287 buoyant downdrafts are warmer than the environment, and tend to show larger  
288 buoyancy values for weaker downdrafts velocities (especially for the intermediate  
289 case). Further analysis also shows that the more unsaturated the downdrafts (indicated  
290 also by low  $B_{Qv}$ ), the larger the positive buoyancy. Moreover, the occurrence during  
291 precipitating stages and for lower aerosol concentrations indicates that slow  
292 evaporation due to larger drop sizes is crucial for downdraft buoyancy production,  
293 enabling a near dry adiabatic lapse rate during descent.

294

### 295 **3.3. The dependency of cloud characteristics on core and margin's processes**

296 Here we evaluate how aerosol effects within the core and margin (using the three core  
297 definitions) affect the cloud characteristics, focusing on two main parameters; size (or  
298 volume) and mass. In Fig. 3 we follow the evolution of cloud, core, and margin mass  
299 and volume for different aerosol concentrations, using only the RHcore definition. We  
300 choose the RHcore since it is the most well behaved out the core types, generally  
301 decreasing monotonically (see Fig. 1). A non-monotonic dependency of total cloud  
302 mass on aerosol concentration is seen, showing a maximum for the intermediate  
303 concentration. This type of dependency has been previously reported for warm cumulus  
304 clouds (Dagan et al., 2015b; Savane et al., 2015).

305 One can generally expect an increase in diffusion and decrease in collision-coalescence  
306 processes efficiency with increase in aerosol concentration (Hudson and Yum, 2001;  
307 Jiang et al., 2009; L'Ecuyer et al., 2009; Pinsky et al., 2013), affecting both  
308 condensation and evaporation processes. The intermediate concentration shows the  
309 highest total mass as a result of being an optimal case with higher condensation  
310 efficiency than the clean case and lower evaporation efficiency than the polluted case.  
311 It is convenient to represent the condensation and evaporation efficiencies by the  
312  $RH_{core}$  and  $RH_{margin}$  mass, respectively. The intermediate cloud has almost identical  
313 core mass as does the polluted cloud, but retains higher mass in its margin as well. The  
314 clean cloud shows the lowest core mass but manages to accumulate the largest mass in  
315 its margin that dissipates slowly in sub-saturated conditions. By comparing the total  
316 cloud mass evolution with the core and margin mass evolutions, it becomes clear that  
317 the total mass is primarily dependent on the cloud core. Another way to see this is by  
318 plotting the core mass fraction (Fig. 3 bottom panel), which shows that clouds are core  
319 dominated (core fraction  $> 0.5$ ) with respect to mass for most of their lifetimes, and for  
320 all aerosol concentrations.

321 With respect to cloud total volume, the lower the concentration, the larger the total  
322 cloud volume. We note that the cloud volume here excludes regions of precipitation  
323 below the initial cloud base height. By separating to core and margin regions, one can  
324 see that the total cloud volume is primarily dependent on the volume of the margin,  
325 which increases significantly with decreasing concentration. This is especially true  
326 during the dissipating stages of cloud lifetime, when the cloud is margin dominated.  
327 Although increasing the aerosol concentration does initially yield an increase in core  
328 volume (as was seen for the mass), the extents of the core size are typically smaller than  
329 those of the margin. There are large differences in the relative core volume percent for  
330 the different clouds. The clean (polluted) cloud is margin (core) dominated with respect  
331 to volume for most of its lifetime. Excluding time of formation, the clean cloud shows  
332 the lowest core volume fractions, but manages to maintain its core for the longest time  
333 span.

334 These results with respect to cloud volume can be attributed to the smaller drop sizes  
335 and higher diffusion efficiencies with increase in aerosol concentration. Additionally,  
336 lower collision-coalescence efficiencies also maintain a narrow droplet spectrum of  
337 small droplets in the polluted cloud. During the growing stage a higher aerosol

338 concentration may permit the cloud to condense more water, release more latent heat,  
339 and promote cloud growth. This explains the larger core volume sizes. However, after  
340 the cloud exhausts its convective potential (i.e. the growth of the convective core  
341 terminates and reaches its peak in mass), its main method of expansion is by mixing  
342 with the environment (i.e. detrainment and dilution). We note that precipitation can also  
343 be considered a method of expansion; however our choice to focus on volume above  
344 initial cloud base excludes this effect. Detrainment and mixing with the environment  
345 result in sub-saturation conditions and evaporation of LWC. A clear indication for  
346 dilution is seen in Fig. 3 where between 30 and 35 mins of simulation time both the  
347 clean and polluted clouds lose total mass but only the clean cloud increases in total  
348 volume. The polluted cloud is composed of small drops, evaporates its margin regions  
349 efficiently, and is thus limited in horizontal growth by detrainment. The clean cloud is  
350 composed of larger drops, less efficient in evaporating its margins, and hence can grow  
351 by dilution of its LWC upon a larger volume. This large margin "shields" the core  
352 during dissipation stages and enables it to live for a longer time.

353 The mechanism behind the results in Fig. 3 is demonstrated in Fig. 4, where horizontal  
354 cross-sections of mean (taken in the vertical dimension) cloud RH are shown for  
355 different stages during the clouds' lifetimes. For the polluted cloud, super- or sub-  
356 saturated conditions are rare. The RH throughout the cloud is near 100% (almost always  
357 between 99.8% and 100.2%) except for a few pixels at its far edges which are a bit  
358 below 99%. The polluted cloud resembles what one would expect to see using a moist  
359 adiabatic approximation (i.e. saturation adjustment), where all excess water vapor  
360 above saturation is converted to liquid water, mimicking infinitely efficient  
361 condensation (and evaporation).

362 The clean cloud shows opposite behavior, with extremes of large super-saturation  
363 during cloud growth (initial stages) and large sub-saturation during cloud dissipation  
364 (final stages). The large super-saturation can be explained by slow diffusional growth,  
365 but the large sub-saturation also takes into consideration the larger drop sizes which  
366 take more time to evaporate. This enables the clean cloud to expand to larger horizontal  
367 extents (by dilution and mixing with the environment without fully evaporating) and  
368 live for longer times. The intermediate aerosol concentration shows a midway scenario,  
369 where the super-saturation is consumed more efficiently than the clean case and at the

370 same time much larger values of sub-saturation may exist than those seen for the  
371 polluted case.

372

#### 373 **4. Results – Cloud field simulations**

374 In the following section we expand our analyses of aerosol effects on cloud core and  
375 margin from the single cloud scale to the cloud field scale. A cloud field can be  
376 considered as composed of many individual clouds and thus can serve to test the  
377 robustness of the aerosol effects seen for a single cloud. Moreover, cloud fields include  
378 the added complexity of interactions between clouds and the clouds' effects on their  
379 thermodynamic environment.

##### 380 **4.1. Sensitivity of different core types to aerosol concentration**

381 Here CvM space representations (see Sect. 2) are used to observe the core volume  
382 fractions of all clouds in BOMEX cloud field simulations. The rows in Fig. 5 represent  
383 different aerosol concentrations while the columns represent different core type  
384 definitions. Different aerosol concentrations produce a vastly different scatter of clouds  
385 in the CvM space, as was previously discussed in depth (Heiblum et al., 2016b). The  
386 clean simulation ( $25 \text{ cm}^{-3}$ ) shows two disconnected regions of cloud scatter: one which  
387 is adjacent to the adiabatic approximation and one of mainly small mass and high COG  
388 clouds. The former region includes both clouds during their growth stages (smaller  
389 masses,  $\text{LWP} < 10 \text{ g m}^{-2}$ ) and large precipitating entities (larger masses,  $\text{LWP} > 10 \text{ g}$   
390  $\text{m}^{-2}$ ) which form due to merging processes (Heiblum et al., 2016b). The latter region  
391 (small mass and high COG) includes clouds at their dissipating stage, which form by  
392 shedding mechanism off the large cloud entities. We note also the existence of small  
393 mass elements well below the adiabat, representing precipitation cloud segments which  
394 shed off large precipitating clouds.

395 The polluted simulation ( $2000 \text{ cm}^{-3}$ ) shows a much more homogeneous scatter of  
396 clouds. The lower part of the scatter (closest to the adiabat) represents the cloud  
397 growing branch while the rest of the scatter represents dissipating clouds, either by  
398 gradual process of rising cloud base or by immediate process of shedding off larger  
399 cloud entity (see Fig. 1, PTI). Precipitating cloud segments below the adiabat are absent  
400 from this simulation. The intermediate simulation ( $250 \text{ cm}^{-3}$ ) shows a scatter which

401 generally more resembles the polluted case. However, the existence of relatively  
402 disconnected (from the main cloud scatter) small mass cloud segments below the  
403 adiabat and near the inversion base height resembles the clean simulation as well. It  
404 should be noted that horizontal dashed lines in Fig. 5 represent the inversion base height  
405 after 5 hours of simulation (approximately middle of simulation), where an increase in  
406 the inversion base height is seen with decrease in aerosol concentration. This is due to  
407 increased net warming in the upper cloudy layer (i.e., release of latent heat during  
408 condensation with reduced local evaporation) with increase in precipitation (Dagan et  
409 al., 2016; Heiblum et al., 2016b), which raises the inversion base.

410 The results in Fig. 5 show a consistent behavior of the core volume fractions for all  
411 aerosol concentrations, where the Wcore type shows the largest fractions and the Bcore  
412 type shows the smallest fractions. The Wcore and RHcore generally show a decrease in  
413 core fractions along the growing branch while the Bcore fraction initially increase with  
414 cloud growth and then decrease for the large mass growing clouds. The percentages in  
415 the panel legends (Fig. 5) indicate the fraction of clouds (out of the scatter) which are  
416 core dominated with respect to volume ( $f_{\text{vol}} > 0.5$ ). For all concentrations, less than 7%  
417 of clouds are Bcore dominated while more than 55% are Wcore dominated (with  
418 RHcore percentages somewhere in between). The Bcore typically occupies a small  
419 portion of a typical cloud volume while the Wcore typically occupies most of the cloud.  
420 The mean cloud area (proportional to scatter point size) shows an increase with increase  
421 in mean clouds LWP.

422 These results are consistent with PTI and the single cloud simulations in Sect. 3.1.  
423 Nevertheless, some significant aerosol effects on the partition to core types can be seen.  
424 Focusing on the growing branch first (i.e. clouds located near the adiabat), we note the  
425 following:

- 426 1) For the RHcore type, the core volume fractions of clouds after formation (i.e.  
427 with small mass) increase with decreasing aerosol concentration. This effect  
428 was also seen for the single cloud simulations and can be explained by the  
429 reduced efficiency of super-saturation consumption for fewer aerosols.
- 430 2) The Bcore volume fraction increases at smaller mass values (or earlier in cloud's  
431 lifetime) and to higher values for increasing aerosol concentration. This effect

432 is complimentary to the previous one, since efficient consumption of super-  
433 saturation should result in more latent heat release and positive buoyancy.

434 3) The core volume fractions of the largest mass clouds increase with increasing  
435 aerosol concentration, for all core types.

436 4) The mean area of large mass clouds increases significantly with decrease in  
437 aerosol concentration.

438 We also note a general increase in the fraction of clouds that are Wcore or RHcore  
439 dominated with increase in aerosol concentration. Meaning adding aerosols shifts a  
440 cloud from being mostly margin to being mostly core. The Bcore is an exception since  
441 the clean case shows the highest fraction of Bcore dominated clouds and both the clean  
442 and polluted cases are more Bcore dominated than the intermediate case. This can be  
443 explained by the different mechanisms of buoyancy production (see Sect. 3.2 and 4.2),  
444 where the polluted case is positively influenced by updraft buoyancy production and a  
445 larger core volume fraction while the frequently precipitating clean case is positively  
446 influenced by downdraft buoyancy production. For the dissipating branch clouds, a  
447 highly variable pattern of core volume fractions can be seen, especially for the small  
448 mass clouds. For all aerosol concentrations, these small cloud fragments can be either  
449 core dominated, margin dominated, or equally partitioned. One can assume that these  
450 differences can be related to the different mechanisms by which cloud fragments form,  
451 either by gradual dissipation of a large cloud and by instantaneous shedding of a large  
452 cloud. As for aerosol effects on the dissipating clouds, we see the following:

453 1) Higher RHcore and Wcore volume fractions for gradually dissipating clouds  
454 (by rising cloud base) with increase in aerosol concentration. This is manifested  
455 by a slower transition from red to blue colors in Fig. 5. It can be explained by  
456 the fact that more aerosols increase the convective intensity and extend the core  
457 size, while efficiently losing the margins, yielding a higher core volume fraction  
458 out of the total cloud.

459 2) The likelihood to find dissipating cloud fragments with a Bcore increases with  
460 decrease in aerosol concentration. For the polluted case most of the dissipating  
461 clouds lack a Bcore. This effect was seen in Fig. 1 and explained in Sect. 3.2,  
462 showing that downdrafts promote heating and positive buoyancy in low aerosol

463 concentration cases where evaporation efficiency (and hence cooling) is limited.

464 This effect is checked for the cloud field scale in Sect. 4.2.

465 As opposed to the single cloud simulations (Sect. 3) where cloud lifetime can be easily  
466 defined, in cloud field simulations (especially the cleaner cases) many clouds do not  
467 live as individual clouds from formation to dissipation but rather split and merge with  
468 other clouds continuously (Heiblum et al., 2016b). Thus, in order to evaluate the  
469 lifetime evolution of cores in cloud fields, we focus on the growing branch and use  
470 cloud mass [kg] as a proxy for the cloud lifetime during its initial and mature stages.  
471 We assume that in the vicinity of the growing branch a larger mass corresponds to a  
472 later stage in lifetime.

473 In Fig. 6 the core mass and volume fractions (using the RH definition) of all growing  
474 branch clouds are sorted by mass for the three aerosol concentrations. We note that the  
475 higher cloud masses reached by lower aerosol concentration simulation can be  
476 explained by cloud field organization effects due to precipitation (i.e. increased merging  
477 of clouds) rather than increased cloud condensation (Heiblum et al., 2016b; Seigel,  
478 2014). The clean case starts off with the highest core fractions (both mass and volume)  
479 which decrease steadily with increase in mass (or increase in lifetime). For all  
480 concentrations, most of the cloud mass is concentrated in the core region. The polluted  
481 case shows a slight increase in core mass fractions with increase in mass, while the  
482 other two cases show decreases in core mass fractions.

483 The core volume fractions show lower values than the mass fractions. The clean clouds  
484 are margin dominated for most masses, and the polluted clouds are core dominated for  
485 all masses. The intermediate case is generally confined to values between the other two  
486 cases. Figure 6 can be considered comparable with the lower panels in Fig. 3, but  
487 excluding the dissipating part of those time series. The similar findings in both figures  
488 indicate the robustness of the aerosol effects on core properties in clouds.

489 Following the analyses of Sect. 3.1, we next test how aerosol concentration affects the  
490 subset properties of one core type within another for all clouds in a field (Fig. 7). We  
491 focus only on the typically smaller sized cores ( $B_{\text{core}}$ ,  $RH_{\text{core}}$ ) within larger sized  
492 cores. Out of the three permutations, the  $RH_{\text{core}}$  inside  $W_{\text{core}}$  shows the lowest  
493 sensitivity to aerosol. All three growing branches (for the different aerosol  
494 concentrations) consistently show that the  $RH_{\text{core}}$  is a subset of  $W_{\text{core}}$  (i.e.  $RH_{\text{core}} \subseteq$



495 Wcore) while the dissipation branches show much lower overlap fraction between the  
496 two cores.

497 Generally, for the dissipating clouds, the lower the mass and the higher the COG, the  
498 smaller the overlap. The dissipating branches do include a scatter of small cloud for  
499 which  $RH_{core} \subseteq W_{core}$ , comprised of small cloud segments which shed off the main  
500 core regions of larger clouds. These findings slightly differ from those of the single  
501 cloud simulations that show  $RH_{core} \subseteq W_{core}$  for their entire lifetimes while for cloud  
502 fields this property breaks down during dissipation. This difference highlights the  
503 importance of cloud interactions (i.e. splitting, merging) and cloud field air flow  
504 patterns (i.e. organized advection, updrafts, and downdrafts) in determining the  
505 relationships between core types, enabling supersaturation and downdrafts to coincide  
506 in small dissipating clouds.

507 The other two permutations (i.e. Bcore inside RHcore, Wcore) show significant  
508 changes due to aerosol. For the polluted case,  $B_{core} \subseteq W_{core}$  for nearly all clouds,  
509 including clouds at initial stages of dissipation. Similar results are seen for Bcore inside  
510 RHcore, but with slightly lower pixel fractions. The polluted case thus illustrates the  
511 case of buoyancy production due to convective updraft. For the lower aerosol  
512 concentrations, two main aerosol effects are seen:

513 1) The lower the concentration, the lower the chance that Bcore is a proper subset  
514 of the other cores for large growing branch clouds.

515 2) The lower the concentration, the more prevalent the independent dissipating  
516 branch Bcore that has little to no overlap with the other cores.

517 For the case of Bcore within RHcore, the lower concentrations show an almost binary  
518 scenario where either  $B_{core} \subseteq RH_{core}$  or  $B_{core} \notin RH_{core}$ . These result bear similarity  
519 with the single cloud simulations, where a quick transition (in time) from  $B_{core} \subseteq$   
520  $RH_{core}$  to  $B_{core} \notin RH_{core}$  was seen. These results imply the existence of two different  
521 buoyancy production processes (more in Sect. 4.2), one associated with supersaturation  
522 and the other with sub-saturation. In contrary, pixels fractions of Bcore inside Wcore  
523 span the entire range of values (i.e. partial overlaps between the core types), as seen for  
524 both single clouds and cloud fields during dissipation. This is to be expected due to the  
525 a more direct physical link and feedbacks between the Bcore and Wcore.

526

## 527 **4.2. Analysis of cloud field buoyancy**

528 In Sect. 3.2 it was seen that for single clouds, positive buoyancy results from two main  
529 mechanisms: i) convective updrafts - where updrafts promote supersaturation and latent  
530 heat release, and thus always positive  $B_{Qv}$  and frequently positive  $B_T$ , and ii) dissipative  
531 downdrafts – where sub-saturated cloudy downdrafts promote a positive  $B_T$  and neutral  
532  $B_{Qv}$ . The latter case is dependent on low evaporation efficiency and hence seen mostly  
533 for precipitating stages of low aerosol concentration simulations. In Fig. 8 we perform  
534 a similar test for the cloud field scale. Instead of analyzing pixel by pixel, we check  
535 whether each buoyancy core within a cloud is  $B_T$  or  $B_{Qv}$  dominated. To quantify this  
536 we use a normalized buoyancy dominance parameter  $\frac{\Sigma pixel_{B_T>0} - \Sigma pixel_{B_{Qv}>0}}{\Sigma pixel_{B>0}}$ , where a  
537 core comprised of only  $B_T > 0$  ( $B_{Qv} > 0$ ) pixels yields 1 (-1). Hence, we expect negative  
538 (positive) values to indicate dominance of updraft buoyancy (downdrafts buoyancy).

539 Analysis of the buoyancy components in the CvM space (right column, Fig. 8) shows  
540 that the large majority of clouds are  $B_{Qv}$  dominated. For all concentrations, clouds  
541 initiate with all pixels showing  $B_{Qv} > 0$ . As clouds develop along the growing branch  
542 the Bcore becomes more abundant with  $B_T > 0$  pixels. This is expected with increasing  
543 release of latent heat during cloud growth. During dissipation  $B_{Qv}$  again becomes the  
544 dominant component for the majority of clouds. The polluted simulation shows an  
545 extreme case where all buoyancy cores in the simulation are  $B_{Qv}$  dominated, while for  
546 the lower concentrations a portion of the dissipating and precipitating clouds are  $B_T$   
547 dominated.

548 Thus, we hypothesize that the polluted simulation only permits buoyancy cores of the  
549 updraft type which intersect with the other core types (i.e.  $B_{core} \in RH_{core}, W_{core}$ ),  
550 while the lower concentrations also permit buoyancy cores of the downdraft type which  
551 do not intersect with the other core types (i.e.  $B_{core} \notin RH_{core}, W_{core}$ ). We test this by  
552 observing the relation of cloud maximum absolute vertical velocity (left column, Fig.  
553 8) and mean drop size (middle column, Fig. 8) with the relative dominance of the  
554 buoyancy terms. Absolute vertical velocity is chosen to represent both updrafts and  
555 downdrafts. The data is further separated to independent ( $B_{core} \notin RH_{core}, W_{core}$ ) and  
556 dependent ( $B_{core} \in RH_{core}, W_{core}$ ) buoyancy subsets of the data, by that separating

557 to buoyant cores within updrafts and downdrafts. Clear aerosol effects are seen on cloud  
558 mean drop size and maximal  $|W|$ . As expected, there is a decrease in drop size with  
559 increase in aerosol concentration and increase in maximal velocity. Regarding cloud  
560 field buoyancy, as predicted the independent buoyancy cores are more frequently  $B_T$   
561 dominated than the dependent buoyancy cores.

562 The polluted case is populated with dependent cores (white scatter) and shows a classic  
563 pre-precipitation convective growth scenario, where relative dominance of the  $B_T$  term  
564 increases linearly with increase in cloud mean drop size. A logarithmic dependence of  
565  $B_T$  dominance on maximal  $|W|$  is seen, which saturates at high maximal  $|W|$ . This can  
566 be explained by the fact increased convection mainly increases the abundance of pixels  
567 with  $B_T > 0$ , but without changing the fact that the entire cloud is  $B_{Qv} > 0$ , so that  $B_T$  is  
568 unlikely to become the dominant term. The lower concentrations show a more complex  
569 scenario. These simulations show a superposition of dependent core convective growth  
570 behavior (i.e. the scatter pattern seen for the polluted case) and additional populations  
571 of both dependent (other white scatter points) and independent (black scatter) cores.

572 The independent cores span all the range of possibilities of  $B_T$  and  $B_{Qv}$  relative  
573 dominances. They tend to have larger cloud mean drop sizes, and near zero maximum  
574  $|W|$ , indicating that they only form at late non-convective stages of cloud development.  
575 Furthermore, a trend is seen for the subset of scatter that is  $B_T$  dominated, where a  
576 positive (negative) correlation between mean drop size (maximal  $|W|$ ) and  $B_T$   
577 dominance is seen. This again stresses the importance of drop size on the formation of  
578 positive buoyancy within downdrafts, and highlights the fact that  $B_T$  should be largest  
579 (and most abundant) below the downdraft equilibrium level, when the  $|W|$  approaches  
580 zero. The independent cores that are  $B_T$  dominated thus fulfill the characteristics of  
581 downdraft buoyancy production process, while the independent cores that are  $B_{Qv}$   
582 dominated may originate from larger clouds (shedding mechanism) with high humidity  
583 content, have weak  $|W|$ , and are slow to evaporate.

584 The intermediate simulation shows an additional scatter area of dependent core clouds  
585 with increasing of  $B_T$  relative dominance for lower maximal  $|W|$ , located between the  
586 independent core clouds and the convective growth core clouds. These clouds may  
587 represent a gradual transition from  $B_{Qv}$  dominance to  $B_T$  dominance during dissipation  
588 which is only possible in the intermediate simulation. This scatter area is absent from

589 the clean and polluted simulation. In the former case due to absence of the gradual  
590 dissipation pathway, and in the latter case due to efficient evaporation eliminating  
591 B<sub>core</sub> during dissipation. We note that the intermediate case shows a slightly higher  
592 percentage of clouds that are B<sub>T</sub> dominated (see legends in Fig. 8) than the clean case.  
593 This can be due to stronger convection in this simulation (i.e. increased |W| range),  
594 which favors increased mixing with the dry environment (see Fig. 9) and the formation  
595 of unsaturated strong downdrafts that descend below the level of neutral buoyancy.

596

### 597 **4.3. Aerosol effects on cloud relative humidity**

598 From Fig. 3 it was learned that a large part of the differences in single cloud  
599 characteristics (such as mass, volume, and the partition of these to core and margin  
600 regions) due to aerosols can be attributed to differences in vapor diffusion efficiencies.  
601 In Fig. 9 we check how these aerosol effects are manifested in the cloud field scale  
602 (using the CvM space) by observing the mean relative humidity (RH) in the cloud core  
603 and margin of all clouds, where the core (margin) mean RH can be taken as a proxy for  
604 condensation (evaporation) efficiency. To gain additional intuition regarding the  
605 distribution of RH values within the clouds, vertical cross-sections (parallel to the  
606 prevailing wind direction) of the most massive clouds from each simulation are shown.

607 The vertical cross-sections demonstrate the large differences in the massive clouds for  
608 each of the simulations. In addition to the increase in precipitation production, lower  
609 aerosol concentrations yield much larger horizontal extents of clouds. The clean,  
610 intermediate, and polluted most massive clouds have a maximum radius of  $\sim 3$ ,  $\sim 1$ , and  
611  $\sim 0.5$  km, respectively. It is clear from the cross-section that the clean cloud is actually  
612 composed of two large clouds which merge together. For the clean case, the highest RH  
613 values are reached slightly below the cloud top. The edges of the clouds show sub-  
614 saturation conditions, with the lowest RH values observed below the LCL (precipitation  
615 regions) and at the upper interface of the cloud with the environments.

616 The intermediate case cloud shows lower maximal and minimal RH values and an  
617 increased dominance of the margin region. This cloud penetrates the inversion layer  
618 and entrains dry air into the cloud. In addition, the cloud produces significant  
619 precipitation which initiates downdrafts of dry entrained air through the cloud center.

620 It can be seen that the increased vertical development of the intermediate case cloud in  
621 comparison with the clean case increases the mixing with the environment. Thus, the  
622 dynamic effect of increased mixing and reduction in cloud RH overcomes the  
623 microphysical effect of increased evaporation and increase in cloud RH. The polluted  
624 case cloud on the other hand shows a homogeneous RH pattern, with most of the cloud  
625 showing around 100% RH and only a thin layer at the cloud edges (mainly at the upper  
626 regions) shows lower RH values. The polluted cloud penetrates the inversion layer as  
627 well, but this case lacks precipitation and the microphysical effect of evaporation  
628 overcomes the dynamical effect of mixing.

629 Keeping in mind the insights obtained from comparisons of individual cloud, we move  
630 on to compare the RH characteristics of all clouds within the field. Looking first at core  
631 mean RH, a robust decrease is seen with increase in aerosol concentration. This  
632 decrease is seen for all cloud types and locations within the CvM space. The polluted  
633 case displays the most homogeneous pattern with all clouds showing core mean RH  
634 values around 100%, indicating efficient consumption of the supersaturation. The  
635 intermediate case displays a slightly less homogeneous pattern with values ranging  
636 from 100% to 101%, the higher values occurring along the growing cloud branch,  
637 especially for the largest clouds. The clean case shows the largest variance in core mean  
638 RH, ranging from 100% for some cloud fragments that soon start to dissipate, to 103%  
639 in the cores of the large cloud entities. In addition to the low efficiency in consuming  
640 supersaturation, the high RH values in clean large clouds are due to the "protection" by  
641 large margin regions surrounding the core region.

642 The CvM patterns of mean margin RH show significant differences between the  
643 polluted case and the other two. The mean margin RH values of the polluted case are  
644 only marginally lower than 100%, since sub-saturated conditions within the cloud are  
645 quickly adjusted by efficient evaporation. Only the largest clouds in the polluted case  
646 permit lower mean margin RH values (~ 95%) due to the entrainment of very dry  
647 environmental pixels near the cloud tops (as seen in the vertical cross-section as well).  
648 The intermediate and clean cases show similar patterns. The smaller mass clouds (both  
649 growing and dissipating) show values above 95%, while the larger mass clouds show  
650 values as low as 85%. The larger clouds are most likely to reach low RH areas near the  
651 inversion base and below the LCL (i.e. sub-cloudy layer) and entrain dry air and by that  
652 reduce the cloud margin RH.

653 As seen in the vertical cross-section examples, the largest clouds in the intermediate  
654 case have even lower margin RH values than for the clean case. This can be explained  
655 by the increased development of the large intermediate clouds to heights with lower RH  
656 and by more intense downdrafts for these large clouds. The lowest RH values in the  
657 domain are seen for the precipitating fragments (i.e. located below the adiabat). These  
658 fragments typically contain low concentrations of large drop sizes (precipitation drops)  
659 which are slow to evaporate and capable of surviving in low RH conditions within the  
660 sub-cloudy layer.

661

## 662 **Summary**

663 In this work we explored how the aerosol effects on warm convective clouds are  
664 reflected in their partition to core and margin regions. Following part I of this work  
665 (PTI), we evaluated three types of core definitions: positive buoyancy ( $B_{core}$ ), super-  
666 saturation ( $RH_{core}$ ), and positive vertical velocity ( $W_{core}$ ). Both single cloud and  
667 cloud field models have been used. For all aerosol concentrations (clean, intermediate,  
668 and polluted) it is shown that the self-contained property of different core types (i.e.  
669  $B_{core} \subseteq RH_{core} \subseteq W_{core}$ ) is maintained for clouds during their growing and mature  
670 stages. This is especially robust for the  $RH_{core} \subseteq W_{core}$  subset. The  $W_{core}$  and  
671  $RH_{core}$  volume fractions decrease monotonically during cloud growth, while  $B_{core}$   
672 initially increases and then decreases after convection ceases. During growth, the  
673  $RH_{core}$  ( $B_{core}$ ) volume fractions are largest for clean (polluted) clouds. This is due to  
674 low (high) diffusion efficiencies, respectively, where efficient condensation promotes  
675  $B_{core}$  at the expense of the  $RH_{core}$ .

676 During dissipation stages cores frequently cease to be subsets of one another and may  
677 either increase or decrease in their volume fractions. In cloud fields we also observe  
678 small cloud fragments which shed off larger cloud entities. This shedding increases for  
679 the lower concentration simulation which produce long-lived large cloud entities due  
680 to cloud merging. These fragments show large variance in volume fraction (for all core  
681 types) magnitudes without any consistent behavior. This is due to the fact that they shed  
682 off various locations of the cloud. The polluted, non-precipitating cases, are unique in

683 that can one expect the Bcore to decrease monotonically and remain the smallest and a  
684 proper subset of the other cores.

685 For low aerosol concentrations, a Bcore may form during dissipation and exist  
686 independently of the other core types. These cores are typically located at the periphery  
687 of large clouds, or throughout small precipitation or dissipating cloud fragments. The  
688 increase in Bcore during dissipation typically coincides with large drop sizes and  
689 precipitation production. The fluctuations in Bcore for low concentrations may also  
690 create a subsequent Wcore, but not of sufficient strength to also create a RHcore. Hence,  
691 the RHcore can be considered the most “well-behaved” and indicative of cloud lifetime,  
692 generally monotonically decreasing in volume fraction irrespective of aerosol  
693 concentration.

694 We show that the Bcore in the warm convective cases considered here may form by  
695 two main processes:

696 1. Convective updrafts: adiabatic cooling within updrafts promotes  
697 supersaturation, condensation, and release of latent heat. These cores are  
698 characterized by both positive temperature ( $B_T > 0$ ) and humidity ( $B_{Q_v} > 0$ )  
699 buoyancy terms.

700 2. Dissipative downdrafts: sub-saturated cloudy downdrafts follow a lapse rate  
701 which is unstable relative to the environmental one. These downdrafts  
702 undershoot the equilibrium level and become positively buoyant. These cores  
703 are characterized by positive temperature ( $B_T > 0$ ) but neutral humidity ( $B_{Q_v} \sim$   
704  $0$ ) buoyancy terms.

705 The updraft buoyancy type is seen for all aerosol concentrations, while the dissipation  
706 buoyancy type is only seen for lower aerosol concentrations. The fact that the downdraft  
707 Bcore is absent from polluted clouds highlights the importance of drop size and its  
708 effect on evaporation rate. The high (low) diffusion (collision coalescence) efficiencies  
709 in polluted clouds maintain a small mean drop size and enable rapid evaporation during  
710 entrainment, causing a negative effect on buoyancy. For lower concentrations, clouds  
711 with a downdraft Bcore only exist during late mature, dissipation, and precipitating  
712 stages after drop size has grown considerably. The larger mean drop sizes reduce

713 evaporation rates and the cloudy downdrafts may thus descend nearly dry adiabatically  
714 and become positively buoyant.

715 Focusing on cores using the RH definition, a cloud's mass (volume) is dependent  
716 primarily on the processes in its core (margin). The core increases cloud mass by  
717 condensation while the margin increases the cloud's volume by mixing with the  
718 environment, or dilution. The magnitude of the effects in each region of the cloud is  
719 strongly dependent on the aerosol concentration. Polluted clouds are core dominated  
720 both in terms of mass and volume, since they can hardly maintain their margins. Clean  
721 clouds are also core dominated in terms of mass, but to a lesser degree. Clean clouds  
722 tend to be margin dominated in terms of volume for most their lifetimes. Thus, despite  
723 weaker convection in the clean clouds, their large, slow evaporating margins enable  
724 their cores (and the entire cloud) to exist for longer time spans by applying a large  
725 "protecting shield" around the core.

726 The different diffusion efficiencies are demonstrated by observing the relative humidity  
727 (RH) values in clouds. Cleaner clouds show larger variance in RH values. During their  
728 growing stages large super-saturation in the core and sub-saturation in the margin can  
729 be seen. During their dissipation stages clouds may exist for minutes without any cloud  
730 core, with the entire cloud at sub-saturation. Polluted clouds show the opposite, with  
731 RH values nearing 100% throughout the cloud, at all stages. Hence, above a certain  
732 aerosol concentration, the saturation adjustment approximation (i.e. instant  
733 condensation of all super-saturation) can be considered valid. However, the transition  
734 from clean to polluted is not always linear. For example, for the largest clouds in the  
735 intermediate case have lower margin RH value than both the clean and polluted cases.  
736 This is due to the fact that the intermediate case manages to develop taller (than the  
737 clean case) clouds with stronger updrafts and downdrafts which entrain drier air from  
738 above the inversion layer base, but at the same time is less efficient in evaporating (than  
739 the polluted case) water and adjusting the RH to 100%.

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741

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743



744 **Author Contributions**

745 RH ran the cloud field simulations, conducted the analyses, and wrote the final draft of  
746 paper. LP participated in writing the first draft and performed single cloud simulations  
747 and relevant analyses. OA, GD, and IK participated in paper editing and discussions.

748

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752

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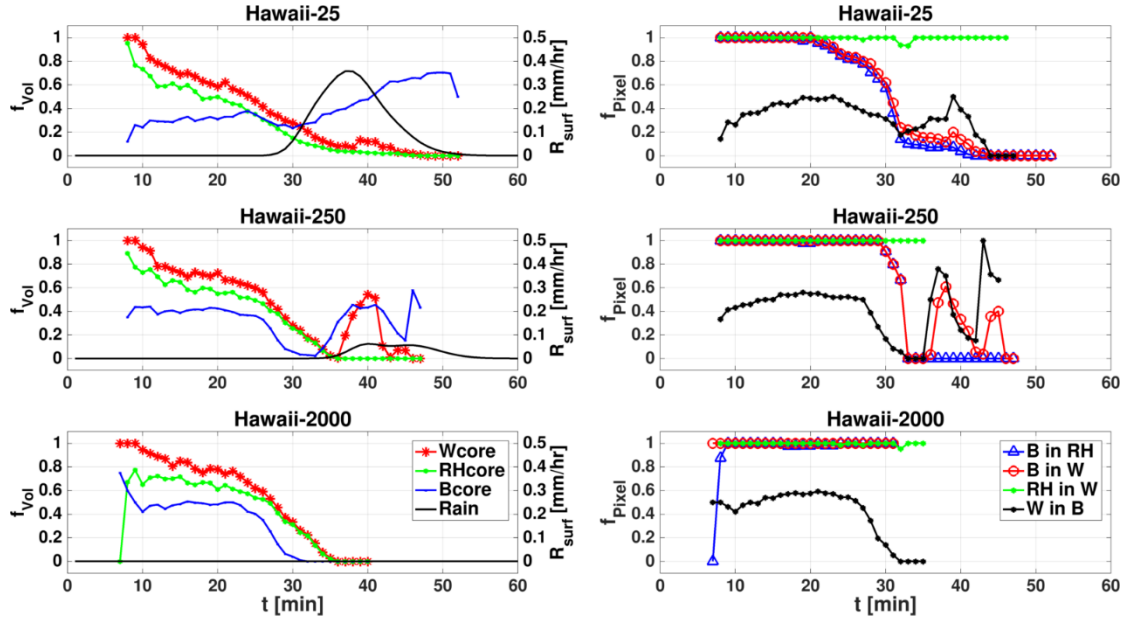
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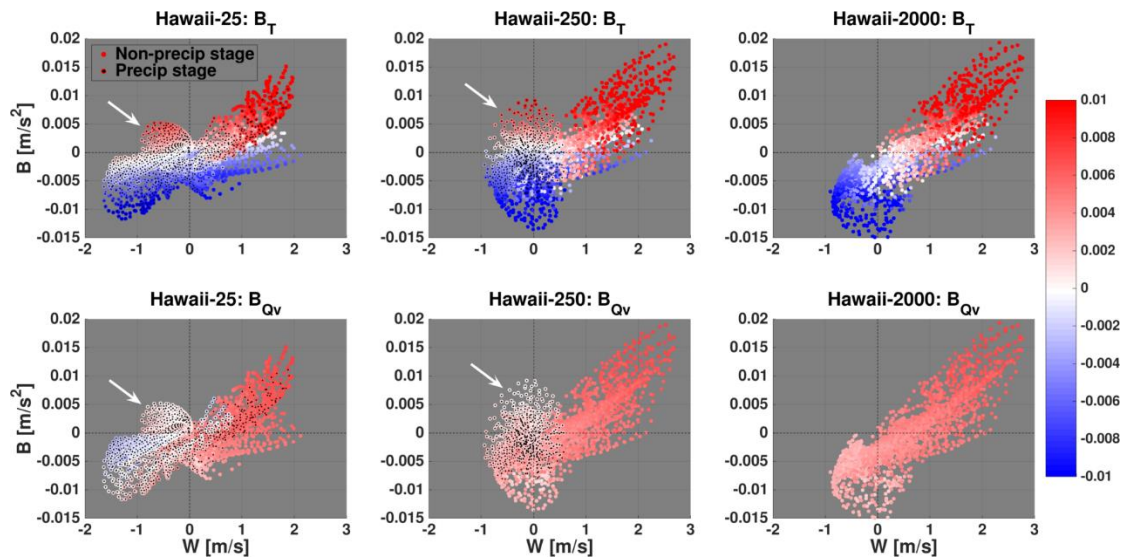
962 **Figures**



963

964 *Figure 1. Left: Time series of core volume fractions ( $f_{vol}$ [%], left axis) and surface rain*  
 965 *rate ( $R_{surf}$ [mm hr<sup>-1</sup>], right axis) for the clean (top panel), intermediate (middle panel),*  
 966 *and polluted (bottom panel) single cloud simulations. Right: Time series of pixel*  
 967 *fractions ( $f_{pixel}$ [%]) of one core type within another, for the respective simulation types.*  
 968 *Core volume and pixel fractions are indicated by different line colors (see legends).*

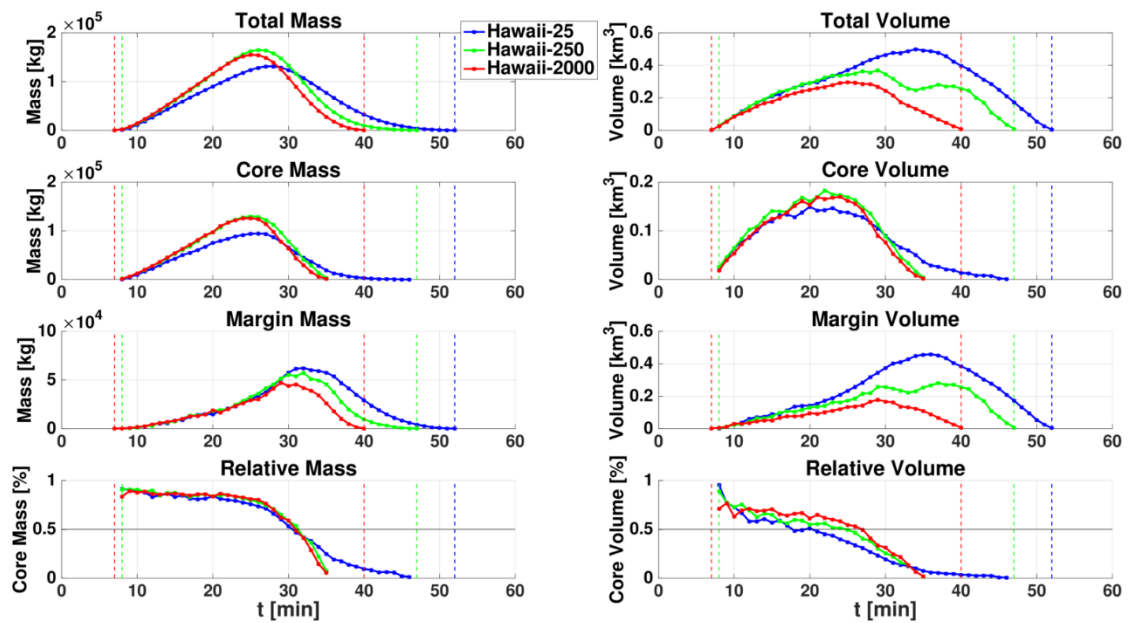
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971 Figure 2. Scatter plots of pixel total buoyancy [ $m s^{-2}$ ] vs. pixel vertical velocity [ $m s^{-1}$ ],  
 972 for the clean (left), intermediate (middle), and polluted (right) simulations. Data  
 973 includes all cloudy pixels during all time steps. Colors represent magnitude of  
 974 buoyancy temperature term ( $B_T$ , upper row) and humidity term ( $B_{Qv}$ , lower row), where  
 975 red (blue) shades indicate positive (negative) values. Markers with black dots  
 976 superimposed represent temporal stages with non-zero surface precipitation. White  
 977 arrows indicate outlier scatter of pixels with positive buoyancy and negative vertical  
 978 velocity.

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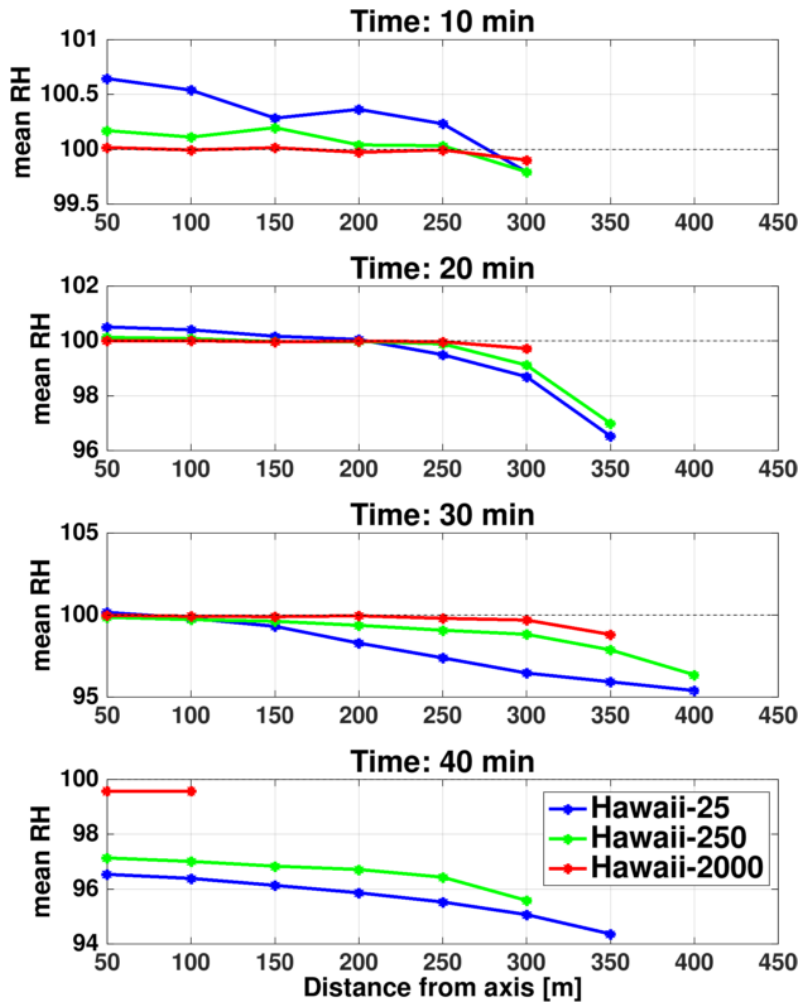


980

981 Figure 3. Time series of cloud mass ([kg], left column) and cloud volume ([ $km^3$ ], right  
 982 column) for the different aerosol concentrations simulations (see legend). The total,  
 983 core, margin, and relative fraction values are shown for each parameter, as indicated  
 984 by panel titles. The core here is defined according to  $RH > 100\%$  definition.

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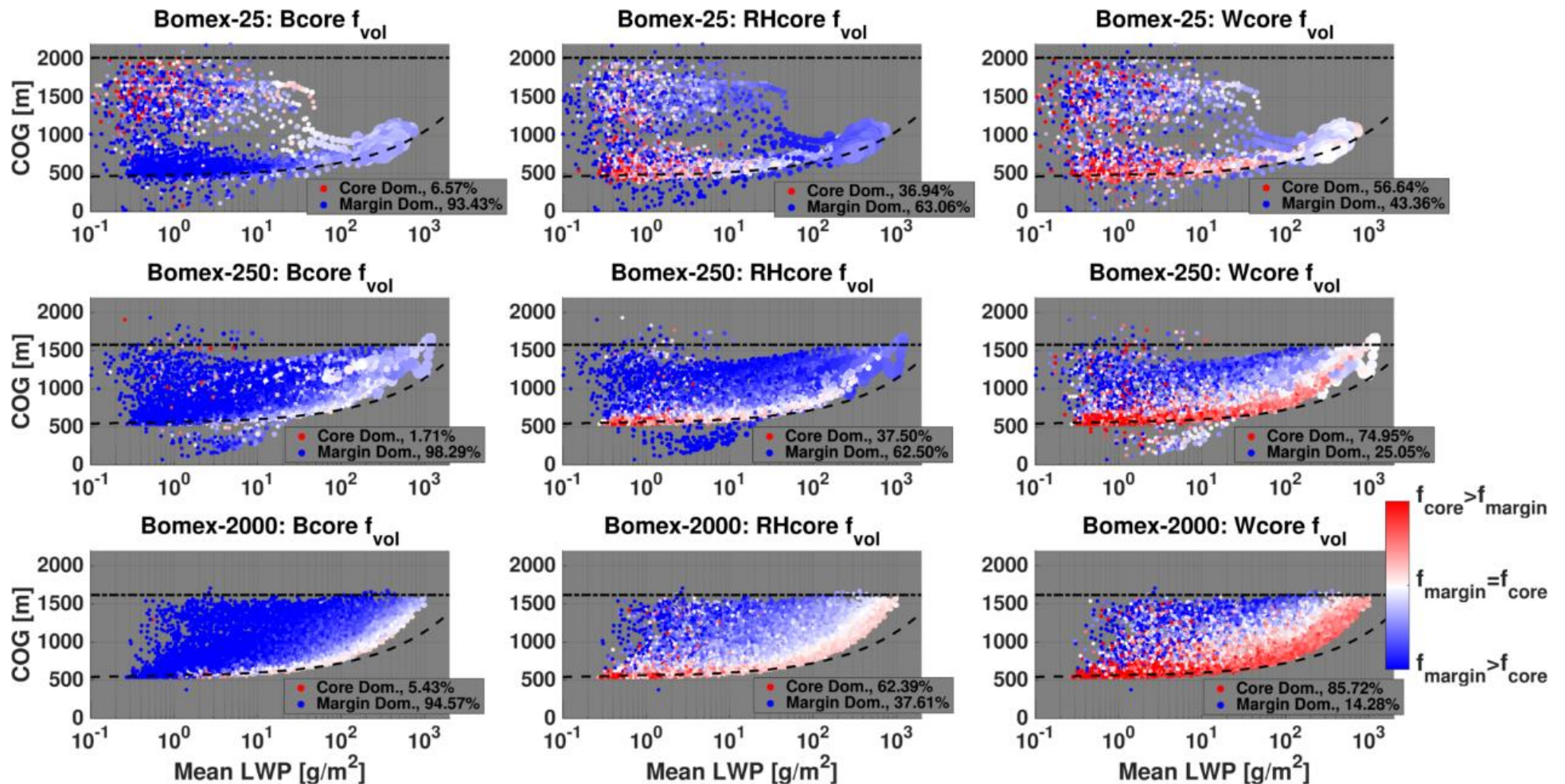
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987 *Figure 4. Four temporal snapshots (see panel titles for times) of RH [%] horizontal*  
 988 *cross-sections. Panels include the results of different aerosol concentrations (see*  
 989 *legend). Cross-sections are obtained by taking the mean RH of all vertical levels for*  
 990 *each horizontal distance from the cloud center axis.*

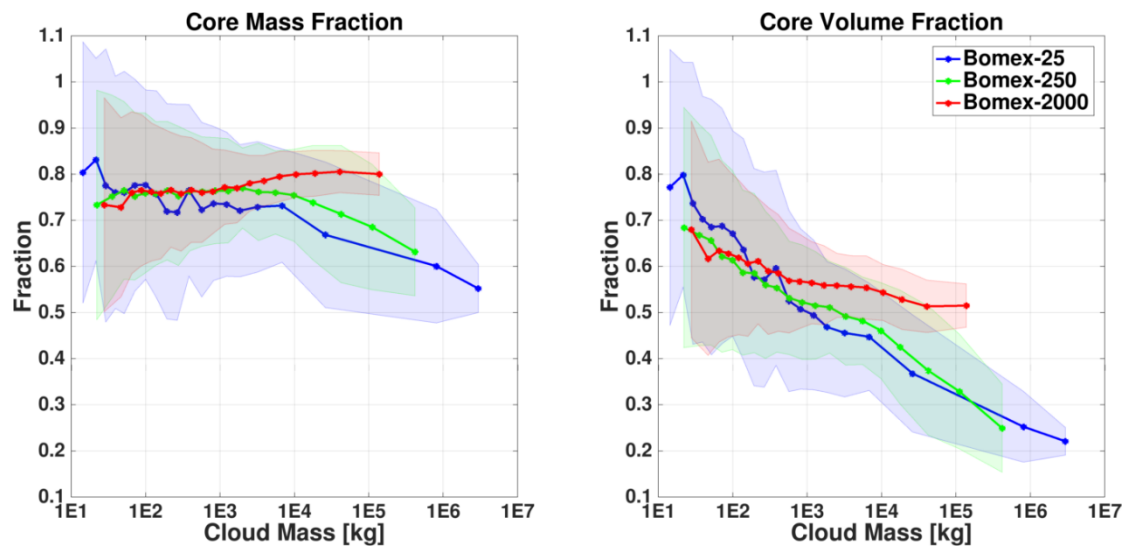
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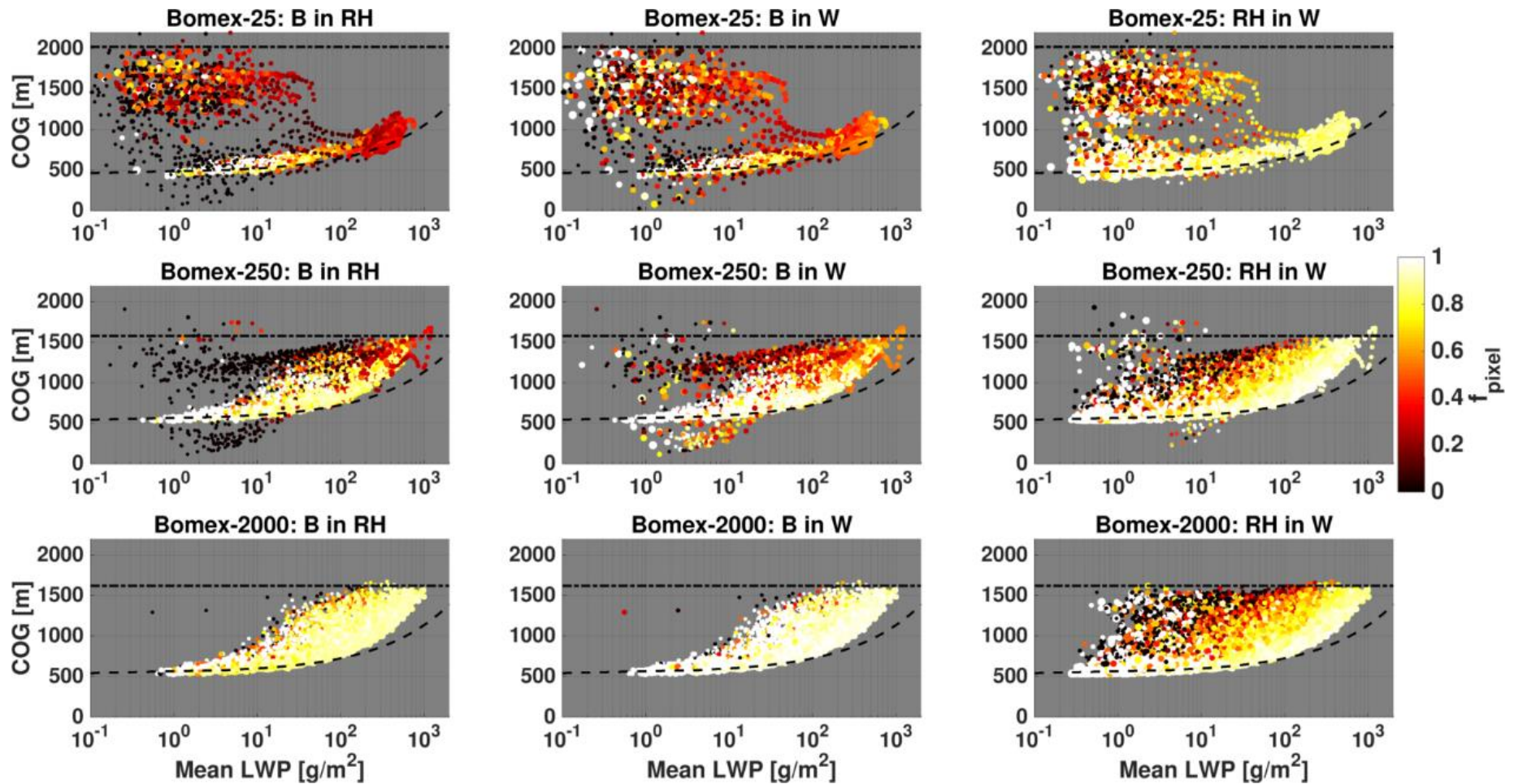


994 Figure 5. CvM phase space diagrams of Bcore (left column), RHcore (middle column), and Wcore (right column) volume fractions ( $f_{vol}$ ) for all  
 995 clouds between 3 h and 8 h in the BOMEX simulations. The upper, middle, and lower rows correspond to the clean, intermediate, and polluted  
 996 aerosol cases. The red (blue) colors indicate a core  $f_{vol}$  above (below) 0.5. The size of each point in the scatter is proportional to the cloud mean  
 997 area, where the smallest (largest) point corresponds to an area of 0.01 (11.4) km<sup>2</sup>. The percentage of clouds that are core dominated ( $f_{vol} > 0.5$ )  
 998 is included in panel legends. For an in-depth description of CvM space characteristics, the reader is referred to Sect. 2.4 in PTI.

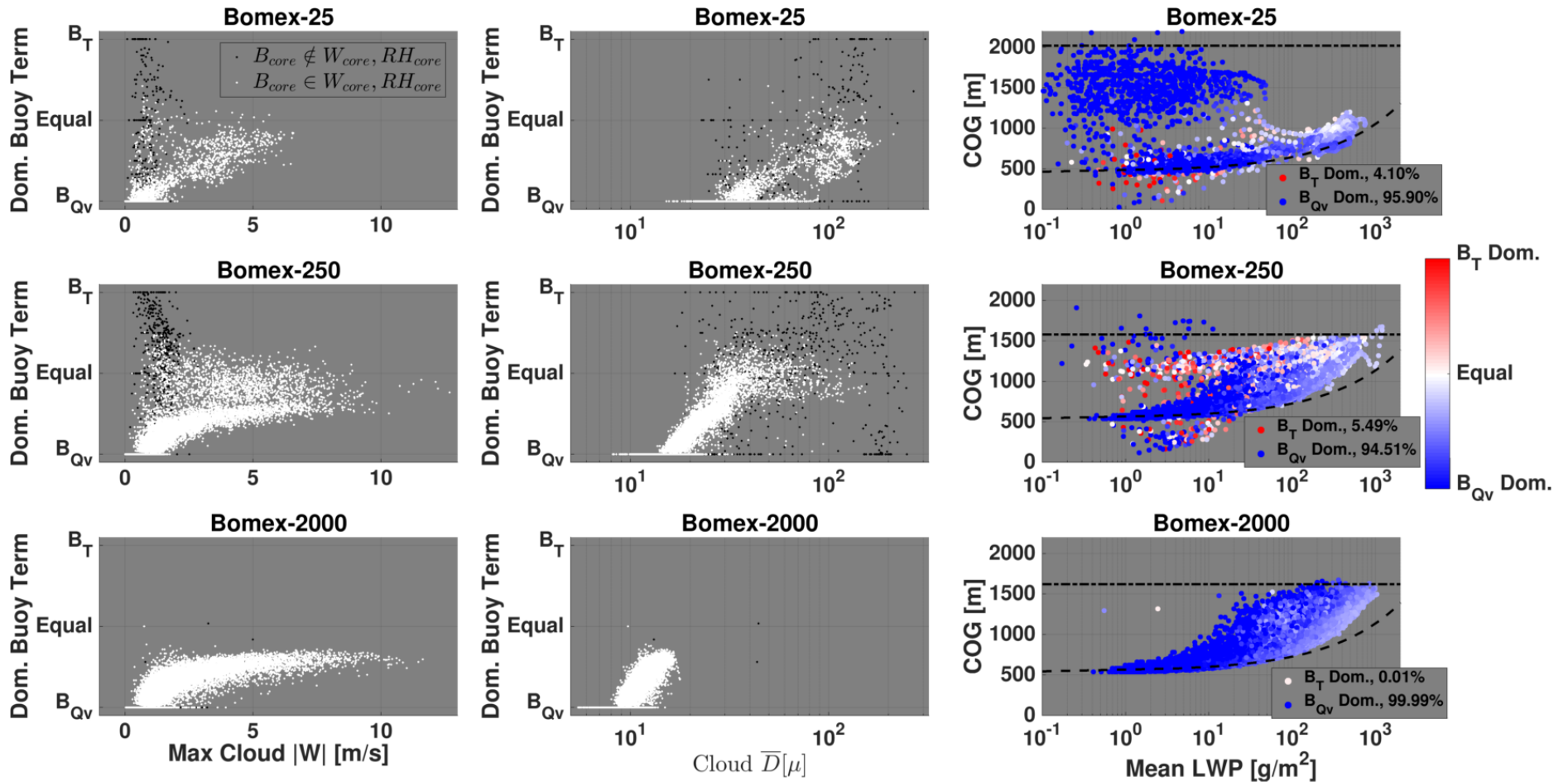


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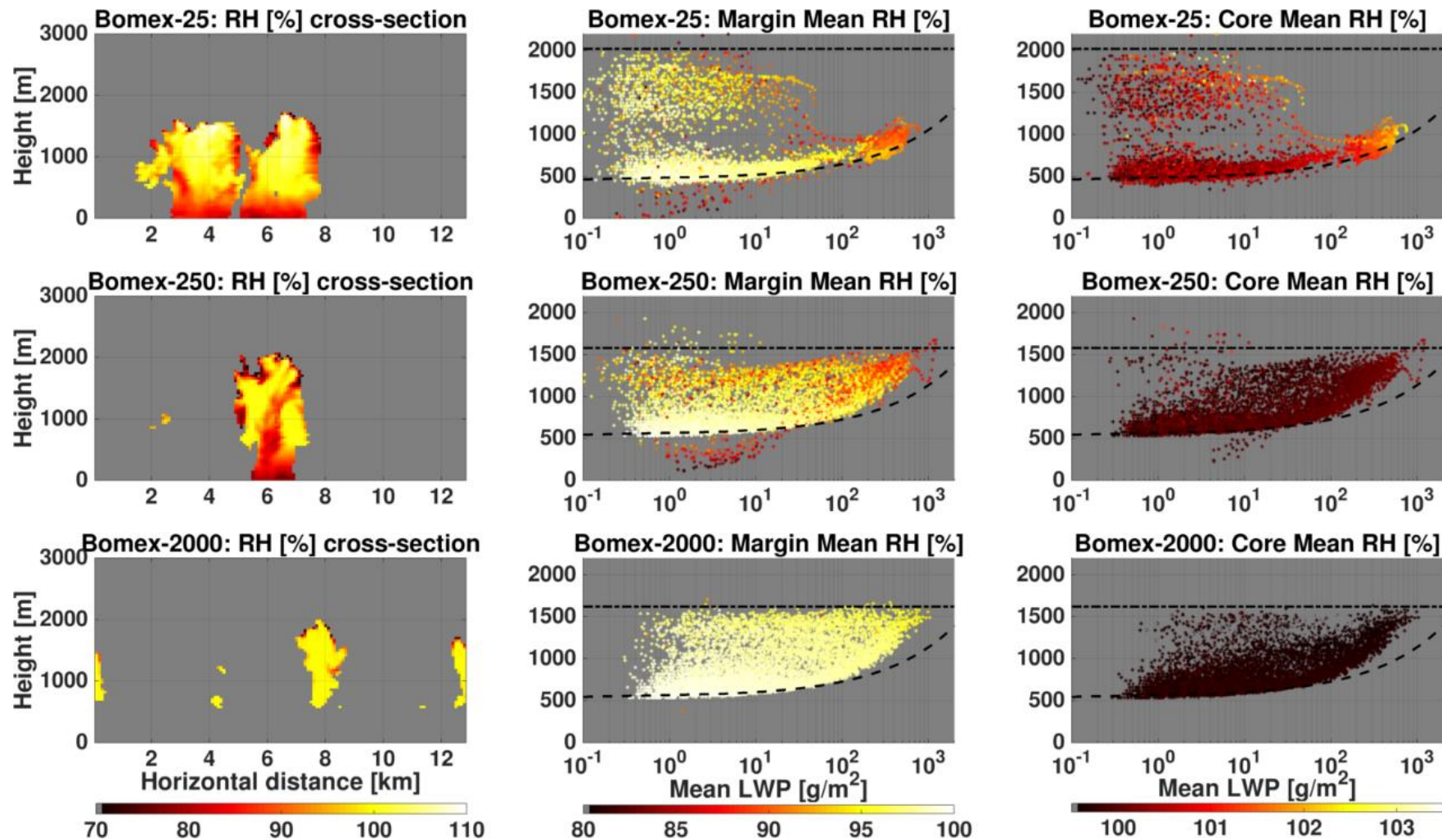
1001 *Figure 6. Average core mass fraction (left) and volume fraction (right) values for*  
 1002 *different aerosol concentrations, as indicated in the legend. The average only includes*  
 1003 *growing branch clouds from within the CvM space (i.e. clouds located in proximity to*  
 1004 *the adiabat). The core here is defined according to  $RH > 100\%$  definition.*



1005 *Figure 7. CvM space diagrams showing the pixel fractions ( $f_{\text{pixel}}$ ) of Bcore within RHcore (left column), Bcore within Wcore (middle column), and*  
 1006 *RHcore within Wcore (right column), for the clean (top row), intermediate (middle row), and polluted (bottom row) simulations. Bright colors*  
 1007 *indicate high pixel fractions (large overlap between two core types) while dark colors indicate low pixel fraction (little overlap between two core*  
 1008 *types). The differences in the scatter density and location for different panels are due to the fact that only clouds which contain a core fraction*  
 1009 *above zero (for the core in question) are considered.*



1010 *Figure 8. Analysis of dominant buoyancy term within  $B_{core}$  of clouds (see text for details). As seen in previous figures, rows represent clean (top),*  
 1011 *intermediate (middle), and polluted (bottom) simulations. Left column: dependence on maximum absolute vertical velocity within cloud. Middle*  
 1012 *column: dependence on partition of total cloud mass to cloud droplets and rain drops. Right column: CvM space diagrams of all clouds with*  
 1013  *$B_{core}$ , where red (blue) shades indicate temperature (humidity) buoyancy terms dominate the cloud. Legends include percentage of clouds that*  
 1014 *are  $B_T$  or  $B_{Qv}$  dominated (see text for explanation).*



1015 *Figure 9. Left column – Relative Humidity (RH [%]) vertical cross-sections slicing through the center of gravity of the most massive cloud in each*  
 1016 *simulation. Middle and right columns display CvM space diagrams of mean cloud margin RH and mean cloud core RH, respectively, using the*  
 1017 *RHcore definition. The upper, middle, and lower panels correspond to the clean, intermediate, and polluted aerosol cases (see panel titles). Notice*  
 1018 *the different color bar ranges for margin and core mean RH panels.*