

**1 Parameterization of cloud droplet size distributions:  
2 comparison with parcel models and observations**

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3 **Abstract.** This work examines the efficacy of various physically-based  
4 approaches derived from 1-D adiabatic parcel model frameworks (a numer-  
5 ical model and a simplified parameterization) to parameterize the cloud droplet  
6 distribution characteristics for computing cloud effective radius and auto-

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7 conversion rate in regional/global atmospheric models. Evaluations are car-  
8 ried out for integrations with single (average) and distributions of updraft  
9 velocity, assuming that a) conditions at  $s_{max}$  are reflective of the cloud col-  
10 umn, or, b) cloud properties vary vertically, in agreement with 1-D parcel  
11 theory. The predicted droplet distributions are then compared against in-  
12 situ cloud droplet observations obtained during the CRYSTAL-FACE and  
13 CSTRIFE missions. Good agreement of droplet relative dispersion between  
14 parcel model frameworks indicates the parameterized parcel model essentially  
15 captures 1-D dynamics; the predicted distributions are overly narrow, with  
16 relative dispersion being a factor of 2 lower than observations. However, if  
17 conditions at cloud maximum supersaturation are used to predict relative  
18 dispersion and applied throughout the cloud column, better agreement is seen  
19 with observations, especially if integrations are carried out over the distri-  
20 bution of updraft velocity. When considering the efficiency of the method,  
21 calculating cloud droplet spectral dispersion at  $s_{max}$  is preferred for linking  
22 aerosol with droplet distributions in large scale models.

## 1. Introduction

23 The greatest uncertainty in assessments of anthropogenic climate change arises from  
24 aerosol-cloud-climate interactions [*Intergovernmental Panel on Climate Change (IPCC)*,  
25 2007], termed “aerosol indirect effects”. Increased aerosol concentrations tend to increase  
26 the number of droplets in warm clouds, which can enhance cloud albedo [*Twomey*, 1977];  
27 increasing droplet number also tends to reduce precipitation efficiency, which can affect  
28 cloud structure, lifetime and radiative properties [*Albrecht*, 1989].

29 Quantifying indirect effects requires a relationship between cloud microphysical prop-  
30 erties (like number and size distribution) and its precursor aerosol. Current treatments  
31 range from empirical correlations between an aerosol proxy (such as mass) and a droplet  
32 distribution moment (typically number) [*Boucher and Lohmann*, 1995], to explicit calcu-  
33 lation of droplet number using a “mechanistic parameterization” [*Ghan et al.*, 1997; *Nenes*  
34 *and Seinfeld*, 2003; *Fountoukis and Nenes*, 2005; *Barahona and Nenes*, 2007]. Although  
35 this is an important step towards addressing issues of aerosol-cloud interactions, calcu-  
36 lation of droplet number and cloud liquid water content alone are not sufficient. Cloud  
37 processes are sufficiently sensitive to droplet size, so parameterizations must also include  
38 some measure of the droplet distribution. For example, autoconversion of cloud water to  
39 rain (i.e., formation of drizzle from self collision of small droplets) is a key cloud process  
40 and very sensitive to droplet size distribution; in fact, the largest uncertainty in assess-  
41 ment of aerosol impacts on precipitation is associated with the treatment of rain formation  
42 in large scale models [*Lohmann and Feichter*, 2005]. Although numerous autoconversion  
43 parameterizations exist (e.g. *Kessler* [1969]; *Manton and Cotton* [1977]; *Rotstayn* [1997];

44 *Khairoutdinov and Kogan* [2000]; *Liu and Daum* [2004], the uncertainty associated with  
45 their application is large, about a factor of ten [*Hsieh et al.*, in press], and largely related  
46 to the treatment of droplet size distribution and the size-dependence of the collection  
47 kernel.

48 Explicit consideration of droplet size distribution is important also for calculation of  
49 cloud radiative properties. The effective radius, required for calculation of cloud optical  
50 depth and radiative forcing, is given by  $r_e = \left(\frac{3L}{4\pi\rho_w kN}\right)^{1/3}$ , where  $\rho_w$  is the density of water,  
51 and the spectral parameter “ $k$ ” expresses the effect of droplet width;  $k=1$  for monodis-  
52 perse droplets, decreasing as the distribution broadens. For example, *Martin et al.* [1994]  
53 proposed  $k=0.80$  for marine clouds (narrow size distribution) and  $k=0.67$  for polluted  
54 clouds (broad size distribution). Although qualitatively correct, these  $k$  values do not  
55 capture the extent of variability in droplet distribution width seen in ambient clouds. *Liu*  
56 *and Daum* [2000, 2002] recognized this and derived formulas for  $r_e$  by assuming droplets  
57 follow a Weibull or gamma size distribution, and explicitly included the effect of relative  
58 dispersion (i.e., the ratio of standard deviation to average radius). When included in GCM  
59 assessments of the indirect effect, variability in droplet spectral broadening decreased in-  
60 direct forcing between 14.3 and 16% [*Peng and Lohmann*, 2003; *Rotstayn and Liu*, 2003],  
61 with an upper limit of 33.1% [*Rotstayn and Liu*, 2003].

62 Understanding the processes that control the width of observed size distributions in  
63 ambient clouds has been the focus of many studies in the past. All agree that single  
64 updrafts, where droplets grow via condensation, tend to produce narrow size distributions;  
65 considering entrainment and mixing can substantially broaden them [*Mason and Chien*,  
66 1962; *Mason and Jonas*, 1974; *Baker et al.*, 1980; *Cooper*, 1989; *Su et al.*, 1998; *Lasher-*

67 *Trapp et al.*, 2005, e.g.,]. Other processes can broaden the distribution as well, such  
68 as collision-coalescence [*Pruppacher and Klett*, 1997, e.g.,], and secondary activation of  
69 droplets above cloud base [*Erlick et al.*, 2005].

70 Although resolving the droplet size distribution is required to reduce the uncertainty of  
71 aerosol indirect effects, explicit cloud droplet microphysics is computationally expensive.  
72 Parameterizations reduce the computational burden, but may (as seen in the previous ex-  
73 amples) introduce significant predictive uncertainty. Efficient parameterization of droplet  
74 spectral width and its dependence on the cloud microphysical state (and changes thereof  
75 from aerosol perturbations) is an active area of research. *Wood* [2005] proposed a gener-  
76 alized droplet distribution derived from observational data; a number of physically-based  
77 alternatives have also been proposed to link aerosol with cloud distribution properties.  
78 *Khvorostyanov and Curry* [1999] also relate  $N$  and droplet distribution width to cloud  
79 parameters such as updraft velocity dispersion and the vertical profile of cloud thermo-  
80 dynamic properties. Similarly, the cloud droplet spectrum tends to broaden when the  
81 updraft velocity decreases [*Peng et al.*, 2007; *Yum and Hudson*, 2005]. *Liu et al.* [2006]  
82 derived an analytical formula that relates the relative dispersion of cloud droplet dis-  
83 tribution to cloud condensation nuclei (CCN) spectra and updraft velocity, based on  
84 adiabatic growth theory of cloud droplets. These approaches apply some form of par-  
85 cel theory towards computing cloud droplet spectral properties. However, few studies  
86 exist that evaluate, through the usage of in-situ cloud observations, the uncertainty in  
87 predicted droplet spectral parameters and autoconversion associated with application of  
88 parcel-based approaches.

89 In this work, we explore the potential of parcel-based approaches for parameterizing  
90 cloud droplet size distribution in regional and global climate models. All approaches  
91 tested assume droplets form adiabatically in individual updrafts (using a parcel model  
92 and a parameterization thereof based on the *Nenes and Seinfeld* [2003] and *Fountoukis  
93 and Nenes* [2005] activation parameterizations) to explicitly compute the size distribu-  
94 tion and growth of an activated droplet population throughout a cloud column. We also  
95 explore another approach, based on computing the relative dispersion at  $s_{max}$  using the  
96 *Nenes and Seinfeld* [2003] parameterization, assuming that it applies to the whole cloud  
97 column. The overall droplet distribution,  $n(D_p)$ , is then computed for either a single up-  
98 draft (corresponding to the average of the measured distribution), or as the superposition  
99 of droplet distributions for each updraft measured in the cloud. Each approach is evalu-  
100 ated by comparing predicted droplet spectral characteristics with in-situ measurements of  
101 cloud droplet size distributions for a wide range of aerosol and cloud forming conditions  
102 sampled during the CRYSTAL-FACE [*Conant et al.*, 2004] and CSTRIFE field compaigns  
103 [*Meskhidze et al.*, 2005]. The importance of predicted size distribution deviations is ex-  
104 pressed in terms of the uncertainty in the predicted spectral dispersion parameter  $k$ , and  
105 autoconversion rate; the latter is done by introducing parameterized and observed size  
106 distributions into the  $R_6$  (i.e., sixth-moment mean radius) parameterization of *Liu and  
107 Daum* [2004], and quantifying the resulting differences in autoconversion.

## 2. Simulating cloud droplet growth

### 2.1. Numerical parcel model

108 Computation of the cloud droplet size distribution is based on the 1-D adiabatic cloud  
109 parcel framework, in which buoyant air parcels develop water vapor supersaturation, and

110 cloud droplets activate upon aerosol particles contained within them. After a maximum  
111 supersaturation,  $s_{max}$ , is reached, all droplets have formed and grow subsequently via  
112 condensation. Since a cloud is characterized by a distribution of updrafts, the parcel  
113 concept can be further extended, so that the average droplet number and size distribution  
114 is the superposition of distributions from each updraft (or some moment thereof, e.g.,  
115 *Meskhidze et al.* [2005]; *Peng et al.* [2005]; *Fountoukis et al.* [2007]).

## 2.2. Parameterization of parcel model

116 Instead of numerically solving the full set of differential equations that describe the  
117 process of activation and condensational growth (e.g., *Nenes et al.* [2001]), we develop  
118 a simplified approach that involves two steps: *i*) calculation of the cloud drop number  
119 concentration and size distribution at the point of  $s_{max}$ , using one of the activation pa-  
120 rameterizations of *Nenes and Seinfeld* [2003], *Fountoukis and Nenes* [2005], or *Barahona*  
121 *and Nenes* [2007], and, *ii*) simulation of the subsequent droplet growth as the cloud parcel  
122 ascends through a simplified treatment of condensational growth.

### 2.2.1. Determination of cloud drop size distribution at $s_{max}$

124 According to Köhler theory, a cloud condensation nucleus (CCN) requires exposure to  
125 a minimum “critical” supersaturation,  $s_c$ , before it can experience unconstrained growth  
126 and transform into a cloud droplet.  $s_c$  depends on particle size and chemical composition;  
127 therefore, the number of droplets forming in a cloud can be computed if the cloud super-  
128 saturation and the aerosol properties are known. In the initial stages of cloud formation,  
129 cooling of the air parcel leads to water vapor supersaturation; CCN then begin to activate  
130 into cloud droplets and rapidly grow. When enough CCN activate, the condensation of  
131 water vapor is strong enough to balance the availability of water vapor for condensation



132 (through cooling); this is the point where supersaturation reaches its maximum value,  
 133  $s_{max}$ , and determines the number of droplets that form [Nenes *et al.*, 2001]. Numerical  
 134 parcel models simulate this process by solving a system of coupled differential equations  
 135 (e.g., Nenes *et al.* [2001]), an approach that is computationally too demanding to be  
 136 included within a global model. Instead, simplified approaches, known as “mechanistic  
 137 activation parameterizations” [e.g., Abdul-Razzak *et al.*, 1998; Nenes and Seinfeld, 2003;  
 138 Fountoukis and Nenes, 2005; Barahona and Nenes, 2007] predict CDNC at the point of  
 139  $s_{max}$  in an ascending parcel; of these, the formulations of Nenes and Seinfeld [2003], Foun-  
 140 toukis and Nenes [2005] and Barahona and Nenes [2007] explicitly predict the droplet size  
 141 distribution (i.e., the concentration of droplets,  $dN$ , within a wet diameter interval  $dD_p$ )  
 142 at  $s_{max}$ , as the size of all activated droplets is known. Droplet size is determined from the  
 143 droplet growth equation, [Pruppacher and Klett, 1997; Seinfeld and Pandis, 1998],

$$\frac{dD_{pi}}{dt} = \frac{G}{D_{pi}} (s - s_{eq}) \tag{1}$$

$$G = \frac{4}{\frac{\rho_w RT}{p^s D_v M_w} + \frac{\Delta H_v \rho_w \left(\frac{\Delta H_v M_w}{RT} - 1\right)}{\kappa_a T}} \tag{2}$$

144 where  $D_{pi}$  is the diameter of droplet size class  $i$ ,  $\rho_w$  is the water density,  $R$  is the universal  
 145 gas constant,  $T$  is the parcel temperature,  $p^s$  is the saturation vapor pressure,  $D_v$  is  
 146 the water vapor diffusivity,  $M_w$  is the molar mass of water,  $\Delta H_v$  is the latent heat of  
 147 condensation of water,  $\kappa_a$  is the thermal conductivity of air,  $s$  is the parcel supersaturation,  
 148 and  $s_{eq}$  is the equilibrium supersaturation of the droplet.

149 Integrating Equation (1) provides the droplet diameter,  $D_p(t_{max})$  of an activated CCN  
 150 at the point of  $s_{max}$ ,

$$D_p^2(t_{max}) = D_p^2(\tau) + 2 \int_{\tau}^{t_{max}} G(s - s_{eq}) dt \quad (3)$$

151 where  $\tau$  is the time at which the CCN activates into a droplet (assumed to occur when the  
 152 parcel supersaturation is equal to the CCN critical supersaturation, *Nenes and Seinfeld*  
 153 [2003]),  $t_{max}$  is the time in the updraft corresponding to  $s_{max}$ , and,  $D_p(\tau)$  is the size of  
 154 CCN at time  $\tau$ .

155 Equation (3) can be simplified if droplet growth is assumed to be unaffected by curvature  
 156 and solute effects (i.e.,  $s_{eq} = 0$ ),

$$D_p^2(t_{max}) = D_p^2(\tau) + 2 \int_{\tau}^{t_{max}} G s dt \quad (4)$$

157 Since  $\int_{\tau}^{t_{max}} s dt \simeq \frac{1}{2\alpha V} [s_{max}^2 - s(\tau)^2]$  [*Twomey, 1959*], substituting into equation (4) gives  
 158 for  $D_p(t_{max})$ ,

$$D_p(t_{max}) = \sqrt{D_p^2(\tau) + \frac{G}{\alpha V} [s_{max}^2 - s(\tau)^2]} \quad (5)$$

159 For most CCN,  $D_p(\tau)$  can be approximated by the critical diameter,  $D_c = \frac{2A}{3s_c}$ , where  
 160  $A = \frac{4M_w\sigma_w}{RT\rho_w}$ , and  $M_w$ ,  $\sigma_w$  are the molar mass and surface tension of water, respectively  
 161 [*Nenes and Seinfeld, 2003*];  $s(\tau)$  can be approximated with the droplet critical supersat-  
 162 uration,  $s_c = \frac{4A^3\rho_w M_s}{27\nu\rho_s M_w d_s^3}$ , where  $M_s$ ,  $\rho_s$ ,  $\nu$ , are the molar mass, density, effective Van't  
 163 Hoff factor of the soluble fraction, and  $d_s$  is the dry diameter of the CCN from which the  
 164 droplet formed.

Depending on the particle  $s_c$ , *i*)  $D_p \gg D_c$ , *ii*)  $D_p \ll D_c$  (i.e., the CCN never strictly  
 activates), or, *iii*)  $D_p \sim D_c$  (i.e., the CCN is very close to the activation point at  $s_{max}$ ).  
 The particles that have  $s_c$  lower than a characteristic “partition supersaturation” [Nenes  
 and Seinfeld, 2003; Fountoukis and Nenes, 2005; Barahona and Nenes, 2007] exhibit  
 behavior type *i* and *ii*, so Equation (5) simplifies to  $D_p(t_{max}) \simeq \sqrt{\frac{G}{\alpha V} [s_{max}^2 - s_c^2]}$ . The  
 remaining CCN exhibit behavior type *iii*, and  $D_p(s_{max}) \simeq D_c$ .

### 2.2.2. Parcel supersaturation profile beyond $s_{max}$

The growth of droplets beyond the point of  $s_{max}$  in the cloud requires the knowledge  
 of the cloud supersaturation profile. Using the droplet spectrum at  $s_{max}$  (section 2.2.1)  
 as an initial condition, we can then compute supersaturation with finite difference over a  
 small time step interval,  $\Delta t$

$$s(t + \Delta t) = s(t) + \left( \frac{ds}{dt} \right) \Delta t \quad (6)$$

where  $s(t)$ ,  $s(t + \Delta t)$  are the supersaturations at time  $t$  and  $t + \Delta t$ , respectively, and  $ds/dt$  is  
 the supersaturation tendency in the parcel. Assuming that the droplets are characterized  
 by  $n$  size classes with diameters  $D_{pi}$ , concentration  $N_i$ , and growth rate  $dD_{pi}/dt$  (from  
 Equation 1),  $ds/dt$  is given by [e.g., Nenes et al., 2001; Nenes and Seinfeld, 2003],

$$\frac{ds}{dt} = \alpha V - \gamma \frac{\pi \rho_w}{2 \rho_a} \sum_{i=1}^n D_{pi}^2 \frac{dD_{pi}}{dt} N_i \quad (7)$$

where  $\alpha = \frac{gM_w \Delta H_v}{c_p RT^2} - \frac{gM_a}{RT}$ ,  $\gamma = \frac{pM_a}{p^s M_w} + \frac{M_w \Delta H_v^2}{c_p RT^2}$ ,  $g$  is the acceleration of gravity,  $M_a$ ,  $c_p$  is the  
 molar mass, heat capacity of air, respectively, and  $p$  is the ambient pressure. The first term  
 at the right hand side of Equation (7) represents the availability of water vapor from the

183 parcel updraft motion (i.e., cooling), and the second term refers to consumption of water  
 184 vapor by condensation on droplets. All properties in Equation (7) are computed for the  
 185 average temperature throughout the cloud column, as simulations with a numerical cloud  
 186 parcel model [Nenes *et al.*, 2001] demonstrate that this assumption does not substantially  
 187 affect the supersaturation profile over a wide range of cloud conditions. With updated  
 188 parcel supersaturation (equation 6), the droplets are then grown by integration of equation  
 189 (1) between time  $t$  and  $t + \Delta t$ ,

$$D_{pi}^2(t + \Delta t) = D_{pi}^2(t) + \Delta t \{2Gs(t + \Delta t)\} \quad (8)$$

190 The liquid water mixing ratio,  $W$ , can be computed as

$$W = \frac{\pi \rho_w}{6 \rho_a} \sum_{i=1}^n D_{pi}^3 N_i \quad (9)$$

191 Equations (6), (8) and (9) can be integrated until the desired liquid water mixing ratio  
 192 has been reached. If the aerosol size distribution is described in terms of lognormal modes,  
 193 they are discretized onto size bins that range from  $D_{gj} - 10\sigma_j$  to  $D_{gj} + 10\sigma_j$ , where  $D_{gj}$   
 194 is the geometric mean diameter of mode  $j$ , and  $\sigma_j$  is the geometric standard deviation for  
 195 mode  $j$ .

196 A series of sensitivity tests were carried out to determine the optimal number of sections  
 197 used in the parameterization. Using 50 sections per mode and a 0.5 s time step ensured  
 198 that droplet number calculated with the parameterization agrees with the numerical parcel  
 199 model predictions to within 5% (not shown).

### 2.3. Relative dispersion at $s_{max}$ represents the cloud column

200 In the initial stages of cloud formation, new (small) droplets are continuously formed and  
 201 grow via condensation. When supersaturation reaches its maximum value, cloud droplet  
 202 formation ceases, and condensational growth, which exhibits a  $D_p^{-1}$  dependency, tends to  
 203 narrow the distribution over time. This means that within the adiabatic condensational  
 204 parcel model framework, relative dispersion of the droplet size distribution is largest at  
 205 the point of maximum supersaturation. If one assumes that relative dispersion at  $s_{max}$  is  
 206 representative of the entire cloud (which implies that the tendency for spectral narrowing  
 207 from condensation growth is compensated by broadening from entrainment), one can use  
 208 the approach described in Section 2.2.1 to determine the cloud spectral dispersion.

### 3. Approaches used to parameterize size distributions

209 Vertical profiles of droplet distribution characteristics, such as size and relative disper-  
 210 sion, can be computed using either of the three approaches described in Section 2. One  
 211 issue still remaining however is the treatment of updraft velocity, as droplet distributions  
 212 can be computed for a single updraft, or a distribution of updrafts. Overall, six approaches  
 213 are evaluated (summarized in Table 1), as combinations of the droplet growth (Section  
 214 2) and updraft distribution treatments (described below), to parameterize droplet size  
 215 distribution characteristics.

#### 3.1. Single updraft

216 The droplet distribution computed for a single updraft at a given height ( $z$ ),  $n(D_p, z)$ ,  
 217 is used to compute the vertical profile of average droplet diameter,  $\overline{D_p}(z)$ ,

$$\overline{D_p}(z) = \frac{\int_0^\infty D_p n(D_p, z) dD_p}{\int_0^\infty n(D_p, z) dD_p} \quad (10)$$

where  $\int_0^\infty n(D_p, z) dD_p = N$  is the total droplet number concentration. The vertical profile of standard deviation,  $\sigma(z)$ , of the size distribution is given by

$$\sigma(z) = \left( \frac{\int_0^\infty n(D_p, z) (D_p - \overline{D_p}(z))^2 dD_p}{\int_0^\infty n(D_p, z) dD_p} \right)^{1/2} \quad (11)$$

218 After  $\overline{D_p}(z)$  and  $\sigma(z)$  are determined, the relative dispersion at any given height,  $\varepsilon(z)$   
219 is

$$\varepsilon(z) = \sigma(z) / \overline{D_p}(z) \quad (12)$$

### 3.2. Distribution of updrafts

220 Clouds are characterized by a range of updrafts, so that the cumulative droplet size  
221 distribution becomes the superposition of distributions from each updraft. Assuming  
222 that the updraft distribution can be described with a probability density function (PDF),  
223  $p(w)$ , the cloud droplet number concentration averaged over  $p(w)$  is then computed as

$$N = \int_0^\infty \int_0^\infty p(w) n(D_p, w, z) dD_p dw \quad (13)$$

224 where by definition  $\int_0^\infty p(w) dw = 1$ , and,  $n(D_p, w, z)$  is the droplet size distribution for a  
225 given updraft,  $w$ , and height,  $z$ . The vertical evolution of average droplet diameter and  
226 standard deviation based on averaging a series of updraft runs can be expressed as

$$\overline{D_p}(z) = \frac{\int_0^\infty \int_0^\infty D_p n(D_p, w, z) p(w) dD_p dw}{\int_0^\infty \int_0^\infty p(w) n(D_p, w, z) dD_p dw} \quad (14)$$

$$\sigma(z) = \left( \frac{\int_0^\infty \int_0^\infty n(D_p, w, z) (D_p - \overline{D_p}(z))^2 p(w) dD_p dw}{\int_0^\infty \int_0^\infty p(w) n(D_p, w, z) dD_p dw} \right)^{1/2} \quad (15)$$

227  $\varepsilon(z)$  is computed in a distribution of updrafts with Equation (12). In this study,  $p(w)$  is  
228 assumed to follow a Gaussian PDF, the moments of which are constrained by the observed  
229 average and standard deviation of updraft velocity.

#### 4. Evaluating droplet growth approaches

230 Each droplet approach is evaluated using in-situ measurements of ambient cloud droplet  
231 size distributions collected during the CRYSTAL-FACE and CSTRIFE field campaigns  
232 [*Conant et al.*, 2004; *Meskhidze et al.*, 2005]. Simulations were carried out for the aerosol  
233 characteristics summarized in Table 2 for CRYSTAL-FACE, and Table 3 for CSTRIFE  
234 data. *Hsieh et al.* [in press] give a description of the cloud conditions, instrumentation and  
235 cloud droplet distribution characteristics for both datasets. To ensure that the observed  
236 distributions used to evaluate each approach were not influenced by the effects of collision-  
237 coalescence, we select horizontal transects for which the droplet distributions are single-  
238 mode and the liquid water content is within a factor of two of the adiabatic value. All of  
239 the CSTRIFE data fit this criterion, while the subset of CRYSTAL-FACE dataset used  
240 is summarized in Table 4.

241 In the sections that follow, we first evaluate the parameterization against the parcel  
242 model for cloud data measured during CRYSTAL-FACE and CSTRIFE. The predictions  
243 are then evaluated against the in-situ data; spectral quantities are compared at the cloud  
244 height where predicted LWC is equal to the measured value. Given that the ability of  
245 each approach to reproduce droplet number was evaluated by *Conant et al.* [2004] and  
246 *Meskhidze et al.* [2005], this study focuses primarily on spectral dispersion.

#### 4.1. Comparison of parcel model and parameterization

247 The parameterized parcel model is first evaluated by comparing predicted droplet mean  
248 size, spectrum width, and relative dispersion against those of a full numerical activation  
249 adiabatic parcel model [Nenes *et al.*, 2001]. Based on the suggestion of Fountoukis *et al.*  
250 [2007], we use an effective water vapor uptake coefficient,  $\alpha$ , of 0.06. Figure 2 shows  
251 the predicted relative dispersion from parameterization (PS approach) and parcel model  
252 (MS approach) for all clouds sampled. The comparison is carried out at the observed  
253 LWC.  $\overline{D_p}$  is always well captured, as the discrepancy between parameterization and parcel  
254 model rarely exceeds 5% (Figure 1). Parameterized relative dispersion agrees with the  
255 parcel model, for most cases, to within 30% (Figure 2). The reasonably good agreement  
256 between numerical and parameterized parcel model suggests that the latter can be used  
257 for predictions of cloud droplet distributions in place of the full parcel model.

#### 4.2. Comparison against observations

258 Predicted droplet spectra using the MS, PS approaches agree reasonably with obser-  
259 vations for transect 4 (T4) when the cloud droplet size distributions are narrow, liquid  
260 water content is close to the adiabatic values, and the distribution is measured close  
261 to cloud base. An example of such a situation is given in Figure 3; uncertainties in  
262 measured cloud base are likely responsible for the shift between observed and measured  
263 distributions. Spectral broadening can also occur from instrument artifacts, such as laser  
264 beam non-homogeneity and coincidence error. Wendisch *et al.* [1996] demonstrated that  
265 beam non-homogeneity in the FSSP overestimates droplet size by 10-15% when in the  
266 15-30 micron size range, and, by 5-10% for 30-50 micron droplets. However, the same  
267 study indicated that spectrum broadening is negligible between 2 and 14 micron radius



(which covers the majority of observed distributions in CSTRIFE and CRYSTAL-FACE).  
*Wendisch et al.* [1996] and *Baumgardner and Spowart* [1990] indicated that instrument response time may cause broadening of the distribution for air speeds higher than 55-60 m s<sup>-1</sup>. Given that the Twin Otter platform operational velocity ranges between 50 - 55 m s<sup>-1</sup>, broadening due to air speed is negligible. Coincidence error can also lead to broadening of the distribution when CDNC > 500 cm<sup>-3</sup> *Brenquier* [1998]. Using this criterion, only a small fraction of the data could be affected, as the 75<sup>th</sup> percentile of CSTRIFE data has CDNC below 370 cm<sup>-3</sup>, and CRYSTAL-FACE, below 590 cm<sup>-3</sup>.

Most often, however, predictions deviate significantly from observations, and is not a result of measurement uncertainty. This is shown in Figure 4, which shows the predicted relative dispersion for all six approaches versus measured values for CRYSTAL-FACE and CSTRIFE distributions. The predicted values from the parameterized parcel model are close to the results based on the numerical parcel model; however, the MS, PS approaches tend to predict narrow distributions relative to those measured. On average, relative dispersion was substantially underestimated by both numerical and parameterized parcel models using single updrafts (MS, PS). Integrating over updraft distributions (MP, PP) tends to increase predicted relative dispersion, although relative dispersion is still underestimated on average by a factor of two (Table 5). SS and SP approaches agree more closely with measurements, suggesting that when considering only condensational growth, the relative dispersion at  $s_{max}$  is a better representation of the cloud droplet size distribution than a full treatment with a 1D parcel model. This implies, to first order, that spectral narrowing from condensation balances broadening from entrainment and mixing processes. Substantial efforts have been made to sample cloud data with positive

291 vertical velocity; nevertheless, a small fraction of downdrafts (especially for the weaker  
292 updrafts in CSTRIFE clouds) may exist in the dataset, and contribute to the discrepancy  
293 between observed and predicted spectral dispersion.

### 4.3. Sensitivity of relative dispersion to the effective water uptake coefficient

294 It is important to assess the sensitivity of predicted spectral dispersion on the water  
295 vapor uptake coefficient,  $\alpha$ , as the latter is a highly uncertain parameter [*Kanakidou et al.*,  
296 2005; *Ruehl et al.*, 2007] that can have a profound impact on droplet number and size.  
297 This sensitivity exercise is shown in Figure 5, where predicted spectral dispersion (using  
298 the SP approach; Table 1) is presented against observations. The range of  $\alpha$  considered  
299 (0.03 to 1.0) is based on the suggestions of *Fountoukis et al.* [2007], whom found that this  
300 range gives CDNC closure to within measurement uncertainty for clouds sampled during  
301 the ICARTT campaign. Overall, the water uptake coefficient has a minimal impact on  
302 relative dispersion since the normalized mean error in  $\varepsilon$  is  $-5.2 \pm 33.8 \%$  ( $0.1 \pm 28.3 \%$ )  
303 for CRYSTAL-FACE (CSTRIFE) and  $\alpha = 1$ ,  $-10.4 \pm 32.1 \%$  ( $-0.7 \pm 26.1 \%$ ) when  $\alpha =$   
304  $0.06$ , and,  $-14.4 \pm 31.6 \%$  ( $-2.9 \pm 25.4 \%$ ) for  $\alpha = 0.03$ .

### 4.4. Relationship between relative dispersion and droplet number concentration

305 *Liu et al.* [2006] show an increase in aerosol loading (with everything else constant)  
306 leads to a positive correlation between dispersion and droplet concentration. An increase,  
307 however, in updraft leads to a negative correlation between dispersion and droplet con-  
308 centration. We attempt to explore which effects (aerosol number or dynamics) dominate  
309 the spectral dispersion in the data of our study. Figure 6 presents observed and predicted  
310 spectral dispersion versus droplet number for all analyzed clouds from (a) CRYSTAL-

311 FACE and (b) CSTRIFE. A negative correlation is found between spectral dispersion  
312 and droplet number. This implies that, in terms of the dispersion-droplet concentration  
313 relationship, the dynamical variation in the analyzed data is stronger than the variation  
314 from aerosol loading.

#### 4.5. Prediction of $k$

315 The six approaches are evaluated in terms of their ability to reproduce the spectral shape  
316 parameter  $k$ , which is the cube of the ratio of volumetric to effective radius (the analysis  
317 could also be done instead terms of the more mathematically consistent  $\beta$  parameter of *Liu*  
318 *and Daum* [2004], defined as  $r_e$  over the droplet volumetric radius; both analysis however  
319 are equivalent). Figure 7 shows the comparison of  $k$  between the predictions and measured  
320 data.  $k$  is substantially overpredicted using the MS, MP, PS and PP approaches for the  
321 majority of the data considered, consistent with the fact that these approaches predict  
322 narrow droplet distributions relative to those measured. Compared to CRYSTAL-FACE  
323 data,  $k$  is further overestimated in the CSTRIFE data (Figure 7, Table 5), consistent  
324 with the complex dynamics in stratocumulus clouds. With the exception of SS and SP  
325 approaches, deviations in predicted  $k$  (Table 5) is too large, being comparable to the range  
326 seen for  $k$  in the whole cloud dataset. The scatter in predicted  $k$  is fairly large, even for  
327 SS, SP; whether it is important for indirect forcing assessments requires the application  
328 of a global model, and is left for a future study.

#### 4.6. Prediction of autoconversion

We now address the uncertainty in autoconversion that results from discrepancy in  
predicted spectral dispersion associated with each approach of Table 1. For this purpose,

the  $R_6$  autoconversion parameterization of *Liu and Daum* [2004] is used,

$$P_6 = \alpha_6 N^{-1/3} L^{7/3} H(R_6 - R_{6c}) \quad (16)$$

where  $P_6$  is the autoconversion rate.  $N$  is the cloud drop number concentration,  $L$  is the liquid water content.  $H$  stands for the Heaviside function which characterizes the threshold process that controls the onset of autoconversion as the sixth moment of the cloud drop distribution,  $R_6$ , is greater than the specified threshold value  $R_{6c}$ . Finally,  $\alpha_6 = \left(\frac{3}{4\pi\rho_w}\right)^2 k_2 \beta_6^6 \left(\frac{L}{N}\right)^{2/3}$ , where  $k_2 = 1.9 \times 10^{11} \text{ cm}^{-3}\text{s}^{-1}$  and  $\beta_6$  is a parameter related to the relative dispersion of a gamma distribution,

$$\beta_6 = \left[ \frac{(1 + 3\varepsilon^2)(1 + 4\varepsilon^2)(1 + 5\varepsilon^2)}{(1 + \varepsilon^2)(1 + 2\varepsilon^2)} \right]^{1/6} \quad (17)$$

The  $R_6$  parameterization is used for two reasons: *i*) it predicts the total coalescence rate and gives an upper limit for autoconversion (and discrepancy thereof), [*Hsieh et al.*, in press; *Wood*, 2005], and, *ii*) total coalescence computed from the kinetic collection equation is in good agreement with  $R_6$  for the data considered in this study [*Hsieh et al.*, in press]. Autoconversion calculations are done only for CRYSTAL-FACE clouds, given that CSTRIFE clouds are far from a precipitating state [*Hsieh et al.*, in press].

Figure 8 shows the  $R_6$  predicted autoconversion rate, calculated based on approaches in Table 1 compared to that computed from measured cloud spectra. A summary of the normalized mean error and standard deviation is given in Table 5. Because the predicted autoconversion rates are computed using the same liquid water content as the measured values, the discrepancy between the prediction and the measurement is due to the difference in cloud droplet number and relative dispersion. On average, MS, MP, PS, PP

347 underestimate  $R_6$  autoconversion rate for CRYSTAL-FACE and CSTRIFE clouds on aver-  
348 age by a factor of 3, mostly because of their underestimation of droplet relative dispersion.  
349 The autoconversion discrepancy can be large as a factor of 10, which is larger than the  
350 inherent variability of the parameterization [Hsieh *et al.*, in press]. The SS, SP tend to be  
351 in better agreement with autoconversion rate predicted from the observed spectra (Figure  
352 8) and tends to be within the estimated uncertainty of the parameterization.

## 5. Summary

353 This work examines the ability of physically-based 1-D adiabatic parcel approaches to  
354 parameterize the cloud droplet distribution characteristics relevant for computation of  
355 cloud effective radius and autoconversion in regional/global atmospheric models. A total  
356 of six approaches is examined, which are combinations of a numerical parcel model, a  
357 simplified parameterization, and their integrations over single updrafts and distributions  
358 thereof. Integrations are applied assuming that *i*) conditions at  $s_{max}$  are reflective of  
359 the cloud column, or, *ii*) cloud properties vary vertically, in agreement with 1-D parcel  
360 theory. Good agreement of droplet relative dispersion between parcel model frameworks  
361 indicates the parameterized parcel model captures most of the 1-D dynamics of the numer-  
362 ical model. When compared against in-situ cloud droplet observations obtained during  
363 the CRYSTAL-FACE and CSTRIFE missions, the distributions predicted with the parcel  
364 model (for single updrafts and distributions thereof) are too narrow, with relative disper-  
365 sion being on average a factor of 2 lower than observations. However, if conditions at cloud  
366 maximum supersaturation are used to predict relative dispersion and applied throughout  
367 the cloud column, a better agreement is seen with observations, especially if integrations  
368 are done over the relevant distribution of updraft velocity; this implies that spectral nar-

369 rowing from condensational growth is largely balanced by broadening from entrainment.  
370 The superiority of the latter method is reflected in predictions of the spectral dispersion  
371 parameter  $k$  (used for calculation of effective radius), but to a lesser degree in calculations  
372 of autoconversion; nevertheless, the simplicity of calculating spectral dispersion at  $s_{max}$   
373 is attractive. Evaluation of this method however with additional in-situ cloud datasets is  
374 required before it could be recommended for usage in large scale models.

375 Although the SS, SP methods outperformed all approaches considered, they are based  
376 on adiabatic cloud parcel theory and may still introduce unacceptable levels of uncertainty  
377 in global modeling. Given that clouds are diabatic, parameterization that accounts for  
378 some degree of entrainment (e.g., *Barahona and Nenes* [2007]) may address this issue and  
379 further improve predictions. Such an application will be the subject of future work.

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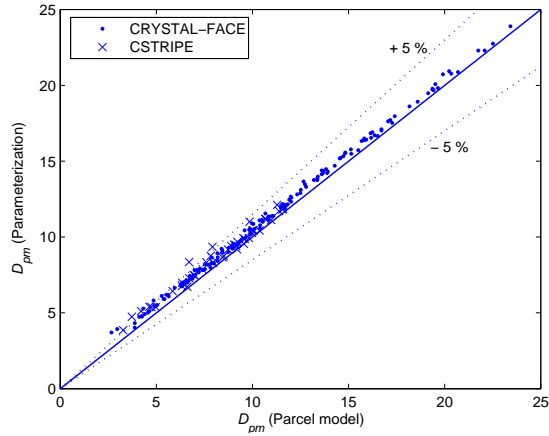


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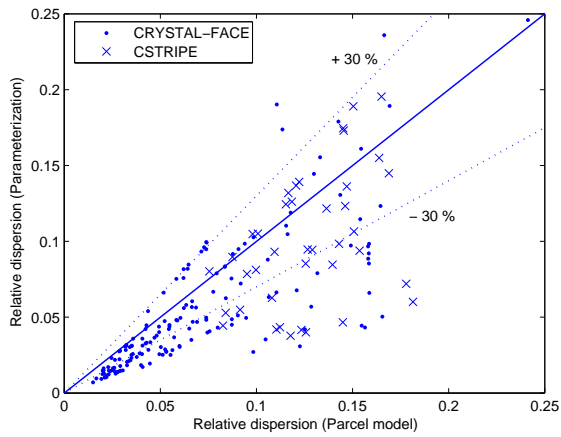
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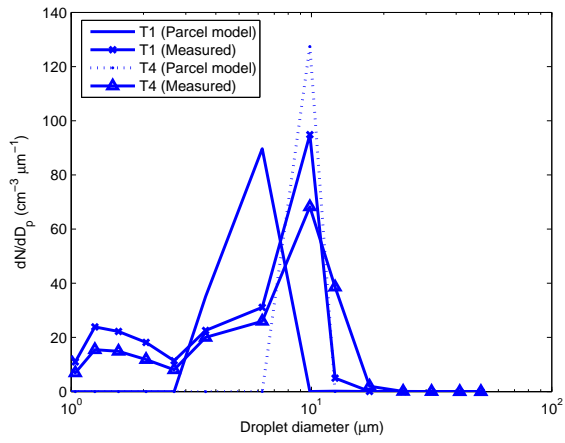
**Figure 1.** Mean droplet diameter ( $\mu\text{m}$ ) as predicted by numerical and parameterized parcel models (MS, PS approaches) based on aerosol characteristics measured during CRYSTAL-FACE and CSTRIFE.



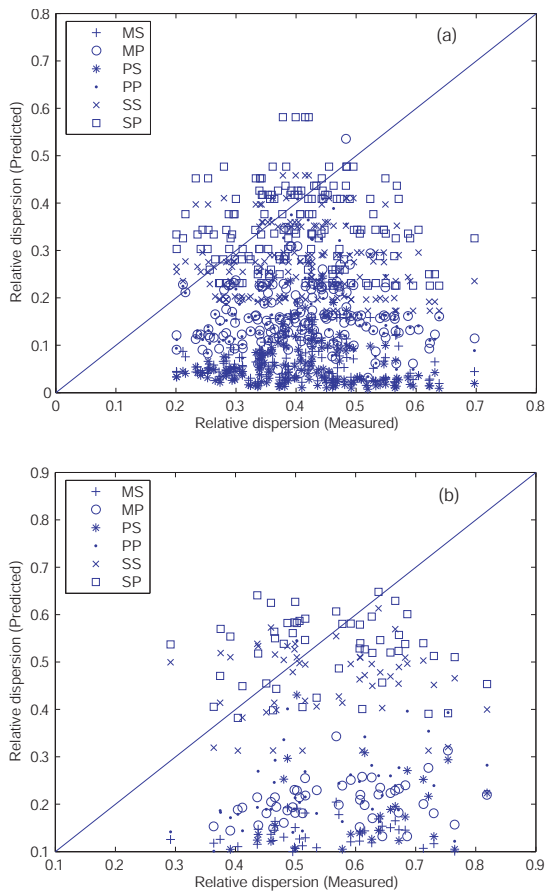
**Figure 2.** Same as Figure 1 but for prediction of relative dispersion.

**Table 1.** Approaches used to parameterize droplet size distribution characteristics.

Symbol	Description of approach
MS	Numerical parcel model, single updraft. $\overline{D_p}(z)$ , $\sigma(z)$ , and $\varepsilon(z)$ computed as described in Sections 2.1, 3.1
MP	Numerical parcel model, distribution of updrafts. $\overline{D_p}(z)$ , $\sigma(z)$ , and $\varepsilon(z)$ computed as described in Sections 2.1, 3.2
PS	Parameterized parcel model, single updraft. $\overline{D_p}(z)$ , $\sigma(z)$ , and $\varepsilon(z)$ computed as described in Sections 2.2, 3.1
PP	Parameterized parcel model, distribution of updrafts. $\overline{D_p}(z)$ , $\sigma(z)$ , and $\varepsilon(z)$ computed as described in Sections 2.2,3.2
SS	$\varepsilon(z)$ at $s_{max}$ for a single updraft apply to the whole cloud column. Parameterized parcel model used.
SP	$\varepsilon(z)$ at $s_{max}$ for a distribution of updrafts apply to the whole cloud column. Parameterized parcel model used.



**Figure 3.** Observed and predicted (using approach PS) droplet spectra for CRYSTAL-FACE cloud C12-1. T1 and T4 refer to transect 1 and 4 of C12-1 [Meskhidze *et al.*, 2005]



**Figure 4.** Prediction of relative dispersion by six approaches summarized in Table 1 compared to measurement for clouds sampled during (a) CRYSTAL-FACE and (b) CSTRIFE.



**Table 2.** Characteristics of aerosol sampled during CRYSTAL-FACE. Size distribution is composed of four lognormal modes, with modal diameter,  $D_{pgi}$  in  $\mu\text{m}$ , geometric standard deviation,  $\sigma_i$  in  $\mu\text{m}$ , and concentration,  $N_i$  in  $\text{cm}^{-3}$ . Flight naming adopted from *Meskhidze et al.* [2005].

Flight	$D_{pg1}$	$\sigma_1$	$N_1$	$D_{pg2}$	$\sigma_2$	$N_2$	$D_{pg3}$	$\sigma_3$	$N_3$	$D_{pg4}$	$\sigma_4$	$N_4$
H4-1	0.011	1.32	321.8	0.052	1.49	296.8	0.15	1.45	190.3	-	-	-
H4-2	0.012	1.35	174.7	0.064	1.83	635.8	0.49	1.23	5.3	-	-	-
H4-3	0.022	1.15	15.1	0.051	1.46	215.5	0.14	1.50	168.9	-	-	-
C4	0.019	1.31	179.4	0.049	1.44	817.7	0.12	1.53	493.0	1.55	1.30	0.5
C6-1	0.012	1.13	21.1	0.038	1.60	287.8	0.13	1.41	117.0	1.66	1.22	2.7
C6-2	0.016	1.19	31.6	0.039	1.53	280.0	0.11	1.39	117.0	1.50	1.31	0.3
C6-3	0.014	1.25	97.6	0.047	1.63	672.8	0.13	1.42	187.3	1.60	1.28	0.3
C8-1	0.019	1.31	21.9	0.104	1.99	1246.0	0.61	1.25	6.2	1.62	1.27	1.5
C8-2	0.014	1.22	68.7	0.114	2.02	1127.0	0.52	1.21	11.6	1.56	1.30	1.6
C10-1	0.015	1.40	459.2	0.035	1.24	421.8	0.11	1.71	3325.0	1.57	1.30	0.5
C10-2	0.011	1.07	47.3	0.033	1.65	3833.0	0.11	1.64	3162.0	1.52	1.30	0.6
C11-1	0.020	1.10	27.6	0.095	2.06	2143.0	0.57	1.25	3.5	1.63	1.27	0.5
C11-2	0.014	1.20	181.7	0.037	1.61	1369.0	0.12	1.78	2493.0	1.77	1.20	2.1
C12-1	0.010	1.08	16.8	0.045	1.44	211.0	0.14	1.57	270.4	1.59	1.29	0.4
C12-2	0.011	1.21	137.9	0.056	1.59	241.4	0.15	1.43	259.8	1.58	1.29	0.8
C16-1	0.013	1.10	37.3	0.031	1.57	355.3	0.12	1.52	133.8	1.51	1.31	0.4
C16-2	0.017	1.26	84.1	0.033	1.54	305.6	0.14	1.35	117.6	1.65	1.25	0.6
C17-1	0.012	1.11	67.6	0.024	1.53	803.4	0.15	1.53	235.6	1.67	1.22	1.5
C17-2	0.011	1.06	51.4	0.021	1.70	494.8	0.15	1.54	226.1	1.63	1.19	1.4
C17-3	0.011	1.05	47.4	0.025	1.79	829.0	0.14	1.61	290.0	1.74	1.21	3.5

**Table 3.** Same as Table 2, but for aerosol sampled during CSTRIFE.

Flight	$D_{pg1}$	$\sigma_1$	$N_1$	$D_{pg2}$	$\sigma_2$	$N_2$	$D_{pg3}$	$\sigma_3$	$N_3$	$D_{pg4}$	$\sigma_4$	$N_4$
CS1	0.012	1.08	29.3	0.059	1.55	1550.0	0.18	1.31	323.7	0.54	1.21	8.6
CS2	0.013	1.19	4.2	0.061	1.32	263.7	0.16	1.53	338.0	0.83	1.06	1.0
CS3	0.029	1.18	15.6	0.064	1.47	1361.0	0.92	1.18	7.0	1.42	1.27	21.7
CS4	0.011	1.03	1.1	0.058	1.40	617.5	0.15	1.46	366.4	0.53	1.44	8.6
CS5	0.014	1.04	2.9	0.060	1.47	871.7	0.15	1.42	362.5	0.66	1.14	6.8
CS6	0.013	1.06	2.2	0.055	1.39	256.4	0.16	1.55	219.4	0.70	1.21	3.0
CS7	0.011	1.03	1.4	0.064	1.43	481.8	0.15	1.44	393.7	1.55	1.25	0.3
CS8	0.014	1.04	1.4	0.095	1.95	650.3	0.58	1.05	1.2	0.72	1.13	0.4

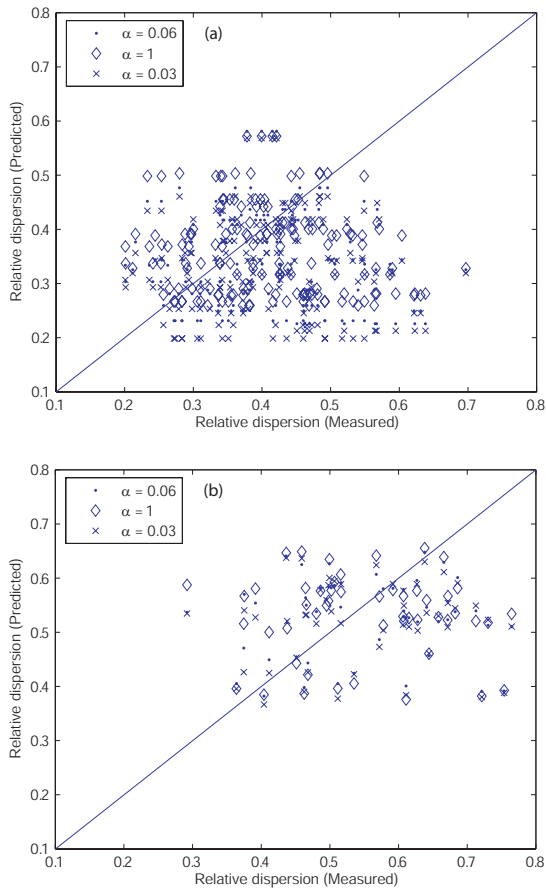
**Table 4.** Observed and modeled  $D_{pavg}$  ( $\mu\text{m}$ ),  $\sigma$  ( $\mu\text{m}$ ),  $\varepsilon$  for CRYSTAL-FACE clouds used in this study. Predictions carried out with the MS approach.

Flight (cloud)	$D_{pavg}$ obs (pred)	$\sigma$ obs (pred)	$\varepsilon$ obs (pred)
C6 (1- 1)	12.48 (11.73)	3.93 (1.45)	0.31 (0.12)
C6 (3- 2)	9.47 ( 6.93)	1.80 (1.43)	0.19 (0.21)
C8 (1- 1)	4.34 ( 4.69)	1.78 (1.45)	0.41 (0.31)
C8 (1- 2)	4.70 ( 5.17)	1.91 (1.40)	0.41 (0.27)
C10 (1-10)	8.57 ( 6.54)	1.99 (0.99)	0.23 (0.15)
C12 (1- 1)	8.47 ( 5.98)	2.19 (0.82)	0.26 (0.14)
C12 (1- 2)	9.58 ( 8.08)	2.42 (1.81)	0.25 (0.22)
C12 (1- 3)	8.53 ( 8.08)	2.27 (1.81)	0.27 (0.22)
C12 (1- 4)	9.43 ( 9.85)	2.88 (0.24)	0.31 (0.02)
C12 (1- 5)	14.47 (13.27)	3.15 (1.75)	0.22 (0.13)
C12 (2- 2)	8.31 ( 9.50)	2.04 (1.12)	0.25 (0.12)
C16 (2- 2)	13.25 (11.99)	3.43 (1.13)	0.26 (0.09)
C16 (2- 3)	14.86 (15.03)	3.94 (2.46)	0.27 (0.16)
C17 (1- 1)	9.53 (11.69)	2.70 (1.32)	0.28 (0.11)
C17 (1- 2)	13.66 (14.12)	3.24 (2.31)	0.24 (0.16)
C17 (2- 4)	11.07 ( 9.97)	3.25 (0.73)	0.29 (0.07)
C17 (2- 5)	13.95 (17.45)	5.13 (0.31)	0.37 (0.02)
C17 (3- 2)	11.49 (11.46)	2.33 (1.43)	0.20 (0.12)
H4 (1- 1)	9.27 ( 9.10)	3.22 (1.46)	0.35 (0.16)
H4 (2- 2)	8.63 ( 9.65)	1.87 (0.93)	0.22 (0.10)
H4 (2- 3)	8.98 ( 9.93)	1.89 (0.50)	0.21 (0.05)
H4 (3- 2)	10.18 (10.57)	1.93 (1.20)	0.19 (0.11)
H4 (3- 3)	9.61 ( 9.88)	1.91 (0.34)	0.20 (0.03)

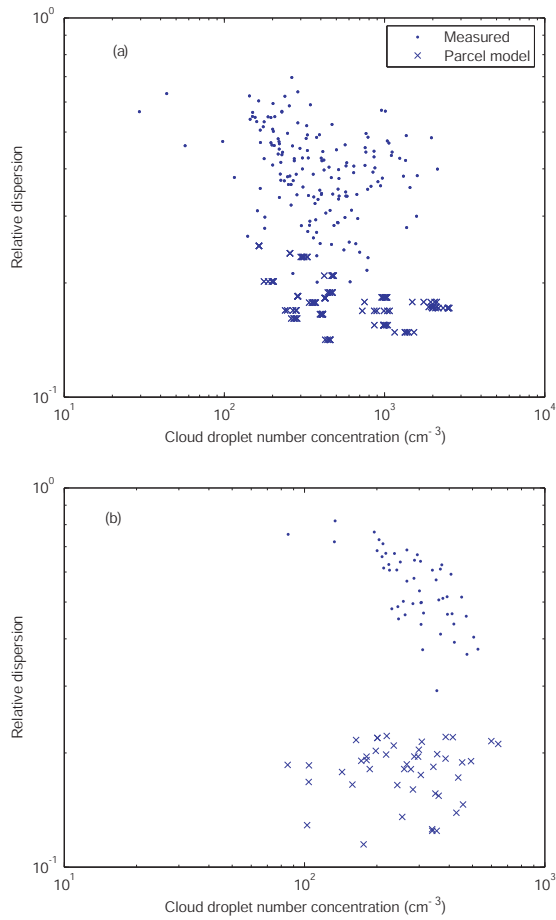
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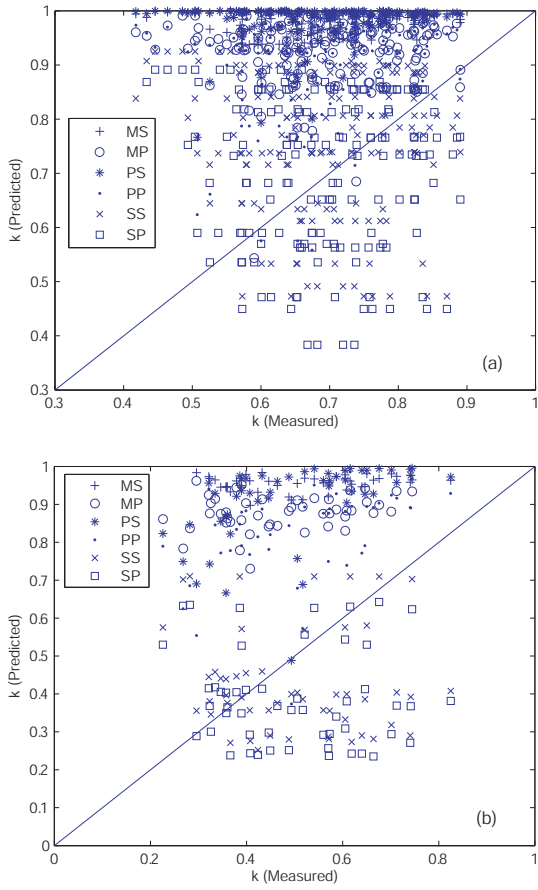
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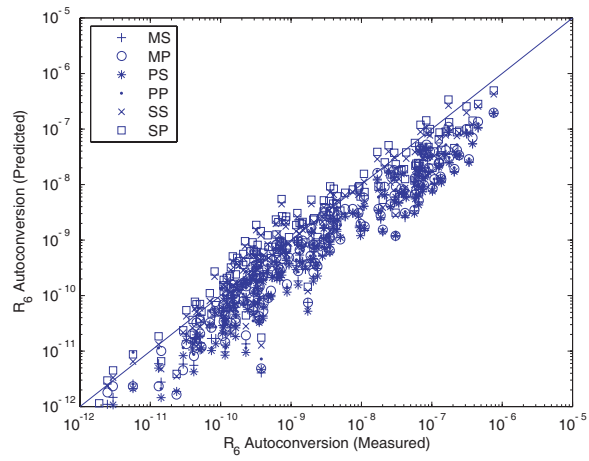
**Figure 5.** Predicted versus measured relative dispersion (SP approach) for a range of  $\alpha$  for (a) CRYSTAL-FACE, and, (b) CSTRIFE clouds.



**Figure 6.** Measured and predicted relative dispersion versus droplet number concentration for (a) CRYSTAL-FACE, and, (b) CSTRIPe clouds. The predicted data is based on droplet size distributions at  $s_{max}$  for single updraft velocity from parcel model simulations.



**Figure 7.** Observed vs. predicted  $k$  using the six approaches of Table 1 for (a) CRYSTAL-FACE, and, (b) CSTRIFE clouds.



**Figure 8.** Observed vs. predicted autoconversion rate [ $\text{kg m}^{-3} \text{s}^{-1}$ ] using the six approaches of Table 1 for CRYSTAL-FACE clouds.

**Table 5.** Normalized (%) mean fractional error (standard deviation) of predicted  $\varepsilon$ ,  $k$  and autoconversion rate for the CRYSTAL-FACE and CSTRIFE droplet distributions.

Data set	Approach	relative dispersion	$k$	$P_6$ autoconversion
CRYSTAL-FACE	MS	-85.37(32.25)	41.30(44.27)	-66.38( 24.63)
CRYSTAL-FACE	MP	-58.92(19.04)	37.04(25.20)	-58.24( 32.99)
CRYSTAL-FACE	PS	-87.42(25.32)	43.37(35.53)	-68.09( 24.19)
CRYSTAL-FACE	PP	-55.67(21.64)	34.12(25.30)	-59.45( 31.48)
CRYSTAL-FACE	SS	-28.51(27.19)	13.79(29.61)	97.26(1278.90)
CRYSTAL-FACE	SP	-10.41(32.09)	4.97(29.62)	181.70(2011.80)
CSTRIFE	MS	-91.51(51.80)	46.84(170.20)	-
CSTRIFE	MP	-62.77(8.29)	99.46( 59.90)	-
CSTRIFE	PS	-83.90(40.35)	76.89(106.70)	-
CSTRIFE	PP	-56.73(16.27)	74.22( 53.38)	-
CSTRIFE	SS	-13.19(24.82)	-3.50( 51.24)	-
CSTRIFE	SP	- 0.71(26.07)	-12.72( 46.61)	-