

1 **Reply to reviewers' comments on "Core and margin in warm convective clouds.**
2 **Part II: aerosol effects on core properties"**

3

4 **Reply to reviewer #1**

5 **General Comment**

6 This paper follows from Part I which sought to examine the various methods of defining
7 the cores and margins of convective clouds by using buoyancy, RH, and vertical
8 velocity to define the core. They showed that these core diagnostics can be subsets of
9 one another, but that this varies in space and time. This follow-on study examines the
10 impacts of varying the aerosol concentration on the core definitions. Given that aerosols
11 can change the cloud DSD, the condensation/evaporation rates can change, and thus the
12 field RH, latent heating, and W. The authors effectively demonstrate the aerosol effects
13 on the evolution of the convective cores and margins.

14

15 **Main Comments:**

16 Two main concerns that can readily be addressed are the need to:

17 MC1) Better state the goals and hypotheses of the study and state what makes this study
18 novel compared to similar ones in the literature.

19 MA1) A paragraph was added to the introduction describing the novelty and goals of
20 the paper: "*As a continuation to Part I of this work (hereafter PTI), in this part we*
21 *analyze aerosols effects on the cloud's partition to core and margin throughout the*
22 *lifetime of a cloud. We report the consequences these effects have on evolution of a*
23 *cloud, in terms of volume, mass, and lifetime. As opposed to other works that typically*
24 *focus on a single cloud core definition, here three different definitions are used (see*
25 *Sect. 2), with emphasis put on the sensitivity of each core definition to aerosol*
26 *concentration. Moreover, the combination of single cloud with large eddy*
27 *simulations enables us to gain process level understanding and test the robustness of*
28 *our findings.*".

29

30 MC2) Better reference past studies in the introduction relative to many of the scientific
31 statements that are made regarding aerosol effects on cloud droplets.

32 MA2) We thank the reviewer for all the suggestions of additional references to this
33 work, which were all implemented into the revised manuscript as described in the
34 specific comments below.

35

36 **Specific Comments:**

37

38 SC1) Line 72-73: The warm phase convective invigoration process has brought about
39 some lively debate in the community in recent years. It does seem however that lately
40 more papers are being published on the matter. I would suggest adding a few additional
41 references that may include the following:

42 Sheffield, A.M., S.M. Saleeby, and S.C. van den Heever, 2015: Aerosol-induced
43 mechanisms for cumulus congestus growth. *J. Geo. Res.*, 120, 8941-8952.

44 Saleeby, S.M., S.R. Herbener, S.C. van den Heever, and T.S. L'Ecuyer, 2015: Impacts
45 of cloud droplet-nucleating aerosols on shallow tropical convection. *J. Atmos. Sci.*, 72,
46 1369-1385.

47 SA1) We thank the reviewer for these additional references. They are now added to the
48 text: "*The processes described above enable the more polluted cloud to condense*
49 *more water and intensify its growth via increased release of latent heat (Kogan and*
50 *Martin, 1994; Koren et al., 2014; Saleeby et al., 2015; Sheffield et al., 2015).*".

51

52 SC2) Lines 80-82: Perhaps add more recent references regarding impacts of aerosol and
53 smaller cloud droplets on condensation and evaporation rates in clouds and along cloud
54 edges.

55 Grant, L.D., and S.C. van den Heever, 2015: Cold pool and precipitation responses to
56 aerosol loading: modulation by dry layers. *J. Atmos. Sci.*, 72, 1398-1408.

57 Storer, R.L., and S.C. van den Heever, 2013: Microphysical processes evident in
58 aerosol forcing of tropical deep convective clouds. *J. Atmos. Sci.*, 70, 430-446.

59 Saleeby, S.M., S.R. Herbener, S.C. van den Heever, and T.S. L'Ecuyer, 2015: Impacts
60 of cloud droplet-nucleating aerosols on shallow tropical convection. *J. Atmos. Sci.*, 72,
61 1369-1385.

62 SA2) Again, we thank the reviewer for bringing these references to our attention. We
63 added them to the revised text: "*An opposite effect should take place in the sub*
64 *saturated regions of the cloud, where more numerous and smaller droplets increase*

65 *the evaporation rate and loss of cloud mass (Grant and van den Heever, 2015;*
66 *Saleeby et al., 2015; Storer and van den Heever, 2013).*”

67

68 SC3) Lines 86-88: This would be a good place to address the concept of “microphysical
69 buffering”. This has become much more prominent of a concept in the past few years.

70 SA3) The concept of microphysical buffering was added to the text, as follows: “A
71 *different approach to aerosol effects suggests that cloud systems are buffered to*
72 *microphysical effects (Stevens and Feingold, 2009). Several studies have shown that*
73 *given enough time for the cloud system to reach steady state, cloud macro-physical*
74 *parameters (e.g. cloud fraction, rain yield) show similar results for various aerosol*
75 *concentrations (Carrió and Cotton, 2014; Glassmeier and Lohmann, 2018; Seifert et*
76 *al., 2015).*”

77

78

79 SC4) Lines 97-99: Figure 17 of Khain et al. (2008, JAS) addresses this aerosol impact
80 on

81 various cloud types and cloud systems. This should be referenced.

82

83 SA4) Thank you for this suggestion, the reference was added to the relevant line.

84

85 SC5) Lines 100-110: This paragraph should cite Reutter et al. (2009) with respect to
86 the “aerosol-limited” vs “updraft-limited” regimes. Looks like you already have this
87 paper in your reference list, but it would be good to add the citation in this paragraph.

88

89 SA5) Agreed, the reference was added to the relevant paragraph as follows: “*Based on*
90 *the idea that clouds can be partitioned to aerosol-limited, updraft-limited, or aerosol*
91 *and updraft sensitive regimes (Reutter et al., 2009), a unified theory for the*
92 *contradicting results regarding aerosol effects was suggested (Dagan et al., 2015)*”.

93

94

95 SC6) Line 140: Here you are transitioning from the paper introduction to the methods
96 section. Your introduction is very thorough, but you haven’t yet stated the goals or
97 science questions of the paper. Please make sure you tell your audience the purpose
98 of the paper and why it is important and novel.

99 SA6) Thank you for this comment, we have added a paragraph explaining the goals of
100 this paper (see MA1 above).

101

102 SC7) Lines 239-244: Things get a bit confusing when you refer to precipitation and
103 evaporation of droplets. It's not clear if you're referring to "cloud droplets" or "rain
104 drops". It would be helpful if cloud hydrometeors are always referred to as "droplets"
105 and rain (precipitation) as "drops". So, are you indicating here that the clean case leads
106 to larger cloud droplets and larger rain drops?

107 SA7) In these sentences we are referring to the entire distribution of drops, which is
108 skewed to larger sizes in clean cases. For clarity, we switched the word droplet to the
109 more general word drop: "*Moreover, the occurrence during precipitating stages and
110 for lower aerosol concentrations indicates that slow evaporation due to larger drop
111 sizes is crucial.*".

112

113

114 SC8) Lines 308-316: This is just a comment, but I appreciate your analysis here and
115 how you allude to polluted clouds essentially mimicking a saturation adjustment with
116 respect to condensation, and how clean clouds allow substantial supersaturation to be
117 carried about. Given that saturation adjustment schemes are still often used in
118 microphysics parameterizations, this re-emphasizes that use of such a scheme can be
119 very inappropriate except under specific circumstances.

120 SA8) Thank you for the comment. We have seen such mimicking effects in previous
121 work as well (Heiblum et al., 2016b).

122

123 SC9) Section 4: Moving into this section reminded me to ask about how your aerosols
124 are treated in the model following initialization. Are the initial aerosol concentrations
125 homogeneous in 3D, do the aerosols advect around the domain, are aerosols removed
126 upon nucleation and regenerated upon droplet evaporation? Are there aerosol sources
127 and sinks? This could certainly be of most importance in a field of clouds.

128 SA9) Thank you for this important comment. To answer your questions, aerosols are
129 initialized homogeneously in 2D (horizontally), maintaining constant mixing ratio with
130 height. They are advected around the domain, and are removed upon nucleation and
131 regenerated upon evaporation. Wet scavenging serves as a sink, while there are no

132 sources. We added some details about how the model treats aerosols and a relevant
133 reference (Heiblum et al., 2016a) for a more complete description: *“To study the effects*
134 *of aerosols on the cloud cores we run each model setup with three different aerosol*
135 *concentrations: clean – 25 cm⁻³, intermediate – 250 cm⁻³, and polluted – 2000 cm⁻³.*
136 *The model domain is initialized using an oceanic size distribution (Altaratz et al.,*
137 *2008; Jaenicke, 1988), maintaining constant mixing ratio with height. Aerosol*
138 *budget includes removal by nucleation and regeneration upon evaporation, while wet*
139 *scavenging by precipitation removes aerosols from the domain. Thus, the aerosol*
140 *concentration may be depleted by 20%–40% (depending on the precipitation amount)*
141 *during the simulation. More on the treatment of aerosols in the cloud field model can*
142 *be found in previous work (Heiblum et al., 2016a).”*.

143

144 SC10) Figure 5: Why is the inversion layer base height higher in the clean case? I don’t
145 recall this being addressed in the paper. Is it initially the same in all cases and then
146 changes over time due to microphysical and dynamical interactions?

147 SA10) As guessed correctly by the reviewer, the inversion base height is initially the
148 same for all cases and evolves differently with time. We now address this point in the
149 paper: *“It should be noted that horizontal dashed lines in Fig. 5 represent the*
150 *inversion base height after 5 hours of simulation (approximately middle of*
151 *simulation), where an increase in the inversion base height is seen with decrease in*
152 *aerosol concentration. This is due to increased net warming in the upper cloudy layer*
153 *(i.e., release of latent heat during condensation with reduced local evaporation) with*
154 *increase in precipitation (Dagan et al., 2016; Heiblum et al., 2016b) , which raises*
155 *the inversion base”*.

156

157 SC11) Line 579: When you refer to evaporation throughout the paper are you referring
158 to partial evaporation as a process or fully evaporated droplets? This could be clarified
159 a bit in the paper. Many past papers including some cited herein often refer to net
160 evaporation of drops/droplets as a distribution without specific concern for full
161 evaporation of droplets.

162 SA11) Thank you for this comment that helped us clarify this point. Throughout the
163 paper we refer to evaporation as a process (i.e. mass evaporated per second [g/s]), and
164 hence many times mention evaporation rates rather than how many droplets were fully
165 evaporated. We added a short sentence to the introduction to clarify this point:

166 ***“Henceforth evaporation will be referred to as a process (i.e. change of mass per unit***
167 ***time) rather than complete evaporation of a water drop.”***

168

169 SC12) Summary section: I find the summary to be a bit over-comprehensive. It’s
170 helpful

171 to the reader to keep this concise and to the point. Keep to the main conclusions and
172 main mechanisms. Details can be seen in the bulk of the paper.

173 SA12) After rereading the summary, we agree with this comment. The summary has
174 been shortened considerably.

175

176 SC13) Figures: My main comment about the figures is that most of the them need to be
177 larger, especially the fonts, so that they are easily readable.

178 SA13) All the figures were redone so that the fonts are larger and readable.

179

180 SC14) Line 302: Should read as “and enables it to live for...”

181 SA14) Thank you, the suggested correction was carried out.

182

183 SC15) Line 426: Should read as “segments which shed off the main...”

184 SA15) Thank you, the suggested correction was carried out.

185

186 SC16) Line 449-451: I find the wording here to make the sentence confusing. Please
187 try clarifying this sentence.

188 SA16) The sentence was rephrased as follows: ***“In contrary, pixels fractions of Bcore***
189 ***inside Wcore span the entire range of values (i.e. partial overlaps between the core***
190 ***types), as seen for both single clouds and cloud fields during dissipation”***

191

192 SC17) Line 521-522: This is a bit of a run-on sentence with a comma separator.

193 SA17) We have changed the sentence as follow: ***“In Fig. 9 we check how these aerosol***
194 ***effects are manifested in the cloud field scale (using the CvM space) by observing the***
195 ***mean relative humidity (RH) in the cloud core and margin of all clouds, where the***
196 ***core (margin) mean RH can be taken as a proxy for condensation (evaporation)***
197 ***efficiency”***.

198

199 SC18) Line 656: This should read as “However, except for the...”

200 SA18) Thank you, we have reformulated the sentence: “*However, excluding the initial*
201 *time of cloud formation where the entire cloud is super-saturated, clean clouds tend*
202 *to be margin dominated in terms of volume for most their lifetimes*”.

203

204

205 **Reply to reviewer #2**

206 **General Comment**

207 In the study effects of aerosols on the structure of small CU are investigated by
208 analyzing

209 the results of axisymmetric cloud model (single cloud simulations), as well as model of
210 cloud ensemble (SAM) (for investigation of general properties of cloud ensembles). To
211 shorten number of parameters, cloud averaged properties are analyzed. Clouds are
212 characterized by core and margin, and effects of aerosols on these regions are
213 investigated. The paper is of interest. However, major revision is necessary.

214

215 **Main Comments:**

216 MC1) Figures 5, 7, 9 indicate that simulation with low aerosol concentration was
217 performed for inversion base at 2000 m, while in other simulations the inversion base
218 was at 1500m. Since the cloud dynamical and microphysical structure as well as cloud
219 size depend on the inversion height, the comparison of the aerosol effects should be
220 performed under similar thermodynamic background conditions.

221 MA1) Thank you for this important comment that helped us clarify our method. All
222 simulations were initialized using the same thermodynamic profile. Figs. 5, 7, 9 show
223 the inversion base height after 5 hours of simulation (and not the initial state), and thus
224 are not equal for different simulations because of microphysical and dynamical
225 interactions between the clouds and their environment that modify the temperature
226 profile of the domain. We choose to display this inversion height (rather than the initial
227 one which is equal for all simulations) since it better reflects the CvM space cloud
228 scatter of the entire simulation. We added to the revised version an explanation
229 clarifying these differences: “*It should be noted that horizontal dashed lines in Fig. 5*
230 *represent the inversion base height after 5 hours of simulation (approximately middle*
231 *of simulation), where an increase in the inversion base height is seen with decrease*
232 *in aerosol concentration. This is due to increased net warming in the upper cloudy*

233 *layer (i.e., release of latent heat during condensation with reduced local evaporation)*
234 *with increase in precipitation (Heiblum et al., 2016b), which raises the inversion*
235 *base”.*

236 MC2) The terminology used in the study is not widely accepted and needs better
237 definition. For instance, it is necessary mathematically define what is “condensation
238 efficiency”, “diffusion efficiency”, etc. (It is possible that such definitions are in Pt 1
239 of the study. Nevertheless, they should be defined in the present study as well). Note
240 that in addition to equation of diffusion growth (“diffusion efficiency”), there is a
241 turbulent diffusion.

242 MA2) We have reviewed the terminology used in the work and defined it when
243 necessary. For the example of diffusion efficiency: “*We note that throughout this work*
244 *the word efficient will be used to describe both the rate and the total change of mass*
245 *attributed to a microphysical process.”. This definition is based on the multiple*
246 *references and descriptions listed in the introduction section.*

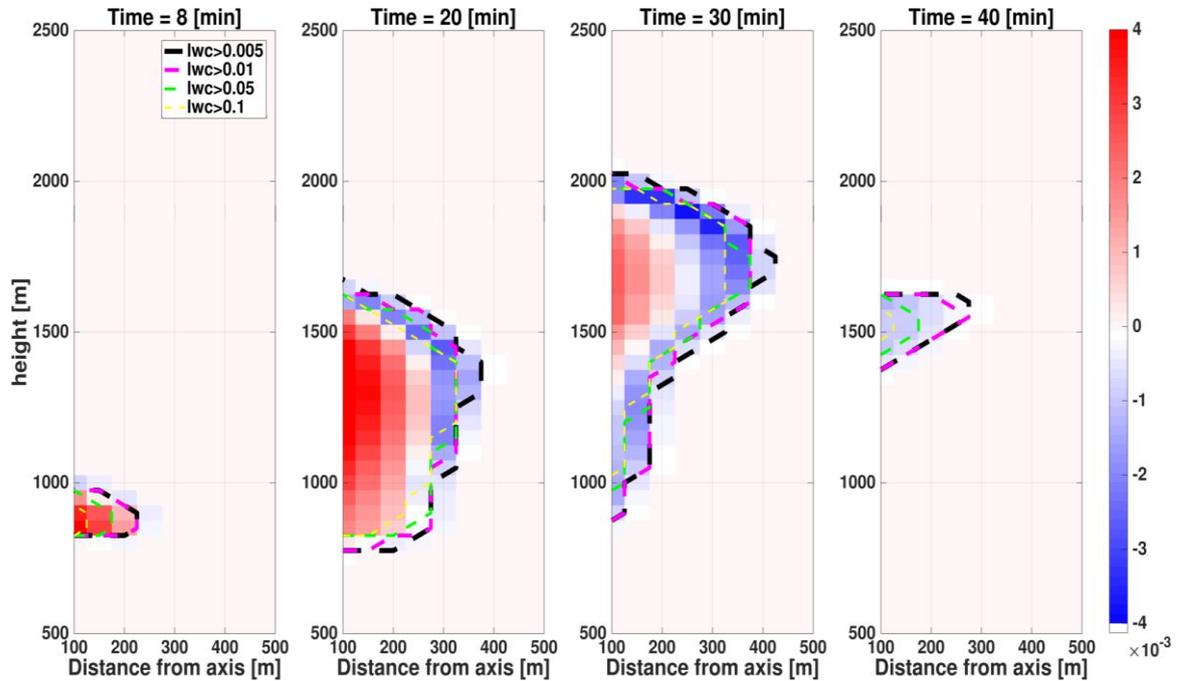
247

248 MC3) Different definitions of cloud cores and cloud margin are interesting. At the same
249 time, these definitions do not agree with the accepted ones. Such definitions lead to a
250 paradox that small dissipating clouds may contain cloud cores. Supposedly, some
251 minimum LWC value should be included into the definition. This will exclude cases,
252 when dissolving cloud with negligible LWC is still considered as combination of cloud
253 core and cloud margin.

254 MA3) As reported in Part I, we do take a minimum threshold of 0.01 g/kg for definition
255 of a cloudy pixel. As a matter of fact, we were questioned about the LWC threshold
256 during the review process of Part I, and showed that this definition best captures the
257 main cloudy processes of condensation and evaporation. We quote our answer here.
258 “*The question of cloud pixel liquid water content (LWC) threshold is something we*
259 *have examined as part of this work. We started by taking an even lower threshold of*
260 *0.005 g/kg (Cohen and Craig, 2006) but eventually raised the threshold to 0.01 g/kg*
261 *based on other works (Jiang et al., 2009; Xue and Feingold, 2006). The impact of*
262 *threshold choice is shown in Fig. RA1 below. The 0.01 and 0.005 g/kg thresholds*
263 *yield similar results with regards to cloud volume, while higher thresholds (0.05 and*
264 *0.1 g/kg) reduce cloud volume significantly. By taking areas of condensation and*
265 *evaporation as indicators of cloudy regions, it can be seen that the higher values*
266 *thresholds “miss” pixels with high evaporation rate (vapor diffusion), in both*

267 *growing and dissipating stages of cloud lifetime. Hence, we find that the 0.01 g/kg*
 268 *threshold best reflects a cloudy volume, without the risk of including insignificant*
 269 *cloud debris as can be seen in some cases for the lower 0.005 g/kg threshold.”.*

270



271

272

273 *Fig. RA1. Four vertical cross-sections (at t=8, 20, 30, 40 minutes) during the single*
 274 *cloud simulation with aerosol concentration of 500 CCN. Y-axis represents height [m]*
 275 *and X-axis represents the distance from the axis [m]. The black, magenta, green and*
 276 *yellow dashed lines represent different LWC thresholds for a cloudy pixel (see legend*
 277 *for values). The background represents the condensation (red) and evaporation rate*
 278 *(blue) [$g\ kg^{-1}\ s^{-1}$].*

279

280

281 **Specific Comments:**

282

283 SC1) line 24 Abstract. The values Bcore, Rcore, Wcore are not defined yet and should
 284 be either defined or excluded from the abstract.

285 SA1) A sentence defining the mentioned values was added to the abstract: “*Three core*
 286 *definitions are examined: positive vertical velocity (Wcore), supersaturation*
 287 *(RHcore), and positive buoyancy (Bcore).”.*

288

289 SC2) Line 45. The text reads: “detrainment while losing mass”. In cloud physics
290 detrainment is “large scale” outflow, typically near cloud top. Interaction of clouds with
291 environment is characterized by entrainment and mixing. The authors supposedly mean
292 that small cloud volumes leaving the parent cloud loss their mass by evaporation.

293 SA2) Based on our understanding detrainment is the same as entrainment, just opposite
294 wind flow (see AMS glossary). A cloud cannot expand horizontally (which is the case
295 here) by entrainment, only by detrainment where the wind vectors are from in the cloud
296 outwards. After detrainment into the non-cloudy environment, mixing occurs and the
297 final result is either a cloudy (with less LWC) or non-cloudy pixel. We try to clarify
298 the sentence as follows: ***“In clean clouds larger droplets evaporate much slower,
299 enabling preservation of cloud size and even increase by detrainment and dilution
300 (volume increase while losing mass)”***.

301

302 SC3) Line 57. what difference of DSD do you mean?

303 SA3) The differences in DSD mentioned in line 57 are explained in the following line
304 in the text. We have reformulated these few sentences in order to avoid confusion (see
305 also SA4 below): ***“Aerosols act as cloud condensation nuclei (CCN) during
306 heterogeneous nucleation of cloud droplets(Köhler, 1936; Mason and Chien,
307 1962).The number, size, and composition of aerosol distribution yields differences in
308 the initial cloud droplet size distribution (DSD). Polluted clouds (i.e. more aerosols)
309 have more, but smaller droplets, and a narrower DSD compared to clean clouds
310 (Andreae et al., 2004; Twomey, 1977).”***.

311

312 SC4) Line 58. The sentence is not clear or not correct. The nucleation itself that takes
313 place at $rN > rN_{crit}$ does not accompanied by decrease of S. S decreases as a result of
314 diffusional growth of nucleated droplets.

315 SA4) Thank you for this comment. We did not intend to say supersaturation is reduced
316 by nucleation, but rather the existence of aerosols enable droplet activation in lower S
317 than in a pristine atmosphere with no aerosols. We have changed this part to be clearer,
318 as seen in SA3 above.

319

320 SC5) Line 77. Strictly speaking, the diffusion growth equation is not symmetric with
321 respect of processes of condensation/evaporation. This asymmetry is considered,

322 sometimes, as a mechanism of DSD broadening (e.g. Korolev, 1995, JAS, 52, 3620-
323 3634).

324 SA5) Thank you for this comment. Analytically speaking, the diffusion equation should
325 be symmetric for condensation and evaporation, but this is only in theory. Thus, we
326 have removed this sentence from the revised text.

327

328 SC6) Line 161. Comment concerning the cloud core definitions. The condition $W > 0$
329 takes place in cloud cores of devolving clouds. Since the time period of cloud
330 developing is relatively short, effects of mixing with surrounding air may not be
331 significant (depending of cloud size and W). At dissolving stage, $W < 0$. So, there is no
332 cloud core in your definition. At the same time LWC in the cloud may has obvious
333 maximum in the cloud center (interior).

334 SA6) We note that our choices of cloud core definitions are based on previous works, as listed
335 in Part I: *“Considering convective clouds, there are several objective measures that*
336 *have been used in previous works for separating a cloud's core from its margins (will*
337 *be referred to as physical cores hereafter). In deep convective cloud simulations the*
338 *core is usually defined by the updrafts' magnitude using a certain threshold, usually*
339 *$W > 1 \text{ m}\cdot\text{s}^{-1}$ (Khairoutdinov et al., 2009; Kumar et al., 2015; Lebo and Seinfeld, 2011;*
340 *Morrison, 2012). Studies on warm cumulus clouds have defined the clouds' core as*
341 *parts with positive buoyancy and positive updrafts (Dawe and Austin, 2012; de Roode*
342 *et al., 2012; Heus and Jonker, 2008; Siebesma and Cuijpers, 1995) or solely regions*
343 *with positively buoyancy (Heus and Seifert, 2013; Seigel, 2014). More recently, cloud*
344 *partition to regions of supersaturation and sub-saturation has been used to define*
345 *the cloud core in single cloud simulations (Dagan et al., 2015).“.*

346 To our knowledge, previous works use a LWC threshold for cloud definition but never
347 for core definition. The case the reviewer describes where LWC has a maximum in the
348 cloud center (or RHcore for example) and there's no Wcore may indeed exist. Due to
349 that, in Part I we define a cloud geometrical core (center of gravity or centroid), and
350 compare its location with the cloud physical core (Wcore, RHcore, Bcore). We quote
351 some of the conclusions here: *“With respect to cloud morphology, the majority of*
352 *clouds are composed from single cores (for all core types), located near the cloud*
353 *centroid/COG, and fit the intuitive core-shell model of decreasing core parameter*
354 *values from cloud center to periphery. This is especially true during cloud growth, as*

355 *during dissipation the cores may decouple from the geometrical core and often*
356 *comprise just a few isolated pixels at the cloud’s edges.* “.

357 Regarding the Wcore definition, we quote from Sect 2.3 in Part I: “*We note that setting*
358 *the core thresholds to positive values (>0) may increase the amount of non-convective*
359 *pixels which are classified as part of a physical core, especially for the Wcore. Indeed,*
360 *taking higher thresholds for the Wcore (e.g. $W > 0.2 \text{ ms}^{-1}$) decreases the Wcore*
361 *extent in the cloud and reduces the variance of Wcore fractions between different*
362 *clouds in a cloud field (as seen in Fig. 4). Nevertheless, any threshold taken is*
363 *subjective in nature, while the positive vertical velocity definition is process based and*
364 *objective.*”.

365 Later in that paper, we show that the Wcore is actually much more “well-behaved” than
366 expected, so that clouds typically have a single Wcore, rather than multiple small
367 Wcores around the cloud. As is written in the text: “*For the Bcore, RHcore, and*
368 *Wcore, 68%, 79%, and 81% of the cloud scatter analyzed (which contain a core) have*
369 *a single core, respectively. Thus, most clouds have a single core. Moreover, it is more*
370 *probable to find multiple buoyancy cores in a cloud than vertical velocity cores. This*
371 *is surprising given our choice of “weak” Wcore thresholds (i.e. positive values) and*
372 *indicates that vertical velocity patterns are relatively well-behaved in cumulus clouds,*
373 *at least for the LES scales chosen here.*”.

374

375 SC7) line 184. How can be explained updrafts at $B < 0$? Gravity waves?

376 SA7) Similar to answer SA6, this issue is also treated in details in Part I. Specifically,
377 updrafts with negative buoyancy are a very common feature in shallow cumulus fields.
378 This issue is discussed in depth in Part I, here we quote some of the relevant text: “*...for*
379 *the adiabatic column case, Bcore is always a proper subset of Wcore (i.e. $B_{\text{core}} \subset$*
380 *Wcore. These effects are commonly seen in warm convective cloud fields where*
381 *permanent vertical layers of negative buoyancy (but with updrafts) within clouds*
382 *typically exist at the bottom and top regions of the cloudy layer (Betts, 1973; de Roode*
383 *and Bretherton, 2003; Garstang and Betts, 1974; Grant and Lock, 2004; Heus et al.,*
384 *2009; Neggers et al., 2007).*“, and also: “*The vertical velocity equation dictates that*
385 *buoyancy is the main production term (de Roode et al., 2012; Romps and Charn,*
386 *2015), and is balanced by perturbation pressure gradients and mixing (on grid and*
387 *sub-grid scales). Thus, all changes of magnitude (and sign) in vertical velocity should*

388 *lag the changes in buoyancy. This is the basis of convective overshooting and*
389 *cumulus formation in the transition layer”.*

390

391 SC8) Line 200. In such case it is difficult to call the condition $W>0$ as definition of
392 cloud core. Small positive W can take place all over cloud just by turbulent fluctuations.
393 Katzwinkel et al. (2014) and Schmeissner et al. 2015 determine cloud interior as
394 $LWC>0.2 \text{ gm}^{-3}$ this condition guaranties that the region chosen is in the cloud interior.
395 SA8) We note that the purpose of these papers (including Part I) is to gain the most
396 general understanding on the partition of cloud to core and margin, using the most
397 general definitions. As mentioned in MA3, applying a $LWC>0.2 \text{ gm}^{-3}$ threshold can
398 exclude much of the cloud, while our goal is to look at the entire cloud. Regarding small
399 W , as explained in SA6 above, adding random thresholds for a core definition is
400 unphysical in our opinion, and can be very sensitive to a specific model or case study.
401 On the other hand, taking positive vs. negative values partitions the cloud based on
402 purely physical considerations, that can also be applied to other works. Nevertheless,
403 as shown in SA6, the occurrence of small positive W pixels is less common than one
404 would think. Thus, for sake of consistency and generality, once a definition is set for
405 cloud core, even a small W_{core} like the one mentioned in line 200 is considered a core.
406 .

407 SC9) line 202. Fig 1. Figure caption. Define LHS axis, RHS axis. What is "other core
408 types"?

409 SA9) Thank you, we have changed LHS and RHS to left axis and right axis. The caption
410 was rephrased as follows: “*Time series of pixel fractions [%] of one core type within*
411 *another, for the respective simulation types*”.

412

413 SC10) lines 236-240 The mechanism proposed requires additional justification. The
414 other option is that in low CCN case drizzle and rain drops rapidly fall down, so LWC
415 is very low in the subsiding of the air. Another possible mechanism is turbulent mixing
416 between warm core and colder margin air. This mixing should lead to an increase in T ,
417 i.e to appearance of positive buoyancy. Can you justify the mechanism that is proposed
418 in the study? What is the cloud stage? Developing or dissolving? Do you see this effect
419 at cloud center or cloud periphery?

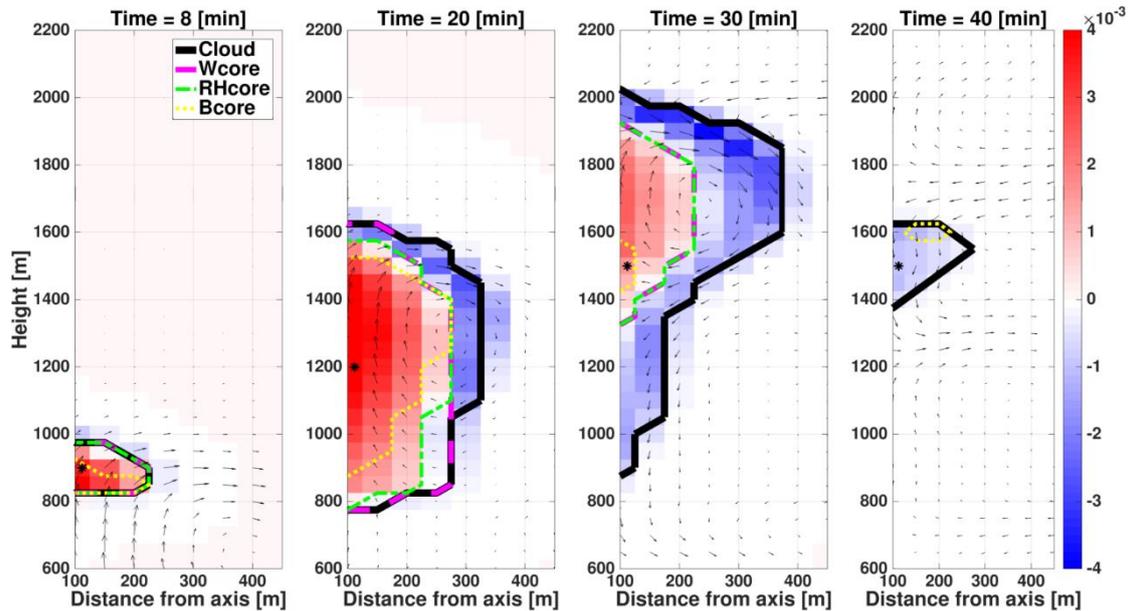
420 SA10) The mechanism we suggest for the pockets of positive buoyancy has been
421 thoroughly checked and is a major part of this paper. We start Sect. 3.2 by noting a

422 main point from Part I, that mixing of cloudy and non-cloudy air or core with margin
423 air almost always (except extreme unlikely cases) reduces buoyancy: “*The theoretical*
424 *arguments in PTI showed that B_{core} should be the smallest of the three. This was*
425 *shown for both the adiabatic cloud column case and also the non-adiabatic case*
426 *where entrainment mixing and consequent evaporation has a strong net negative*
427 *effect has on cloud buoyancy”*. The other option that the reviewer is referring to is the
428 depletion of LWC and hence a smaller water loading term in the buoyancy equation,
429 however, this was checked and the water loading term is as large in the positive
430 buoyancy pockets as in their surroundings. We added this sentence to the text: “*The*
431 *liquid water content buoyancy term (not shown here) is always negative and typically*
432 *increases (in absolute value) with increase in vertical velocity or total buoyancy”*.
433 Throughout the paper (see Figs. 5, 7) and also in Part I (see Fig. RA2 below), we find
434 these pockets of positive buoyancy mostly during late mature and dissipation stages of
435 the cloud, after the initial main convective core has disappeared. This effect is mostly
436 attributed to the cloud periphery but can also be in the cloud center for small dissipating
437 clouds. Both sections 3.2 (for single cloud) and 4.2 (for cloud field) provide proof and
438 attempt to justify the assumption that positive buoyancy is formed due to heating in
439 downdrafts during cloud dissipation.
440 Finally, we have altered the definition of this effect from “adiabatic heating” to “downdraft
441 buoyancy production” and added a more rigorous description of the effect based on previous
442 theoretical and observational works. The description is as follows: “*Although not usually*
443 *the focus of studies, the existence of positively buoyant downdrafts in convective*
444 *clouds has been reported in both observations (Igau et al., 1999; Wei et al., 1998) and*
445 *simulations (Xu and Randall, 2001; Zhao and Austin, 2005a, 2005b). A possible*
446 *explanation for this can be deduced from previous theoretical studies predicting*
447 *mixing induced downdrafts in cumulus clouds (Betts and Silva Dias, 1979; Betts,*
448 *1982). It was shown that in some cases cloud - environment mixtures are negatively*
449 *buoyant (while still containing liquid water) and the consequent downdrafts can*
450 *sometimes descend only part way down to the cloud base before reaching neutral*
451 *buoyancy. Similar to convective overshooting, parcels with negative vertical*
452 *momentum may then “undershoot” the downdraft equilibrium level and turn*
453 *positively buoyant while the downdraft weakens. One can therefore expect the*
454 *magnitude of positive buoyancy within the downdraft to reach a maximum when the*

455 *velocity approached zero. Hereafter we refer to positive buoyancy production within*
 456 *downrafts as downdraft buoyancy.*

457 *Downdraft buoyancy production occurs frequently in cumulus fields because the*
 458 *negatively buoyant downrafts follow a warming lapse rate which is more unstable*
 459 *than the environmental one, which is typically between the dry adiabat and moist*
 460 *adiabat (as is the case for the Hawaiian and BOMEX profiles simulated in this work).*
 461 *On one extreme, a descending parcel is least buoyant (i.e. coolest) when evaporation*
 462 *(after mixing) keeps it just barely saturated (Paluch and Breed, 1984)(also PT1) so*
 463 *that the lapse rate of descent tends to moist adiabatic and may remain negatively*
 464 *buoyant. On the other extreme, if little to no evaporation of liquid water occurs, the*
 465 *descent will follow the dry adiabat and switch to neutral (and then positive) buoyancy*
 466 *rapidly. Thus, the ability of a negatively buoyant cloudy downdraft to sustain itself*
 467 *depends on continuous inflow of liquid water (by mixing) and its consequent*
 468 *evaporation (Knupp and Cotton, 1985). “.*

469



470

471 *Figure RA2. Four vertical cross-sections (at t=8, 20, 30, 40 minutes) during the single*
 472 *cloud simulation. Y-axis represents height [m] and X-axis represents the distance from*
 473 *the axis [m]. The black, magenta, green and yellow lines represent the cloud,*
 474 *W_{core} , RH_{core} and B_{core} , respectively. The black arrows represent the wind, the*
 475 *background represents the condensation (red) and evaporation rate (blue) [$g\ kg^{-1}\ s^{-1}$],*
 476 *and the black asterisks indicate the vertical location of the cloud centroid. Note that in*

477 *some cases the lines indicating core boundaries overlap (mainly seen for RH and W*
478 *cores).*

479

480

481 SC11) Line 248 How is cloud margin region defined and calculated?

482 SA11) Core and margin are defined according to the three definitions used throughout
483 the paper, Wcore, Bcore, and RHcore. We reformulated the sentence to clarify this
484 point: “***Here we evaluate how aerosol effects within the core and margin (using the***
485 ***three core definitions) affect the cloud characteristics*”.**

486

487 SC12) line 254. It is difficult to see the non-monotonic dependence. We see that the
488 maximum cloud mass takes place at high CCN concentration, but lifetime is larger for
489 the low CCN concentration case.

490 SA12) The non-monotonic dependence referred to in that specific sentence is only that
491 of total cloud mass, as quoted from the text: “***A non-monotonic dependency of total***
492 ***cloud mass on aerosol concentration is seen, showing a maximum for the***
493 ***intermediate concentration. This type of dependency has been previously reported for***
494 ***warm cumulus clouds (Dagan et al., 2015; Savane et al., 2015).*”**. The effect isn’t very
495 large, but in Fig. 3 it is clearly seen that the intermediate concentration reaches the
496 maximum total mass. References for this behavior are given.

497

498 SC13) line 285. I do not fully agree with the interpretation. In case of high CCN
499 concentration droplets are small and mixing with surrounding leads to fast complete
500 evaporation of the droplets. Moreover, small droplets fully and easily evaporate also at
501 $W < 0$. At the same time, larger droplets formed at low CCN concentration evaporate
502 only partially. Why is it necessary to focus on the weak effect of the differences in the
503 evaporation rates?

504 SA13) Thank you for this comment. We agree with the reviewer that there are
505 additional parts in the interpretation that should be explained in more details beside the
506 part of differences in diffusion efficiencies (which includes different rates and different
507 droplet surface areas for diffusion process to occur). We now also emphasize the
508 different DSD before evaporation starts which impact the cloud lifetime and cloud
509 volume. Here are a few changes in the text: “***These results with respect to cloud volume***

510 *can be attributed to the smaller drop sizes and higher diffusion efficiencies with*
511 *increase in aerosol concentration.”*, and: *“The polluted cloud is composed of small*
512 *drops, evaporates its margin regions efficiently, and is thus limited in dilution growth.*
513 *The clean cloud is composed of larger drops, less efficient in evaporating its margins,*
514 *and hence can grow by dilution of its LWC upon a larger volume.”*, and: *“The clean*
515 *cloud shows opposite behavior, with extremes of large super-saturation during cloud*
516 *growth (initial stages) and large sub-saturation during cloud dissipation (final*
517 *stages). The large super-saturation can be explained by the low diffusion efficiency,*
518 *but the large sub-saturation also takes into consideration the larger drop sizes which*
519 *take more time to evaporate”*

520

521 SC14) line 293 At the dissolving stage cloud air descends, i.e. $W < 0$ within cloud body
522 (Schmeisner et al. 2015). The subsiding dramatically decreases RH and leads to droplet
523 evaporation. It is natural, that small droplets evaporate first. This decreases the life time
524 of clouds in polluted air. It would be important to separate two effects: turbulent mixing
525 of clouds with surrounding and their evaporation at $W < 0$. Note that small C_u often
526 dissipate and evaporate within the inversion layer, where turbulence (i.e. mixing is
527 weak). In such case, namely subsiding place dominating role in cloud dissolving.

528 SA14) The aforementioned line raises the point that once dissipation commences the
529 only method of cloud volume growth is by dilution via mixing with the environment.
530 If precipitation below the LCL (lifting condensation level) is excluded, this dilution can
531 only be attributed to mixing and not subsidence.

532

533 SC15) line 295. Which effect? How can precipitation be considered as a method of...?
534 please reword the sentence. What kind of expansion can be induced by precipitation?
535 Why the "choice to focus on volume above initial cloud base excludes this effect"? If
536 the precipitation-induced cooling leads to the formation of new clouds, it is impossible
537 to exclude the effect by the choice of the altitude, above which cloud properties are
538 considered.

539 SA15) Continuing the previous answer (SA14), we wanted to differentiate between
540 cloud volume expansions due to dilution versus cloud volume expansion due to
541 precipitation below the cloud base. Since we have significant precipitation in the clean
542 case, cloud mass descends below the initial cloud base (approximately the LCL) and
543 increases the cloud volume significantly. For better comparison with the other more

544 polluted cases, we took only the cloudy pixels above the initial cloud base (which is
545 equal for all simulations) and thus volume changes can be attributed to other effects
546 than precipitation below the cloud base.

547

548 SC16) line 295 Detrainment is the outflow of cloud mass from the cloud. It cannot
549 change the cloud properties. Entrainment of dry environment air into the cloud indeed
550 can lead to subsaturation. Regular (non-turbulent) entrainment takes place near cloud
551 base. At later cloud edges lateral turbulent entrainment and mixing takes place.

552 SA16) Please see SA2 on this issue. We define detrainment as the opposite of
553 entrainment (i.e. air flowing out of cloud). The outflow of air from the cloud can then
554 mix with the surrounding similarly to when air is entrained into a cloud and mixes.

555

556 SC17) line 298. The effect of dilution depends on cloud width. The larger width the
557 lower the effect of lateral mixing is. The increase or decrease of cloud width depends
558 also on LWC.

559 SA17) In this section we are comparing three axi-symmetric single clouds which
560 initially have the exact same width. In line 298 we just want to illustrate that the effect
561 of dilution occurs, meaning increase in cloud volume at the same time there is loss of
562 cloud mass: ***“A clear indication for dilution is seen in Fig. 3 where between 30 and
563 35 mins of simulation time both the clean and polluted clouds lose total mass but only
564 the clean cloud increases in total volume”***.

565

566 SC18) line 300. What is "detrainment growth"?

567 SA18) The sentence was changed slightly to read: ***“...limited in horizontal growth by
568 detrainment”***. As explained above in SA14, SA15, and SA16, growth by detrainment
569 is when clouds may expand in volume after cloudy air is mixed with surrounding
570 environmental air and the droplets do not fully evaporate.

571

572 SC19) line 314. To define “diffusion efficiency”.

573 SA19) See MA2 for the issue to defining diffusion efficiency. Specifically, in the
574 mentioned line we removed this term and replaced with “slow diffusion”.

575

576 SC20) line 327. It is interesting to see the RH (r) profiles in the humid shell around
577 cloud. SA20) Calculating and presenting the RH(r) profiles in the humid shell around

578 all (or a subset of) clouds requires an extensive analysis which is beyond the scope of
579 this work. Moreover, our focus here is on in-cloud processes and cloud properties rather
580 than the effects on the environment adjacent to the cloud. Nevertheless, we note the
581 previous works showing RH(r) have been done, one of them by a member of our
582 research group , showing the distance scale for which RH decreases to the
583 environmental mean.

584

585 SC21) line 375. Do you suppose that dissipating clouds may contain dominating cores?

586 How does it agree with observations?

587 SA21) As can be seen in Fig. 5, many of the dissipating branch clouds (both larger and
588 smaller ones) can be core dominated, mostly for the Wcore definition but also for a
589 small percentage of clouds using the Bcore definition. We define the dissipation branch
590 according to the COG height so that most dissipating clouds have a cloud base above
591 the LCL and may still be mostly with updrafts. As for observations, according to our
592 knowledge, most observations are biased to larger clouds with cloud base near the LCL
593 and not the smaller cloud fragment which occupy the cloudy layer. Nevertheless,
594 although not for small cumulus clouds, studies have shown frequent Bcore in
595 downdrafts (Igau et al., 1999; Wei et al., 1998).

596

597 SC22) Line 375. Figure 5 shows that simulations with low CCN concentration were
598 performed for the case of 2000 m inversion altitude. Two other simulations were
599 performed for 1500 m altitude. The clouds should be quite different geometrically and
600 microphysically in such cases. How can such clouds be compared?

601 SA22) Please see MA1 on this issue. The simulations were initialized with the same
602 profile (and same inversion base height) but evolved differently due to different
603 microphysical effects of the clouds on the thermodynamic conditions. In Fig. 5 we
604 present the thermodynamic conditions after 5 h of simulations because we want them
605 to reflect the actual state during the simulation and not the initial state. These different
606 thermodynamic conditions are among the aerosol effects on clouds that are only seen
607 in cloud field simulations.

608

609 SC23) Line 417. 1) we see again that there the difference in the inversion level in the
610 simulations. Higher clouds can have larger cloud cover and longer life time etc. 2) It is

611 necessary to add to the figure caption the conditions corresponding to the rows and
612 columns or refer notations in fig 5.

613 SA23) For (1), see MA1 and SA22 above explaining the different inversion level
614 heights. For (2), we have added description to the figure caption: “*CvM space diagrams*
615 *showing the pixel fractions of Bcore within RHcore (left column), Bcore within*
616 *Wcore (middle column), and RHcore within Wcore (right column), for the clean (top*
617 *row), intermediate (middle row), and polluted (bottom row) simulations.”.*

618

619 SC24) Line 458. I still wonder, how weak downdraft can lead to the temperature of
620 subsiding air higher than in surrounding. Such subsidence should be actually along the
621 moist adiabat. Why the downdrafts should be weak? It seems that subsidence
622 accompanied by evaporation leads to cold pool that accelerates formation of new
623 clouds. It seems that positive buoyancy in the area of weak downdraft is the results of
624 horizontal mixing between warm zone with $W > 0$ (with high buoyancy) and the cloud
625 periphery.

626 SA24) A rigorous explanation for buoyant downdrafts is now added to revised
627 manuscript (see SA10). Basically, theory shows that cloudy downdrafts follow a lapse
628 rate more unstable than the environment, meaning that a level of neutral buoyancy is
629 reached above the cloud base. Since downdrafts have negative vertical momentum
630 during descent, they will “undershoot” the equilibrium level and become positively
631 buoyant. We show that this effect is highly dependent on aerosol concentration since
632 the evaporation rates (and thus determine the lapse rate of descent. As shown in PT1
633 (and explained in SA10), mixing between positively buoyant and negatively buoyant
634 regions is unlikely to create positively buoyant mixed parcels.

635

636 SC25) line 531. One can suppose that many clouds are isolated even in the clean case.
637 Why do you illustrate the clean case by merging clouds?

638 SA25) Section 4.3 in the paper deals with the different relative humidity seen in the
639 clouds for different aerosol concentrations. As part of this analysis, in Fig. 9 we present
640 cross-sections of the most massive clouds for each simulation. In line 531 we just
641 explain to the reader that the most massive clean cloud is actually composed to two
642 large clouds that merge together and are connected by a few pixels (as can be clearly
643 seen in Fig. 9). We choose to mention this because this is very typical of the clean case
644 and a characteristic worth knowing (in our opinion), where precipitation promotes cold

645 pools and later significant merging. We also mention this in the beginning of Sect. 4.1:
646 *“The clean simulation (25 cm^{-3}) shows two disconnected regions of cloud scatter: one*
647 *which is adjacent to the adiabatic approximation and one of mainly small mass and*
648 *high COG clouds. The former region includes both clouds during their growth stages*
649 *(smaller masses, $LWP < 10\text{ g m}^{-2}$) and large precipitating entities (larger masses,*
650 *$LWP > 10\text{ g m}^{-2}$) which form due to merging processes (Heiblum et al., 2016b).”*, and
651 later in that section: *“We note that the higher cloud masses reached by lower aerosol*
652 *concentration simulation can be explained by cloud field organization effects due to*
653 *precipitation (i.e. increased merging of clouds) rather than increased cloud*
654 *condensation (Heiblum et al., 2016b; Seigel, 2014).”*.

655

656 SC26) Line 572. So, the formation of low RH is the result of averaging over wider
657 layers which contain the inversion layer and layer below LCL. Please confirm.

658 SA26) Exactly, the large clouds’ margin regions may include areas in the inversion
659 layer and layer below the LCL, and thus we may get low mean margin RH.

660

661 SC27) Line 619. Term convection is not suitable. Besides, if you want to compare T
662 with surrounding, moist adiabatic cooling results in heating as compared with the
663 surrounding.

664 SA27) In the revised manuscript we have changed the terms of the two positive
665 buoyancy production processes to updraft buoyancy production and downdraft
666 buoyancy production.

667

668 SC28) Line 621. Adiabatic heating is also not exact term. In the situation considered
669 adiabatic heating is accompanied by turbulent mixing and droplet evaporation. So,
670 many factors determine T in this area, so process is not adiabatic.

671 SA28) Thank you for this comment, it is true that a cloudy downdraft will likely not
672 descend purely adiabatically. As described in the revised manuscript (and SA10), the
673 descent of a cloudy parcel (during entrainment) will be following a lapse rate
674 somewhere between the moist adiabat and dry adiabat, which represent the two extreme
675 cases. We have removed the term “adiabatic heating” from the revised text.

676

677

678

679

References

680 Altaratz, O., Koren, I., Reisin, T., Kostinski, A., Feingold, G., Levin, Z. and Yin, Y.:
681 Aerosols' influence on the interplay between condensation, evaporation and rain in
682 warm cumulus cloud, *Atmos. Chem. Phys.*, 8(1), 15–24, doi:10.5194/acp-8-15-2008,
683 2008.

684 Andreae, M. O., Rosenfeld, D., Artaxo, P., Costa, A. A., Frank, G. P., Longo, K. M.,
685 and Silva-Dias, M. A. F.: Smoking rain clouds over the Amazon., *Science*, 303(5662),
686 1337–1342, doi:10.1126/science.1092779, 2004.

687 Betts, A. K.: Non-precipitating cumulus convection and its parameterization, *Q.J*
688 *Royal Met. Soc.*, 99(419), 178–196, doi:10.1002/qj.49709941915, 1973.

689 Betts, A. K.: Saturation point analysis of moist convective overturning, *J. Atmos. Sci.*,
690 39(7), 1484–1505, doi:10.1175/1520-0469(1982)039<1484:SPAOMC>2.0.CO;2,
691 1982.

692 Betts, A. K. and Silva Dias, M. F.: Unsaturated downdraft thermodynamics in
693 cumulonimbus, *J. Atmos. Sci.*, 36(6), 1061–1071, doi:10.1175/1520-
694 0469(1979)036<1061:UDTIC>2.0.CO;2, 1979.

695 Carrió, G. G. and Cotton, W. R.: On the buffering of CCN impacts on wintertime
696 orographic clouds: An idealized examination, *Atmos. Res.*, 137, 136–144,
697 doi:10.1016/j.atmosres.2013.09.011, 2014.

698 Dagan, G., Koren, I. and Altaratz, O.: Competition between core and periphery-based
699 processes in warm convective clouds – from invigoration to suppression, *Atmos.*
700 *Chem. Phys.*, 15(5), 2749–2760, doi:10.5194/acp-15-2749-2015, 2015.

701 Dagan, G., Koren, I., Altaratz, O. and Heiblum, R. H.: Aerosol effect on the evolution
702 of the thermodynamic properties of warm convective cloud fields., *Sci. Rep.*, 6,
703 38769, doi:10.1038/srep38769, 2016.

704 Dawe, J. T. and Austin, P. H.: Statistical analysis of an LES shallow cumulus cloud
705 ensemble using a ' ' cloud tracking algorithm, *Atmos. Chem. Phys.*, 12(2), 1101–1119,
706 doi:10.5194/acp-12-1101-2012, 2012.

707 de Roode, S. R. and Bretherton, C. S.: Mass-Flux Budgets of Shallow Cumulus
708 Clouds, *J. Atmos. Sci.*, 60(1), 137–151, doi:10.1175/1520-
709 0469(2003)060<0137:MFBOSC>2.0.CO;2, 2003.

710 de Roode, S. R., Siebesma, A. P., Jonker, H. J. J. and de Voogd, Y.: Parameterization
711 of the vertical velocity equation for shallow cumulus clouds, *Mon. Wea. Rev.*, 140(8),
712 2424–2436, doi:10.1175/MWR-D-11-00277.1, 2012.

713 Garstang, M. and Betts, A. K.: A review of the tropical boundary layer and cumulus
714 convection: structure, parameterization, and modeling, *Bull. Amer. Meteor. Soc.*,
715 55(10), 1195–1205, doi:10.1175/1520-0477(1974)055<1195:AROTTB>2.0.CO;2,
716 1974.

717 Glassmeier, F. and Lohmann, U.: Precipitation Susceptibility and Aerosol Buffering
718 of Warm- and Mixed-Phase Orographic Clouds in Idealized Simulations, *J. Atmos.*
719 *Sci.*, 75(4), 1173–1194, doi:10.1175/JAS-D-17-0254.1, 2018.

720 Grant, A. L. M. and Lock, A. P.: The turbulent kinetic energy budget for shallow
721 cumulus convection, *Q.J Royal Met. Soc.*, 130(597), 401–422, doi:10.1256/qj.03.50,
722 2004.

723 Grant, L. D. and van den Heever, S. C.: Cold pool and precipitation responses to
724 aerosol loading: modulation by dry layers, *J. Atmos. Sci.*, 72(4), 1398–1408,
725 doi:10.1175/JAS-D-14-0260.1, 2015.

726 Heiblum, R. H., Altaratz, O., Koren, I., Feingold, G., Kostinski, A. B., Khain, A. P.,
727 Ovchinnikov, M., Fredj, E., Dagan, G., Pinto, L., Yaish, R. and Chen, Q.:
728 Characterization of cumulus cloud fields using trajectories in the center of gravity
729 versus water mass phase space: 1. Cloud tracking and phase space description, *J.*
730 *Geophys. Res. Atmos.*, 121(11), 6336–6355, doi:10.1002/2015JD024186, 2016a.

731 Heiblum, R. H., Altaratz, O., Koren, I., Feingold, G., Kostinski, A. B., Khain, A. P.,
732 Ovchinnikov, M., Fredj, E., Dagan, G., Pinto, L., Yaish, R. and Chen, Q.:
733 Characterization of cumulus cloud fields using trajectories in the center of gravity
734 versus water mass phase space: 2. Aerosol effects on warm convective clouds, *J.*
735 *Geophys. Res. Atmos.*, 121(11), 6356–6373, doi:10.1002/2015JD024193, 2016b.

736 Heus, T. and Jonker, H. J. J.: Subsiding Shells around Shallow Cumulus Clouds, *J.*
737 *Atmos. Sci.*, 65(3), 1003–1018, doi:10.1175/2007JAS2322.1, 2008.

738 Heus, T., Jonker, H. J. J., Van den Akker, H. E. A., Griffith, E. J., Koutek, M. and
739 Post, F. H.: A statistical approach to the life cycle analysis of cumulus clouds selected
740 in a virtual reality environment, *J. Geophys. Res.*, 114(D6),
741 doi:10.1029/2008JD010917, 2009.

742 Heus, T. and Seifert, A.: Automated tracking of shallow cumulus clouds in large
743 domain, long duration large eddy simulations, *Geosci. Model Dev.*, 6(4), 1261–1273,
744 doi:10.5194/gmd-6-1261-2013, 2013.

745 Igau, R. C., LeMone, M. A. and Wei, D.: Updraft and downdraft cores in TOGA
746 COARE: why so many buoyant downdraft cores?, *J. Atmos. Sci.*, 56(13), 2232–2245,
747 doi:10.1175/1520-0469(1999)056<2232:UADCIT>2.0.CO;2, 1999.

748 Jaenicke, R.: 9.3.1 Physical properties, in *Physical and chemical properties of the air*,
749 edited by G. Fischer, pp. 405–420, Springer-Verlag, Berlin/Heidelberg., 1988.

750 Khairoutdinov, M. F., Krueger, S. K., Moeng, C.-H., Bogenschutz, P. A. and Randall,
751 D. A.: Large-eddy simulation of maritime deep tropical convection, *J. Adv. Model.*
752 *Earth Syst.*, 2, 15, doi:10.3894/JAMES.2009.1.15, 2009.

753 Knupp, K. R. and Cotton, W. R.: Convective cloud downdraft structure: An
754 interpretive survey, *Rev. Geophys.*, 23(2), 183, doi:10.1029/RG023i002p00183, 1985.

755 Kogan, Y. L. and Martin, W. J.: Parameterization of bulk condensation in numerical
756 cloud models, *J. Atmos. Sci.*, 51(12), 1728–1739, doi:10.1175/1520-
757 0469(1994)051<1728:POBCIN>2.0.CO;2, 1994.

758 Köhler, H.: The nucleus in and the growth of hygroscopic droplets, *Transactions of*
759 *the Faraday Society*, 32, 1152–1161, 1936.

760 Koren, I., Dagan, G. and Altaratz, O.: From aerosol-limited to invigoration of warm
761 convective clouds., *Science*, 344(6188), 1143–1146, doi:10.1126/science.1252595,
762 2014.

763 Kumar, V. V., Jakob, C., Protat, A., Williams, C. R. and May, P. T.: Mass-Flux
764 Characteristics of Tropical Cumulus Clouds from Wind Profiler Observations at
765 Darwin, Australia, *J. Atmos. Sci.*, 72(5), 1837–1855, doi:10.1175/JAS-D-14-0259.1,
766 2015.

767 Lebo, Z. J. and Seinfeld, J. H.: Theoretical basis for convective invigoration due to
768 increased aerosol concentration, *Atmos. Chem. Phys.*, 11(11), 5407–5429,
769 doi:10.5194/acp-11-5407-2011, 2011.

770 Mason, B. J. and Chien, C. W.: Cloud-droplet growth by condensation in cumulus,
771 *Quarterly Journal of the Royal Meteorological Society*, 88(376), 136–142, 1962.

772 Morrison, H.: On the robustness of aerosol effects on an idealized supercell storm
773 simulated with a cloud system-resolving model, *Atmos. Chem. Phys.*, 12(16), 7689–
774 7705, doi:10.5194/acp-12-7689-2012, 2012.

775 Neggers, R. A. J., Stevens, B. and Neelin, J. D.: Variance scaling in shallow-cumulus-
776 topped mixed layers, *Q.J Royal Met. Soc.*, 133(628), 1629–1641, doi:10.1002/qj.105,
777 2007.

778 Paluch, I. R. and Breed, D. W.: A Continental Storm with a Steady, Adiabatic Updraft
779 and High Concentrations of Small Ice Particles: 6 July 1976 Case Study, *J. Atmos.*
780 *Sci.*, 41(6), 1008–1024, doi:10.1175/1520-
781 0469(1984)041<1008:ACSWAS>2.0.CO;2, 1984.

782 Reutter, P., Su, H., Trentmann, J., Simmel, M., Rose, D., Gunthe, S. S., Wernli, H.,
783 Andreae, M. O. and Pöschl, U.: Aerosol- and updraft-limited regimes of cloud droplet
784 formation: influence of particle number, size and hygroscopicity on the activation of
785 cloud condensation nuclei (CCN), *Atmos. Chem. Phys.*, 9(18), 7067–7080,
786 doi:10.5194/acp-9-7067-2009, 2009.

787 Romps, D. M. and Charn, A. B.: Sticky Thermals: Evidence for a Dominant Balance
788 between Buoyancy and Drag in Cloud Updrafts, *J. Atmos. Sci.*, 72(8), 2890–2901,
789 doi:10.1175/JAS-D-15-0042.1, 2015.

790 Saleeby, S. M., Herbener, S. R., van den Heever, S. C. and L’Ecuyer, T.: Impacts of
791 cloud droplet–nucleating aerosols on shallow tropical convection, *J. Atmos. Sci.*,
792 72(4), 1369–1385, doi:10.1175/JAS-D-14-0153.1, 2015.

793 Savane, O., Vant-Hull, B., Mahani, S. and Khanbilvardi, R.: Effects of aerosol on
794 cloud liquid water path: statistical method a potential source for divergence in past
795 observation based correlative studies, *Atmosphere*, 6(3), 273–298,
796 doi:10.3390/atmos6030273, 2015.

797 Seifert, A., Heus, T., Pincus, R. and Stevens, B.: Large-eddy simulation of the
798 transient and near-equilibrium behavior of precipitating shallow convection, *J. Adv.*
799 *Model. Earth Syst.*, 7(4), 1918–1937, doi:10.1002/2015MS000489, 2015.

800 Seigel, R. B.: Shallow Cumulus Mixing and Subcloud-Layer Responses to Variations
801 in Aerosol Loading, *J. Atmos. Sci.*, 71(7), 2581–2603, doi:10.1175/JAS-D-13-0352.1,
802 2014.

803 Sheffield, A. M., Saleeby, S. M. and van den Heever, S. C.: Aerosol-induced
804 mechanisms for cumulus congestus growth, *J. Geophys. Res. Atmos.*, 120(17), 8941–
805 8952, doi:10.1002/2015JD023743, 2015.

806 Siebesma, A. P. and Cuijpers, J. W. M.: Evaluation of parametric assumptions for
807 shallow cumulus convection, *J. Atmos. Sci.*, 52(6), 650–666, doi:10.1175/1520-
808 0469(1995)052<0650:EOPAFS>2.0.CO;2, 1995.

809 Stevens, B. and Feingold, G.: Untangling aerosol effects on clouds and precipitation
810 in a buffered system., *Nature*, 461(7264), 607–613, doi:10.1038/nature08281, 2009.

811 Storer, R. L. and van den Heever, S. C.: Microphysical processes evident in aerosol
812 forcing of tropical deep convective clouds, *J. Atmos. Sci.*, 70(2), 430–446,
813 doi:10.1175/JAS-D-12-076.1, 2013.

814 Twomey, S.: The influence of pollution on the shortwave albedo of clouds, *J. Atmos.*
815 *Sci.*, 34(7), 1149–1152, doi:10.1175/1520-0469(1977)034<1149:TIOPOT>2.0.CO;2,
816 1977.

817 Wei, D., Blyth, A. M. and Raymond, D. J.: Buoyancy of convective clouds in TOGA
818 COARE, *J. Atmos. Sci.*, 55(22), 3381–3391, doi:10.1175/1520-
819 0469(1998)055<3381:BOCCIT>2.0.CO;2, 1998.

820 Xu, K.-M. and Randall, D. A.: Updraft and downdraft statistics of simulated tropical
821 and midlatitude cumulus convection, *J. Atmos. Sci.*, 58(13), 1630–1649,
822 doi:10.1175/1520-0469(2001)058<1630:UADSOS>2.0.CO;2, 2001.

823 Zhao, M. and Austin, P. H.: Life cycle of numerically simulated shallow cumulus
824 clouds. part I: transport, *J. Atmos. Sci.*, 62(5), 1269–1290, doi:10.1175/JAS3414.1,
825 2005a.

826 Zhao, M. and Austin, P. H.: Life cycle of numerically simulated shallow cumulus
827 clouds. part II: mixing dynamics, *J. Atmos. Sci.*, 62(5), 1291–1310,
828 doi:10.1175/JAS3415.1, 2005b.

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

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868 **Abstract:**

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

879 It is shown that a B_{core} forms due to two processes: i) ~~Convection~~Convective updrafts
880 – condensation within supersaturated updrafts and release of latent heat, ii) ~~Adiabatic~~
881 heating due to weakDissipative downdrafts – sub-saturated cloudy downdrafts that
882 warm during descent “undershoot” the level of neutral buoyancy. The former process
883 occurs during cloud growth for all aerosol concentrations. The latter process only
884 occurs for low aerosol concentrations during dissipation and precipitation stages where
885 large mean drop sizes permit slow evaporation rates: and sub-saturation during descent.

886 The aerosol effect on the diffusion efficiencies play a crucial role in the development
887 of the cloud and its partition to core and margin. Using the RH_{core} definition, it is
888 shown that the total cloud mass is mostly dictated by core processes, while the total
889 cloud volume is mostly dictated by margin processes. Increase in aerosol concentration
890 increases the core (mass and volume) due to enhanced condensation but also decreases
891 the margin due to evaporation. In clean clouds larger droplets evaporate much slower,
892 enabling preservation of cloud ~~volume~~size and even increase by ~~dilution~~
893 and dilution (volume increase while losing mass). This explains how despite having
894 smaller cores and less mass, cleaner clouds may live longer and grow to larger sizes.

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900 1. Introduction

901 Aerosols remain one of the largest sources of uncertainty in climate predictions, mainly
902 via their effects on clouds (IPCC, 2013). Here we focus on the aerosol effects on warm
903 clouds. Aerosols act as cloud condensation nuclei (CCN) during heterogeneous
904 nucleation ~~by reducing the supersaturation required for droplet activation of cloud~~
905 ~~droplets~~ (Köhler, 1936; Mason and Chien, 1962), ~~yielding. The number, size, and~~
906 ~~composition of aerosol distribution yields~~ differences in the initial cloud droplet size
907 distribution (DSD). Polluted clouds ~~(i.e. more aerosols)~~ have more, but smaller
908 droplets, and a narrower DSD compared to clean clouds (Andreae et al., 2004; Twomey,
909 1977). Changes in the initial DSD drive various effects and feedbacks on the cloud's
910 evolution and key processes, such as: droplet mobility, condensation/evaporation
911 budgets, collision-coalescence, and entrainment (Jiang et al., 2006; Koren et al., 2015;
912 Small et al., 2009; Xue and Feingold, 2006).

913 It is well known that an abundance of small droplets in a cloud (a narrow DSD) reduces
914 the efficiency of the collision-coalescence process (Squires, 1958; Twomey, 1977;
915 Warner, 1968), prolongs the diffusional growth time (Khain et al., 2005; Wang, 2005),
916 and delays or even completely suppresses the initiation of precipitation (Albrecht, 1989;
917 Hudson and Mishra, 2007; Hudson and Yum, 2001; L'Ecuyer et al., 2009). Moreover,
918 in-cloud condensational growth is more efficient in consuming supersaturation because
919 of the larger surface area-to-volume ratio of droplets (Dagan et al., 2015a, 2015b;
920 Mordy, 1959; Pinsky et al., 2013; Reutter et al., 2009; Seiki and Nakajima, 2014). ~~These~~
921 ~~processes~~ We note that throughout this work the word *efficient* will be used to describe
922 both the rate and the total change of mass attributed to a microphysical process. The
923 processes described above enable the more polluted cloud to condense more water and
924 intensify its growth via increased release of latent heat (Kogan and Martin, 1994; Koren
925 et al., 2014; Saleeby et al., 2015; Sheffield et al., 2015). The smaller droplets are also

926 pushed higher in the atmosphere due to larger droplet mobility (Koren et al., 2014,
927 2015).

928 However, the increase in aerosol amount yields suppressing effects as well. ~~The~~
929 ~~symmetry of the diffusion equation dictates that an~~An opposite effect should take place
930 in the sub saturated regions of the cloud, where more numerous and smaller droplets
931 increase the evaporation rate and loss of cloud mass (Grant and van den Heever, 2015;
932 Saleeby et al., 2015; Storer and van den Heever, 2013). Henceforth evaporation will be
933 referred to as a process (i.e. change of mass per unit time) rather than complete
934 evaporation of a water drop. Increased evaporation can promote entrainment mixing
935 which in turn mixes more sub saturated air into the cloud and further promotes
936 evaporation (Jiang et al., 2006; Small et al., 2009; Xue and Feingold, 2006), effectively
937 initiating a positive feedback between evaporation and mixing with the eventual
938 suppression of cloud growth. This effect may also be accompanied by a suppressing
939 effect of the larger water loading in polluted clouds which contain more liquid water
940 mass.

941 The competition between those opposing processes that are driven by enhanced aerosol
942 loading determines the net aerosol effect on cloud properties such as cloud fraction,
943 lifetime, albedo, mass, size, and precipitation amount. However, the sign and magnitude
944 of such effects are non-trivial (Jiang and Feingold, 2006). Previous studies report
945 opposing findings regarding the total aerosol effects on warm clouds (Altaratz et al.,
946 2014). Some studies suggest cloud invigoration by aerosols (bigger and deeper clouds)
947 (Dey et al., 2011; Kaufman et al., 2005; Koren et al., 2014; Yuan et al., 2011) while
948 some suggest cloud suppression or no effect at all (Jiang and Feingold, 2006; Li et al.,
949 2011; Savane et al., 2015; Xue et al., 2008). Moreover, other work has shown that the
950 precipitation susceptibility (i.e. quantifies the sensitivity of precipitation to the aerosol
951 increase) has a non-monotonic behavior that reaches its maximum at intermediate LWP
952 values (Sorooshian et al., 2009), implying that the resultant aerosol effects are heavily
953 dependent on cloud type and environmental conditions (Khain et al., 2008). ~~(Stevens~~
954 ~~and Feingold, 2009)~~ ~~(Carrió and Cotton, 2014; Glassmeier and Lohmann, 2018; Seifert~~
955 ~~et al., 2015).~~

956 A unified theory for the contradicting results regarding aerosol effectsA different
957 approach to aerosol effects suggests that cloud systems can be buffered to

958 microphysical effects (Stevens and Feingold, 2009). Several studies have shown that
959 given enough time for the cloud system to reach steady state, cloud macro-physical
960 parameters (e.g. cloud fraction, rain yield) show similar results for various aerosol
961 concentrations (Carrió and Cotton, 2014; Glassmeier and Lohmann, 2018; Seifert et al.,
962 2015). Based on the idea that clouds can be partitioned to aerosol-limited, updraft-
963 limited, or aerosol and updraft sensitive regimes (Reutter et al., 2009), a unified theory
964 for the contradicting results regarding aerosol effects was suggested (Dagan et al.,
965 2015b)was shown in recent work (Dagan et al., 2015b). It was shown that. Given an
966 aerosol range that covers all three regimes, the competition between opposite processes
967 leads to an optimum value of aerosol concentration regarding various cloud properties
968 like total mass, cloud top, or rain_(Dagan et al., 2015b). A cloud that develops under
969 low aerosol concentration is aerosol limited, as it does not have enough collective
970 droplet surface area to consume the available water vapor. On the other side of the trend,
971 a cloud that develops in polluted environment (with more aerosols than the optimum)
972 is influenced significantly by enhanced entrainment and larger water loading, causing
973 suppression of cloud development. The optimal concentration is a function of the
974 thermodynamic conditions (temperature and humidity profiles) and cloud size.

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

985 In addition, it was shown that clouds impact differently the environmental
986 thermodynamics according to the aerosol level in the field (Dagan et al., 2016; Seifert
987 and Heus, 2013; Seifert et al., 2015). For example changes in aerosol loading impact
988 the amount of precipitation reaching the surface and subsequently the evaporative
989 cooling below cloud base and the organization patterns (Seifert and Heus, 2013; Seigel,
990 2014; Xue et al., 2008). Moreover, an increase in aerosol loading may increase

991 evaporation rates around the margins and tops of clouds (Seigel, 2014; Stevens, 2007;
992 Xue and Feingold, 2006), cooling the upper cloudy layer and increasing the convective
993 instability. Therefore aerosol effects on phase changes and precipitation result in
994 vertical redistribution of heat and moisture, which may either stabilize or destabilize
995 the environment in which subsequent clouds grow (Seifert and Heus, 2013).

996 Irrespective of the definition chosen, the cloud's core and margin are dominated by
997 different processes (Dagan et al., 2015b). These processes often compete with each
998 other, with the dominant one changing along the cloud's evolution. For example, at the
999 initial stage of cloud formation, a cloud is more adiabatic and is controlled by the core's
1000 processes (condensation), and when it dissipates the margin processes are more
1001 dominant (entrainment and evaporation). Aerosols affect each of these processes and
1002 thus each stage in the cloud's lifetime. As a continuation to Part I of this work (hereafter
1003 PTI), in this part we analyze aerosols effects on the cloud's partition to core and margin
1004 throughout the lifetime of a cloud. We report the consequences these effects have on
1005 evolution of a cloud, in terms of volume, mass, and lifetime. As opposed to other works
1006 that typically focus on a single cloud core definition, here three different definitions are
1007 used (see Sect. 2), with emphasis put on the sensitivity of each core definition to aerosol
1008 concentration. Moreover, the combination of single cloud with large eddy simulations
1009 enables us to gain process level understanding and test the robustness of our findings.

1010

1011 **2. Methods**

1012 The analyses performed here are to the most part identical to those described in Part
1013 IPTI of this work. In this section we shall thus only give a brief review of the methods
1014 used. For single cloud simulations we use the Tel-Aviv University axisymmetric cloud
1015 model (TAU-CM (Reisin et al., 1996)), and for cloud field simulations we use the
1016 System for Atmospheric Modeling (SAM) Model (version 6.10.3, for details see
1017 webpage: <http://rossby.msrc.sunysb.edu/~marat/SAM.html>, (Khairoutdinov and
1018 Randall, 2003)).

1019 Both models utilize explicit bin microphysics schemes (Khain et al., 2004; Tzivion et
1020 al., 1987), solving nucleation, diffusion (i.e. condensation and evaporation), collisional
1021 coalescence, breakup, and sedimentation microphysical processes. The single cloud

1022 model is initialized using a Hawaiian thermodynamic profile, based on the 91285
1023 PHTO Hilo radiosonde at 00Z, 21 Aug, 2007. The cloud field model is setup based on
1024 the BOMEX case study, including an initialization setup (sounding, surface fluxes, and
1025 surface roughness) and large scale forcing setup (Siebesma et al., 2003). More details
1026 on the model setups and definitions can be found in PTI.

1027 To study the effects of aerosols on the cloud cores we run each model setup with three
1028 different aerosol concentrations: clean – 25 cm^{-3} , intermediate – 250 cm^{-3} , and polluted
1029 – 2000 cm^{-3} . ~~(Heiblum et al., 2016a) As defined in Part I. The model domain is~~
1030 initialized using an oceanic size distribution (Altaratz et al., 2008; Jaenicke, 1988),
1031 maintaining constant mixing ratio with height. Aerosol budget includes removal by
1032 nucleation and regeneration upon evaporation, while wet scavenging by precipitation
1033 removes aerosols from the domain. Thus, the aerosol concentration may be depleted by
1034 20%–40% (depending on the precipitation amount) during the simulation. More on the
1035 treatment of aerosols in the cloud field model can be found in previous work (Heiblum
1036 et al., 2016a). As defined in PTI, all pixels with at least 0.01 g kg^{-1} of liquid water are
1037 considered cloudy. Cloud cores are defined using three definitions: -1) RHcore: relative
1038 humidity $> 100\%$, 2) ~~B_{core}~~: Bcore: buoyancy > 0 , and 3) ~~W_{core}~~: Wcore: vertical velocity
1039 > 0 . Relative humidity (RH) and vertical velocity (W) are standard outputs of the model,
1040 while the buoyancy (B) is calculated based on eq. 1 in PTI, where each cloudy pixel is
1041 compared with the mean non-cloudy thermodynamic reference state per height.

1042 In order to reduce the problem's dimensionality and distill signals in a cloud field
1043 system governed by high variance, we use the Gravity vs. Mass (CvM) phase space in
1044 combination with an automated 3D cloud tracking algorithm (Heiblum et al., 2016a).
1045 The CvM phase space enables a compact view of all clouds in the simulation, by
1046 projecting only their Center-of-Gravity (COG) height and mass at each output time step.
1047 Using the cloud tracking, it was shown that the lifetime of a cloud can be described by
1048 a trajectory on this phase space. Hence, the different locations in the CvM space are
1049 associated with different stages in a cloud's lifetime (i.e. growing, precipitating, and
1050 dissipating). For an in-depth explanation of the CvM space, the reader is referred to
1051 Sect. 2.4 in PTI (see schematic illustration - Fig. 1, PTI).

1052

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1054 3. Results – Single cloud simulations

1055 3.1. Sensitivity of different core types to aerosol concentration

1056 Figure 1 presents time series of single cloud core volume fractions and cores'
1057 properties, for three aerosol concentrations (clean, intermediate, and polluted). Also
1058 included are time series of instantaneous rain-rates [mm hr⁻¹] at the domain surface. For
1059 all aerosol concentrations and during most of the clouds' lifetimes, the volume fraction
1060 of W_{core} tends to be the largest and of B_{core} the smallest. Exceptions to this
1061 finding are seen either at the initial time step for the polluted cloud or the later stages
1062 of cloud lifetime for the lower concentration clouds. In addition, we find that $RH_{core} \subset$
1063 $W_{core} RH_{core} \subset W_{core}$ for all stages of cloud lifetime while $B_{core} \subset$
1064 $W_{core}, RH_{core} B_{core} \subset W_{core}, RH_{core}$ for all stages of the polluted cloud but only
1065 applies to the growing stages of lower concentration clouds before precipitation
1066 production. Thus, the main finding from PTI (i.e. $B_{core} \subset RH_{core} \subset W_{core}$) $B_{core} \subset$
1067 $RH_{core} \subset W_{core}$) generally applies to all aerosol concentrations during the pre-
1068 precipitation stages of the clouds' lifetimes.

1069 Lower aerosol concentration simulations produce more rain, and at earlier stages of
1070 cloud lifetime due to efficient collision coalescence. The increase in B_{core} volume
1071 fraction at later stages of cloud lifetime in those simulations (clean and intermediate)
1072 coincides with initiation of precipitation production, followed by a consequent increase
1073 in W_{core} volume fraction as well (more so for the intermediate concentration).
1074 This dissipating W_{core} is mostly contained within the B_{core} . The possible mechanism
1075 behind the increase in prevalence of buoyant parcels during precipitation is explored in
1076 Sect. 3.2. The lack of RH_{core} pixels at these stages indicates that the
1077 W_{core} is composed of pixels with small vertical velocities, insufficient for
1078 supersaturation production. The RH_{core} is the only one which is not sensitive to
1079 rain and monotonically decreases during all clouds' lifetimes. Another clear aerosol
1080 effect seen in Fig. 1 is an increase in cloud lifetime with decrease in aerosol
1081 concentration. This point will be further explored in Sect. 3.3.

1082

1083 3.2. Mechanisms governing positive buoyancy

1084 The theoretical arguments in PTI showed that B_{core} should be the smallest of the
1085 three. This was shown for both the adiabatic cloud column case and also the non-
1086 adiabatic case where entrainment mixing and consequent evaporation has a strong net
1087 negative effect has on cloud buoyancy. Despite this fact, results show (see Fig. 1, and
1088 Fig. 2 in PTI) that pockets of positive buoyancy may form independent of the other
1089 cores during dissipation and precipitation stages, even though evaporation is to be
1090 expected then. Since positive buoyancy is the result of either higher temperature or
1091 vapor content (or both) than the surrounding environment, we choose to analyze these
1092 two terms during different stages of the single cloud lifetimes: clouds' lifetimes. The
1093 liquid water content buoyancy term (not shown here) is always negative and typically
1094 increases (in absolute value) with increase in vertical velocity or total buoyancy.

1095 Figure 2 shows the values of the temperature (B_T) and humidity (B_{Q_v}) buoyancy terms
1096 in pixel buoyancy vs. pixel vertical velocity phase space. The scatter plots include all
1097 cloudy pixels during all time steps, for the three different aerosol concentration
1098 simulations. The distribution of points for the polluted simulation shows a positive
1099 linear dependence of buoyancy on vertical velocity. Negative vertical velocity is
1100 associated with negative buoyancy and positive vertical velocity shows a transition
1101 from negative to positive buoyancy with increase in magnitude. For this case both B_T
1102 and B_{Q_v} increase with increase in vertical velocity, as is generally expected in
1103 convective clouds. The sign of pixel buoyancy is mostly dependent on B_T since all
1104 pixels have positive B_{Q_v} and a negative water loading term. This behavior is also seen
1105 for lower aerosol concentrations, where the sign of buoyancy is to the most part
1106 determined by B_T . Hereafter, we refer to positive buoyancy (both B_T and B_{Q_v})
1107 production within updrafts as updraft buoyancy.

1108 The clean and intermediate simulations show a similar dependence of buoyancy on
1109 vertical velocity; however, it is apparent that these simulations also include an outlier
1110 scatter region of pixels with positive buoyancy and weak negative vertical velocity
1111 which is absent in the polluted simulation (see white arrows, Fig. 2). Consistent with
1112 the rest of the cloudy pixels, these outlier pixels have positive B_T , but differ in that they
1113 show neutral B_{Q_v} . It can also be seen that these pixels are only attributed to the stages
1114 after surface precipitation has commenced (indicated by black dots in markers).
1115 Precipitation is indicative of both downdraft motion and abundance of large droplet
1116 sizes.

1117 Thus, we hypothesize that pockets of positive buoyancy may form due to transport of
1118 parcels with higher potential temperature from above, namely adiabatic heating. The
1119 weak downdrafts also transport lower mixing ratio (Q_v) values, as is indicated by the
1120 neutral B_{Q_v} . Although not usually the focus of studies, the existence of positively
1121 buoyant downdrafts in convective clouds has been reported in both observations (Igau
1122 et al., 1999; Wei et al., 1998) and simulations (Xu and Randall, 2001; Zhao and Austin,
1123 2005a, 2005b). A possible explanation for this can be deduced from previous theoretical
1124 studies predicting mixing induced downdrafts in cumulus clouds (Betts and Silva Dias,
1125 1979; Betts, 1982). It was shown that in some cases cloud - environment mixtures are
1126 negatively buoyant (while still containing liquid water) and the consequent downdrafts
1127 can sometimes descend only part way down to the cloud base before reaching neutral
1128 buoyancy. Similar to convective overshooting, parcels with negative vertical
1129 momentum may then “undershoot” the downdraft equilibrium level and turn positively
1130 buoyant while the downdraft weakens. One can therefore expect the magnitude of
1131 positive buoyancy within the downdraft to reach a maximum when the velocity
1132 approaches zero. Hereafter we refer to positive buoyancy production within downdrafts
1133 as downdraft buoyancy.

1134 Downdraft buoyancy production occurs frequently in cumulus fields because the
1135 negatively buoyant downdrafts follow a warming lapse rate which is more unstable than
1136 the environmental one, which is typically between the dry adiabat and moist adiabat (as
1137 is the case for the Hawaiian and BOMEX profiles simulated in this work). On one
1138 extreme, a descending parcel is least buoyant (i.e. coolest) when evaporation (after
1139 mixing) keeps it just barely saturated (Paluch and Breed, 1984, also PTI) so that the
1140 lapse rate of descent tends to moist adiabatic and may remain negatively buoyant. On
1141 the other extreme, if little to no evaporation of liquid water occurs, the descent will
1142 follow the dry adiabat and switch to neutral (and then positive) buoyancy rapidly. Thus,
1143 the ability of a negatively buoyant cloudy downdraft to sustain itself depends on
1144 continuous inflow of liquid water (by mixing) and its consequent evaporation (Knupp
1145 and Cotton, 1985).

1146 Indeed, the results in Fig. 2 match the hypothesis explained above, where positively
1147 buoyant downdrafts are warmer than the environment, and tend to show larger
1148 buoyancy values for weaker downdrafts velocities (especially for the intermediate
1149 case). Further analysis also shows that the more unsaturated the downdrafts (indicated

1150 also by low B_{Qv}), the larger the positive buoyancy. Moreover, the occurrence during
1151 precipitating stages and for lower aerosol concentrations indicates that slow
1152 evaporation due to larger ~~droplet sizes is crucial. Indeed, most pixels with negative~~
1153 ~~buoyancy show positive B_{Qv} except for the clean case where rain pixels from the cloudy~~
1154 ~~layer sediment well below the cloud base and experience higher environmental Q_v~~
1155 ~~(while evaporating slowly), resulting in negative B_{Qv} .~~ drop sizes is crucial for downdraft
1156 buoyancy production, enabling a near dry adiabatic lapse rate during descent.

1157

1158 3.3. The dependency of cloud characteristics on core and margin's processes

1159 Here we evaluate how aerosol effects within the core and margin (using the three core
1160 definitions) affect the cloud characteristics, focusing on two main parameters; size (or
1161 volume) and mass. In Fig. 3 we follow the evolution of cloud, core, and margin mass
1162 and volume for different aerosol concentrations, using only the ~~RH_{core}~~ RH_{core}
1163 definition. We choose the ~~RH_{core}~~ RH_{core} since it is the most well behaved out the core
1164 types, generally decreasing monotonically (see Fig. 1). A non-monotonic dependency
1165 of total cloud mass on aerosol concentration is seen, showing a maximum for the
1166 intermediate concentration. This type of dependency has been previously reported for
1167 warm cumulus clouds (Dagan et al., 2015b; Savane et al., 2015).

1168 One can generally expect an increase in diffusion and decrease in collision-coalescence
1169 processes efficiency with increase in aerosol concentration (Hudson and Yum, 2001;
1170 Jiang et al., 2009; L'Ecuyer et al., 2009; Pinsky et al., 2013), affecting both
1171 condensation and evaporation processes. The intermediate concentration shows the
1172 highest total mass as a result of being an optimal case with higher condensation
1173 efficiency than the clean case and lower evaporation efficiency than the polluted case.
1174 It is convenient to represent the condensation and evaporation efficiencies by the
1175 ~~RH_{core}~~ RH_{core} and ~~RH_{margin}~~ RH_{margin} mass, respectively. The intermediate cloud has
1176 almost identical core mass as does the polluted cloud, but retains higher mass in its
1177 margin as well. The clean cloud shows the lowest core mass but manages to accumulate
1178 the largest mass in its margin that dissipates slowly in ~~sub-saturated~~ sub-saturated
1179 conditions. By comparing the total cloud mass evolution with the core and margin mass
1180 evolutions, it becomes clear that the total mass is primarily dependent on the cloud core.
1181 Another way to see this is by plotting the core mass fraction (Fig. 3 bottom panel),

1182 which shows that clouds are core dominated (core fraction > 0.5) with respect to mass
1183 for most of their lifetimes, and for all aerosol concentrations.

1184 With respect to cloud total volume, the lower the concentration, the larger the total
1185 cloud volume. We note that the cloud volume here excludes regions of precipitation
1186 below the initial cloud base height. By separating to core and margin regions, one can
1187 see that the total cloud volume is primarily dependent on the volume of the margin,
1188 which increases significantly with decreasing concentration. This is especially true
1189 during the dissipating stages of cloud lifetime, when the cloud is margin dominated.
1190 Although increasing the aerosol concentration does initially yield an increase in core
1191 volume (as was seen for the mass), the extents of the core size are typically smaller than
1192 those of the margin. There are large differences in the relative core volume percent for
1193 the different clouds. The clean (polluted) cloud is margin (core) dominated with respect
1194 to volume for most of its lifetime. Excluding time of formation, the clean cloud shows
1195 the lowest core volume fractions, but manages to maintain its core for the longest time
1196 span.

1197 These results with respect to cloud volume can ~~again~~ be attributed to the smaller drop
1198 sizes and higher diffusion efficiencies with increase in aerosol concentration.
1199 Additionally, lower collision-coalescence efficiencies also maintain a narrow droplet
1200 spectrum of small droplets in the polluted cloud. During the growing stage a higher
1201 aerosol concentration may permit the cloud to condense more water, release more latent
1202 heat, and promote cloud growth. This explains the larger core volume sizes. However,
1203 after the cloud exhausts its convective potential (i.e. the growth of the convective core
1204 terminates and reaches its peak in mass), its main method of expansion is by mixing
1205 with the environment (i.e. detrainment and dilution). We note that precipitation can also
1206 be considered a method of expansion; however our choice to focus on volume above
1207 initial cloud base excludes this effect. –Detrainment results and mixing with the
1208 environment result in sub-saturation conditions and evaporation of LWC. A clear
1209 indication for dilution is seen in Fig. 3 where between 30 and 35 mins of simulation
1210 time both the clean and polluted clouds lose total mass but only the clean cloud
1211 increases in total volume. The polluted cloud is composed of small drops, evaporates
1212 its margin regions efficiently, and is thus limited in horizontal growth by detrainment
1213 growth. The clean cloud is composed of larger drops, less efficient in evaporating its
1214 margins, and hence can grow by dilution of its LWC upon a larger volume. This large

1215 margin "shields" the core during dissipation stages and enables it to live for a longer
1216 time.

1217 The mechanism behind the results in Fig. 3 is demonstrated in Fig. 4, where horizontal
1218 cross-sections of mean (taken in the vertical dimension) cloud RH are shown for
1219 different stages during the clouds' lifetimes. For the polluted cloud, super- or sub-
1220 saturated conditions are rare. The RH throughout the cloud is near 100% (almost always
1221 between 99.8% and 100.2%) except for a few pixels at its far edges which are a bit
1222 below 99%. The polluted cloud resembles what one would expect to see using a moist
1223 adiabatic approximation (i.e. saturation adjustment), where all excess water vapor
1224 above saturation is converted to liquid water, mimicking infinitely efficient
1225 condensation (and evaporation).

1226 The clean cloud shows opposite behavior, with extremes of large super-saturation
1227 during cloud growth (initial stages) and large sub-saturation during cloud dissipation
1228 (final stages). ~~Both extremes can be explained by the low diffusion efficiency in this~~
1229 ~~ease.~~The large super-saturation can be explained by slow diffusional growth, but the
1230 large sub-saturation also takes into consideration the larger drop sizes which take more
1231 time to evaporate. This enables the clean cloud to expand to larger horizontal extents
1232 (by dilution and mixing with the environment without fully evaporating) and live for
1233 longer times. The intermediate aerosol concentration shows a midway scenario, where
1234 the super-saturation is consumed more efficiently than the clean case and at the same
1235 time much larger values of sub-saturation may exist than those seen for the polluted
1236 case.

1237

1238 **4. Results – Cloud field simulations**

1239 In the following section we expand our analyses of aerosol effects on cloud core and
1240 margin from the single cloud scale to the cloud field scale. A cloud field can be
1241 considered as composed of many individual clouds and thus can serve to test the
1242 robustness of the aerosol effects seen for a single cloud. Moreover, cloud fields include
1243 the added complexity of interactions between clouds and the clouds' effects on their
1244 thermodynamic environment.

1245 **4.1. Sensitivity of different core types to aerosol concentration**

1246 Here CvM space representations (see Sect. 2) are used to observe the core volume
1247 fractions of all clouds in BOMEX cloud field simulations. The rows in Fig. 5 represent
1248 different aerosol concentrations while the columns represent different core type
1249 definitions. Different aerosol concentrations produce a vastly different scatter of clouds
1250 in the CvM space, as was previously discussed in depth (Heiblum et al., 2016b). The
1251 clean simulation (25 cm^{-3}) shows two disconnected regions of cloud scatter: one which
1252 is adjacent to the adiabatic approximation and one of mainly small mass and high COG
1253 clouds. The former region includes both clouds during their growth stages (smaller
1254 masses, $\text{LWP} < 10 \text{ g m}^{-2}$) and large precipitating entities (larger masses, $\text{LWP} > 10 \text{ g}$
1255 m^{-2}) which form due to merging processes (Heiblum et al., 2016b). The latter region
1256 (small mass and high COG) includes clouds at their dissipating stage, which form by
1257 shedding mechanism off the large cloud entities. We note also the existence of small
1258 mass elements well below the adiabat, representing precipitation cloud segments which
1259 shed off large precipitating clouds.

1260 The polluted simulation (2000 cm^{-3}) shows a much more homogeneous scatter of
1261 clouds. The lower part of the scatter (closest to the adiabat) represents the cloud
1262 growing branch while the rest of the scatter represents dissipating clouds, either by
1263 gradual process of rising cloud base or by immediate process of shedding off larger
1264 cloud entity (see Fig. 1, PTI). Precipitating cloud segments below the adiabat are absent
1265 from this simulation. The intermediate simulation (250 cm^{-3}) shows a scatter which
1266 generally more resembles the polluted case. However, the existence of relatively
1267 disconnected (from the main cloud scatter) small mass cloud segments below the
1268 adiabat and near the inversion base height resembles the clean simulation as well.
1269 (Heiblum et al., 2016b) It should be noted that horizontal dashed lines in Fig. 5 represent
1270 the inversion base height after 5 hours of simulation (approximately middle of
1271 simulation), where an increase in the inversion base height is seen with decrease in
1272 aerosol concentration. This is due to increased net warming in the upper cloudy layer
1273 (i.e., release of latent heat during condensation with reduced local evaporation) with
1274 increase in precipitation (Dagan et al., 2016; Heiblum et al., 2016b), which raises the
1275 inversion base.

1276 The results in Fig. 5 show a consistent behavior of the core volume fractions for all
1277 aerosol concentrations, where the W_{core} type shows the largest fractions and the
1278 B_{core} type shows the smallest fractions. The W_{core} and RH_{core}

1279 generally show a decrease in core fractions along the growing branch while the
1280 B_{core} fraction initially increase with cloud growth and then decrease for the large
1281 mass growing clouds. The percentages in the panel legends (Fig. 5) indicate the fraction
1282 of clouds (out of the scatter) which are core dominated with respect to volume ($f_{vol} >$
1283 0.5). For all concentrations, less than 7% of clouds are Bcore dominated while more
1284 than 55% are Wcore dominated (with RHcore percentages somewhere in between). The
1285 Bcore typically occupies a small portion of a typical cloud volume while the Wcore
1286 typically occupies most of the cloud. The mean cloud area (proportional to scatter point
1287 size) shows an increase with increase in mean clouds LWP.

1288 These results are consistent with PTI and the single cloud simulations in Sect. 3.1.
1289 Nevertheless, some significant aerosol effects on the partition to core types can be seen.
1290 Focusing on the growing branch first (i.e. clouds located near the adiabat), we note the
1291 following:

1292 1) For the RH_{core} type, the core volume fractions of clouds after formation
1293 (i.e. with small mass) increase with decreasing aerosol concentration. This
1294 effect was also seen for the single cloud simulations and can be explained by
1295 the reduced efficiency of super-saturation consumption for fewer aerosols.

1296 2) The B_{core} volume fraction increases at smaller mass values (or earlier in
1297 cloud's lifetime) and to higher values for increasing aerosol concentration. This
1298 effect is complimentary to the previous one, since efficient consumption of
1299 super-saturation should result in more latent heat release and positive buoyancy.

1300 3) The core volume fractions of the largest mass clouds increase with increasing
1301 aerosol concentration, for all core types.

1302 4) For the dissipating branch clouds The mean area of large mass clouds increases
1303 significantly with decrease in aerosol concentration.

1304 We also note a general increase in the fraction of clouds that are Wcore or RHcore
1305 dominated with increase in aerosol concentration. Meaning adding aerosols shifts a
1306 cloud from being mostly margin to being mostly core. The Bcore is an exception since
1307 the clean case shows the highest fraction of Bcore dominated clouds and both the clean
1308 and polluted cases are more Bcore dominated than the intermediate case. This can be
1309 explained by the different mechanisms of buoyancy production (see Sect. 3.2 and 4.2),

1310 where the polluted case is positively influenced by updraft buoyancy production and a
1311 larger core volume fraction while the frequently precipitating clean case is positively
1312 influenced by downdraft buoyancy production. For the dissipating branch clouds, a
1313 highly variable pattern of core volume fractions can be seen, especially for the small
1314 mass clouds. For all aerosol concentrations, these small cloud fragments can be either
1315 core dominated, margin dominated, or equally partitioned. One can assume that these
1316 differences can be related to the different mechanisms by which cloud fragments form,
1317 either by gradual dissipation of a large cloud and by instantaneous shedding of a large
1318 cloud. As for aerosol effects on the dissipating clouds, we see the following:

1319 1) Higher $\frac{RH_{core}}{RH_{core}}$ and $\frac{W_{core}}{W_{core}}$ volume fractions for gradually
1320 dissipating clouds (by rising cloud base) with increase in aerosol concentration.
1321 This is manifested by a slower transition from red to blue colors in Fig. 5. It can
1322 be explained by the fact that more aerosols increase the convective intensity and
1323 extend the core size, while efficiently losing the margins, yielding a higher core
1324 volume fraction out of the total cloud.

1325 2) The likelihood to find dissipating cloud fragments with a $\frac{B_{core}}{B_{core}}$ increases
1326 with decrease in aerosol concentration. For the polluted case most of the
1327 dissipating clouds lack a $\frac{B_{core}}{B_{core}}$. This effect was seen in Fig. 1 and
1328 explained in Sect. 3.2, showing that ~~weak~~ downdrafts promote heating and
1329 positive buoyancy in low aerosol concentration cases where evaporation
1330 efficiency (and hence cooling) is limited. This ~~hypothesis~~ effect is checked for
1331 the cloud field scale in Sect. 4.2.

1332 As opposed to the single cloud simulations (Sect. 3) where cloud lifetime can be easily
1333 defined, in cloud field simulations (especially the cleaner cases) many clouds do not
1334 live as individual clouds from formation to dissipation but rather split and merge with
1335 other clouds continuously (Heiblum et al., 2016b). Thus, in order to evaluate the
1336 lifetime evolution of cores in cloud fields, we focus on the growing branch and use
1337 cloud mass [kg] as a proxy for the cloud lifetime during its initial and mature stages.
1338 We assume that in the vicinity of the growing branch a larger mass corresponds to a
1339 later stage in lifetime.

1340 In Fig. 6 the core mass and volume fractions (using the RH definition) of all growing
1341 branch clouds are sorted by mass for the three aerosol concentrations. We note that the

1342 higher cloud masses reached by lower aerosol concentration simulation can be
 1343 explained by cloud field organization effects due to precipitation (i.e. increased merging
 1344 of clouds) rather than increased cloud condensation (Heiblum et al., 2016b; Seigel,
 1345 2014). The clean case starts off with the highest core fractions (both mass and volume)
 1346 which decrease steadily with increase in mass (or increase in lifetime). For all
 1347 concentrations, most of the cloud mass is concentrated in the core region. The polluted
 1348 case shows a slight increase in core mass fractions with increase in mass, while the
 1349 other two cases show decreases in core mass fractions.

1350 The core volume fractions show lower values than the mass fractions. The clean clouds
 1351 are margin dominated for most masses, and the polluted clouds are core dominated for
 1352 all masses. The intermediate case is generally confined to values between the other two
 1353 cases. Figure 6 can be considered comparable with the lower panels in Fig. 3, but
 1354 excluding the dissipating part of those time series. The similar findings in both figures
 1355 indicate the robustness of the aerosol effects on core properties in clouds.

1356 Following the analyses of Sect. 3.1, we next test how aerosol concentration affects the
 1357 subset properties of one core type within another for all clouds in a field (Fig. 7). We
 1358 focus only on the typically smaller sized cores ((B_{core}, RH_{core})) within
 1359 larger sized cores. Out of the three permutations, the RH_{core} inside W_{core}
 1360 shows the lowest sensitivity to aerosol. All three growing branches (for the different
 1361 aerosol concentrations) consistently show that the RH_{core} is a subset of
 1362 W_{core} (i.e. $RH_{core} \subseteq W_{core}$) while the dissipation branches
 1363 show much lower overlap fraction between the two cores.

1364 Generally, for the dissipating clouds, the lower the mass and the higher the COG, the
 1365 smaller the overlap. The dissipating branches do include a scatter of small cloud for
 1366 which $RH_{core} \subseteq W_{core}$, comprised of small cloud segments which
 1367 shed off the main core regions of larger clouds. These findings slightly differ from
 1368 those of the single cloud simulations that show $RH_{core} \subseteq W_{core}$ for
 1369 their entire lifetimes while for cloud fields this property breaks down during
 1370 dissipation. This difference highlights the importance of cloud interactions (i.e.
 1371 splitting, merging) and cloud field air flow patterns (i.e. organized advection, updrafts,
 1372 and downdrafts) in determining the relationships between core types, enabling
 1373 supersaturation and downdrafts to coincide in small dissipating clouds.

1374 The other two permutations (i.e. $B_{core} \subseteq B_{core}$ inside RH_{core}, W_{core})
 1375 show significant changes due to aerosol. For the polluted case, $B_{core} \subseteq W_{core} \subseteq B_{core} \subseteq$
 1376 W_{core} for nearly all clouds, including clouds at initial stages of dissipation. Similar
 1377 results are seen for $B_{core} \subseteq B_{core}$ inside RH_{core}, RH_{core} , but with slightly lower pixel
 1378 fractions. The polluted case thus illustrates the case of buoyancy production due to
 1379 convective processes. For the lower aerosol concentrations, two main aerosol
 1380 effects are seen:

- 1381 1) The lower the concentration, the lower the chance that $B_{core} \subseteq B_{core}$ is a proper
 1382 subset of the other cores for large growing branch clouds.
- 1383 2) The lower the concentration, the more prevalent the independent dissipating
 1384 branch $B_{core} \subseteq B_{core}$ that has little to no overlap with the other cores.

1385 For the case of $B_{core} \subseteq B_{core}$ within RH_{core}, RH_{core} , the lower concentrations show an
 1386 almost binary scenario where either $B_{core} \subseteq RH_{core} \subseteq B_{core} \subseteq RH_{core}$ or $B_{core} \not\subseteq$
 1387 $RH_{core} \subseteq B_{core} \not\subseteq RH_{core}$. These results bear similarity with the single cloud simulations,
 1388 where a quick transition (in time) from $B_{core} \subseteq RH_{core} \subseteq B_{core} \subseteq RH_{core}$ to $B_{core} \not\subseteq$
 1389 $RH_{core} \subseteq B_{core} \not\subseteq RH_{core}$ was seen. These results imply the existence of two
 1390 different buoyancy production processes (as will be shown more in Sect. 4.2), one
 1391 associated with supersaturation and the other with subsaturation. In
 1392 contrary, inside, which shows higher values and more fluctuations in pixels fractions of
 1393 B_{core} inside W_{core} span the entire range of values (i.e. partial overlaps between the
 1394 core types), as seen for both single clouds and cloud fields during dissipation.
 1395 This is to be expected due to the a more direct physical link and feedbacks between the
 1396 $B_{core} \subseteq B_{core}$ and $W_{core} \subseteq W_{core}$.

1397

1398 4.2. Analysis of cloud field buoyancy

1399 In Sect. 3.2 it was seen that for single clouds, positive buoyancy results from two main
 1400 mechanisms: i) convection - convective updrafts - where updrafts promote
 1401 supersaturation and latent heat release, and thus always positive B_{Qv} and frequently
 1402 positive B_T and B_{Qv} , and ii) adiabatic heating/dissipative downdrafts - where weak sub-
 1403 saturated cloudy downdrafts promote a positive B_T and neutral B_{Qv} . The latter case is

1404 dependent on low evaporation efficiency and hence seen mostly for precipitating stages
 1405 of low aerosol concentration simulations.

1406 In Fig. 8 we perform a similar test for the cloud field scale. Instead of analyzing pixel
 1407 by pixel, we check whether each buoyancy core within a cloud is B_T or B_{Qv} dominated.
 1408 To quantify this we use a normalized buoyancy dominance parameter
 1409 $\frac{\Sigma pixel_{B_T > 0} - \Sigma pixel_{B_{Qv} > 0}}{\Sigma pixel_{B > 0}}$, where a core comprised of only $B_T \geq 0$ ($B_{Qv} \geq 0$) pixels yields
 1410 1 (-1). Hence, we expect negative (positive) values to indicate dominance of
 1411 convective updraft buoyancy (adiabatic heating downdrafts buoyancy).

1412 Analysis of the buoyancy components in the CvM space (right column, Fig. 8) shows
 1413 that the large majority of clouds are B_{Qv} dominated. For all concentrations, clouds
 1414 initiate with all pixels showing $B_{Qv} \geq 0$. As clouds develop along the growing branch
 1415 the B_{core} becomes more abundant with $B_T \geq 0$ pixels. This is expected with
 1416 increasing release of latent heat during cloud growth. During dissipation B_{Qv} again
 1417 becomes the dominant component for the majority of clouds. The polluted simulation
 1418 shows an extreme case where all buoyancy cores in the simulation are B_{Qv} dominated,
 1419 while for the lower concentrations a portion of the dissipating and precipitating clouds
 1420 are B_T dominated.

1421 Thus, we hypothesize that the polluted simulation only permits buoyancy cores of the
 1422 convective updraft type which intersect with the other cores types (i.e. $B_{core} \in$
 1423 RH_{core}, W_{core}), $B_{core} \in RH_{core}, W_{core}$), while the lower concentrations also permit
 1424 buoyancy cores of the adiabatic heating downdraft type which do not intersect with the
 1425 other core types (i.e. $B_{core} \notin RH_{core}, W_{core}$). This hypothesis is tested $B_{core} \notin RH_{core},$
 1426 W_{core}). We test this by observing the effects relation of cloud maximum absolute
 1427 vertical velocity (left column, Fig. 8) and mean drop size (middle column, Fig. 8)
 1428 with the relative dominance of the buoyancy terms. Absolute vertical velocity is
 1429 chosen to represent both updrafts and downdrafts. The data is further separated to
 1430 independent ($B_{core} \notin RH_{core}, W_{core}$) $B_{core} \notin RH_{core}, W_{core}$) and dependent ($B_{core} \in$
 1431 RH_{core}, W_{core}) $B_{core} \in RH_{core}, W_{core}$) buoyancy subsets of the data, by that
 1432 separating to buoyant cores within updrafts and downdrafts. Clear aerosol effects are
 1433 seen on cloud mean drop size and maximal $|W_{-}|$. As expected, there is a decrease in
 1434 drop size with increase in aerosol concentration and increase in maximal velocity.

1435 Regarding cloud field buoyancy, as predicted the independent buoyancy cores are more
1436 frequently $B_T B_T$ dominated than the dependent buoyancy cores.

1437 The polluted case is populated with dependent cores (white scatter) and shows a classic
1438 pre-precipitation convective growth scenario, where relative dominance of the $B_T B_T$
1439 term increases linearly with increase in cloud mean drop size. A logarithmic
1440 dependence of $B_T B_T$ dominance on maximal $|W|$ is seen, which saturates at high
1441 maximal $|W|$. This can be explained by the fact increased convection mainly increases
1442 the abundance of pixels with $B_T > 0, B_T > 0$, but without changing the fact that the entire
1443 cloud is $B_{Qv} > 0, B_{Qv} > 0$, so that $B_T B_T$ is unlikely to become the dominant term.

1444 The lower concentrations show a more complex scenario. These simulations show a
1445 superposition of dependent core convective growth behavior (i.e. the scatter pattern
1446 seen for the polluted case) and additional populations of both dependent (other white
1447 scatter points) and independent (black scatter) cores. ~~The independent cores span all the
1448 range of possibilities of B_T and B_{Qv} relative dominances. They tend to have larger cloud
1449 mean drop sizes, and near zero maximum W , indicating that they only form at late non-
1450 convective stages of cloud development. The independent cores that are B_T dominated
1451 thus fulfill the characteristics of adiabatic heating process, while the independent cores
1452 that are B_{Qv} dominated may originate from larger clouds (shedding mechanism) with
1453 high humidity content and are slow to evaporate.~~

1454 The independent cores span all the range of possibilities of B_T and B_{Qv} relative
1455 dominances. They tend to have larger cloud mean drop sizes, and near zero maximum
1456 $|W|$, indicating that they only form at late non-convective stages of cloud development.
1457 Furthermore, a trend is seen for the subset of scatter that is B_T dominated, where a
1458 positive (negative) correlation between mean drop size (maximal $|W|$) and B_T
1459 dominance is seen. This again stresses the importance of drop size on the formation of
1460 positive buoyancy within downdrafts, and highlights the fact that B_T should be largest
1461 (and most abundant) below the downdraft equilibrium level, when the $|W|$ approaches
1462 zero. The independent cores that are B_T dominated thus fulfill the characteristics of
1463 downdraft buoyancy production process, while the independent cores that are B_{Qv}
1464 dominated may originate from larger clouds (shedding mechanism) with high humidity
1465 content, have weak $|W|$, and are slow to evaporate.

1466 The intermediate simulation shows an additional scatter area of dependent core clouds
1467 with increasing of $B_T B_T$ relative dominance for lower maximal $|W|$, located between
1468 the independent core clouds and the convective growth core clouds. These clouds may
1469 represent a gradual transition from B_{QV} dominance to $B_T B_T$ dominance during
1470 dissipation which is only possible in the intermediate simulation. This scatter area is
1471 absent from the clean and polluted simulation. In the former case due to absence of the
1472 gradual dissipation pathway, and in the latter case due to efficient evaporation
1473 eliminating B_{core} during dissipation. We note that the
1474 intermediate case shows a slightly higher percentage of clouds that are B_T dominated
1475 (see legends in Fig. 8) than the clean case. This can be due to stronger convection in
1476 this simulation (i.e. increased $|W|$ range), which favors increased mixing with the dry
1477 environment (see Fig. 9) and the formation of unsaturated strong downdrafts that
1478 descend below the level of neutral buoyancy.

1479

1480 **4.3. Aerosol effects on cloud relative humidity**

1481 From Fig. 3 it was learned that a large part of the differences in single cloud
1482 characteristics (such as mass, volume, and the partition of these to core and margin
1483 regions) due to aerosols can be attributed to differences in vapor diffusion efficiencies.
1484 In Fig. 9 we check how these aerosol effects are manifested in the cloud field scale
1485 (using the CvM space) by observing the mean relative humidity (RH) in the cloud core
1486 and margin of all clouds. ~~The, where the~~ core (margin) mean RH can be taken as a
1487 proxy for condensation ~~efficiency, the margin mean RH as a proxy for~~ (evaporation)
1488 efficiency. To gain additional intuition regarding the distribution of RH values within
1489 the clouds, vertical cross-sections (parallel to the prevailing wind direction) of the most
1490 massive clouds from each simulation are shown.

1491 The vertical cross-sections demonstrate the large differences in the massive clouds for
1492 each of the simulations. In addition to the increase in precipitation production, lower
1493 aerosol concentrations yield much larger horizontal extents of clouds. The clean,
1494 intermediate, and polluted most massive clouds have a maximum radius of ~ 3 , ~ 1 , and
1495 ~ 0.5 km, respectively. It is clear from the cross-section that the clean cloud is actually
1496 composed of two large clouds which merge together. For the clean case, the highest RH
1497 values are reached slightly below the cloud top. The edges of the clouds show sub-

1498 saturation conditions, with the lowest RH values observed below the LCL (precipitation
1499 regions) and at the upper interface of the cloud with the environments.

1500 The intermediate case cloud shows lower maximal and minimal RH values and an
1501 increased dominance of the margin region. This cloud penetrates the inversion layer
1502 and entrains dry air into the cloud. In addition, the cloud produces significant
1503 precipitation which initiates downdrafts of dry entrained air through the cloud center.
1504 It can be seen that the increased vertical development of the intermediate case cloud in
1505 comparison with the clean case increases the mixing with the environment. Thus, the
1506 dynamic effect of increased mixing and reduction in cloud RH overcomes the
1507 microphysical effect of increased evaporation and increase in cloud RH. The polluted
1508 case cloud on the other hand shows a homogeneous RH pattern, with most of the cloud
1509 showing around 100% RH and only a thin layer at the cloud edges (mainly at the upper
1510 regions) shows lower RH values. The polluted cloud penetrates the inversion layer as
1511 well, but this case lacks precipitation and the microphysical effect of evaporation
1512 overcomes the dynamical effect of mixing.

1513 Keeping in mind the insights obtained from comparisons of individual cloud, we move
1514 on to compare the RH characteristics of all clouds within the field. Looking first at core
1515 mean RH, a robust decrease is seen with increase in aerosol concentration. This
1516 decrease is seen for all cloud types and locations within the CvM space. The polluted
1517 case displays the most homogeneous pattern with all clouds showing core mean RH
1518 values around 100%, indicating efficient consumption of the supersaturation. The
1519 intermediate case displays a slightly less homogeneous pattern with values ranging
1520 from 100% to 101%, the higher values occurring along the growing cloud branch,
1521 especially for the largest clouds. The clean case shows the largest variance in core mean
1522 RH, ranging from 100% for some cloud fragments that soon start to dissipate, to 103%
1523 in the cores of the large cloud entities. In addition to the low efficiency in consuming
1524 supersaturation, the high RH values in clean large clouds are due to the "protection" by
1525 large margin regions surrounding the core region.

1526 The CvM patterns of mean margin RH show significant differences between the
1527 polluted case and the other two. The mean margin RH values of the polluted case are
1528 only marginally lower than 100%, since sub-saturated conditions within the cloud are
1529 quickly adjusted by efficient evaporation. Only the largest clouds in the polluted case

1530 permit lower mean margin RH values (~ 95%) due to the entrainment of very dry
1531 environmental pixels near the cloud tops (as seen in the vertical cross-section as well).
1532 The intermediate and clean cases show similar patterns. The smaller mass clouds (both
1533 growing and dissipating) show values above 95%, while the larger mass clouds show
1534 values as low as 85%. The larger clouds are most likely to reach low RH areas near the
1535 inversion base and below the LCL (i.e. sub-cloudy layer) and entrain dry air and by that
1536 reduce the cloud margin RH.

1537 As seen in the vertical cross-section examples, the largest clouds in the intermediate
1538 case have even lower margin RH values than for the clean case. This can be explained
1539 by the increased development of the large intermediate clouds to heights with lower RH
1540 and by more intense downdrafts for these large clouds. The lowest RH values in the
1541 domain are seen for the precipitating fragments (i.e. located below the adiabat). These
1542 fragments typically contain low concentrations of large drop sizes (precipitation drops)
1543 which are slow to evaporate and capable of surviving in low RH conditions within the
1544 sub-cloudy layer.

1545

1546

1547 Summary

1548 In this work we explored how the aerosol effects on warm convective clouds are
1549 reflected in their partition to core and margin regions. Following part I of this work
1550 (PTI), we evaluated three types of core definitions: positive buoyancy (B_{core}),
1551 super-saturation (RH_{core}), and positive vertical velocity (W_{core}).
1552 Both single cloud and cloud field models have been used. ~~The former distills the
1553 dominant in-cloud processes affected by aerosols while the latter also takes into
1554 consideration the multiple temporal cloud evolution pathways and the additional effects
1555 of cloud field organization and interactions between clouds.~~

1556 For all aerosol concentrations, (clean, intermediate, and polluted) it is shown that the
1557 self-contained property of different core types (i.e. $B_{core} \subseteq RH_{core} \subseteq W_{core}$, $B_{core} \subseteq$
1558 $RH_{core} \subseteq W_{core}$) is maintained for clouds during their growing and mature stages.
1559 This is especially robust for the $RH_{core} \subseteq W_{core}$ subset. The

1560 W_{core} and RH_{core} volume fractions decrease monotonically during cloud
1561 growth, while B_{core} initially increases and then decreases after convection ceases.
1562 During growth, the RH_{core} (B_{core}) volume fractions are largest for
1563 clean (polluted) clouds. This is due to low (high) diffusion efficiencies, respectively,
1564 where efficient condensation promotes B_{core} at the expense of ~~the~~
1565 RH_{core} .

1566 During dissipation stages cores frequently cease to be subsets of one another and may
1567 either increase or decrease in their volume fractions. In cloud fields we also observe
1568 small cloud fragments which shed off larger cloud entities. This shedding increases for
1569 the lower concentration simulation which produce long-lived large cloud entities due
1570 to cloud merging. These fragments show large variance in volume fraction (for all core
1571 types) magnitudes without any consistent behavior. This is due to the fact that they shed
1572 off various locations of the cloud. The polluted, non-precipitating cases, are unique in
1573 that can one expect the B_{core} to decrease monotonically and remain the smallest
1574 and a proper subset of the other cores.

1575 For low aerosol ~~concentration, clouds which are capable of producing~~
1576 ~~precipitation~~ concentrations, a B_{core} may form during dissipation and exist
1577 independently of the other core types. These cores are typically located at the periphery
1578 of large clouds, or throughout small precipitation or dissipating cloud fragments. The
1579 increase in B_{core} during dissipation typically coincides with large drop sizes and
1580 precipitation production. The fluctuations in B_{core} for low concentrations may
1581 also create a subsequent W_{core} , but not of sufficient strength to also create a
1582 RH_{core} . Hence, the RH_{core} can be considered the most “well-behaved”
1583 and indicative of cloud lifetime, generally monotonically decreasing in volume fraction
1584 irrespective of aerosol concentration.

1585 We show that the B_{core} in the warm convective cases considered here may form
1586 by two main processes:

- 1587 1. Convection Convective updrafts: adiabatic cooling within updrafts promotes
1588 supersaturation, condensation, and release of latent heat. These cores are
1589 characterized by both positive temperature ($B_T > 0$) and humidity ($B_{Qv} > 0$)
1590 buoyancy terms.

1591 ~~2. Adiabatic heating: weak~~Dissipative ~~downrafts during dissipation or~~
1592 ~~precipitation transport higher potential temperatures from above.~~

1593 2. The convective case: sub-saturated cloudy downrafts follow a lapse rate which
1594 is seen for all aerosol concentrations, and is unstable relative to the
1595 environmental one. These downrafts undershoot the equilibrium level and
1596 become positively buoyant. These cores are characterized by a dependent B_{core}
1597 (i.e. $B_{core} \in RH_{core}, W_{core}$). During convection B_{core} pixels have a positive
1598 humidity term (B_{qv}), with an increasing abundance of a positive temperature
1599 term (B_T) pixels with increase in cloud maximum vertical velocities. During
1600 dissipation this type of B_{core} shrinks rapidly due to negative B_T . The adiabatic
1601 heating case ($B_T > 0$) but neutral humidity ($B_{qv} \sim 0$) buoyancy terms.

1602 The updraft buoyancy type is seen for all aerosol concentrations, while the dissipation
1603 buoyancy type is only seen for lower aerosol concentrations, and is characterized by
1604 independent B_{core} (i.e. $B_{core} \notin RH_{core}, W_{core}$). In this case B_T is the dominant term in
1605 the cloud. The clouds with independent B_{core} experience near neutral vertical velocities
1606 for all pixels, and typically show larger cloud mean drop sizes than for the dependent
1607 type ones.

1608 ~~The.~~ The fact that the adiabatic heating B_{core} downdraft B_{core} is absent from polluted
1609 clouds highlights the importance of mean drop size and its effect on evaporation rate.
1610 The high (low) diffusion (collision coalescence) efficiencies in polluted clouds
1611 maintain a small mean drop size and efficientenable rapid evaporation during
1612 entrainment. In PTI we saw that evaporation always has, causing a strong negative
1613 effect on buoyancy. In the polluted case the convective B_{core} disappear rapidly during
1614 dissipation and cannot form in small cloud fragments even if they experience weak
1615 downrafts. The importance of drop size is illustrated by the fact that even for For lower
1616 concentrations, clouds with independent B_{core} a downdraft B_{core} only exist during late
1617 mature, dissipation, and precipitating stages after drop size has grown considerably.
1618 The larger mean drop sizes reduce evaporation rates and the cloudy downrafts may
1619 thus descend nearly dry adiabatically and become positively buoyant.

1620 Focusing on cores using the RH definition, a cloud's mass (volume) is dependent
1621 primarily on the processes in its core (margin). The core increases cloud mass by

1622 condensation while the margin increases the cloud's volume by mixing with the
1623 environment, or dilution. The magnitude of the effects in each region of the cloud is
1624 strongly dependent on the aerosol concentration. ~~Increasing the aerosol concentration~~
1625 ~~increases the vapor diffusion rate, minimizing both the super saturation and sub-~~
1626 ~~saturation (absolute) values in the cloud. Thus, polluted clouds are efficient in~~
1627 ~~accumulating water mass but also in losing it. This competition between the core mass~~
1628 ~~gain and margin mass loss regions is what brings about the concept of an optimal~~
1629 ~~aerosol concentration (Dagan et al., 2015b), and explains why more polluted clouds are~~
1630 ~~not necessarily more massive.~~

1631 Polluted clouds are core dominated both in terms of mass and volume, since they can
1632 hardly maintain their margins. Clean clouds are also core dominated in terms of mass,
1633 but to a lesser degree. ~~However, expect for the initial time of cloud formation where the~~
1634 ~~entire cloud is super saturated, clean~~Clean clouds tend to be margin dominated in terms
1635 of volume for most their lifetimes. Thus, despite weaker convection in the clean clouds,
1636 their large, slow evaporating margins enable their cores (and the entire cloud) to exist
1637 for longer time spans by applying a large "protecting shield" around the core.

1638 The different diffusion efficiencies are demonstrated by observing the relative humidity
1639 (RH) values in clouds. Cleaner clouds show larger variance in RH values. During their
1640 growing stages large super-saturation in the core and sub-saturation in the margin can
1641 be seen. During their dissipation stages clouds may exist for minutes without any cloud
1642 core, with the entire cloud at sub-saturation. Polluted clouds show the opposite, with
1643 RH values nearing 100% throughout the cloud, at all stages. Hence, above a certain
1644 aerosol concentration, the saturation adjustment approximation (i.e. instant
1645 condensation of all super-saturation) can be considered valid. However, the transition
1646 from clean to polluted is not always linear. For example, for the largest clouds in the
1647 intermediate case have lower margin RH value than both the clean and polluted cases.
1648 This is due to the fact that the intermediate case manages to develop taller (than the
1649 clean case) clouds with stronger updrafts and downdrafts which entrain drier air from
1650 above the inversion layer base, but at the same time is less efficient in evaporating (than
1651 the polluted case) water and adjusting the RH to 100%.

1652 ~~Finally, we note that the cloud organization also changes with aerosol concentration,~~
1653 ~~and thus serves as an additional factor affecting the cloud partition to core and margin.~~

1654 Decreasing the aerosol concentration increases the precipitation yield, which alters the
1655 sub-cloudy layer organization and promotes merging between different clouds
1656 (Heiblum et al., 2016b; Seifert and Heus, 2013; Seigel, 2014). These effects are minimal
1657 in the polluted cases. Hence, to a first approximation polluted cloud fields can be
1658 considered as a superposition of many single clouds while clean cloud fields behave
1659 very differently than a collection of single clean clouds. The continuous merging
1660 between clean clouds creates large cloud entities that evolve along relatively long times.
1661 These large precipitating entities also frequently shed small cloud fragments into the
1662 upper cloudy layer. This effect, combined with the low vapor diffusion, explains why
1663 clean clouds tend to be even more margin dominated (in terms of volume) during
1664 growth, while showing larger core fractions (especially B_{core}) during dissipation.

1665 Author Contributions

1666 RH ran cloud field simulations and conducted the analyses, and wrote the final draft of
1667 paper. LP participated in writing the first draft, and performed single cloud simulations
1668 and relevant analyses. OA, GD, and IK participated in paper editing and discussions.

1669

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1673

1674 **References**

1675 Albrecht, B. A.: Aerosols, cloud microphysics, and fractional cloudiness., Science,
1676 245(4923), 1227–1230, doi:10.1126/science.245.4923.1227, 1989.

1677 Altaratz, O., Koren, I., Reisin, T., Kostinski, A., Feingold, G., Levin, Z. and Yin, Y.:
1678 Aerosols' influence on the interplay between condensation, evaporation and rain in
1679 warm cumulus cloud, Atmos. Chem. Phys., 8(1), 15–24, doi:10.5194/acp-8-15-2008,
1680 2008.

1681 Altaratz, O., Koren, I., Remer, L. A. and Hirsch, E.: Review: Cloud invigoration by
1682 aerosols—Coupling between microphysics and dynamics, Atmos. Res., 140-141, 38–
1683 60, doi:10.1016/j.atmosres.2014.01.009, 2014.

1684 Andreae, M. O., Rosenfeld, D., Artaxo, P., Costa, A. A., Frank, G. P., Longo, K. M.
1685 and Silva-Dias, M. A. F.: Smoking rain clouds over the Amazon., Science, 303(5662),
1686 1337–1342, doi:10.1126/science.1092779, 2004.

1687 Betts, A. K.: Saturation point analysis of moist convective overturning, *J. Atmos. Sci.*,
1688 39(7), 1484–1505, doi:10.1175/1520-0469(1982)039<1484:SPAOMC>2.0.CO;2,
1689 1982.

1690 Betts, A. K. and Silva Dias, M. F.: Unsaturated downdraft thermodynamics in
1691 cumulonimbus, *J. Atmos. Sci.*, 36(6), 1061–1071, doi:10.1175/1520-
1692 0469(1979)036<1061:UDTIC>2.0.CO;2, 1979.

1693 Carrió, G. G. and Cotton, W. R.: On the buffering of CCN impacts on wintertime
1694 orographic clouds: An idealized examination, *Atmos. Res.*, 137, 136–144,
1695 doi:10.1016/j.atmosres.2013.09.011, 2014.

1696 Dagan, G., Koren, I. and Altaratz, O.: Aerosol effects on the timing of warm rain
1697 processes, *Geophys. Res. Lett.*, 42(11), 4590–4598, doi:10.1002/2015GL063839,
1698 2015a.

1699 Dagan, G., Koren, I. and Altaratz, O.: Competition between core and periphery-based
1700 processes in warm convective clouds – from invigoration to suppression, *Atmos.*
1701 *Chem. Phys.*, 15(5), 2749–2760, doi:10.5194/acp-15-2749-2015, 2015b.

1702 Dagan, G., Koren, I., Altaratz, O. and Heiblum, R. H.: Aerosol effect on the evolution
1703 of the thermodynamic properties of warm convective cloud fields., *Sci. Rep.*, 6,
1704 38769, doi:10.1038/srep38769, 2016.

1705 Dagan, G., Koren, I., Altaratz, O. and Heiblum, R. H.: Time-dependent, non-
1706 monotonic response of warm convective cloud fields to changes in aerosol loading,
1707 *Atmos. Chem. Phys.*, 17(12), 7435–7444, doi:10.5194/acp-17-7435-2017, 2017.

1708 Dey, S., Di Girolamo, L., Zhao, G., Jones, A. L. and McFarquhar, G. M.: Satellite-
1709 observed relationships between aerosol and trade-wind cumulus cloud properties over
1710 the Indian Ocean, *Geophys. Res. Lett.*, 38(1), doi:10.1029/2010GL045588, 2011.

1711 Glassmeier, F. and Lohmann, U.: Precipitation Susceptibility and Aerosol Buffering
1712 of Warm- and Mixed-Phase Orographic Clouds in Idealized Simulations, *J. Atmos.*
1713 *Sci.*, 75(4), 1173–1194, doi:10.1175/JAS-D-17-0254.1, 2018.

1714 Grant, L. D. and van den Heever, S. C.: Cold pool and precipitation responses to
1715 aerosol loading: modulation by dry layers, *J. Atmos. Sci.*, 72(4), 1398–1408,
1716 doi:10.1175/JAS-D-14-0260.1, 2015.

1717 Heiblum, R. H., Altaratz, O., Koren, I., Feingold, G., Kostinski, A. B., Khain, A. P.,
1718 Ovchinnikov, M., Fredj, E., Dagan, G., Pinto, L., Yaish, R. and Chen, Q.:
1719 Characterization of cumulus cloud fields using trajectories in the center of gravity
1720 versus water mass phase space: 1. Cloud tracking and phase space description, *J.*
1721 *Geophys. Res. Atmos.*, 121(11), 6336–6355, doi:10.1002/2015JD024186, 2016a.

1722 Heiblum, R. H., Altaratz, O., Koren, I., Feingold, G., Kostinski, A. B., Khain, A. P.,
1723 Ovchinnikov, M., Fredj, E., Dagan, G., Pinto, L., Yaish, R. and Chen, Q.:

1724 Characterization of cumulus cloud fields using trajectories in the center of gravity
1725 versus water mass phase space: 2. Aerosol effects on warm convective clouds, J.
1726 Geophys. Res. Atmos., 121(11), 6356–6373, doi:10.1002/2015JD024193, 2016b.

1727 Hudson, J. G. and Mishra, S.: Relationships between CCN and cloud microphysics
1728 variations in clean maritime air, Geophys. Res. Lett., 34(16),
1729 doi:10.1029/2007GL030044, 2007.

1730 Hudson, J. G. and Yum, S. S.: Maritime–continental drizzle contrasts in small cumuli,
1731 J. Atmos. Sci., 58(8), 915–926, doi:10.1175/1520-
1732 0469(2001)058<0915:MCDCIS>2.0.CO;2, 2001.

1733 Igau, R. C., LeMone, M. A. and Wei, D.: Updraft and downdraft cores in TOGA
1734 COARE: why so many buoyant downdraft cores?, J. Atmos. Sci., 56(13), 2232–2245,
1735 doi:10.1175/1520-0469(1999)056<2232:UADCIT>2.0.CO;2, 1999.

1736 IPCC: Climate Change 2013: The Physical Science Basis. Working Group I
1737 Contribution to the Fifth Assessment Report of the IPCC, Cambridge Univ. Press,
1738 New York., 2013.

1739 Jaenicke, R.: 9.3.1 Physical properties, in Physical and chemical properties of the air,
1740 edited by G. Fischer, pp. 405–420, Springer-Verlag, Berlin/Heidelberg., 1988.

1741 Jiang, H. and Feingold, G.: Effect of aerosol on warm convective clouds: Aerosol-
1742 cloud-surface flux feedbacks in a new coupled large eddy model, J. Geophys. Res.,
1743 111(D1), doi:10.1029/2005JD006138, 2006.

1744 Jiang, H., Feingold, G. and Koren, I.: Effect of aerosol on trade cumulus cloud
1745 morphology, J. Geophys. Res., 114(D11), doi:10.1029/2009JD011750, 2009.

1746 Jiang, H., Xue, H., Teller, A., Feingold, G. and Levin, Z.: Aerosol effects on the
1747 lifetime of shallow cumulus, Geophys. Res. Lett., 33(14),
1748 doi:10.1029/2006GL026024, 2006.

1749 Kaufman, Y. J., Koren, I., Remer, L. A., Rosenfeld, D. and Rudich, Y.: The effect of
1750 smoke, dust, and pollution aerosol on shallow cloud development over the Atlantic
1751 Ocean., Proc. Natl. Acad. Sci. USA, 102(32), 11207–11212,
1752 doi:10.1073/pnas.0505191102, 2005.

1753 Khain, A. P., BenMoshe, N. and Pokrovsky, A.: Factors Determining the Impact of
1754 Aerosols on Surface Precipitation from Clouds: An Attempt at Classification, J.
1755 Atmos. Sci., 65(6), 1721–1748, doi:10.1175/2007JAS2515.1, 2008.

1756 Khain, A., Pokrovsky, A., Pinsky, M., Seifert, A. and Phillips, V.: Simulation of
1757 Effects of Atmospheric Aerosols on Deep Turbulent Convective Clouds Using a
1758 Spectral Microphysics Mixed-Phase Cumulus Cloud Model. Part I: Model
1759 Description and Possible Applications, J. Atmos. Sci., 61(24), 2963–2982,
1760 doi:10.1175/JAS-3350.1, 2004.

- 1761 Khain, A., Rosenfeld, D. and Pokrovsky, A.: Aerosol impact on the dynamics and
 1762 microphysics of deep convective clouds, *Q.J Royal Met. Soc.*, 131(611), 2639–2663,
 1763 doi:10.1256/qj.04.62, 2005.
- 1764 Khairoutdinov, M. F. and Randall, D. A.: Cloud resolving modeling of the ARM
 1765 summer 1997 IOP: model formulation, results, uncertainties, and sensitivities, *J.*
 1766 *Atmos. Sci.*, 60(4), 607–625, doi:10.1175/1520-
 1767 0469(2003)060<0607:CRMOTA>2.0.CO;2, 2003.
- 1768 Knupp, K. R. and Cotton, W. R.: Convective cloud downdraft structure: An
 1769 interpretive survey, *Rev. Geophys.*, 23(2), 183, doi:10.1029/RG023i002p00183, 1985.
- 1770 Kogan, Y. L. and Martin, W. J.: Parameterization of bulk condensation in numerical
 1771 cloud models, *J. Atmos. Sci.*, 51(12), 1728–1739, doi:10.1175/1520-
 1772 0469(1994)051<1728:POBCIN>2.0.CO;2, 1994.
- 1773 Köhler, H.: The nucleus in and the growth of hygroscopic droplets, *Transactions of*
 1774 *the Faraday Society*, 32, 1152–1161, 1936.
- 1775 Koren, I., Altaratz, O. and Dagan, G.: Aerosol effect on the mobility of cloud
 1776 droplets, *Environmental Research Letters*, 10(10), 104011, doi:10.1088/1748-
 1777 9326/10/10/104011, 2015.
- 1778 Koren, I., Dagan, G. and Altaratz, O.: From aerosol-limited to invigoration of warm
 1779 convective clouds., *Science*, 344(6188), 1143–1146, doi:10.1126/science.1252595,
 1780 2014.
- 1781 L’Ecuyer, T. S., Berg, W., Haynes, J., Lebsock, M. and Takemura, T.: Global
 1782 observations of aerosol impacts on precipitation occurrence in warm maritime clouds,
 1783 *J. Geophys. Res.*, 114(D9), doi:10.1029/2008JD011273, 2009.
- 1784 Li, Z., Niu, F., Fan, J., Liu, Y., Rosenfeld, D. and Ding, Y.: Long-term impacts of
 1785 aerosols on the vertical development of clouds and precipitation, *Nat. Geosci.*, 4(12),
 1786 888–894, doi:10.1038/ngeo1313, 2011.
- 1787 Mason, B. J. and Chien, C. W.: Cloud-droplet growth by condensation in cumulus,
 1788 *Quarterly Journal of the Royal Meteorological Society*, 88(376), 136–142, 1962.
- 1789 Mordy, W.: Computations of the growth by condensation of a population of cloud
 1790 droplets, *Tellus*, 11(1), 16–44, doi:10.1111/j.2153-3490.1959.tb00003.x, 1959.
- 1791 Paluch, I. R. and Breed, D. W.: A Continental Storm with a Steady, Adiabatic Updraft
 1792 and High Concentrations of Small Ice Particles: 6 July 1976 Case Study, *J. Atmos.*
 1793 *Sci.*, 41(6), 1008–1024, doi:10.1175/1520-
 1794 0469(1984)041<1008:ACSWAS>2.0.CO;2, 1984.

1795 Pinsky, M., Mazin, I. P., Korolev, A. and Khain, A.: Supersaturation and diffusional
1796 droplet growth in liquid clouds, *J. Atmos. Sci.*, 70(9), 2778–2793, doi:10.1175/JAS-
1797 D-12-077.1, 2013.

1798 Reisin, T., Levin, Z. and Tzivion, S.: Rain Production in Convective Clouds As
1799 Simulated in an Axisymmetric Model with Detailed Microphysics. Part I: Description
1800 of the Model, *J. Atmos. Sci.*, 53(3), 497–519, doi:10.1175/1520-
1801 0469(1996)053<0497:RPICCA>2.0.CO;2, 1996.

1802 Reutter, P., Su, H., Trentmann, J., Simmel, M., Rose, D., Gunthe, S. S., Wernli, H.,
1803 Andreae, M. O. and Pöschl, U.: Aerosol- and updraft-limited regimes of cloud droplet
1804 formation: influence of particle number, size and hygroscopicity on the activation of
1805 cloud condensation nuclei (CCN), *Atmos. Chem. Phys.*, 9(18), 7067–7080,
1806 doi:10.5194/acp-9-7067-2009, 2009.

1807 Saleeby, S. M., Herbener, S. R., van den Heever, S. C. and L’Ecuyer, T.: Impacts of
1808 cloud droplet–nucleating aerosols on shallow tropical convection, *J. Atmos. Sci.*,
1809 72(4), 1369–1385, doi:10.1175/JAS-D-14-0153.1, 2015.

1810 Savane, O., Vant-Hull, B., Mahani, S. and Khanbilvardi, R.: Effects of aerosol on
1811 cloud liquid water path: statistical method a potential source for divergence in past
1812 observation based correlative studies, *Atmosphere*, 6(3), 273–298,
1813 doi:10.3390/atmos6030273, 2015.

1814 Seifert, A. and Heus, T.: Large-eddy simulation of organized precipitating trade wind
1815 cumulus clouds, *Atmos. Chem. Phys.*, 13(11), 5631–5645, doi:10.5194/acp-13-5631-
1816 2013, 2013.

1817 Seifert, A., Heus, T., Pincus, R. and Stevens, B.: Large-eddy simulation of the
1818 transient and near-equilibrium behavior of precipitating shallow convection, *J. Adv.*
1819 *Model. Earth Syst.*, 7(4), 1918–1937, doi:10.1002/2015MS000489, 2015.

1820 Seigel, R. B.: Shallow Cumulus Mixing and Subcloud-Layer Responses to Variations
1821 in Aerosol Loading, *J. Atmos. Sci.*, 71(7), 2581–2603, doi:10.1175/JAS-D-13-0352.1,
1822 2014.

1823 Seiki, T. and Nakajima, T.: Aerosol effects of the condensation process on a
1824 convective cloud simulation, *J. Atmos. Sci.*, 71(2), 833–853, doi:10.1175/JAS-D-12-
1825 0195.1, 2014.

1826 Sheffield, A. M., Saleeby, S. M. and van den Heever, S. C.: Aerosol-induced
1827 mechanisms for cumulus congestus growth, *J. Geophys. Res. Atmos.*, 120(17), 8941–
1828 8952, doi:10.1002/2015JD023743, 2015.

1829 Siebesma, A. P., Bretherton, C. S., Brown, A., Chlond, A., Cuxart, J., Duynkerke, P.
1830 G., Jiang, H., Khairoutdinov, M., Lewellen, D., Moeng, C.-H., Sanchez, E., Stevens,
1831 B. and Stevens, D. E.: A large eddy simulation intercomparison study of shallow

- 1832 cumulus convection, *J. Atmos. Sci.*, 60(10), 1201–1219, doi:10.1175/1520-
1833 0469(2003)60<1201:ALESIS>2.0.CO;2, 2003.
- 1834 Small, J. D., Chuang, P. Y., Feingold, G. and Jiang, H.: Can aerosol decrease cloud
1835 lifetime?, *Geophys. Res. Lett.*, 36(16), doi:10.1029/2009GL038888, 2009.
- 1836 Sorooshian, A., Feingold, G., Lebsock, M. D., Jiang, H. and Stephens, G. L.: On the
1837 precipitation susceptibility of clouds to aerosol perturbations, *Geophys. Res. Lett.*,
1838 36(13), doi:10.1029/2009GL038993, 2009.
- 1839 Squires, P.: The microstructure and colloidal stability of warm clouds, *Tellus*, 10(2),
1840 256–261, doi:10.1111/j.2153-3490.1958.tb02011.x, 1958.
- 1841 Stevens, B.: On the growth of layers of nonprecipitating cumulus convection, *J.*
1842 *Atmos. Sci.*, 64(8), 2916–2931, doi:10.1175/JAS3983.1, 2007.
- 1843 Stevens, B. and Feingold, G.: Untangling aerosol effects on clouds and precipitation
1844 in a buffered system., *Nature*, 461(7264), 607–613, doi:10.1038/nature08281, 2009.
- 1845 Storer, R. L. and van den Heever, S. C.: Microphysical processes evident in aerosol
1846 forcing of tropical deep convective clouds, *J. Atmos. Sci.*, 70(2), 430–446,
1847 doi:10.1175/JAS-D-12-076.1, 2013.
- 1848 Twomey, S.: The influence of pollution on the shortwave albedo of clouds, *J. Atmos.*
1849 *Sci.*, 34(7), 1149–1152, doi:10.1175/1520-0469(1977)034<1149:TIOPOT>2.0.CO;2,
1850 1977.
- 1851 Tzivion, S., Feingold, G. and Levin, Z.: An efficient numerical solution to the
1852 stochastic collection equation, *J. Atmos. Sci.*, 44(21), 3139–3149, doi:10.1175/1520-
1853 0469(1987)044<3139:AENSTT>2.0.CO;2, 1987.
- 1854 Wang, C.: A modeling study of the response of tropical deep convection to the
1855 increase of cloud condensation nuclei concentration: 1. Dynamics and microphysics,
1856 *J. Geophys. Res.*, 110(D21), doi:10.1029/2004JD005720, 2005.
- 1857 Warner, J.: A Reduction in Rainfall Associated with Smoke from Sugar-Cane Fires—
1858 An Inadvertent Weather Modification?, *J. Appl. Meteor.*, 7(2), 247–251,
1859 doi:10.1175/1520-0450(1968)007<0247:ARIRAW>2.0.CO;2, 1968.
- 1860 Wei, D., Blyth, A. M. and Raymond, D. J.: Buoyancy of convective clouds in TOGA
1861 COARE, *J. Atmos. Sci.*, 55(22), 3381–3391, doi:10.1175/1520-
1862 0469(1998)055<3381:BOCCIT>2.0.CO;2, 1998.
- 1863 Xu, K.-M. and Randall, D. A.: Updraft and downdraft statistics of simulated tropical
1864 and midlatitude cumulus convection, *J. Atmos. Sci.*, 58(13), 1630–1649,
1865 doi:10.1175/1520-0469(2001)058<1630:UADSOS>2.0.CO;2, 2001.

1866 Xue, H. and Feingold, G.: Large-Eddy Simulations of Trade Wind Cumuli:
 1867 Investigation of Aerosol Indirect Effects, *J. Atmos. Sci.*, 63(6), 1605–1622,
 1868 doi:10.1175/JAS3706.1, 2006.

1869 Xue, H., Feingold, G. and Stevens, B.: Aerosol effects on clouds, precipitation, and
 1870 the organization of shallow cumulus convection, *J. Atmos. Sci.*, 65(2), 392–406,
 1871 doi:10.1175/2007JAS2428.1, 2008.

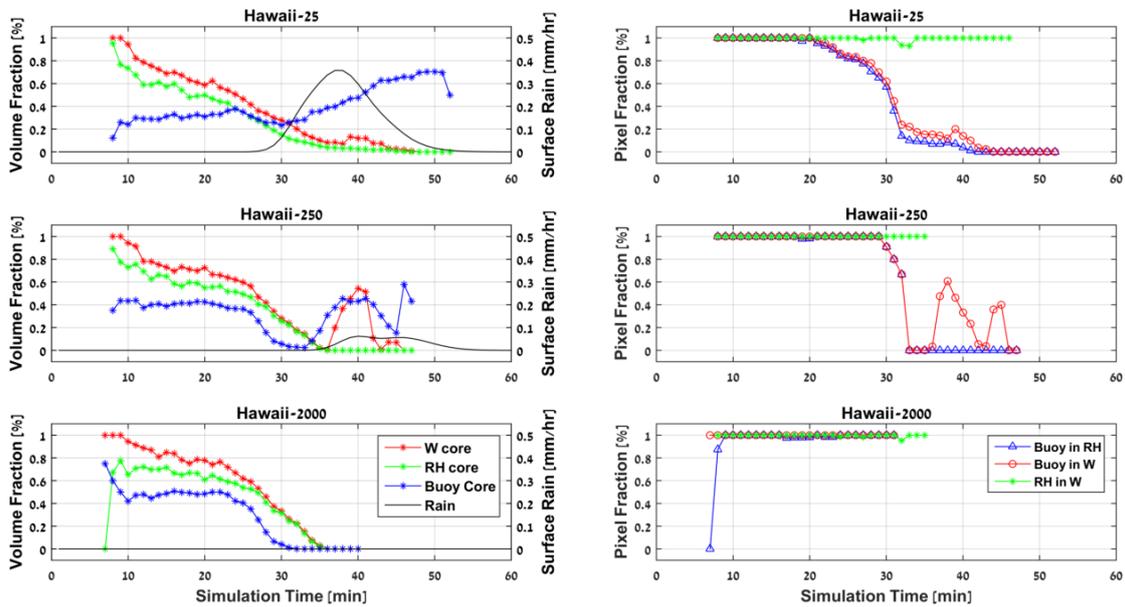
1872 Yuan, T., Remer, L. A. and Yu, H.: Microphysical, macrophysical and radiative
 1873 signatures of volcanic aerosols in trade wind cumulus observed by the A-Train,
 1874 *Atmos. Chem. Phys.*, 11(14), 7119–7132, doi:10.5194/acp-11-7119-2011, 2011.

1875 Zhao, M. and Austin, P. H.: Life cycle of numerically simulated shallow cumulus
 1876 clouds. part I: transport, *J. Atmos. Sci.*, 62(5), 1269–1290, doi:10.1175/JAS3414.1,
 1877 2005a.

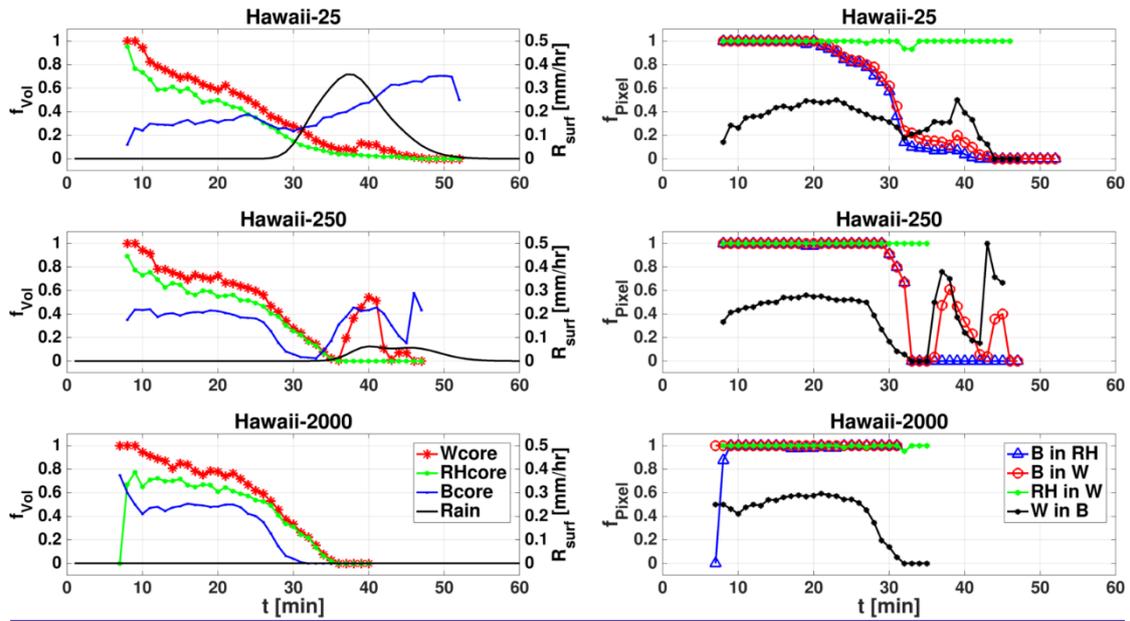
1878 Zhao, M. and Austin, P. H.: Life cycle of numerically simulated shallow cumulus
 1879 clouds. part II: mixing dynamics, *J. Atmos. Sci.*, 62(5), 1291–1310,
 1880 doi:10.1175/JAS3415.1, 2005b.

1881
 1882

1883 **Figures**



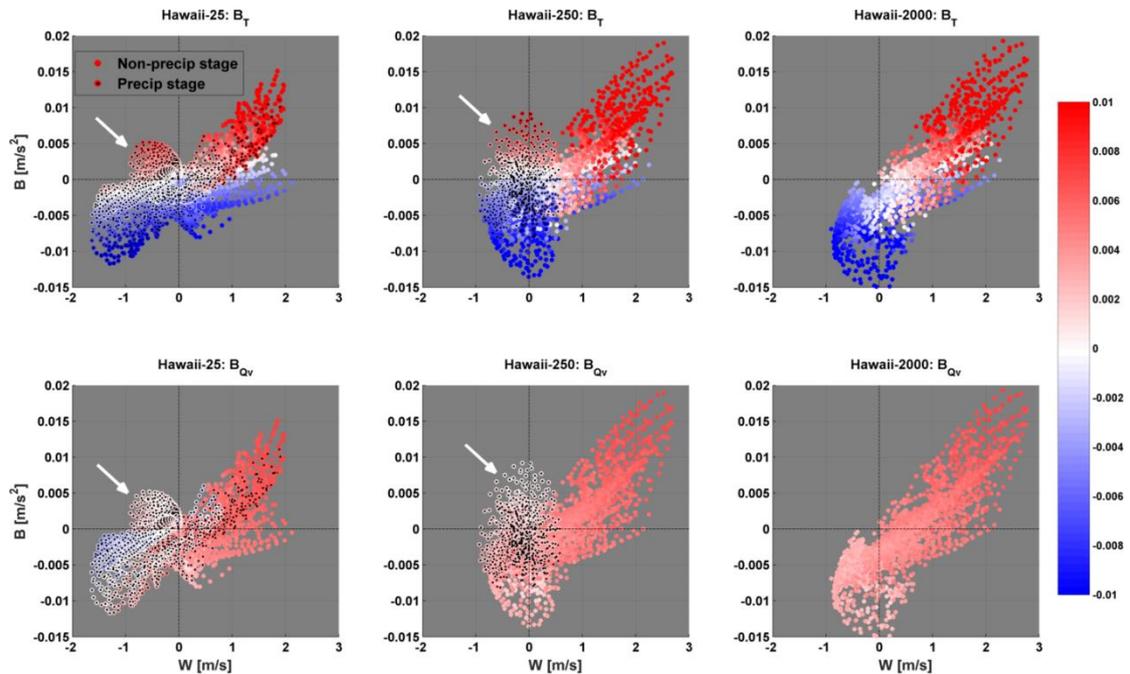
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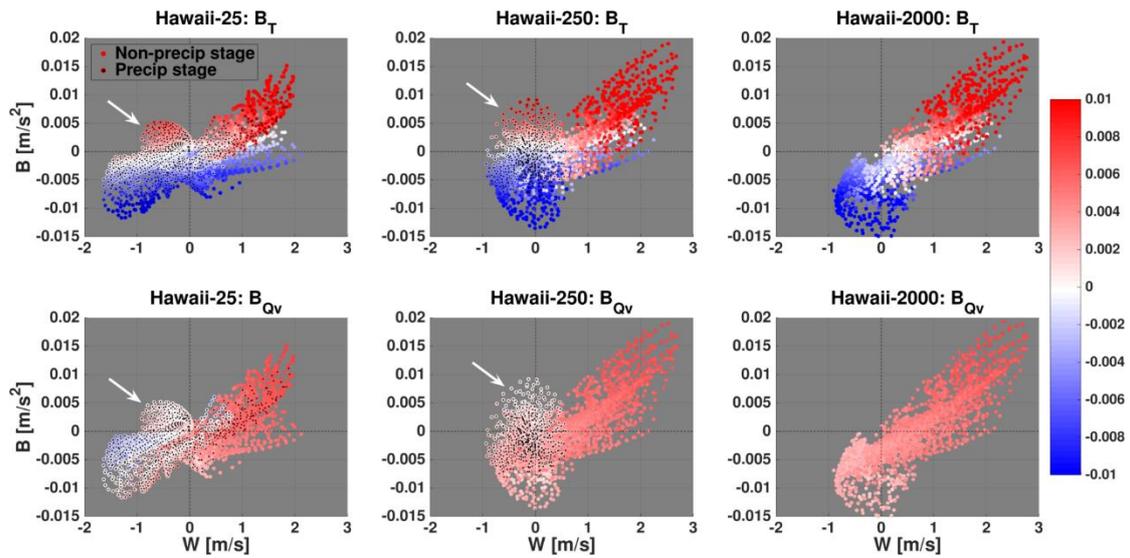
1885

1886 Figure 1. Left: Time series of core volume fractions (f_{vol} [%], LHS (left axis) and
 1887 surface rain-rate (R_{surf} [mm hr⁻¹], RHS (right axis) for the clean (top panel),
 1888 intermediate (middle panel), and polluted (bottom panel) single cloud simulations.
 1889 Right: Time series of core-pixel fractions (f_{pixel} [%]) of one core type within other core
 1890 types [%], another, for the respective simulation types. Core volume and pixel fractions
 1891 are indicated by different line colors (see legends).

1892



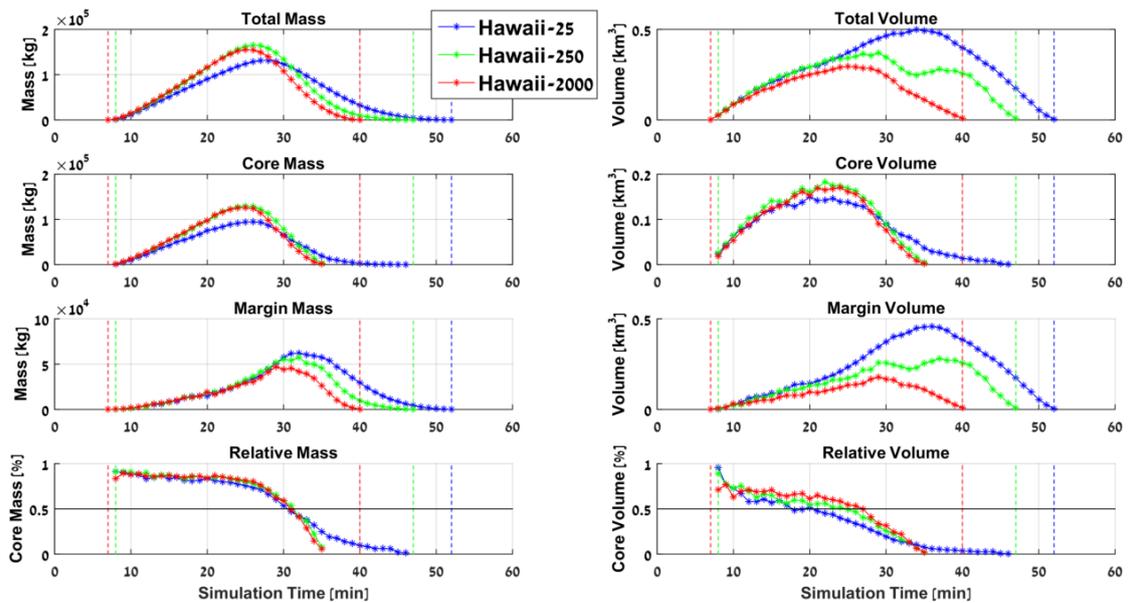
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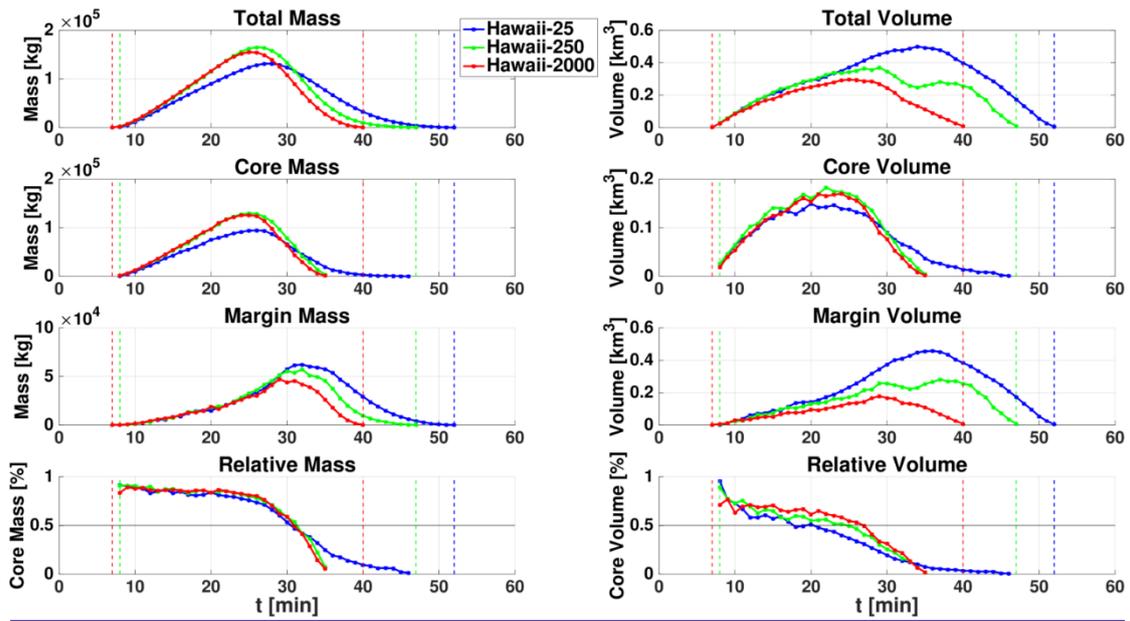
1894

1895 *Figure 2. Scatter plots of pixel total buoyancy [$m s^{-2}$] vs. pixel vertical velocity [$m s^{-1}$],*
 1896 *for the clean (left), intermediate (middle), and polluted (right) simulations. Data*
 1897 *includes all cloudy pixels during all time steps. Colors represent magnitude of*
 1898 *buoyancy temperature term (B_T , upper row) and humidity term (B_{Qv} , lower row), where*
 1899 *red (blue) shades indicate positive (negative) values. Markers with black dots*
 1900 *superimposed represent temporal stages with non-zero surface precipitation. White*
 1901 *arrows indicate outlier scatter of pixels with positive buoyancy and negative*
 1902 *velocity.*

1903



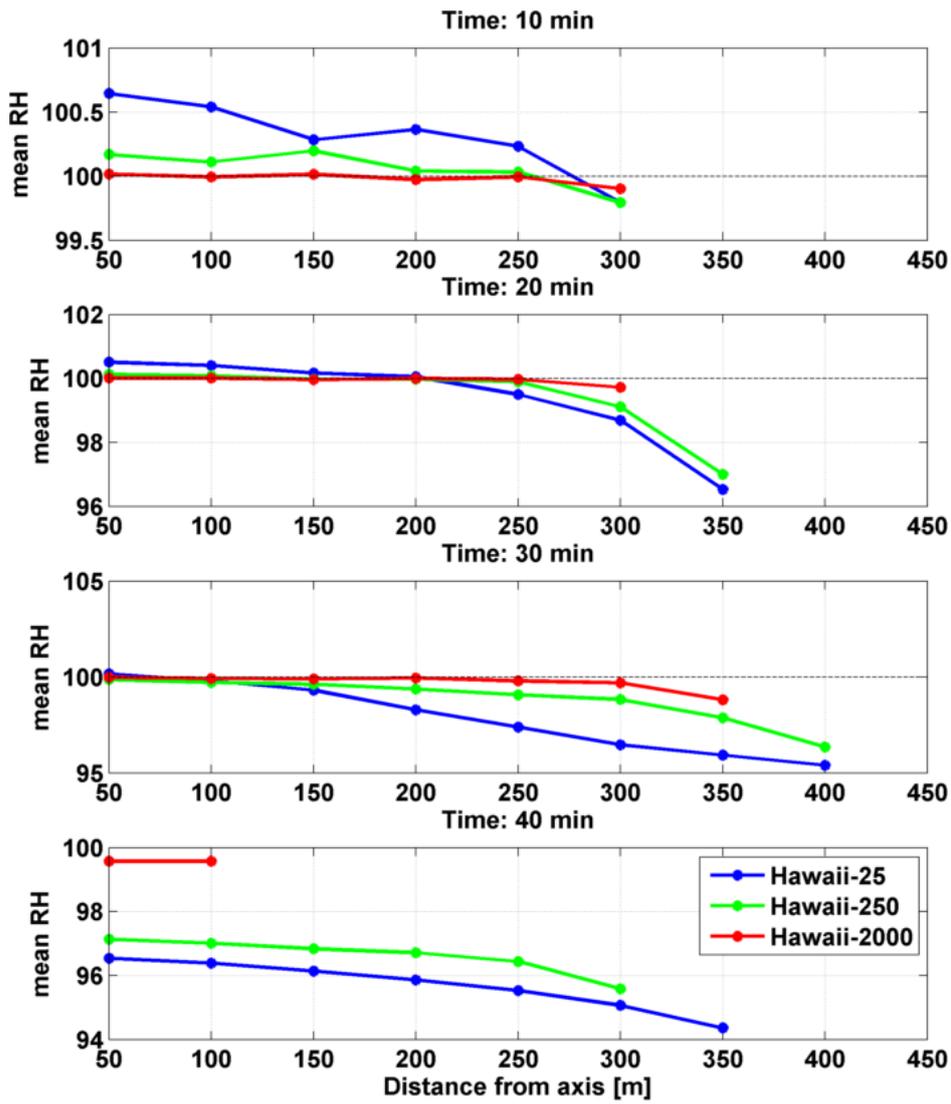
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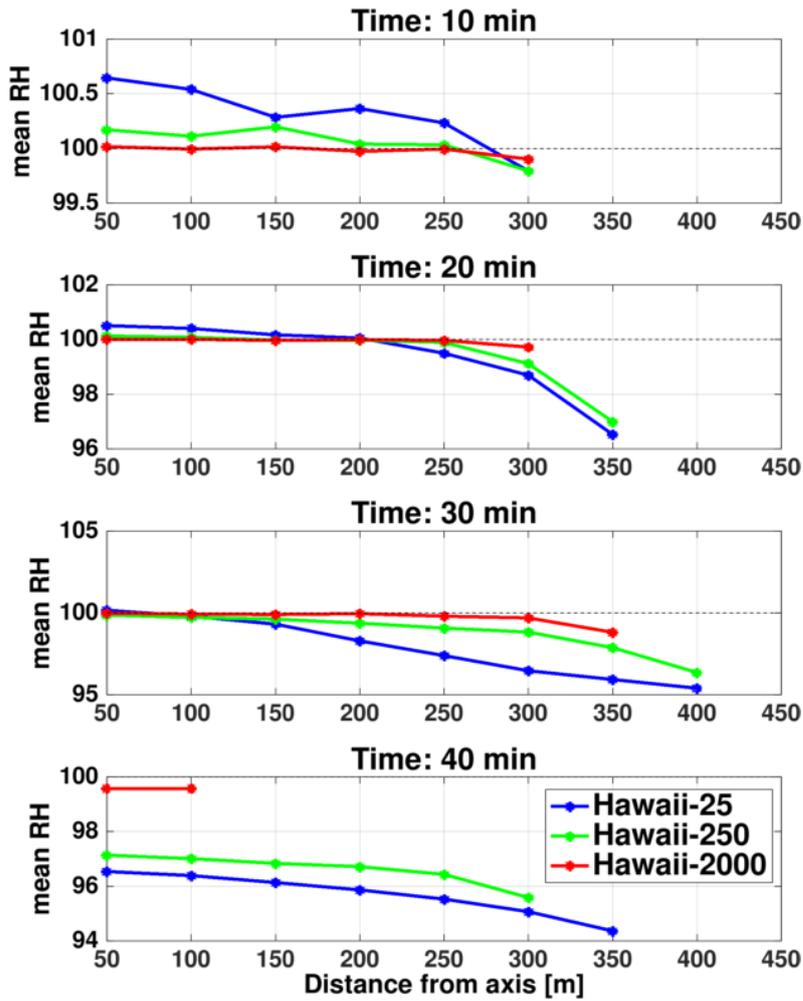
1905

1906 *Figure 3. Time series of cloud mass ([kg], left column) and cloud volume ([km³], right*
 1907 *column) for the different aerosol concentrations simulations (see legend). The total,*
 1908 *core, margin, and relative fraction values are shown for each parameter, as indicated*
 1909 *by panel titles. The core here is defined according to RH > 100% definition.*

1910



1911



1912

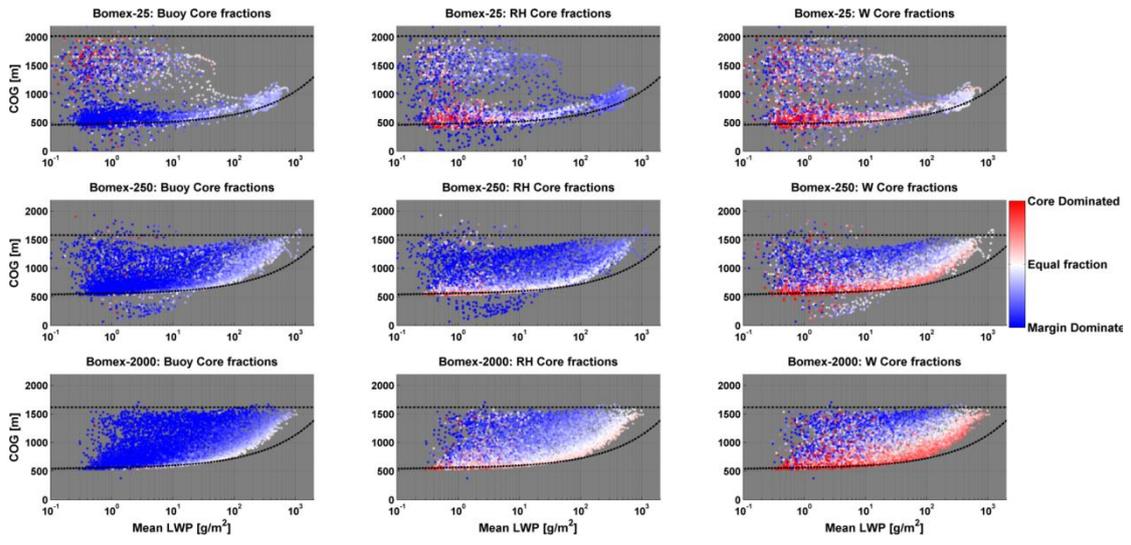
1913 *Figure 4. Four temporal snapshots (see panel titles for times) of RH [%] horizontal*
 1914 *cross-sections. Panels include the results of different aerosol concentrations (see*
 1915 *legend). Cross-sections are obtained by taking the mean RH of all vertical levels for*
 1916 *each horizontal distance from the cloud center axis.*

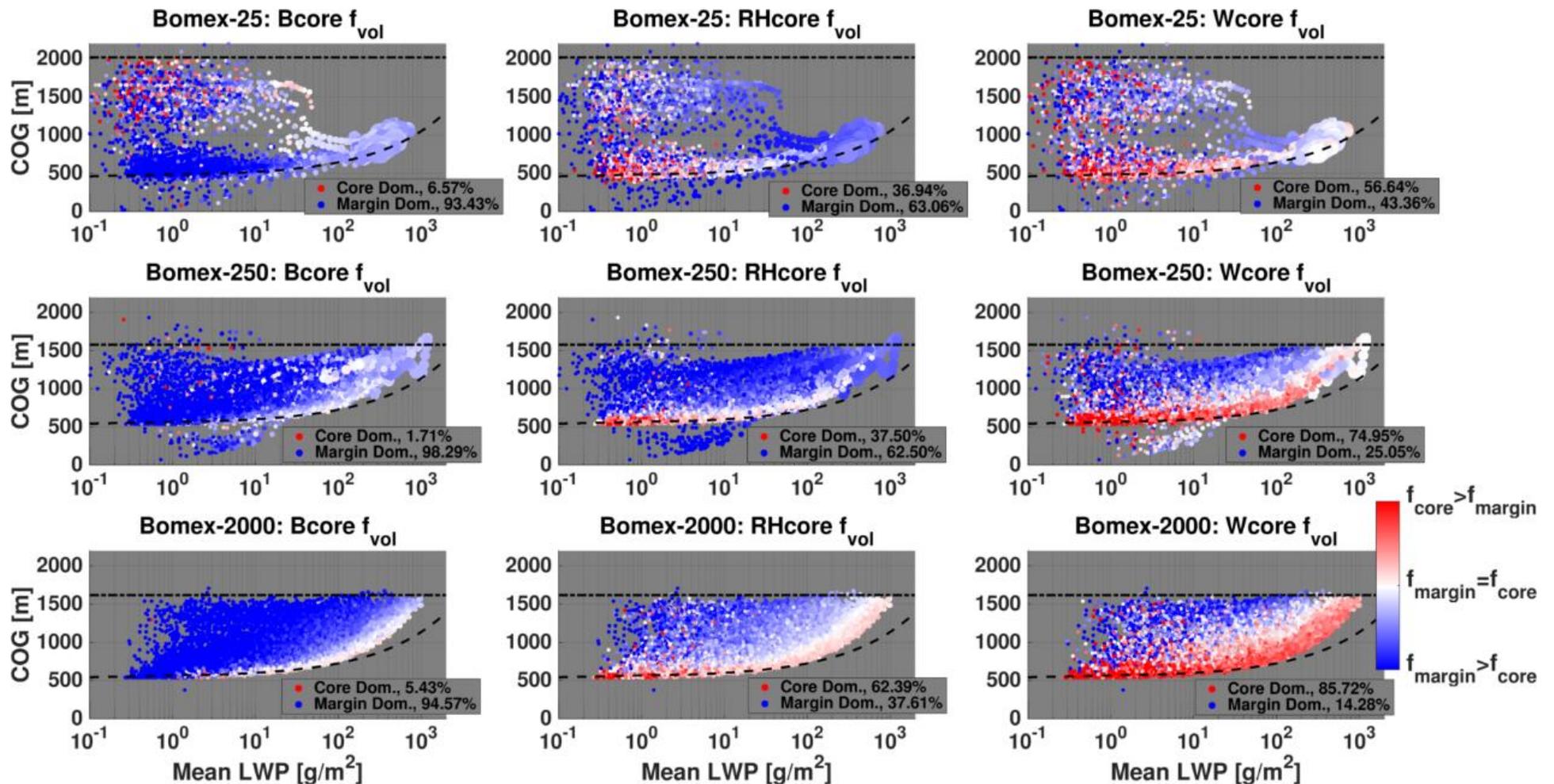
1917

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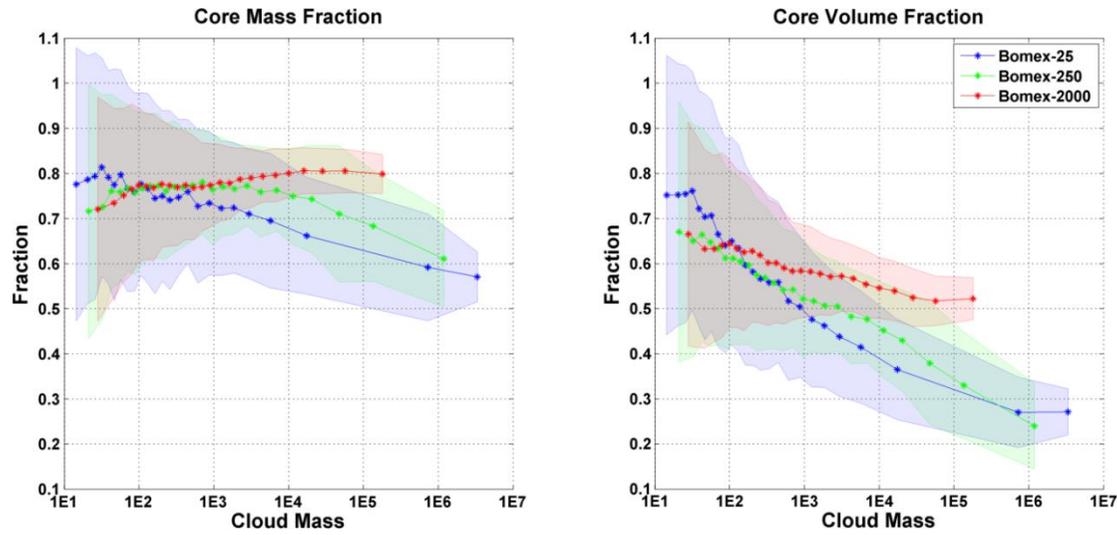




1921 Figure 5. CvM phase space diagrams of Bcore (left column), RHcore (middle column), and Wcore (right column) volume fractions (f_{vol}) for all
 1922 clouds between 3 h and 8 h in the BOMEX simulations. The upper, middle, and lower rows correspond to the clean, intermediate, and polluted
 1923 aerosol cases. -The red (blue) colors indicate a core volume fraction f_{vol} above (below) 0.5. The majority of clouds are confined size of each point
 1924 in the scatter is proportional to the region between the adiabatic cloud growth approximation (curved dashed line) and mean area, where the
 1925 inversion layer base height (horizontal dashed line). smallest (largest) point corresponds to an area of 0.01 (11.4) km^2 . The percentage of clouds

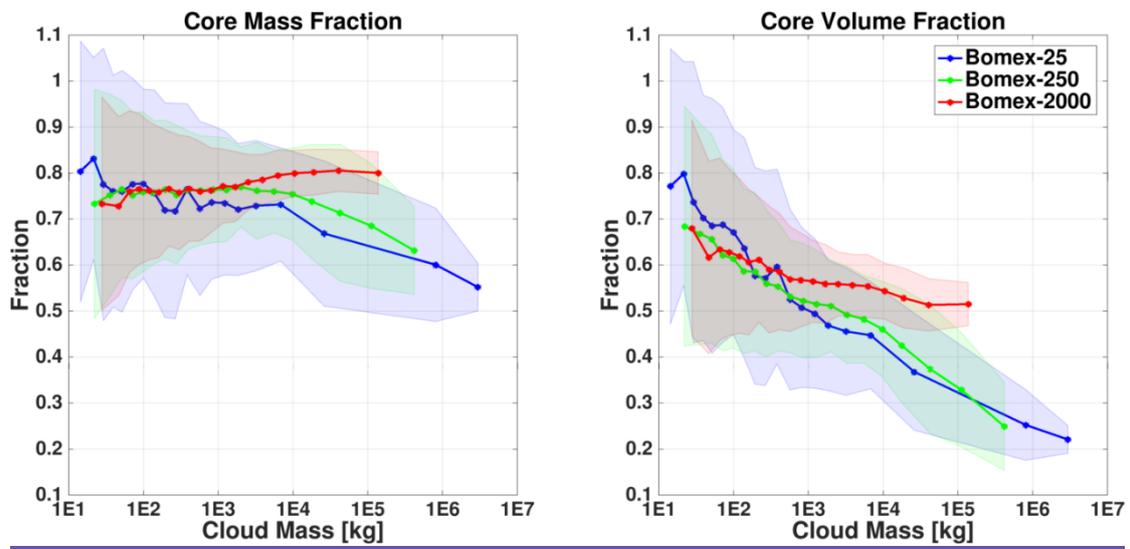
1926 *that are core dominated ($f_{vol} > 0.5$) is included in panel legends. For an in-depth description of CvM space characteristics, the reader is referred*
1927 *to Sect. 2.4 in PTI.*

1928



1929

1930

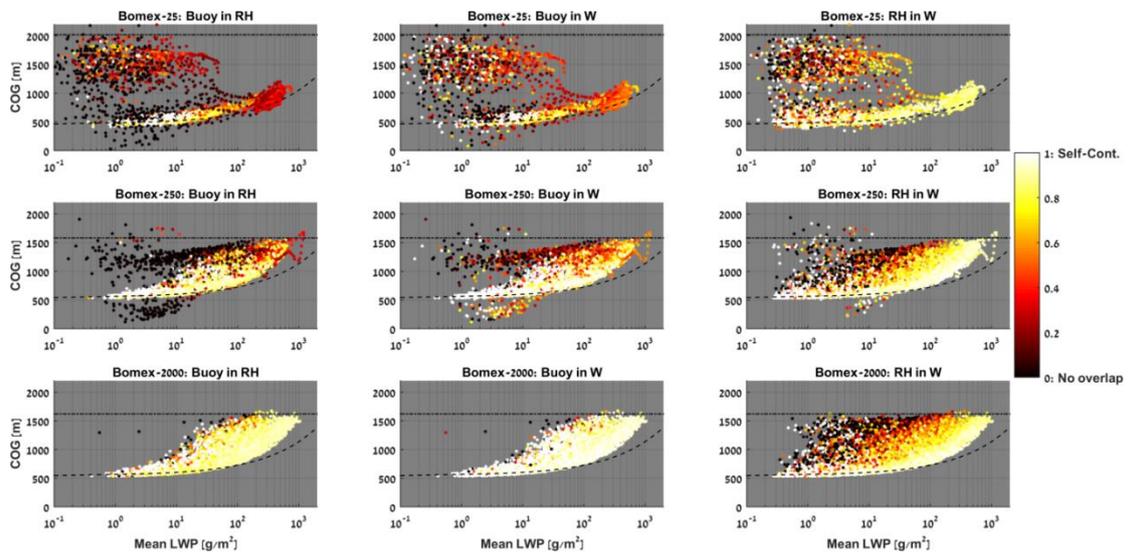


1931

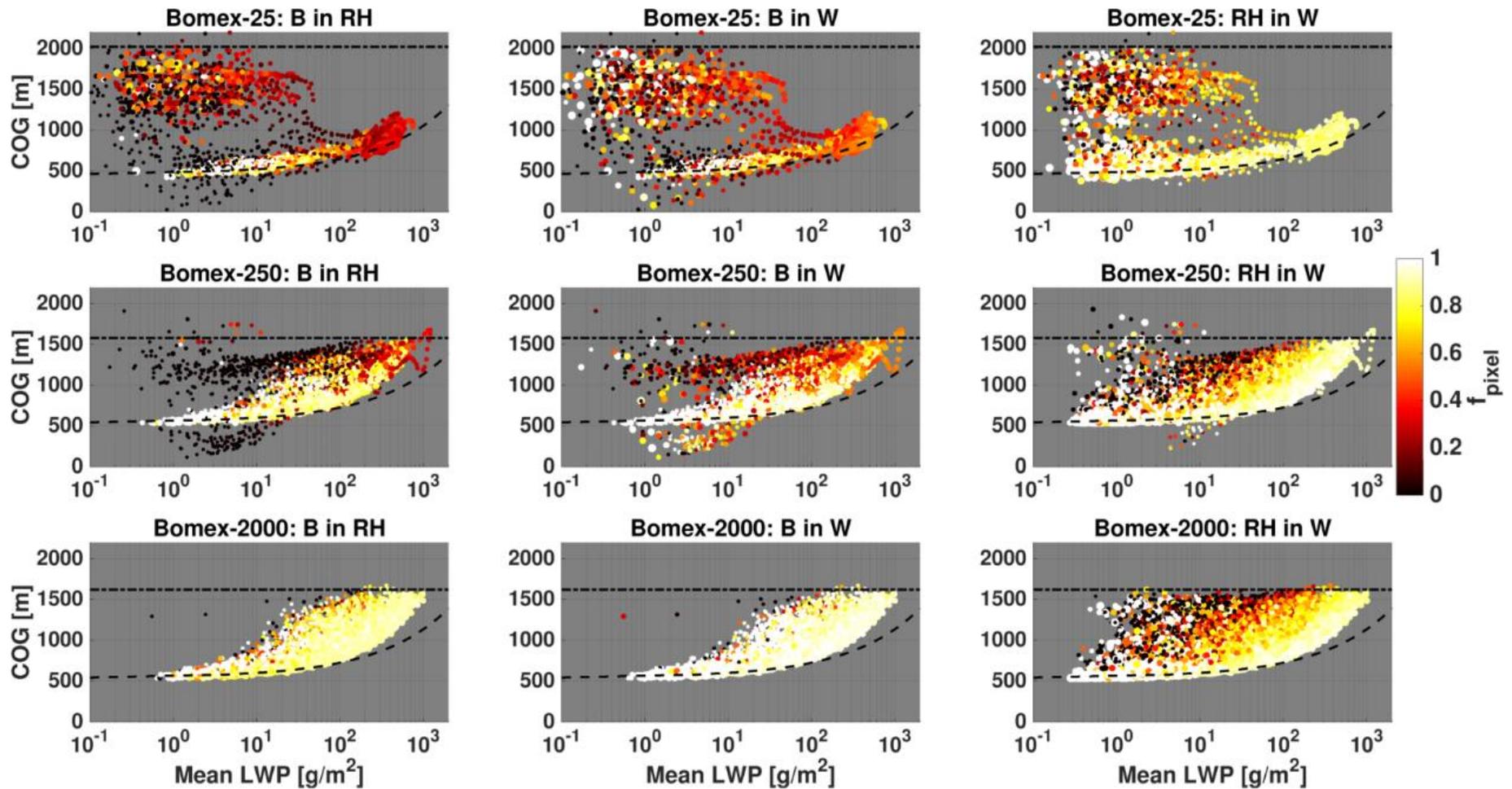
1932 *Figure 6. Average core mass fraction (left) and volume fraction (right) values for*
 1933 *different aerosol concentrations, as indicated in the legend. The average only includes*
 1934 *growing branch clouds from within the CvM space (i.e. clouds located in proximity to*
 1935 *the adiabat). The core here is defined according to $RH > 100\%$ definition.*

1936

1937



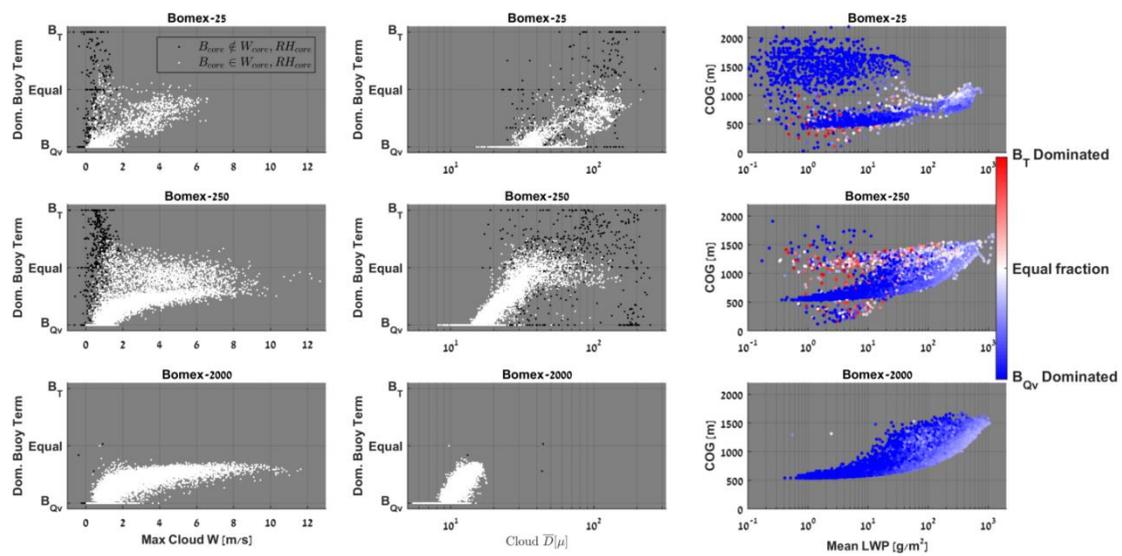
1938



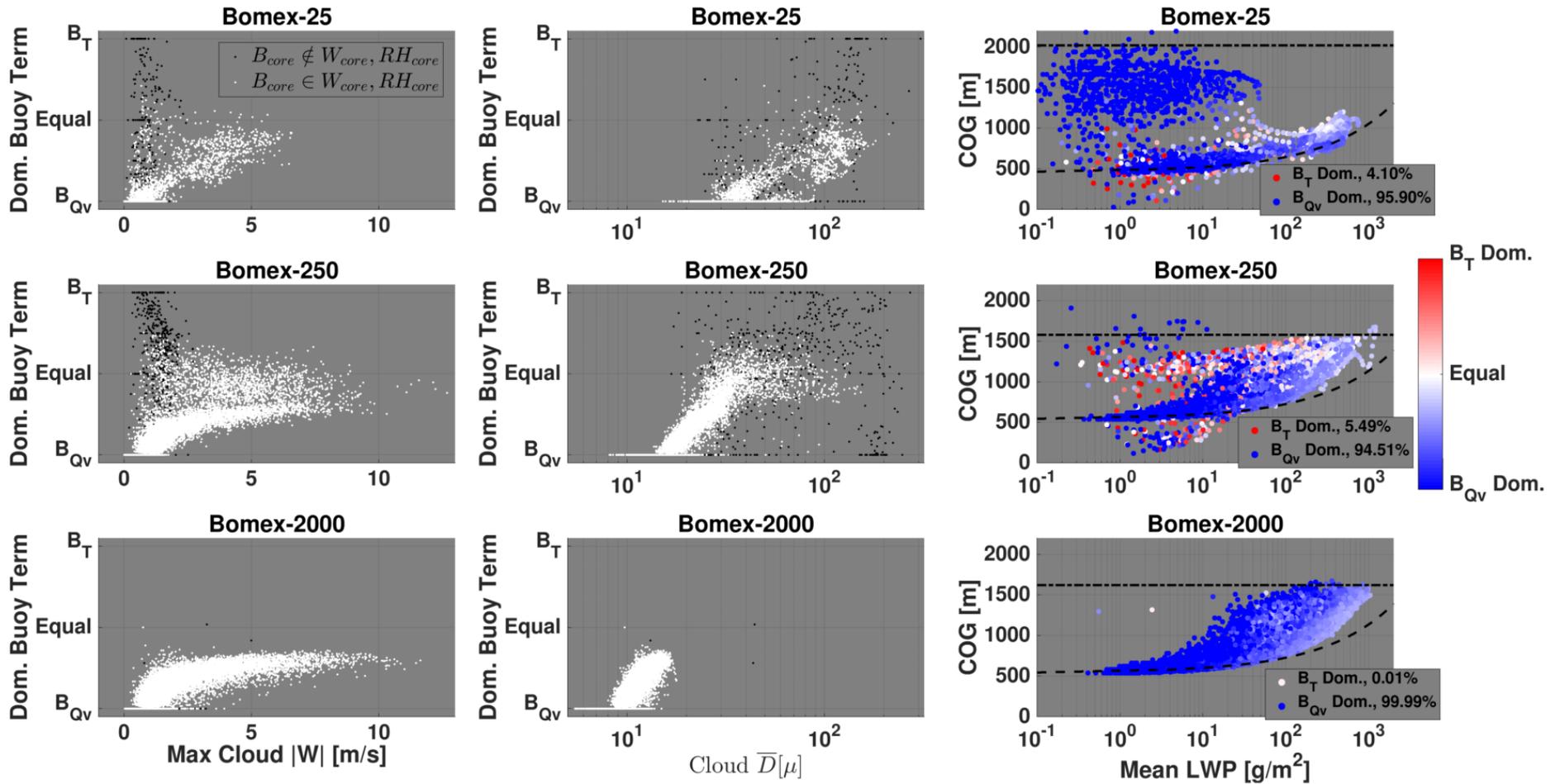
1939 Figure 7. CvM space diagrams showing the pixel fractions (f_{pixel}) of Bcore within RHcore (left column), Bcore within Wcore (middle column), and
 1940 RHcore within Wcore (right column), for the clean (top row), intermediate (middle row), and polluted (bottom row) simulations. Bright colors
 1941 indicate high pixel fractions (large overlap between two core types) while dark colors indicate low pixel fraction (little overlap between two core
 1942 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
 1943 above zero (for the core in question) are considered.

1944

1945



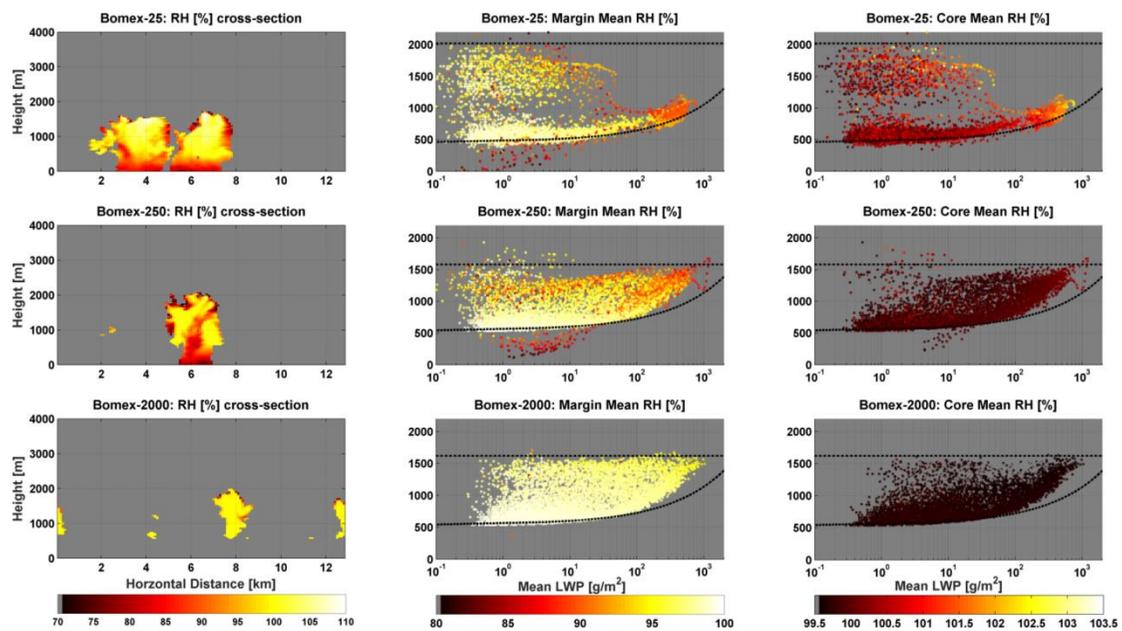
1946



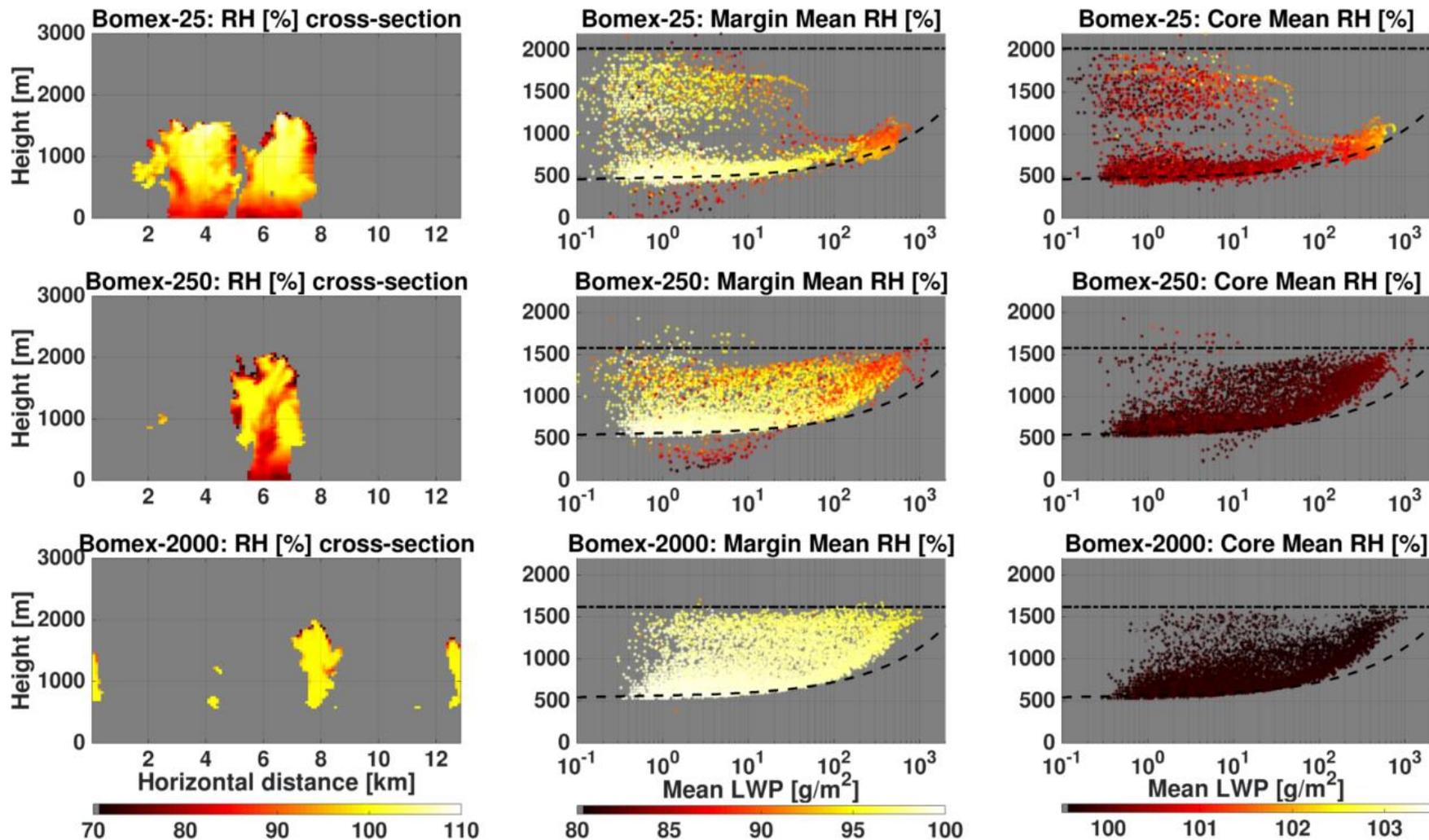
1947 Figure 8. Analysis of dominant buoyancy term within B_{core} of clouds (see text for details). As seen in previous figures, rows represent clean
 1948 (top), intermediate (middle), and polluted (bottom) simulations. Left column: dependence on maximum absolute vertical velocity within cloud.
 1949 Middle column: dependence on partition of total cloud mass to cloud droplets and rain drops. Right column: CvM space diagrams of all clouds
 1950 with B_{core} , where red (blue) shades indicate temperature (humidity) buoyancy terms dominate the cloud. Legends include percentage of
 1951 clouds that are B_T or B_{Qv} dominated (see text for explanation).

1952

1953



1954



1955 Figure 9. Left column – Relative Humidity (RH [%]) vertical cross-sections slicing through the center of gravity of the most massive cloud in each
 1956 simulation. Middle and right columns display CvM space diagrams of mean cloud margin RH and mean cloud core RH, respectively, using the
 1957 RH_{core} definition. The upper, middle, and lower panels correspond to the clean, intermediate, and polluted aerosol cases (see panel titles).
 1958 Notice the different color bar ranges for margin and core mean RH panels.