



36 **Abstract**

37 Flight data measured in warm convective clouds near Istanbul in June 2008 were used to  
38 investigate the relative dispersion of cloud droplet size distribution. The relative dispersion  
39 ( $\varepsilon$ ), defined as the ratio between the standard deviation ( $\sigma$ ) of the cloud droplet size  
40 distribution and cloud droplet average radius ( $\langle r \rangle$ ), is a key factor in regional and global  
41 models. The relationship between  $\varepsilon$  and the clouds' microphysical and thermodynamic  
42 characteristics is examined. The results show that  $\varepsilon$  is constrained with average values in the  
43 range of  $\sim 0.25$ – $0.35$ .  $\varepsilon$  is shown not to be correlated with cloud droplet concentration or  
44 liquid water content (LWC). However,  $\varepsilon$  variance is shown to be sensitive to droplet  
45 concentration and LWC, suggesting smaller variability of  $\varepsilon$  in the clouds' most adiabatic  
46 regions. A criterion for use of in-situ airborne measurement data for calculations of statistical  
47 moments (used in bulk microphysical schemes), based on the evaluation of  $\varepsilon$ , is suggested.

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51 **1. Introduction**

52 Droplet size distribution is one of the most important variables in the study of cloud  
53 physics. The size distribution properties are controlled by the thermodynamic conditions and  
54 by the microphysical and dynamic state of the cloud. Near the cloud base at the first stage of  
55 droplet formation, the size distribution is determined by the supersaturation (determined by  
56 the thermodynamic conditions and the updraft) and the aerosol properties. Higher in the  
57 cloud, at later stages of the cloud's development, additional processes, such as collision–  
58 coalescence, raindrop sedimentation, entrainment and mixing, further modify the drops' size  
59 distribution. On the other hand, the droplet size distribution determines the timing and  
60 magnitude of microphysical processes, which affect the cloud's dynamics through  
61 determination of terminal velocities, drag of the falling raindrops, and the release of latent  
62 heat.

63 The relative dispersion of cloud droplets ( $\epsilon$ ) is a parameter that represents droplet size  
64 distribution. It is defined as the ratio between the standard deviation ( $\sigma$ ) and the mean radius  
65 ( $\langle r \rangle$ ) of the clouds' droplet distribution. Both  $\sigma$  and  $\langle r \rangle$  are key variables used in various  
66 parameterization schemes, such as reflectivity of clouds (Hansen and Travis, 1974; Slingo,  
67 1989; Liu and Daum, 2000a, b; Daum and Liu, 2003) and autoconversion processes (e.g.,  
68 Liu et al., 2005, 2006a; Hsieh et al., 2009). However, instead of using both  $\sigma$  and  $\langle r \rangle$ , their  
69 ratio (i.e. the relative dispersion,  $\epsilon$ ) is often used. This is done in atmospheric models that  
70 span a wide scale from cloud resolution (CRMs) to global climate models (GCMs). These  
71 models are used to explore aerosol effects on clouds, such as the first and second indirect  
72 aerosol effects: the first effect links higher aerosol loading to the formation of numerous but  
73 smaller cloud droplets and higher cloud reflectivity (Twomey, 1977), and the second effect  
74 links the increase in aerosol loading with an increase in cloud lifetime (Albrecht, 1989).  
75 Another effect that can be potentially explored using the relative dispersion is the convective  
76 cloud invigoration effect (Koren et al., 2005, Andreae et al., 2004, Tao et al., 2012, Altaratz  
77 et al., 2014).

78 Considering the importance of  $\epsilon$ , many studies have been conducted to analyze the  
79 sensitivity of this parameter to environmental conditions and to key microphysical and  
80 thermodynamic cloud properties. This has been done in stratiform clouds (Peng and  
81 Lohmann, 2003; Rotstajn and Liu, 2003; Peng et al., 2007; Miles et al., 2000; Martin et al.  
82 1994; Ma et al., 2010; Lu and Seinfeld, 2006; Pawlowska et al., 2006). Fewer studies have  
83 examined this parameter for convective clouds, and the reported results are quite diverse. For

84 example, Lu et al. (2008) and Berg et al. (2011) analyzed airborne measurements of shallow  
85 cumuli under various levels of anthropogenic pollution and found an average  $\varepsilon$  of around  
86 0.3. In Berg et al. (2012), the pollution levels were assessed using CO concentrations (up to  
87 170 ppbv) and in Lu et al. (2008), the highest accumulation mode aerosol concentration was  
88  $1,650 \text{ cm}^{-3}$ . Zhao et al. (2006) analyzed data collected in 135 flights in different  
89 environments and found that  $\varepsilon$  values tend to converge to a range of  $\sim 0.4$  to  $0.5$  for droplet  
90 concentrations ( $N_c$ ) higher than  $50 \text{ cm}^{-3}$ . Deng et al. (2009) also indicated similar  
91 convergence of  $\varepsilon$  with  $N_c$ . Martins and Silva Dias (2009) studied cumulus clouds in the  
92 Amazonian dry season and found  $\varepsilon$  values in the range of 0.38 to 0.59.

93 Some studies have examined the sensitivity of  $\varepsilon$  to aerosol loading. By compiling  
94 measured data from different field studies, including warm cumulus clouds, Liu and Daum  
95 (2002) suggested that  $\varepsilon$  is sensitive to and positively correlated with aerosol loading at the  
96 cloud base. This conclusion was subsequently supported by modeling (Yum and Hudson,  
97 2005), observational data (McFarquhar and Heymsfield 2001) and theoretical studies (Liu et  
98 al., 2006b). Other investigations using observational data (Martins and Silva Dias, 2009;  
99 Hsieh et al., 2009) have suggested a negative relationship between  $\varepsilon$  and aerosol loading.

100  $\varepsilon$  is influenced by other factors as well. Yum and Hudson (2005) and Liu et al. (2006b)  
101 studied the combined effect of updraft values and aerosol loading (as they both determine  
102 supersaturation values) on  $\varepsilon$  using adiabatic condensational growth theory. These two factors  
103 are mainly influential at early stages of cloud development, before the processes of  
104 collection, sedimentation, entrainment and mixing become dominant. They found opposite  
105 effects: an increase in  $\varepsilon$  with the increase in aerosol loading and a decrease in  $\varepsilon$  with the  
106 increase in updraft velocity. They suggested that continental clouds have smaller  $\langle r \rangle$  and  
107 therefore, larger  $\varepsilon$ .

108 The chemical composition of aerosols is another influential factor that should be taken  
109 into account when studying  $\varepsilon$ . It determines the activation process and growth by  
110 condensation, and therefore affects the spectral distribution of the drops and the behavior of  
111  $\varepsilon$  (Martins and Silva Dias, 2009).

112 Zhao et al. (2006) and Liu et al. (2008) indicated that the so-called dispersion effect (higher  $\varepsilon$   
113 in a high aerosol loading environment) may account, at least in part, for the discrepancies  
114 between different estimations of the first indirect effect in different studies (Feingold et al.,  
115 2003; Rosenfeld and Feingold, 2003). This effect is linked to the impact of  $\varepsilon$  on the  
116 calculated effective radius. According to the dispersion effect (Liu and Daum, 2002)  $\varepsilon$  is

117 positively correlated with the aerosol loading, resulting in a larger effective radius and lower  
118 cloud reflectivity (Slingo, 1989), which can reduce the first indirect effect (e.g., Feingold et  
119 al., 2003; Rosenfeld and Feingold, 2003). Therefore the impact of aerosol loading on  $\epsilon$   
120 should be well understood for enabling suitable use of climate models in quantifying the  
121 impact of aerosol loading on the droplet size distribution, effective radius, and clouds'  
122 reflectivity.

123 Xie et al. (2013) studied four types of parameterizations for treating the relationship  
124 between  $N_c$  and  $\epsilon$ . They implemented these schemes into the Weather Research and  
125 Forecasting (WRF) model, aimed at studying the effects of aerosol on cloud microphysics  
126 and ground precipitation. They concluded that the  $N_c$ - $\epsilon$  relationship (positive or negative  
127 change of  $\epsilon$  with  $N_c$ ) influences the autoconversion process (i.e conversion of cloud droplets  
128 to raindrops), and therefore affects the response of ground precipitation to a change in  
129 aerosols. Xie et al. (2013) suggested that for a positive  $N_c$ - $\epsilon$  relationship, the large-sized rain  
130 drops at high aerosol concentrations enhance the efficiency of the surface precipitation. The  
131 diversity of schemes for the  $N_c$ - $\epsilon$  relationship (as shown in Fig. 1 of Xie et al., 2013)  
132 suggests that much more research is needed to understand the physics behind the properties  
133 of  $\epsilon$ .

134 Recently, Tas et al. (2012) monitored the response of  $\epsilon$  in warm cumulus clouds to  
135 changes in thermodynamic conditions and aerosol loading, per cloud evolutionary stage. In  
136 that work, the cloud lifetime was divided into three stages based on the dominant  
137 microphysical processes. Using a detailed microphysical model, a different pattern of  $\epsilon$  was  
138 shown for each stage. Their results indicated that  $\epsilon$  has a narrow range around  $\sim 0.25$ – $0.35$   
139 during the mature stage of the cloud's lifetime (defined as the stage when the total water  
140 mass is around its maximum with only minor changes). They claimed that trends in  $\epsilon$  can be  
141 explained by the balance between the two main growth processes that dictate the droplet size  
142 distribution, condensation and collision–coalescence (before the initiation of significant  
143 rain). At the mature stage, the relative importance of the collision–coalescence-induced  
144 growth slowly increases, such that  $\epsilon$  growth is relatively slow.

145 In this study, we use detailed airborne measurements carried out near Istanbul, Turkey in  
146 June 2008, to explore  $\epsilon$  in non-precipitating continental convective clouds under various  
147 conditions of aerosol loading.

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## 150 2. Measurements and instrumentation

151 The 2007–2008 Cloud and Aerosol Research in Istanbul (CARI) project was aimed at  
152 exploring cloud and precipitation characteristics as a feasibility study for cloud-seeding  
153 operations in the area of Istanbul (Teller et al., 2008). A Piper Cheyenne II research aircraft  
154 (see Axisa et al., 2005 for details of the aircraft) was equipped with a Droplet Measurement  
155 Technologies (DMT) cloud droplet probe (CDP) to measure the concentration and size  
156 distribution of cloud droplets in the radius range of 1.5–25  $\mu\text{m}$ . In addition, aerosol  
157 concentrations and size distributions in the radius range of 0.055–1.5  $\mu\text{m}$  were measured  
158 using a DMT passive cavity aerosol spectrometer probe (PCASP, SPP200). This work  
159 focuses on the measurements that were carried out on 6–7 Jun 2008.

160 Each flight focused on one single cloud with penetrations at different altitudes (the  
161 aircraft ascended or descended at height steps of approximately 150 m). As can be inferred  
162 from Fig. 1c, the duration of each penetration was about 15–25 s, corresponding to  
163 horizontal flight distances of approximately 1–2 km (the aircraft speed was 70–90  $\text{m s}^{-1}$   
164 depending on the wind speed and direction). The information about cloud top height  
165 presented in this paper is based on verification that no cloudy region was present above a  
166 specific height. This was done by visual inspection of the visibility around the aircraft,  
167 combined with the measured cloud droplet concentration and LWC above this height. Cloud  
168 top height was set as the highest altitude for which measured cloud droplet concentration and  
169 LWC were higher than 10  $\text{cm}^{-3}$  and 0.01  $\text{g kg}^{-1}$ , respectively, in agreement with the criteria  
170 of Deng et al. (2009) for the determination of a cloudy region.

171 A shallow frontal system passed over the area of Istanbul on the night of 6 Jun 2008,  
172 bringing some rain showers to the area. Figure 1a shows an image of the Eastern  
173 Mediterranean region, taken by the Moderate Resolution Imaging Spectroradiometer  
174 (MODIS) sensor, on 7 Jun 2008, showing the area west of Istanbul after passage of the front.  
175 The airborne measurements in five warm cumulus clouds were conducted before (clouds  
176 TRK1 and 2) and after (TRK3, 4 and 5) the passage of the front (see the flight tracks in Fig.  
177 1b). There was a slight decrease in temperature after the passage of the front (this can be  
178 seen in the minor differences between the temperature levels of TRK1, 2 compared to those  
179 of TRK3, 4, 5 in Table 1). Such measurements provide a unique opportunity to study the  
180 relationships between relative dispersion ( $\epsilon$ ) and different cloud properties (e.g. height above  
181 the cloud base, LWC,  $N_c$ ).

182 Table 1 presents some details about each measured cloud: the time of the measurement,  
183 the top and base height levels and the corresponding temperature and pressure levels, and

184 aerosol loading. Further details are provided below.

185 Flights of 6 Jun 2008: On this day, the research aircraft conducted two flights to measure  
186 two cumulus clouds that developed west of the urban area of Istanbul. These clouds are  
187 referred to as TRK1 and TRK2 (see Fig. 1). The thickness of the two clouds was around  
188 2,000 m, with a cloud base temperature of about 15°C and cloud top temperature between 0  
189 and -2°C. Cloud imaging probe (CIP) measurements carried out onboard the aircraft showed  
190 that the clouds did not contain ice hydrometeors.

191 Flights of 7 Jun 2008: Three flights were conducted on this day in three cumulus clouds,  
192 west of Istanbul—TRK3, TRK4 and TRK5 (see Fig. 1). The environmental conditions of  
193 clouds TRK3 and TRK4 and their physical sizes were quite similar to TRK1 and TRK2.  
194 TRK5 was a shallower cloud of only 1,000 m depth and a cloud top temperature of 8°C.

195 To ensure statistically significant results, we analyzed only cloud measurements with  
196 droplet concentrations larger than 10 cm<sup>-3</sup> and LWC > 0.01 g kg<sup>-1</sup> as in Deng et al. (2009).  
197 Sensitivity tests revealed that the results do not change significantly by applying other  
198 threshold values to define the clouds' boundaries, within a droplet concentration range of 5–  
199 50 cm<sup>-3</sup>, or LWC range of 0.001–0.1 g kg<sup>-1</sup>. Analyses were performed for two types of cloud  
200 regions: (i) inner cloud and (ii) cloud boundary.

201 The more adiabatic, inner cloud dataset required that not only the sampling point itself,  
202 but also two neighboring sampling points (representing in total 2 s or ~140 m) all be  
203 associated with measured concentrations higher than 10 cm<sup>-3</sup> and LWC higher than 0.01 g  
204 kg<sup>-1</sup>. The cloud boundaries were defined as those data intervals that met the condition of  
205 concentration > 10 cm<sup>-3</sup> and LWC > 0.01 g kg<sup>-1</sup>, but one of their neighboring points did not  
206 meet this criterion.

207

### 208 **3. Results**

209 Fig. 1c shows some differences between the clouds that were investigated on June 6<sup>th</sup> and 7<sup>th</sup>.  
210 The measurements of TRK1 and TRK2 (6 Jun 2008) show an increase and then a decrease in  
211 the maximum total cloud droplet concentration as a function of altitude (each penetration  
212 was 15–25 s, corresponding to a flight distance of 1–2 km). A maximum of 1,650 (1,400)  
213 cm<sup>-3</sup> was measured at the cloud base and 1,100 (700) cm<sup>-3</sup> at the cloud top in cloud TRK1  
214 (TRK2). The ambient aerosol concentration in the diameter range of 0.11–3 μm below the  
215 cloud base was about 1,800 cm<sup>-3</sup> in the case of cloud TRK1 and 900 cm<sup>-3</sup> for cloud TRK2.

216 The cloud droplet concentrations measured on 7 Jun 2008 were smaller than those  
217 measured on June 6<sup>th</sup>, and most of the penetrations were smaller than 1,000 cm<sup>-3</sup> (see Figs. 1

218 and 2). The average aerosol concentration for the morning flight (TRK3) was  $700 \text{ cm}^{-3}$ ,  
219 whereas for the afternoon flights (TRK4 and TRK5) it increased to  $1,000 \text{ cm}^{-3}$ .

220 Fig. 2 shows the average droplet size distribution per height level. The height above  
221 ground level is binned into 10 intervals. The mean droplet radius ( $\langle r \rangle$ ) and the standard  
222 deviation ( $\sigma$ ) at each height interval is shown by the yellow and red lines, respectively. Note  
223 that due to instrumental limitations of the CDP, the maximal measured drop size was equal  
224 to a radius of  $\sim 25 \mu\text{m}$ .

225 Such representation of the size distribution (see Fig. 2) allowed us to investigate the  
226 impact of the instrumental limitation on our analysis. It revealed that in all cases except  
227 TRK3, the upper limit cut-off droplet size was below  $25 \mu\text{m}$ , implying that no bigger  
228 droplets were present. TRK3, having a lower aerosol loading (see Table 1), had a lower  
229 cloud droplet concentration (see Fig. 3b) and a broader size distribution (Fig. 2). Thus,  
230 although a contribution from larger droplets was expected, it could not be included in the  
231 analysis. This limitation suggests that calculations of moments of the size distribution might  
232 be biased in the case of TRK3.

233 Figure 2 reveals additional information about the sampled clouds: (i) cases TRK1 and  
234 TRK2 had larger droplet concentrations compared to the other clouds. In these cases,  $\langle r \rangle$   
235 was  $\sim 5\text{--}6 \mu\text{m}$  at the cloud base and  $8\text{--}9 \mu\text{m}$  near the cloud top. As case TRK3 had the lowest  
236 droplet concentration, the average radius at the cloud base was  $6.5 \mu\text{m}$  and increased to  
237 about  $8.5 \mu\text{m}$  close to the cloud top; (ii) in cases TRK4 and TRK5, the width of the droplet  
238 size distributions decreased with altitude. In general, near the cloud tops there was a decrease  
239 in droplet concentration; (iii) TRK5 had the lowest LWC value (below  $1.5 \text{ g kg}^{-1}$ ).

240 As can be seen in Fig. 2, the changes in  $\sigma$  and  $\langle r \rangle$  as a function of height above the cloud  
241 base (see the red and yellow lines in the figure) were similar for all clouds except cloud  
242 TRK3. This observation suggests that, except for TRK3, the relative dispersion value ( $\varepsilon =$   
243  $\sigma/\langle r \rangle$ ) is not sensitive to the vertical height above the cloud base. The reason for the  
244 exception in case TRK3 is discussed in Sect. 4.

245 Figure 3 shows the relationship of the relative dispersion with height (Fig. 3a) and LWC  
246 (Fig. 3b). In Fig. 3a, the colors of the points for the average  $\varepsilon$  represent the average LWC,  
247 and it can be seen that the average relative dispersion changes very little as a function of  
248 height. In Fig. 3b, the colors of the points for the average  $\varepsilon$  represent the droplet  
249 concentrations. The black lines in both figure panels represent the average values of  $\varepsilon$ ,  
250 obtained for each of the 10 different bins, and sorted in the figure according to height (Fig.

251 3a) or LWC (Fig. 3b). The error bars represent the 95% confidence interval for the mean  $\epsilon$ .  
252 While it is clear that on average for each flight, the droplet concentration increases with  
253 LWC (see colors of the average  $\epsilon$  points), the average relative dispersion falls into a narrow  
254 range and does not depend on LWC. Figure 4 is similar to Fig. 3, but is based only on  
255 measurements in the cloud boundaries where LWC and  $N_c$  are below the threshold values of  
256  $0.01 \text{ g kg}^{-1}$  and  $10 \text{ cm}^{-3}$ , respectively. Figures 3 and 4 demonstrate that for both the inner  
257 cloud and its boundaries, the droplet concentration increases with LWC, while the average  
258 relative dispersion remains almost constant. Moreover, apart from differences in the total  
259 number of data points, the results near the cloud boundary (entrainment zones) are similar to  
260 those near the inner parts (the more adiabatic regions of the cloud). The similar relative  
261 dispersion values when comparing Figs. 3 and 4 and the decrease in LWC and  $N_c$  suggest  
262 that a fraction of the droplets were totally evaporated due to mixing with the outside  
263 environmental air, but the shape of the droplet size distribution did not change. This implies  
264 that non-homogeneous entrainment mixing was the dominant process at the cloud  
265 boundaries, similar to the findings of Small et al. (2013).

266 It should be noted that although the error bars in Fig. 4 are significantly larger than in  
267 Fig. 3, both figures demonstrate invariant values of  $\epsilon$  as a function of vertical height above  
268 the cloud base and LWC. It can also be noted that the trend for the TRK3 case is different. A  
269 clear decrease in  $\epsilon$  is observed near the top of the cloud associated with higher LWC values.  
270 This issue will be further discussed in Section 4.

271 Figure 5 presents  $\epsilon$  as a function of  $\langle r \rangle$ . The  $\langle r \rangle$  values are binned such that each point  
272 represents different heights range, similar to the height binning that is shown in Fig. 2. This  
273 representation suggests that the relative dispersion is invariant to changes in average droplet  
274 radius (which by itself is highly correlated to the height within the cloud as explained in Fig.  
275 2). This reinforces the conclusions drawn from Figs. 3 and 4.

276 Figure 6 combines all of the clouds' data together (except TRK3) and shows the  $\epsilon$  and its  
277 variance values as a function of LWC (Fig. 6a) and droplet concentration ( $N_c$ , Fig. 6b). The  
278 gray crosses represent  $\epsilon$  values, while the blue circles and red crosses represent the binned  
279 (number-based) mean of  $\epsilon$  and its standard deviation, respectively.

280 The results show that  $\epsilon$  values vary significantly ( $\epsilon \sim 0.1\text{--}1.25$ ) only in cloud segments  
281 with very low LWC and low drop concentrations ( $\text{LWC} < \sim 0.01 \text{ g kg}^{-1}$  and  $N_c < \sim 5 \text{ cm}^{-3}$ ).  
282 For higher LWC and  $N_c$  values, the  $\epsilon$  fits within a relatively narrow range of values between  
283 0.24 and 0.37.

284 Figure 7 presents an additional analysis for the combined dataset of all clouds together  
285 except TRK3. Figure 7a presents separate histograms of  $\varepsilon$  for the measured cloud data  
286 obtained during each flight. This figure demonstrates that  $\varepsilon$  variance decreases for flights  
287 associated with higher aerosol loading, which may be related to increasing  $N_c$  and/or LWC,  
288 and extension of the relative duration of the cloud mature stage with increasing aerosol  
289 loading as suggested by the second indirect effect (Albrecht et al., 1989). Figure 7b presents  
290  $\varepsilon$  histograms for different vertical parts of the clouds. This graph indicates that the variance  
291 of  $\varepsilon$  tends to be smaller near the cloud base, compared to higher levels in the cloud. Possible  
292 reasons for this difference are discussed in the next section. This figure further suggests that  
293  $\varepsilon$  does not show any significant trend with increasing height above the cloud base.

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#### 295 **4. Discussion and summary**

296 Using in-situ flight measurements of droplet size distributions in warm continental cumulus  
297 clouds, we investigated the dependence of  $\varepsilon$  on cloud microphysical properties (LWC,  $\langle r \rangle$   
298 and  $N_c$ ).

299 The results suggest that the mean values of relative dispersion estimated for those  
300 cumulus clouds do not show any significant trend with LWC, height within the cloud,  
301 droplet concentration, aerosol loading or average droplet radius. On the other hand, a  
302 second-order effect on  $\varepsilon$  distribution is clearly seen as a decrease in the variance of  $\varepsilon$  with an  
303 increase in LWC and  $N_c$  (see Fig. 6).

304 Overall, the mean  $\varepsilon$  values vary in the range of 0.24 to 0.37. This is in agreement with  
305 previous studies which indicated that  $\varepsilon$  tends to be bounded in a similar narrow range in  
306 warm cumuli (Pandithurai et al., 2012; Berg et al., 2011), stratus clouds (Peng et al., 2007)  
307 and stratocumulus clouds (Pawlowska et al., 2006).

308 Our findings also showed that the more scattered  $\varepsilon$  values ( $\sim 0.1$ – $1.25$ ) were associated  
309 with very low LWC and  $N_c$ , below threshold values of  $\sim 0.01$  g kg<sup>-1</sup> and  $\sim 5$  cm<sup>-3</sup>, respectively  
310 (similar to the findings of Pandithurai et al., 2012; Zhao et al., 2006; and Deng et al., 2009).  
311 Measurement quality is low in those cloud regions, and this may be reflected as an increase  
312 in  $\varepsilon$  variance. However, Tas et al. (2012) also showed, using detailed microphysical model,  
313 that  $\varepsilon$  tends to be more scattered during the non-mature cloud development stages and for  
314 entrainment zones in the cloud, which are also associated with low LWC and  $N_c$  values.

315 Above the threshold levels of  $N_c$  and LWC,  $\varepsilon$  showed fast convergence to average values.  
316 Deng et al. (2009) and Zhao et al. (2006) also indicated convergence of  $\varepsilon$  to a narrow range  
317 (0.4–0.5) with increasing  $N_c$  associated with higher pollution levels. Tas et al. (2012) showed

318 that  $\varepsilon$  fits into a narrow range for the core of a cumulus cloud in its mature stage, and for  
319 high LWC. In the present study, we also observed convergence of  $\varepsilon$  with aerosol loading,  
320 which might be related to an increase in  $N_c$ , LWC, or both. Note that an increase in aerosol  
321 loading can lead to extension of the mature stage, as a result of the second indirect effect  
322 (Albrecht et al., 1989). Therefore, the convergence of  $\varepsilon$  due to either an increase in aerosol  
323 loading or an extension of the mature stage might be related to the same basic mechanism.

324 How reliable are the  $\varepsilon$  estimations based on the CDP measurements? To estimate the  $\varepsilon$   
325 values correctly, one needs a full description of the droplet size distribution. Our  
326 measurements were limited to a range of radii between 1.5 and 25  $\mu\text{m}$ . Clearly,  $\varepsilon$  estimations  
327 deviate when the tail of the size distribution exceeds 25  $\mu\text{m}$  in radius, i.e., the estimated  
328 variance will be smaller than the real one (see TRK3 in Fig. 2) and as a consequence, the  $\varepsilon$   
329 values as well (see TRK3 in Figs. 3 and 4).

330 The droplet size distributions for different vertical levels in each cloud are shown in Fig.  
331 2, and it is evident that except for case TRK3, the concentration of droplets  $>25 \mu\text{m}$  is  
332 negligible. The relative dispersion values for the TRK3 case tended to decrease in the upper  
333 parts of the cloud, characterized by larger LWC values. As indicated above, TRK3 was the  
334 cleanest case and it probably contained larger drops that were not measured by the probe.  
335 This suggests that in such cases, the estimation of  $\varepsilon$  might be incorrect. Specifically, the  
336 contribution of the larger droplets is expected to be more significant for the case of larger  
337 LWC higher up in the cloud (see case TRK3 in Fig. 2). Therefore, the decrease in  $\varepsilon$  for such  
338 data points might be an artifact due to incomplete representation of the large drops.

339 Our analysis suggests that a bias in  $\varepsilon$  due to failure to detect the entire droplet size  
340 distribution, including the tail, of large drops, may serve as a criterion for the reliability of  
341 the measurement data for application in microphysical analyses. We are currently in the  
342 process of validating this hypothesis using datasets from other campaigns.

343 Regarding all of the other clouds, based on the relatively small  $\langle r \rangle$  values (see Fig. 2),  
344 the sparse population of large droplets (for all clouds except TRK3) and the relatively high  
345 aerosol loading, we assume that drop growth in all of the measured clouds was dominated by  
346 the condensation process. It is well known that growth by condensation leads to an increase  
347 in  $\langle r \rangle$  but a decrease in the width of the size distribution (smaller  $\sigma$ ) (e.g. Rogers and Yau,  
348 1989). However, the invariant nature of  $\varepsilon$  values in this and some other studies suggests that  
349 additional processes occur simultaneously with condensation. These additional processes act  
350 to increase  $\sigma$ , such that the ratio of  $\sigma$  to  $\langle r \rangle$  remains relatively constant. Such processes may

351 include drop growth by collision–coalescence or the formation of new droplets by activation  
352 of cloud condensation nuclei (CCN) (increasing the number of the smaller droplets) or  
353 activation of giant CCN (which may increase the number of the larger drops). These  
354 scenarios act to broaden the droplet spectrum. In this study, we cannot determine which of  
355 these processes is more significant. Moreover, the contribution of each of the two processes  
356 to maintaining a relatively constant range of  $\varepsilon$  may vary at different locations and stages of  
357 cloud evolution. Collection-based processes are more important higher in the cloud and at  
358 later stages in the cloud's evolution, while activation of new particles is more important near  
359 the cloud base and in the early stages of its development.

360 Autoconversion and radiation parameterizations in many GCMs and CRMs are currently  
361 based on the estimated impact of aerosol loading on the magnitude of  $\varepsilon$  (see Section 1). The  
362 present study uses airborne measurements to demonstrate that  $\varepsilon$  is not correlated with LWC,  
363  $N_c$  or  $\langle r \rangle$ , suggesting that  $\varepsilon$  is relatively invariant to changes in the cloud's microphysical  
364 properties. On the other hand, variance in  $\varepsilon$  was found to be correlated with LWC and  $N_c$ ,  
365 suggesting that  $\varepsilon$  variance, rather than  $\varepsilon$ , does depend on the cloud's microphysical  
366 properties. This finding may pave the way for improving autoconversion and radiation  
367 parameterizations, which rely on  $\varepsilon$  values in CRMs and GCMs. However, further testing of  
368 the correlation of  $\varepsilon$  with these parameters under different ambient conditions and adiabatic  
369 and non-adiabatic cloud conditions is warranted.

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385 **References**

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387 Ackerman, A.S., Toon, O.B., Taylor, J.P., Johnson, D.W., Hobbs, P.V., and Ferek, R.J.:  
388 Effects of aerosols on cloud albedo: evaluation of Twomey's parameterization of  
389 clouds susceptibility using measurements of ship tracks. *J. Atmos. Sci.*, 57, 2684–2695,  
390 doi:10.1175/1520-0469 (2000).  
391  
392 Altaratz, O., Koren, I., Remer, L. A., and Hirsch, E.: Review: Cloud invigoration by  
393 aerosols—Coupling between microphysics and dynamics, *Atmospheric*  
394 *Research*, 140–141(0), 38–60, 2014.  
395  
396 Albrecht, B.: Aerosols, cloud microphysics, and fractional cloudiness, *Science*, 245,  
397 1227–1230, 1989.  
398  
399 Andreae, M.O., Rosenfeld, D., Artaxo, P., Costa, A.A., Frank, G.P., Longo, K.M.,  
400 Silva-Dias, M.A.F.: Smoking rain clouds over the Amazon. *Science* 303, 1337-  
401 1342. <http://dx.doi.org/10.1126/science.1092779>, 2004.  
402  
403 Axisa, D., D., Rosenfeld, J., Santarpia, W., Woodley, and Collins, D.: The Southern Plains  
404 Experiment in Cloud Seeding of Thunderstorms for Rainfall Augmentation  
405 (SPECTRA) project: Operational tools used towards verifying glaciogenic and  
406 hygroscopic seeding conceptual models, case studies and preliminary results.  
407 Preprints, *16th Conf. on Planned and Inadvertent Weather Modification*, San Diego,  
408 CA, Amer. Meteor. Soc., Paper 6.6, 2005.  
409  
410 Berg, L. K., C. M. Berkowitz, J. C. Barnard, G. Senum, and Springston, S. R.: Observations  
411 of the first aerosol indirect effect in shallow cumuli, *Geophys. Res. Lett.*, 38, L03809,  
412 doi:10.1029/2010GL046047, 2011.  
413  
414 Daum, P., and Liu, Y.: Dispersion of cloud droplet size distributions, cloud  
415 parameterizations, and indirect aerosol effects, paper presented at 13th ARM Science  
416 Team Meeting, Atmos. Radiat. Meas. Clim. Res. Facility, U.S. Dep. of Energy, Daytona  
417 Beach, Fla., 14–18 March, 2003.  
418  
419 Deng, Z., Zhao, C., Zhang, Q., Haung, M., and Ma, X.: Statistical analysis of properties and  
420 parameterization of effective radius of warm cloud in Beijing area, *Atmos. Res.*, 93,  
421 888–896, 2009.  
422  
423 Feingold, G., Eberhard, W. L., Veron, D. E. and Previdi, M.: First measurements of the  
424 Twomey indirect effect using ground-based remote sensors, *Geophys. Res. Lett.*, 30(6),  
425 1287, doi:10.1029/2002GL016633, 2003.  
426  
427 Hansen, J. E., and Travis, L. D.: Light-scattering in planetary atmospheres, *Space Sci. Rev.*,  
428 16(4), 527–610, doi:10.1007/BF00168069, 1974.  
429  
430 Hsieh, W. C., Nenes, A., Flagan, R. C., Seinfeld, J. H., Buzorius, G., and Jonsson, H.:  
431 Parameterization of cloud droplet size distributions: Comparison with parcel models and  
432 observations, *J. Geophys. Res.*, 114, D11205, doi:10.1029/2008JD011387, 2009.  
433  
434 Koren, I., Kaufman, Y.J., Rosenfeld, D., Remer, L.A., Rudich, Y.: Aerosol

435 invigoration and restructuring of Atlantic convective clouds. *Geophys. Res. Lett.*  
436 32 (14), L14828, 2005.  
437

438 Liu, Y., and Daum, P. H.: Spectral dispersion of cloud droplet size distributions and the  
439 parameterization of cloud droplet effective radius, *Geophys. Res. Lett.*, 27, 1903–1906,  
440 doi:10.1029/1999GL011011, 2000a.  
441

442 Liu, Y., and Daum, P. H.: Which size distribution function to use in parameterization of  
443 effective radius, paper presented at 13th International Conference on Clouds and  
444 Precipitation, Int. Comm. on Clouds and Precip. Reno, Nev., 14–18, 2000b.  
445

446 Liu, Y., and Daum, P. H.: Anthropogenic aerosols: indirect warming effect from dispersion  
447 forcing, *Nature*, 419, 580–581, doi:10.1038/419580a, 2002.  
448

449 Liu Y, Daum P. H, McGraw R.: Size truncation effect, threshold behavior, and a new type of  
450 autoconversion parameterization. *Geophys Res Lett*, 32: L11811, 2005.  
451

452 Liu, Y., Daum, P. H, McGraw, R., and Miller M.: Generalized threshold function  
453 accounting for effect of relative dispersion on threshold behavior of autoconversion  
454 process. *Geophys. Res. Lett.* 33 L11804 2006a.  
455

456 Liu, Y., Daum, P. H., and Yum, S. S.: Analytical expression for the relative dispersion of the  
457 cloud droplet size distribution, *Geophys. Res. Lett.*, 33, L02810,  
458 doi:10.1029/2005GL024052, 2006b.  
459

460 Liu, Y., P. H. Daum, H. Guo, and Peng Y.: Dispersion bias, dispersion effect and aerosol-  
461 cloud conundrum, *Environ. Res. Lett.*, 3, 045021, doi:10.1088/1748-9326/3/4/045021,  
462 2008.  
463

464 Lu, M.-L., and Seinfeld, J. H.: Effect of aerosol number concentration on cloud droplet  
465 dispersion: A large-eddy simulation study and implications for aerosol indirect forcing, *J.*  
466 *Geophys. Res.*, 111, D02207, doi:10.1029/2005JD006419, 2006.  
467

468 Lu, M.-L., G. Feingold, H. H. Jonsson, P. Y. Chuang, H. Gates, R. C. Flagan, and Seinfeld,  
469 J. H.: Aerosol-cloud relationships in continental shallow clouds, *J. Geophys. Res.*, 113,  
470 D15201, doi:10.1029/2007JD009354, 2008.  
471

472 Ma, J., A. Chen, W. Wang, P. Yan, H. Liu, S. Yang, Z. Hu, and Lelieveld J.: Strong air  
473 pollution causes widespread haze-clouds over China, *J. Geophys. Res.*, 115, D18204,  
474 doi:10.1029/2009JD013065, 2010.  
475

476 Martin, G. M., Johnson, D. W., and Spice, A.: The measurement and parameterization of  
477 effective radius of droplets in warm stratocumulus clouds, *J. Atmos. Sci.*, 51, 1823-  
478 1842, doi:10.1175/1520-0469(1994)051<1823:TMAPOE>2.0.CO;2, 1994.  
479

480 McFarquhar, G. M., and Heymsfield, A. J.: Parameterization of INDOEX microphysical  
481 measurements and calculations of cloud susceptibility: Applications for climate studies,  
482 *J. Geophys. Res.*, 106, 28,675-28,698, doi:10.1029/2000JD900777, 2001.  
483

484 Martins, J. A., and Silva Dias, M. A. F.: The impact of smoke from forest fires on the  
485 spectral dispersion of cloud droplet size distributions in the Amazonian region, *Environ.*  
486 *Res. Lett.*, 4, 015002, doi:10.1088/1748- 9326/4/1/015002, 2009.

487

488 Miles, N. L., Verlinde, J., and Clothiaux, E. E.: Cloud droplet size distributions in low-level  
489 stratiform clouds, *J. Atmos. Sci.*, 57, 295–311, 2000.

490

491 Pandithurai, G., Dipu, S., Prabha, T. V., Maheskumar, R. S., Kulkarni J. R., and Goswami  
492 B. N.: Aerosol effect on droplet spectral dispersion in warm continental cumuli, *J.*  
493 *Geophys. Res.*, 117, D16202, doi:10.1029/2011JD016532, 2012.

494

495 Pawlowska, H., Grabowski, W.W., and Brenguier, J.-L.: Observations of the width of cloud  
496 droplet spectra in stratocumulus. *Geophysical Research Letters* 33, L29810,  
497 <http://dx.doi.org/10.1029/2006GL026841>, 2006.

498

499 Peng, Y., and Lohmann, U.: Sensitivity study of the spectral dispersion of the cloud droplet  
500 size distribution on the indirect aerosol effect, *Geophys. Res. Lett.*, 30(10), 1507,  
501 doi:10.1029/2003GL017192, 2003.

502

503 Peng Y., Lohmann U., Leaitch R., and Kulmala M.: An investigation into the aerosol  
504 dispersion effect through the activation process in marine stratus clouds, *J. Geophys.*  
505 *Res.*, 112, D11117. DOI: 10.1029/2006JD007401, 2007.

506

507 Rogers, R.R. and Yau, M.K.: A short course in cloud physics (3rd edition), Pergamon  
508 Press, 1989.

509

510 Rosenfeld, D., and Feingold, G.: Explanation of the discrepancies among satellite  
511 observations of the aerosol indirect effects, *Geophys. Res. Lett.*, 30(14), 1776,  
512 doi:10.1029/2003GL017684, 2003.

513

514 Rotstayn, L. D., and Liu, Y.: Sensitivity of the first indirect aerosol effect to an increase of  
515 cloud droplet spectral dispersion with droplet number concentration, *J. Clim.*, 16, 3476–  
516 3481, doi:10.1175/1520-0442, 2003.

517

518 Slingo, A.: A GCM parameterization for the shortwave Radiative properties of water clouds,  
519 *J. Atmos. Sci.*, 46, 1419–1427, doi:10.1175/1520-0469, 1989.

520

521 Small J. D., Chuang, P. Y., and H. H. Jonsson, Microphysical imprint of entrainment in  
522 warm cumulus, *Tellus B* 2013, 65, 19922, <http://dx.doi.org/10.3402/tellusb.v65i0.19922>.

523

524 Tao, W.-K., Chen, J.-P., Li, Z., Wang, C., Zhang, C.: Impact of aerosols on convective  
525 clouds and precipitation. *Rev. Geophys.* 50, RG2001. [http://](http://dx.doi.org/10.1029/2011RG000369)  
526 [dx.doi.org/10.1029/2011RG000369](http://dx.doi.org/10.1029/2011RG000369), 2012.

527

528 Tas, E., Koren, I., and Altaratz, O.: On the sensitivity of droplet size relative dispersion to  
529 warm cumulus cloud evolution, *Geophys. Res. Lett.*, 39, L13807,  
530 doi:10.1029/2012GL052157, 2012.

531

532 Teller A., D. Axisa, R. Bruintjes, D. Collins, A. Mutlu, C. Kluzek, M. Pocerlich, P. R.  
533 Buseck, E. Freney, S. Tessororf, S. Hunter, O. Şen, K. Kocak, H. Toros, A. Köse,

534 Tunc, M.: Cloud and Aerosol Research in Istanbul Final Report 2007–2008,  
535 NCAR/RAL, 2008.  
536  
537 Twomey, S.: The influence of pollution on the short wave albedo of clouds, *J. Atmos. Sci.*,  
538 34, 1149–1152, doi:10.1175/1520-0469, 1977.  
539  
540 Xie, X., X. Liu, Y. Peng, Y. Wan, Z. Yue, and Li., X.: Numerical simulation of clouds and  
541 precipitation depending on different relationships between aerosol and cloud droplet  
542 spectral dispersion, *Tellus B*, 65, 1–17, 2013.  
543  
544 Yum, S. S., and Hudson, J. G.: Adiabatic predictions and observations of cloud droplet  
545 spectral broadness, *Atmos. Res.*, 73, 203–223, doi:10.1016/j.atmosres.2004.10.006,  
546 2005.  
547  
548 Zhao, C., Tie, X., Brasseur, G., Noone, K. J., Nakajima, T., Noone, K. J., Nakajima, T.,  
549 Zhang, Q., Zhang, R., Huang, M., Duan, Y., Li, G., and Ishizaka, Y.: Aircraft  
550 measurements of cloud droplet spectral dispersion and implications for indirect aerosol  
551 radiative forcing. *Geophys. Res. Lett.*, 33, L16809. DOI: 10.1029/2006GL026653, 2006.  
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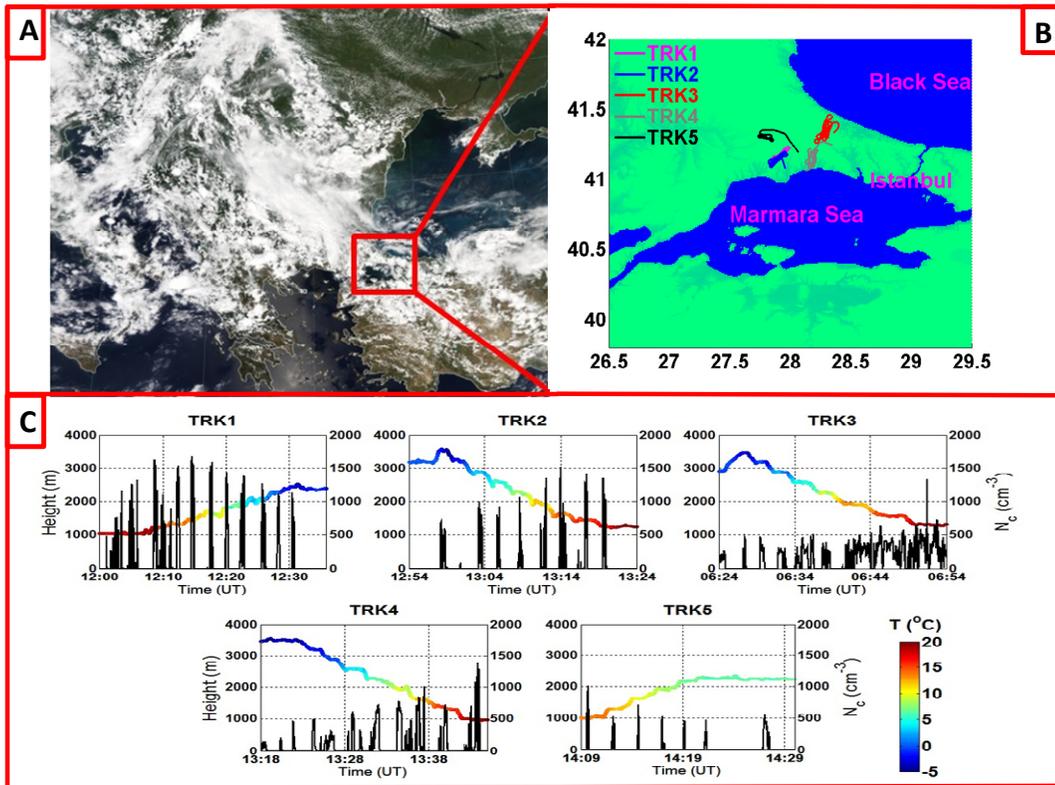
561 *Table 1. Airborne measurements used for the present study. For each of the five airborne*  
 562 *measurements used in the present study, the flight date and the corresponding abbreviation*  
 563 *used in this paper, number of data points, aerosol loading at the cloud base (see Section 2)*  
 564 *minimum and maximum temperature, minimum and maximum pressure, cloud base and*  
 565 *(estimated) cloud top height are indicated.*  
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<b>Flight date/ time (LT)</b>	<b>Abbr.</b>	<b>No. of data points (rounded)</b>	<b>Aerosol loading (cm<sup>-3</sup>) (0.11–3 µm)</b>	<b>Min.–Max. Temp. (°C)</b>	<b>Min.–Max. Height AGL (m)</b>	<b>Min.– Max. pressure (mb)</b>
6 Jun 2008/ 12:00-12:36	TRK1	380	1,800	5.7–16	1,000–2,500	750–901
6 Jun 2008/ 12:54-13:24	TRK2	240	900	-2.1–15.4	1,200–3,550	655–882
7 Jun 2008/ 06:24-06:54	TRK3	1,040	700	-1.3–9.8	1,250–3,450	661–874
7 Jun 2008/ 13:18-13:45	TRK4	450	800	-1.4–13.6	950–3,550	660–907
7 Jun 2008/ 14:09-14:30	TRK5	110	1,000	5.7–13.9	1,000–2,350	767–905

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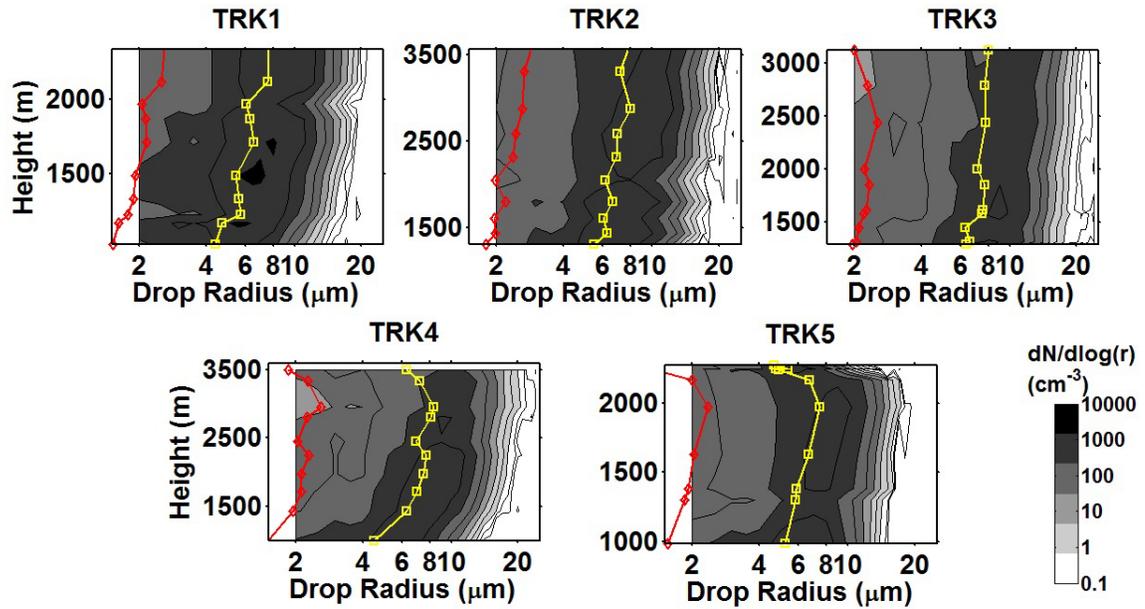
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Fig. 1. (a) MODIS image of the Eastern Mediterranean region on 7 Jun 2008. (b) The tracks of the five flights. (c) A summary of flight profiles and cloud droplet concentration in airborne measurements carried out on 6–7 Jun 2008 around Istanbul, Turkey. Black line shows the droplet concentration and colored line shows the height above ground and the temperature.

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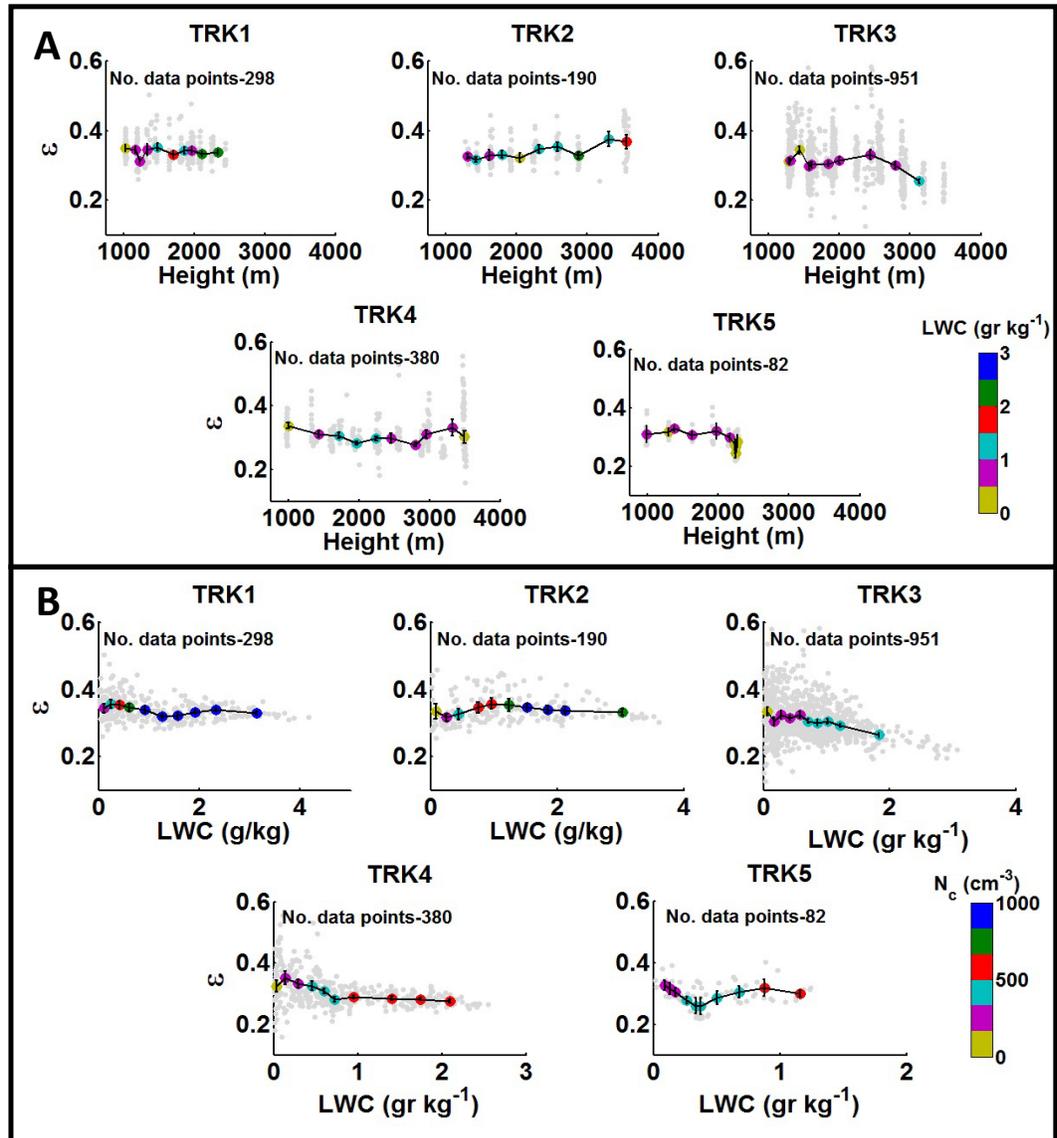
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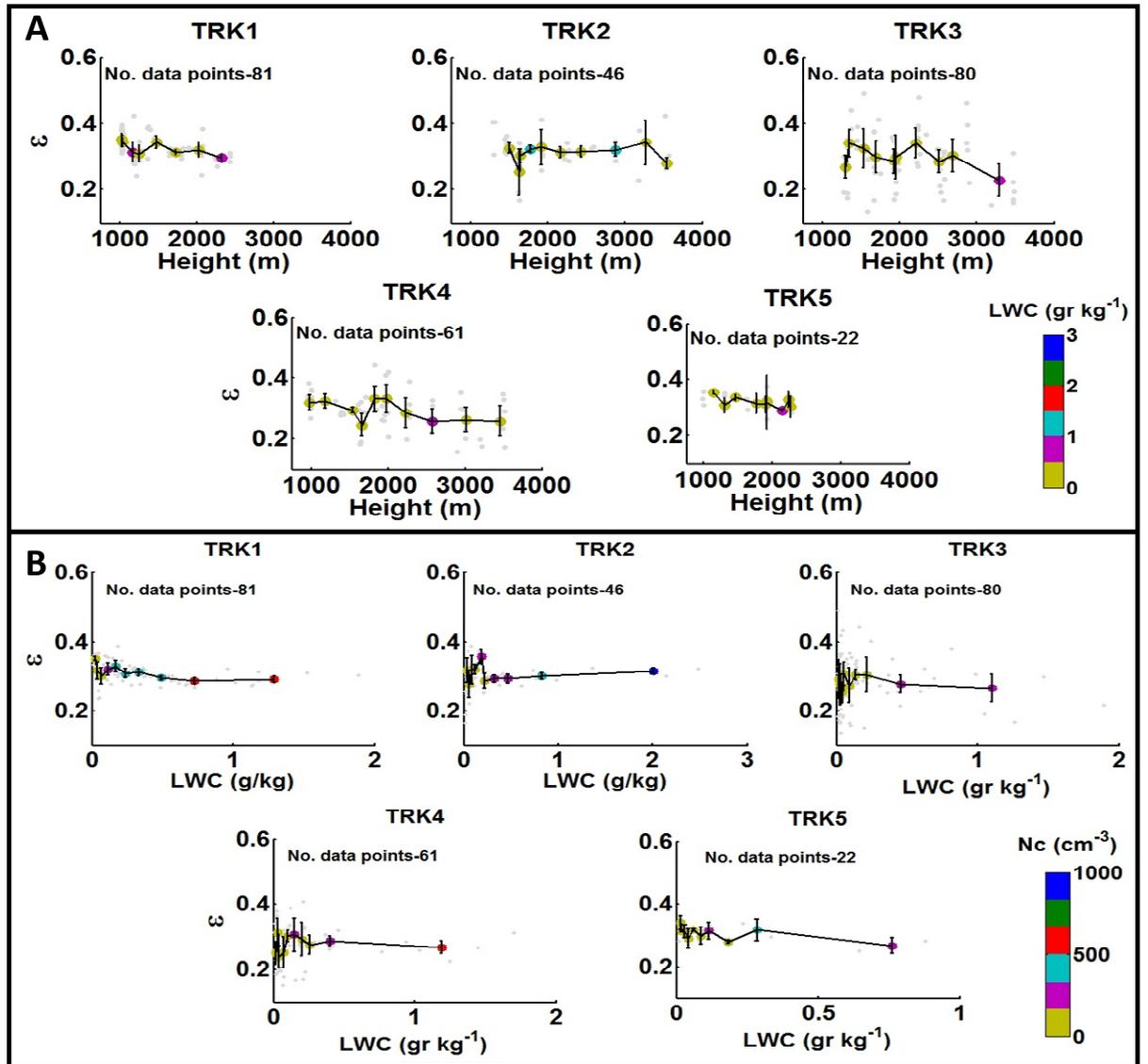
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Fig. 2. Cloud droplet size distribution as a function of height above the ground. The contours show the distribution ( $dN/d\log(r)$ ). The yellow and red lines represent the average and standard deviation of the radius over the entire measurements, respectively. For the purpose of constructing the lines of the average radius and the standard deviation, we divided the measurements into 10 height bins and for each bin the average was calculated. Note that the vertical axes are not uniform, accounting for the different cloud tops observed in the different flights.

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672 *Fig. 3. (a) Relative dispersion ( $\epsilon$ ) vs. height above the ground with colors representing the*  
 673 *liquid water content (LWC) and (b)  $\epsilon$  vs. LWC with colors representing the droplet*  
 674 *concentration for the inner cloud data points. Error bars represent standard error of the*  
 675 *average  $\epsilon$  for each height level (in a) and LWC (in b) with a confidence level of 95%.*



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Fig. 4. (a) Relative dispersion ( $\epsilon$ ) vs. height above the ground with colors representing the liquid water content (LWC) and (b)  $\epsilon$  vs. LWC with colors representing the droplet concentration for the cloud boundary data points. Error bars represent the standard error of the average  $\epsilon$  for each height level (in a) and LWC (in b) with a confidence level of 95%.

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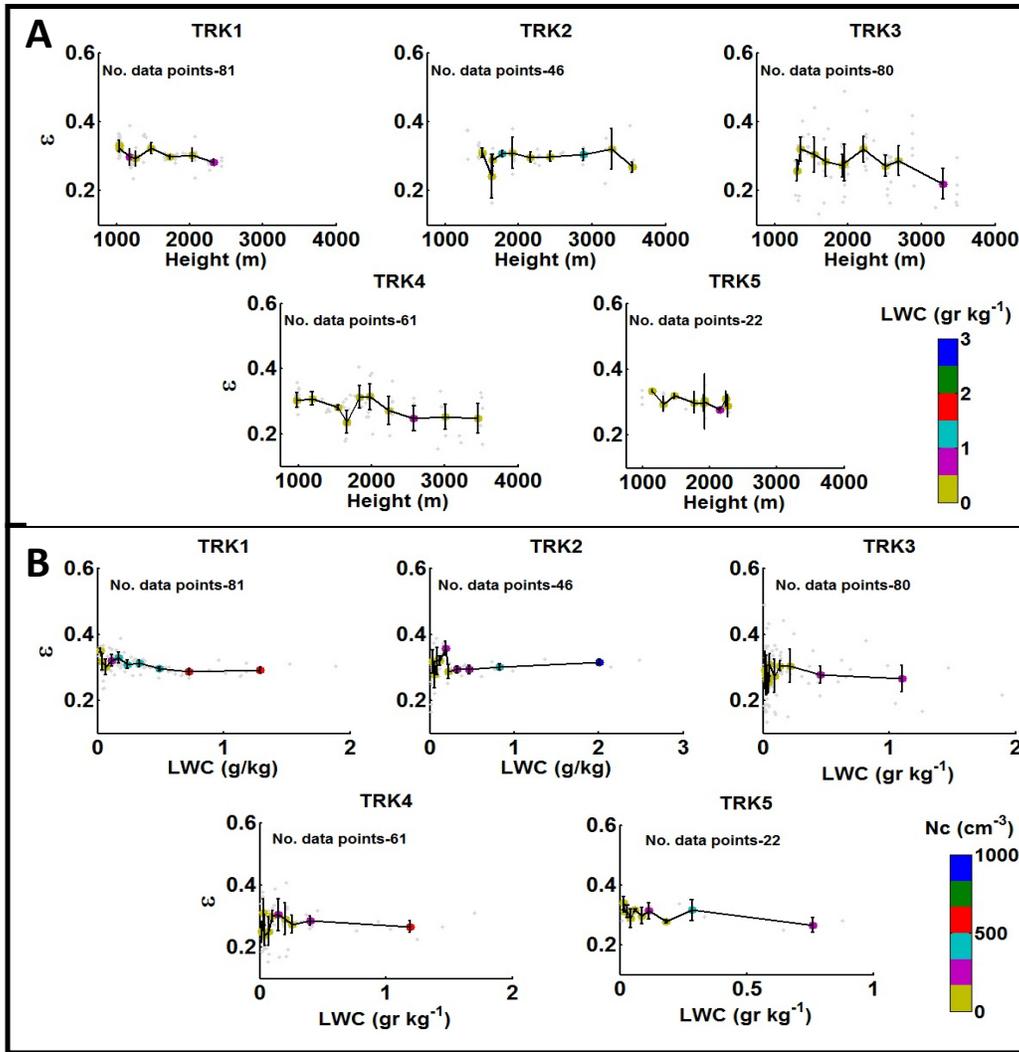
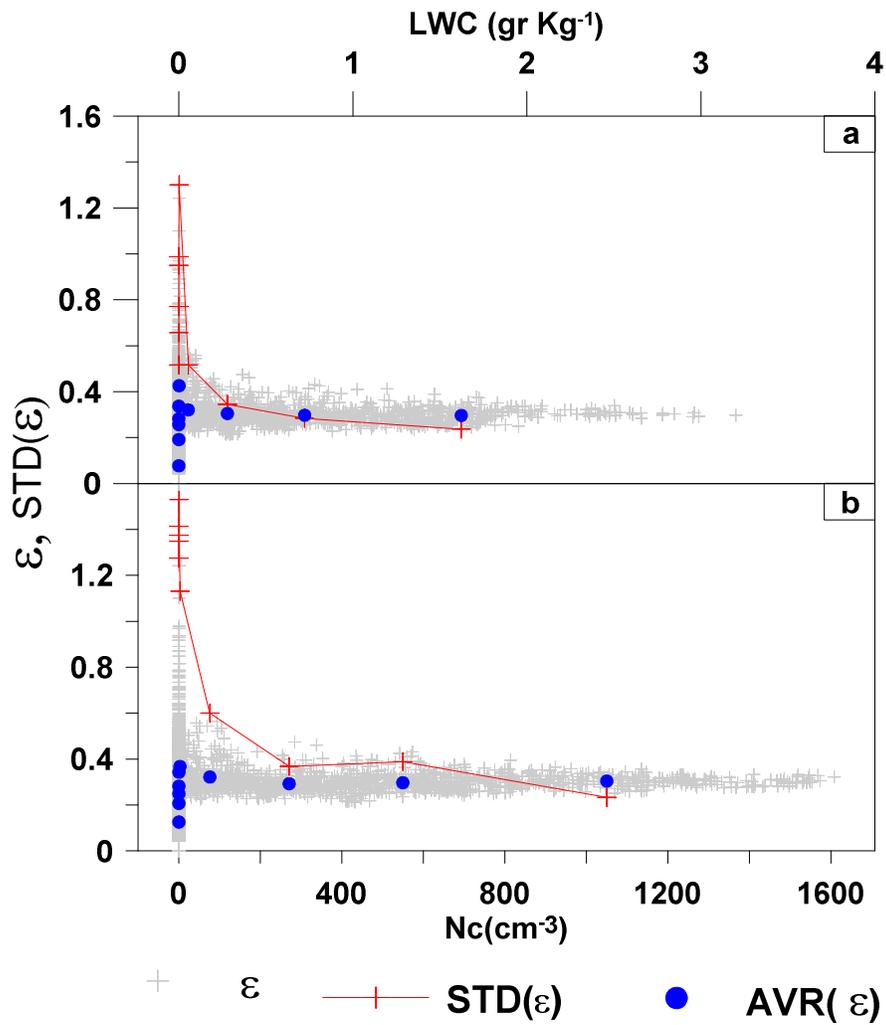


Fig. 5. Relative dispersion vs. average radius for (a) the inner cloud data, and (b) the cloud boundaries. Error bars represent the standard error of the average  $\epsilon$  for each  $\langle r \rangle$  level with a confidence level of 95%.



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727 *Fig. 6. Relative dispersion and its variance as a function of cloud liquid water content*  
 728 *(LWC) and droplet number. Relative dispersion ( $\epsilon$ ), relative dispersion average ( $AVR(\epsilon)$ )*  
 729 *and relative dispersion variance ( $STD(\epsilon)$ ) are presented vs. LWC (a) and  $N_c$  (b).  $AVR(\epsilon)$  and*  
 730 *( $STD(\epsilon)$ ) are presented as the average values of 10 number-based size bins.*

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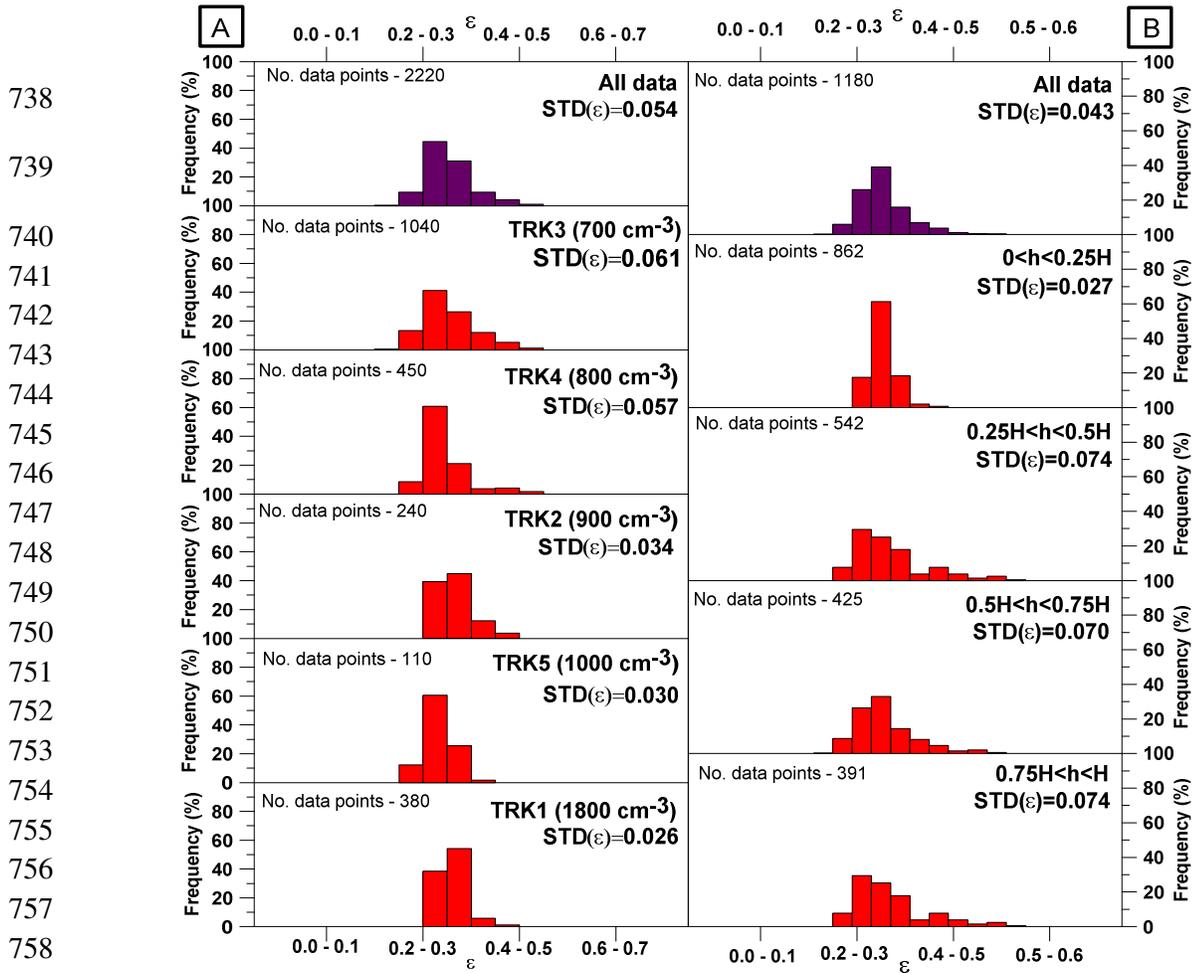
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760 *Figure 7. (a) Histograms of  $\epsilon$  for different aerosol loadings. The average aerosol loading for*  
761 *each flight (calculated at cloud base height) is presented. All histograms are based only on*  
762 *measured data associated with  $N_c > 10 \text{ cm}^{-3}$ . (b) Histogram of  $\epsilon$  for different height ranges*  
763 *above the cloud base (indicated individually for each histogram by “h” range of the total*  
764 *cloud depth, “H”), excluding data collected during flight TRK3. All histograms are based*  
765 *only on measured data associated with  $N_c > 10 \text{ cm}^{-3}$ . The top panel (All data) is based on*  
766 *data collected during all flights. Data collected during flight TRK3 were not used for any of*  
767 *the histograms.*