Aggregation and Scaling of Ice Crystal Size Distributions

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ABSTRACT

Ice particle size distributions (PSDs) can be scaled onto a single exponential distribution for a wide range of observed conditions as demonstrated using data from Atmospheric Research Measurement (ARM) cirrus uncinus, Tropical Rainfall Measurement Mission (TRMM) tropical anvils, and First International Satellite Cloud Climatology Project (ISCCP) Regional Experiment (FIRE1) midlatitude cirrus field programs. The successful scaling of the PSDs is the result of the domination of the aggregation process. The PSD is found to be a function of mean particle size and precipitation rate only. A correlation between precipitation rate and particle mass and fall speed relations is demonstrated and made use of in a semiempirical model of ice cloud that predicts the evolution of PSDs.

1. Introduction

To understand and predict the evolution of cloud systems, knowledge of the particle size distribution (PSD) is necessary. The successful representation of ice PSDs by exponential size distributions for particle diameters greater than a few hundred microns has been well documented (e.g., Ohtake 1970; Lo and Passarelli 1982; Gordon and Marwitz 1986; Houze et al. 1976; Field 1999). Particles smaller than a few hundred microns have PSDs that usually differ from that of the large particles and this has led to some workers choosing to use a modified gamma distribution to represent these distributions to capture the behavior of the smaller particles (Mitchell 1991; Heymsfield et al. 2002a). We believe that differences in the PSD behavior at small and large sizes indicates a difference in the physics controlling the evolution of the PSD. For small particles, particle production (nucleation and/or breakup) and diffusional growth dominates the evolution of the PSD, while for larger particles the evolution of the PSD is dominated by aggregation (Field 2000). In this paper we will demonstrate that the large particle ice PSD mode satisfies a scaling relation exhibited universally by aggregating systems and that the exponential distribution is a natural consequence of aggregation.

The descent of ice crystals is controlled by aerodynamic effects that lead to differential sedimentation throughout the cloud layer. This allows ice crystals to come into contact and aggregate to form larger crystals. Ice crystals tend to approach each other on ballistic or straight line trajectories, but every collision does not necessarily result in a sticking event. Therefore, to some extent aggregation of ice crystals may be limited by the number of collisions it requires to form an aggregate rather than the time required for the particles to come into contact with one another. This type of aggregation is known as reaction-limited aggregation (RLA). If the sticking efficiency upon contact between ice crystals were unity, then it would be the time it takes for the particles to come into contact with one another that would be important in controlling the aggregation rate. This type of aggregation is known as diffusion-limited aggregation (DLA; Meakin 1992, and references therein provide an overview). Computer simulation and experimental evidence (Lin et al. 1990) suggest that aggregates generate fractal structures that have a fractal dimension—$d_f = 2.1$ for RLA and 1.8 for DLA. This value is equivalent to the exponent in ice crystal mass–dimension relationships and is in good agreement with value of $\approx 2$ found for ice aggregates (e.g., Brown and Francis 1995). Researchers (see Meakin 1992) looking at colloidal systems and computer simulations also discovered that the PSDs studied in those cases evolving through the aggregation process could be scaled successfully using

$$N(M, t) = M_0(t)^{-2} f[M/M_0(t)], \quad (1)$$
where $N(M, t)$ is the time-dependent PSD as a function of particle mass, $M_0(t)$ is a mean mass representative of the PSD (e.g., the number-weighted mean mass), $\theta$ is a scaling exponent, and $f(M/M_0)$ is a scaling function that is common to all $N(M, t)$ and independent of time. Furthermore, they find that by multiplying Eq. (1) by $M_0^\theta$ and integrating over all $M$, for conservation of mass, $\theta = 2$. So, given $M_0(t)$ and the scaling function $f(M/M_0)$, the PSD $N(M, t)$ can be found.

Before attempting to apply these findings to the case of ice crystal aggregation it is important to look at the differences between the systems under examination. The computer simulations and idealized experimental exploration of aggregation involve the combination of many thousands of primary particles under average zero gravity conditions that approach the scaling form given in Eq. (1). Computer simulations of three-dimensional cluster–cluster aggregation (which is most appropriate for ice crystal aggregation) are usually carried out on a cubic lattice that is initially populated with a parent distribution of particles that can be either mono- or polydisperse. For DLA, particles are allowed to follow a random walk on the lattice until they come into contact with another particle. When this occurs the particles are joined and form a larger cluster. This is continued until the largest clusters consist of in excess of $10^6$ monomers resulting in size distributions extending over ~6 orders of magnitude. Other types of aggregation can be simulated by modifying the way clusters move on the lattice (straight lines for ballistic aggregation) or by reducing the chances of collisions resulting in a sticking event (RLA). In laboratory experiments colloids such as silica are suspended in liquid within a vessel. To mitigate the problems of sedimentation the vessel is inverted at regular intervals. Again, size distributions in the laboratory studies can extend over 5 or 6 orders of magnitude. In contrast ice crystal aggregates only contain ~50 primary particles at most after falling through several kilometers of cloud (e.g., Harimayo and Kawasato 2001). Thus, the aggregation of ice particles is only in the early stages compared to colloidal systems, and consequently, the change in size of ice crystal aggregates from cloud top to cloud base tends to only vary by 1 or 2 orders of magnitude compared to 5 or more orders of magnitude seen in the idealized cases throughout the evolution of the colloidal aggregates.

Colloidal aggregates consisting of many thousands of monomers display properties that would suggest that they are fractal structures such as a power-law density function (Witten and Cates 1986). But what is the minimum number of monomers that need to be combined before fractal properties begin to become evident? Warren and Ball’s (1989) results suggest that combinations of only a handful of monomers are required before the aggregate begins to exhibit selfsimilar properties. Ice crystals aggregates are composed of fewer monomers than their colloidal counterparts and they will be observed when they are possibly in a “pre-fractal” stage.

Fig. 1. Schematic diagram of the evolution of a spatially and temporally varying cloud consisting of two populations of ice crystals at different times indicated by the time above each rendition. The particles represented by the horizontal and vertical hatching have a fall speed of 0.8 and 1.3 m s$^{-1}$, respectively. The wind shear is 0.003 s$^{-1}$. The filled circle represents an aircraft that is descending with a fall speed of 1 m s$^{-1}$ and drifting with the wind.

The most important difference is the influence of differential sedimentation and diffusional growth effects that are present in ice cloud. The effect of sedimentation in a closed vessel would be to deplete the larger particles relative to a nonsedimenting case, but we will show that although this affects the evolution of the PSDs a scaling function can still be found. The diffusional growth effects are a more intransigent problem because of the large variation in possible growth histories that may mask the effect of aggregation at the small particle end of the PSDs.

In reality, clouds vary both spatially, temporally, and are affected by sedimentation, but “... for a source which varies slowly in time, there can be a region below that behaves as if the source were infinite and steady” (Lo and Passarelli 1982). The analysis presented in this paper makes use of the observations made during Lagrangian spiral descents and treats the data as if they were from a steady-state infinite cloud. Here, we reiterate Lo and Passarelli’s argument to defend our steady-state treatment of the data. We can introduce spatial and temporal variability in the following manner. Consider the trajectories of two sets of particles with different fall speeds (0.8 and 1.3 m s$^{-1}$; aggregate crystal sizes in the range of hundreds to thousands of microns) emanating from a steady source, 15 km in extent, that acts for 2000 s and then turns off. Figure 1 shows the regions covered by the two populations (represented by vertical or horizontal hatching) at regular intervals for a further 2000 s. The wind shear is 0.003 s$^{-1}$, and each time interval pictured has been offset horizontally for clarity. The regions where both populations are present are marked by crosshatching. If the source had been infinite, steady, and acted for a long time, the crosshatched region would have been very extensive, allowing an aircraft to sample the cloud at leisure. However, for the spatial and temporally varying case, as time goes on,
the region of overlap diminishes in size. But it can be seen that, in principle, if an aircraft samples with a descent rate close to the average particle fall speed and advects with the ambient wind field, then it can remain in the crosshatched region (filled circle). In this way the effects of spatial and temporal variability can be minimized and we can treat the observations obtained from the Lagrangian descent as if they were obtained in a steady-state cloud. Diffusional growth of ice crystals or new nucleation of ice particles should not affect this argument as long as the change in mean particle size due to these effects is smaller than that due to aggregation. The working hypothesis of this paper is that, for the observations presented here, aggregation dominates the evolution of the ice PSD. The ability to scale the PSDs and success of the semiempirical modeling presented later are a test of this hypothesis.

In this paper, section 2 will explore the scaling idea in the context of ice clouds. Section 3 will introduce the datasets used. Section 4 will apply the scaling arguments to a single case study. Section 5 will extend the analysis to incorporate data from other flights. In section 6 the mass and fall speed power-laws will be presented. Sections 7 and 8 will introduce and test a semiempirical model of PSD evolution. Section 9 will provide a summary and conclusions.

2. Scaling and steady-state ice mass flux density

Bearing in mind the argument that we are able to treat regions of spatial and temporally varying cloud as if it were in steady state when we sample it with a Lagrangian descent, then we can proceed as follows. Consider the following scenario: in an ideal steady-state cloud, mass is continually replaced at cloud top at a constant rate and transported to lower altitudes by sedimentation while aggregation is occurring. For this scenario no diffusional growth, sublimation or melting is considered. It can be seen that aggregation will lead to larger particles and hence greater fall speeds lower down in the cloud and so ice water content (IWC) will decrease with increasing depth, but ice mass flux density will be conserved.

Aggregation phenomena have been studied extensively using computer simulations [see Meakin (1992) for an overview] and it has been shown that the evolution of PSDs controlled by aggregation can be scaled successfully using Eq. (1) which can be rewritten as

$$N(D, t) = D_1(t)^{-\theta} g[D/D_1(t)],$$

(2)

where $N(D, t)$ is the PSD at time $t$, $D_1(t)$ is an average diameter for the PSD at time $t$, $\theta$ is a scaling exponent, and $g[D/D_1(t)]$ is the scaled function. In this paper the average diameter $D_1(t)$ is defined as half the ratio of the second and first moments of the PSD. In a nonsettling system a value for $\theta$ can be found by conservation of mass, but in a settling system that has reached steady state it is ice mass flux density that is conserved. So, plotting $D_1(t)^{\theta} N(D, t)$ versus $D/D_1(t)$ will result in the collapse of the PSDs onto a single curve. Sekhon and Srivastava (1970) originally attempted a scaling of PSDs of the few spectra available to them but used conservation of IWC to facilitate the scaling.

Given the scaling relation the exponent $\theta$ can be found from conservation of ice mass flux density $\phi$:

$$\phi(t) = \int_0^\infty N(D, t) m(D) \nu(D) \, dD,$$

(3)

where $m(D)$ and $\nu(D)$ are the mass and fall velocity of a particle with diameter $D$. Assuming the following power laws represent the mass and fall velocity: $m = aD^b$, $\nu = aD^b$, and letting $aa = \eta$. Eq. (3) becomes

$$\phi(t) = \int_0^\infty \eta D_1(t)^{-\theta} D^{b+\theta} g \left[ \frac{D}{D_1(t)} \right] \, dD,$$

(4)

and on letting $x = D/D_1$ and rearranging we obtain

$$\phi(t) = D_1(t)^{\theta+b+1-\theta} \int_0^\infty \eta x^{\theta+b} g(x) \, dx.$$  

(5)

The integral on the right-hand side is constant and so the mass flux density $\phi(t)$ will be constant if

$$\theta = b + 1.$$  

(6)

The function $g(x)$ is a solution to the Smoluchowski equations. It is known from numerical simulation (e.g., Leighton 1980) that $g(x)$ can have the form

$$g[D/D_1(t)] = AD^\nu \exp[-D/D_1(t)],$$

(7)

where $\nu$ controls the shape of the PSD at small sizes. The normalization factor $A$ can be obtained by substituting Eqs. (6) and (7) into (4) to give

$$A = \frac{\phi D_1(t)^{-\nu}}{\eta \Gamma(\theta + \nu)},$$

(8)

which results in the following expression for $N(D, t)$ when Eqs. (2), (7), and (8) are combined:

$$N(D, t) = \frac{\phi}{\eta \Gamma(\theta + \nu)} D_1(t)^{-\theta} \left[ \frac{D}{D_1(t)} \right]^{\nu} \exp \left[ -\frac{D}{D_1(t)} \right].$$

(9)

where $\Gamma$ is the gamma function. Unfortunately, the small end of the PSD is affected by other physical processes that often result in the small end of the PSD being much steeper than the larger end so it is difficult to assign a value for $\nu$. Estimates of $D_1$ for an arbitrary value of $\nu$ are hard to obtain because of the lack of knowledge of the PSD of the aggregating population for small particle sizes. Setting $\nu = 0$ allows a consistent estimate of $D_1$ obtained from an exponential distribution. Therefore, for simplicity we set $\nu = 0$ to obtain

$$N(D, t) = \frac{\phi}{\eta \Gamma(\theta)} D_1(t)^{-\theta} \exp \left[ -\frac{D}{D_1(t)} \right].$$

(10)
Equation (10) can be compared directly with standard form of the exponential distribution used for PSDs that was introduced by Gunn and Marshall (1958) to represent observed snow PSDs:

\[ N(D, t) = N_0(t) \exp[-\lambda(t)D], \]

where \( N_0 \) is the y intercept of the exponential distribution and \( \lambda \) is the gradient in loglinear space. By inspection it can be seen that

\[ \lambda = \frac{1}{D_1(t)} \quad \text{and} \quad N_0 = \frac{\phi D_1(t)^{-\theta}}{\eta \Gamma(\theta)}. \]

If we had initially assumed an exponential distribution [Eq. (11)] for the PSD, and integrated to obtain the ice mass flux density, we would have arrived directly at \( N_0 \) [cf. Mitchell 1988, Eq. (15)], but by not assuming a form for the PSD we were still able to determine the value that is required for the exponent \( \theta \) to conserve ice mass flux density. It will also be shown in section 4 that no assumption needs to be made about the form of the PSD we were still able to determine the form for the exponential distribution used for PSDs that couples \( \lambda \) and \( N_0 \) together and hence concentration \( (N_0/\lambda) \) and mean particle diameter \((D_0)\), to obtain a scaling of the PSDs.

Combining Eqs. (12) and (13) provides the following relation:

\[ d \ln N_0 \propto d \ln \phi + \theta d \ln \lambda. \]

Therefore, for constant ice mass flux density \((d \ln \phi = 0)\) the gradient of \( N_0 \) versus \( \lambda \) in log–log space will yield the value of \( \theta \). The aggregation process strongly couples \( \lambda \) and \( N_0 \) together and hence concentration \((N_0/\lambda)\) and mean particle diameter \((D_0)\).

Up to now we have used the term ice mass flux density because we wished to emphasise the physical quantity being conserved, but from now on we will use the term precipitation rate instead of ice mass flux density because it is a more familiar term when we deal with the observations. Ice mass flux density \( \phi \) is linked to precipitation rate \( P \) by \( P = \phi \rho_v \), where \( \rho_v \) is the density of liquid water.

Lo and Passarelli (1982) pointed out that straight lines in \( \lambda-N_0 \) space represented a constant moment of an exponential distribution. Hence, a family of lines representing constant precipitation rate \( (P) \) can be plotted. Ideal steady-state aggregating-only clouds would have \( \lambda-N_0 \) trajectories that followed these lines. In practice there is usually some large-scale atmospheric motion that will lead to diffusional growth or loss within the cloud. The diffusional growth will lead to motion in \( \lambda-N_0 \) to cross from one locus of constant precipitation rate to another.

3. Datasets used

Data from five spirals were acquired in midlatitude ice clouds during the First International Satellite Cloud Climatology Project (ISCCP) Research Experiment (FIRE I) in Wisconsin (fall 1986). Cloud-top temperatures were in the \( -35^\circ \text{C} \) to \( -40^\circ \text{C} \) range and base temperatures were close to \(-20^\circ \text{C}\). Data from seven spirals were collected during the tropical rain measuring mission (TRMM), six in a campaign near Kwajalein [Kwajalein Experiment (KWAJEX), summer 1999] and a seventh in Brazil (spring 1999). Cloud top temperatures ranged from about \(-50^\circ \text{C} \) to \(-15^\circ \text{C} \) and data were collected to temperatures above \(0^\circ \text{C}\). One spiral was used from the midlatitude Atmospheric Research Measurement (ARM) intensive observation period (IOP) in Oklahoma (spring 2000). Cloud-top temperatures were close to \(-50^\circ \text{C} \). The reader is referred to the following recent studies that describe these datasets in more detail: Heymsfield et al. (2002b) for the ARM campaign, Heymsfield and Miloshevich (2003) for the FIRE I and ARM, and Heymsfield et al. (2002a) for the TRMM campaigns.

Particle size distributions used for this study spanned a wide range of sizes, from tens of microns to centimeters, and included direct measurements of IWC and high-resolution particle imagery (see references above for more details of the instruments). The PSD from the airborne observations were measured by Particle Measuring Systems (PMS) 2D-C probes (nominally \( \sim \)50 to 1000 \( \mu \text{m} \) in 25- \( \mu \text{m} \) intervals for FIRE I and 30- \( \mu \text{m} \) intervals for TRMM and ARM), 2D-P probes (100 \( \mu \text{m} \) to \( > \)3000 \( \mu \text{m} \) in 100- \( \mu \text{m} \) intervals for FIRE and 200- \( \mu \text{m} \) for ARM), and a Stratton Park Engineering Company (SPEC) high-volume precipitation spectrometer (HVS) probe (1 mm to \( > \)3 cm in 0.2-mm intervals; TRMM). All of the parameters used (IWC, \( P \), \( D_0 \)) were estimated from average PSDs that were obtained over each 0.5 km (5 s) of flight distance for the FIRE dataset, 1 km (about 7 s) of flight distance for the TRMM flights, and about 0.7 km (5 s) for the ARM flights.

Slope \( (\lambda) \) and intercept \( (N_0) \) parameters were fitted to the \( -1 \) km PSDs using a moment method described in Heymsfield et al. (2002a). This method has the advantage of weighting the fit to the larger end of the PSD and minimizing any contamination effects from the smaller end of the PSD, which is believed to be dominated by different physical processes.

Heymsfield et al. (2002b) describe a method of estimating crystal mass and fall speed by utilizing cross-sectional area as well as maximum dimension information. The computation of precipitation rate using this method is crucial because it decouples the estimates of mass and fall speed from the power-law relations assumed in section 2. They propose that the effective density of an ice particle \( (\rho_v = 6 \rho m / \pi D^3) \), where \( m \) is the crystal mass and \( D \) is the crystal maximum dimension) is related to the both the maximum crystal dimension and the crystal’s projected area ratio \( A_a \) (\( = 4 A / \pi D^2 \), where \( A \) is the projected cross-sectional area). For example a column will have a smaller \( A_a \) than a plate for the same maximum dimension and also has a lower
effective density. For each particle the mass and fall speed can be estimated and then summed to find the IWC or precipitation rate of a measured PSD. Heymsfield et al. (2002b) showed through a variety of data sources that included theoretical particle models, estimates of aggregate crystal volume from detailed inspection of cloud particle imager (CPI) images, and collection of ice crystals at ground level, relationships between the equivalent density drag coefficients $D$ and $A_e$. Subsequent comparison of derived ice water contents from 2D probes using this two parameter method with that obtained from direct measurements made with a counterflow virtual impactor (CVI) probe showed agreement to within 20%. Similarly, good agreement was found between estimates and measurements of fall speed when using the two-parameter method.

The two-parameter method of estimating crystal mass and fall speed is different to the standard power-law relations $(m - D, v - D)$ in that it is not necessary to make any assumption about the habit of the ice crystals or the shape of the PSD. In this analysis we have used the estimates of IWC and precipitation rate obtained from the two-parameter method as being accurate indicators of the actual values. In the scaling analysis, we have made use of the power-law relations $(m - D, v - D)$ in combination with an exponential PSD to independently describe the ice crystal properties because they are believed to be a natural consequence of aggregate formation.

In the following sections the distribution variable $N$ in Eq. (10) is identical to the binwidth normalized concentration $dN/dD$. We are unable to measure the elapsed time experienced by different PSDs, so altitude must be used as a proxy; that is, older aggregates are found lower down in the cloud. Therefore, all of the $t$ subscripts are now replaced by height $z$. Unfortunately, we have to accept that there are problems associated with assuming this because of differential sedimentation, but as we argued in the introduction, the effects should be minimized by the Lagrangian spiral sampling technique.

### 4. ARM case example

Data (0.7-km averages) from a Lagrangian spiral descent carried out on 9 March 2000 for the ARM field program are presented to demonstrate the scalability of ice PSDs. This flight was carried out in a line of cirrus uncinus and started at cloud top ($-50^\circ C$, 9500 m) and descended to cloud base ($-28^\circ C$, 6600 m). Figure 2a shows horizontal position data for this Lagrangian spiral descent. The solid line in Fig. 2b is the IWC measured by a CVI probe and the dotted line is the IWC estimated from the two-parameter method, which are depicted showing good agreement in Fig. 2c. The IWC– and precipitation–altitude (Fig. 2d) plots are oscillatory, indicating a variation in the horizontal structure encountered. This structure is best visualized by ordering the IWC and precipitation rate $P$ as a function of the aircraft heading and altitude (Figs. 2e and 2f). The coherent vertical structure exemplified by the sharp edge in IWC and $P$ at $140^\circ$ indicates that the aircraft was successfully sampling in the air parcel relative frame and that the PSDs sampled near cloud top evolve down through the cloud into the PSDs sampled at cloud base.

Qualitative evidence for the action of the aggregation process can be seen in the CPI imagery (Fig. 3). Example images are arranged as a function of height and size, but no attempt has been made to represent the relative abundances. There are some aggregates visible in the middle column (particle diameters between 400 and 600 μm), but the largest aggregates appear in the right-hand column (diameters greater than 800 μm). It should be borne in mind that bigger aggregates exist but the CPI can only image particles up to 2000 μm in diameter. These images indicate that, for this case, bullet rosette crystals are aggregating together to form larger more complex particles.

Below 9000 m for this ARM case, $P$ changes only slightly and so provides a test of the PSD scaling. Figure 4a shows PSDs averaged over each loop below 9000 m where the aircraft heading was greater than 300° or smaller than $140^\circ$ (−15 km of aircraft track). Similarly, IWC, $P$, $D_1$, and $N$ were averaged over the same interval. The individual PSDs display bimodal spectra with a mode that increased from 200 to 500 μm with depth from cloud top. Likewise, the PSDs above 600 μm become broader with increasing depth from cloud top. Figure 4b shows that the mean value of $P$ appears to increase slightly with depth. The variation indicated by the bars representing one standard deviation is quite broad, and to demonstrate how the data can be collapsed using the scaling arguments given in section 2, we will assume that $P$ is approximately constant over the depth of cloud considered. Figure 4c shows the $\lambda$–$N_1$ plot for this case that has the gradient $\theta = 3.9 \pm 0.2$ in log-log space. Using this value for $\theta$ the scaled spectra collapse onto a well-defined function and even the mode between 200–500 μm lines up quite well (Fig. 4d). It can be seen that just using the scaling factor $D_1^x$ results in the data collapse onto a well-defined exponential curve for $D/D_1 > 2$ confirming that the choosing $g(x)$ to be an exponential function [Eq. (7)] is satisfactory. Further evidence of the effects of aggregation in this case can be seen in the evolution of $D_1$ and number concentration as a function of altitude (Figs. 5a,b). Again the data are loop average values carried out between a heading of $300^\circ$ and $140^\circ$. The number-weighted mean diameter shows an increase in size by a factor of $\sim 3$ over the depth of the cloud, coincident with a decrease in concentration by a factor of $\sim 10$.

For constant $P$ we know that $\theta = 1$ is equal to the sum of the exponent in the mass–diameter relation, and the exponent in the fall speed–diameter relation. Heymsfield et al. (2002b) show that for aggregates of rosettes, the equivalent particle density is $\sim D^{-0.96}$ and hence, the mass $\sim D^{2.04}$. The exponent in the fall speed relation
is estimated to be 0.68 and so the predicted value of $\theta$ is 3.7, in agreement with the observed value.

5. Application to other data

If regions where the precipitation rate is constant can be located the $\theta$ value can be estimated using the gradient found from the $\lambda-N_0$ plot [Eq. (14)]. In practice the precipitation rate usually varies throughout the depth of the cloud because of diffusional growth effects and so, to overcome this complicating factor, $\sim 1$ km PSDs (and hence estimates of $\lambda$ and $N_0$) were logarithmically binned [$P$ bin ends: 0.001(1.7)$^x$ mm h$^{-1}$; $x = 0, 1, 2, \ldots$] according to precipitation rate obtained from the two-
parameter method (Heymsfield et al. 2002b) for each flight. Values of $\theta$ and an error estimate were obtained from least squares fits to $\lambda$ and $N_0$ in log space for each precipitation rate bin and flight.

In the following analysis of the data two assumptions have been made: (i) when selecting PSDs with the same precipitation rate from the same descent it is assumed that it is equivalent to observing a single PSD at dif-
ferent stages undergoing aggregation only; (ii) $\beta$ and $b$ are constant over a large range of $D_1$; that is, aggregates of two or three crystals have similar (same) $\beta$ and $b$ values to larger aggregates. Assumptions (i) and (ii) imply that for constant precipitation rate plots of $\lambda$ and $N_0$ in log–log space will lie on straight lines.

Figure 6 shows the $\theta$ values as a function of $P$ for the 14 ARM, TRMM, and FIRE I cases considered. The solid line is an exponential function fitted in semilog space (given in Table 1) capped at $\theta = 5$ (spherical particles: $\beta = 3$, $b \approx 1$) for all of the data. An exponential form was chosen because we wanted a function that would asymptote to a constant value of $\theta$ for large precipitation rates. Figure 7 shows all the data collected together with the fitted exponential function (same as in Fig. 6). It is noted that the $\theta$ values obtained for the ARM flight when constant $P$ values are selected are greater than the one obtained in section 4. This is because, although there was not much diffusional growth in the lower part of the ARM flight, it was still enough
to slightly lower the value of $\theta$ obtained. The residual variation in $\theta$ from the line fit has a standard deviation of 0.6.

What Fig. 7 shows is that it is possible to choose a $\theta$ value derived from a set of $m - D, n - D$ power-law relations combined with an exponential PSD that will give estimates of $P$ consistent with those determined from the two-parameter method using the actual measured PSD described in section 3. We know that $P$ and $\theta$ are physically linked because the value of $\theta$ is dependent upon the conservation of ice mass flux density. However, we cannot necessarily say beforehand how $\theta$ will vary with $P$ if at all. Figure 7 shows that $\theta$ is correlated with $P$ and that $\theta$ values start off high at low $P$ values and eventually attain a value of 3–4 for $P > 0.5$ mm h$^{-1}$. We suggest that the value of $\theta$ is controlled directly by the particle habit, which may be selected to some extent by the process of binning $P$ (e.g., synoptically generated cirrus has low $P$ values) and the amount of aggregation that has occurred. The scatter in $\theta$ probably reflects the variation in primary crystal habit and initial growth conditions soon after nucleation. An additional factor is that high rainfall rates tend to be correlated with larger $D_1$ values. Hence, mass and fall velocities are weighted by the larger particles that also tend to have the lower $\beta$ and $b$ values.

Given the precipitation rates and the parameters derived from the exponential fits to the PSDs, Eq. (13) can be used to obtain the product of the prefactors to the mass–diameter and fall speed–diameter relations. This combined prefactor $\eta$ is depicted in Fig. 8 as a function of $\theta$. There is very good correlation between $\theta$ and $\eta$ due to the fact that the degree of aggregation and habit influences the prefactor as well as the exponent in the mass–diameter and fall speed–diameter relations.

The variation in the residual values of $\eta$ after the line fit is removed is a factor of 2.

Now it can be seen that, because $\eta$ and $\theta$ can be represented by functions of precipitation rate, then Eq. (10) describing the time-dependent evolution of the PSD is a function of precipitation rate and $D_1$ only. This has implications for radar reflectivity estimates, $Z$, of $P$ and means that $Z$ is a function of $P$ and $D_1$, and not $P$ alone, just as Jameson and Kostinski (2001) point out.

Using the expressions for $\eta$, $\theta$ (Table 1) in combination with estimates of $P$ obtained from the two-parameter method for each of ~2500 ~1 km PSDs we have attempted to scale all of the measured spectra onto a single curve by multiplying $N$ by $\eta f^\theta D_1/\phi$. We can also obtain the best possible scaled function, given the inherent natural variability, by multiplying $N$ by the measured $\eta f^\theta D_1/\phi$, that is, $1/N_0$. Figure 9 is a two-dimensional histogram of all the scaled PSDs. The binned frequencies have been normalized for each $D/D_1$ interval. The dashed line is the overplotted exponential function $\exp(-D/D_1)$ that onto which the scaled PSDs are expected to collapse. The contour encompassing the greatest area contains 80% of the data for each $D/D_1$ interval and each subsequent contour contains 20% less. The exponential function fits well for $1 < D/D_1 < 9$. For $D/D_1 < 1$ the small end of the PSD is not dominated by the effects of aggregation and so the scaled function is not expected to apply. For $D/D_1 > 9$, the data has collapsed onto a curve, which appears slightly superexponential at these large sizes. The lower panel shows the same data but with the exponential trend removed to leave the residual values. For $D/D_1 \approx 10$, the particle diameters are >6 mm. Choosing different values for $\nu$ in Eq. (9) does not result in a significantly better fit to the data. The reason for the deviation of the

![Fig. 5](http://journals.ametsoc.org/doi/abs/10.1175/1520-0469(2003)060<0544:AASOIC>2.0.CO;2?journalCode=josd)
scaled PSDs from the exponential at large $D/D_1$ is not clear but it may be a sampling problem at low particle concentrations, an instrumental effect or an artifact of the method used to obtain the slope of the PSD in log-linear space. Another possible problem is that diffusional growth is affecting the form of the PSD and contaminating the effect of aggregation alone. Overplotted as a thick solid contour is the 80th percentile of the data using the measured value of $N_0$ to scale the data. It can be seen that scaling using the parameterized $\theta$ values gives very similar results to the best possible scaling. Accuracy in estimating IWC and radar reflectivity depends upon the how well the PSD is represented in the parts of the PSD that contribute most to the IWC and radar reflectivity. For IWC, it is where $N(D)D^\beta$ reaches a maximum and where $N(D)D^{2\beta}$ reaches a maximum for radar reflectivity. Because $\beta$ can be found in the range 2–3, the accuracy of the ice PSD at $D/D_1 = 2–3$...
TABLE 1. Useful generalized relationships; $P$ (mm h$^{-1}$), $m = aD^b$, $v = aD^b$, cgs units.

<table>
<thead>
<tr>
<th>Variable</th>
<th>Expression</th>
<th>Range</th>
<th>Uncertainty*</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\theta$</td>
<td>$= \beta + b + 1$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\beta$</td>
<td>$= \exp[-1.43 \log_{10}(P)] + 2.8^{**}$</td>
<td>2.8–5</td>
<td>$1\sigma = \pm 0.6$</td>
</tr>
<tr>
<td>$\eta$</td>
<td>$= \exp(3.01\theta - 10.5)$</td>
<td>0.1–400</td>
<td>Factor of 2</td>
</tr>
<tr>
<td>$b_f$</td>
<td>$= 0.638$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\beta$</td>
<td>$= \theta(1 + b_f)$</td>
<td>1.8–3.1</td>
<td>$1\sigma = \pm 0.4$</td>
</tr>
<tr>
<td>$b$</td>
<td>$= b_f\theta(1 + b_f) - 1$</td>
<td>0.0–1.0</td>
<td>$1\sigma = \pm 0.4$</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>$= \exp[4(\beta - 3.5)]$</td>
<td>0.0003–0.01</td>
<td>Factor of 2</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>$= \eta/a$</td>
<td>100–700</td>
<td></td>
</tr>
</tbody>
</table>

* Uncertainty in $\theta$ divided equally between $\beta$ and $b$.

** For $P < 0.04$, $\theta = 5$.

6. Useful generalized parameters

To utilize the power-law equations relating the particle diameter to particle mass and fall speed the four parameters, $\alpha$, $\beta$, $a$, $b$ need to be distinguished. It should be borne in mind that disentangling $\alpha$, $\beta$, $a$, $b$ will produce values that are subject to quite large uncertainties (see Table 1). Nevertheless, following the treatment by Mitchell (1996), in which the Reynolds Re and Best $X$ numbers are related through a power law of the form $Re = a_fX^b$ ($a_f$, $b_f$ are constants), an ice crystal fall speed relation was developed:

$$v = a_f\zeta \left(\frac{2ag}{\rho_a\zeta^2}\right)^{b_f} D^{b_f(\beta + 2 - \sigma) - 1},$$  \hspace{1cm} (15)

where $\zeta$ is the viscosity of air, $g$ is the acceleration due to gravity, $\rho_a$ is the density of air, and $\gamma$ and $\sigma$ are constants in a power law relating the particle diameter to cross-sectional area ($= \gamma D^\sigma$). It can be seen that there exists relationships between the exponents and prefactors of the $m - D$ and $v - D$ power laws:

$$b = b_f(\beta + 2 - \sigma) - 1 \hspace{1cm} \text{and} \hspace{1cm} (16)$$

$$a = a_f\zeta \left(\frac{2ag}{\rho_a\zeta^2}\right)^{b_f} D^{b_f(\beta + 2 - \sigma) - 1},$$  \hspace{1cm} (17)

If we set $\sigma = 2$, which empirical relations find it close to (e.g., Mitchell 1996), we find that

$$b = b_f, \hspace{1cm} (18)$$

and hence

$$\beta = \frac{\theta}{1 + b_f}, \hspace{1cm} (19)$$

$$b = \frac{\theta b_f}{1 + b_f} - 1. \hspace{1cm} (20)$$

Figure 10 is a plot of $\theta$, $\beta$, and $b$ as a function of $P$ when Eqs. (19) and (20) are used in conjunction with the expression for $\theta$ given in Table 1.
Mitchell also predicts a link between $\alpha$ and $\sigma$ [Eq. (17)], but it is a more complicated function of environmental variables. A simpler approach to disentangling $\alpha$ and $\sigma$ from $\eta$ is to find a relation between one of the prefactors and exponents. We have attempted to do this with some data from Kajikawa (1989) and Heymsfield et al. (2002b) that present mass–diameter power laws for aggregates. A curve was fitted to a scatterplot of the exponents and prefactors in the $m – D$ power law (see Table 1).

With the separation of the variables required for the mass–diameter and fall speed–diameter power-law relations (see Table 1 for expressions for $\alpha$, $a$, $\beta$, $b$ and other useful parameters), conversion between IWC and precipitation rate can be found as a function of $D_1$ (Fig. 11):

$$IWC = \int_0^\infty \alpha D^\theta N_0 \exp \left(-\frac{D}{D_1}\right) dD = \frac{\alpha \eta \Gamma(\beta + 1)D_1^{-b}}{\eta \Gamma(\theta)}.$$

(21)

The ability to convert between IWC and precipitation rate should be of use to cloud modelers, but can only
be accomplished if the variable $D_1$ is prognosed along with IWC.

7. Semiempirical model of aggregation and diffusional growth

Lo and Passarelli (1982) first noted that the derived constants from the exponential fit, $N_0$ and $\lambda$, could be plotted as a trajectory in $\lambda$–$N_0$ phase space to chart the evolution of the PSD. Typically, they noted that when $N_0$ decreased, so did $\lambda$ until either cloud base, the melting layer, or a value for $\lambda$ of $10 \text{ cm}^{-1}$ was reached. Passarelli suggested that the simultaneous decrease of $N_0$ and $\lambda$ was due to aggregation. Wolde and Vali (2002) recently collated a range of studies and showed that the evolution trajectory of PSDs in $\lambda$–$N_0$ space followed a power law of the form $N_0 \propto \lambda^p$, where $2.0 < p < 3.0$. The $\lambda$–$N_0$ phase space is a useful way to visualize a PSD and we would like to build on the work of Lo and Passarelli and Mitchell in trying to understand the variation in observed $\lambda$–$N_0$ trajectories.

Plots of spectral evolution in $\lambda$–$N_0$ space are useful because these parameters can be derived directly from the measurement of the PSD, but perhaps more fundamental is $P$–$D_1$ space in which the ordinates are the two variables controlling the PSD [Eq. (10)]. Both types of plots will be shown in the next section.

In order to compute how $\lambda$ and hence $N_0$ change we need to predict how $\lambda$ will change due to aggregation and diffusional growth. For the change in $\lambda$ due to diffusional growth and aggregation, we have adopted the results of Mitchell (1988) who provided a solution to the equations governing the evolution of snow PSDs through diffusion, sedimentation, and aggregation by utilizing a conservation of moments method for a steady-state conditions. We have not considered explicitly the role of primary or secondary multiplication, but if any ice is generated by this route it will increase the ice water content and consequently the precipitation rate. The subsequent changes in the $\lambda$ and $N_0$ result in an increase in the total number concentration of the exponential size distribution that can accommodate any production of new particles. However, as we maintain, even if new particle production is occurring the form of the size distribution will still be dominated by the action of aggregation.

Mitchell’s solution requires the values for $\beta$ and $b$ to be chosen to best suit the case. Separation of the variables discussed in section 6 as a function of precipitation rate means that a steady-state cloud can be defined by the precipitation rate profile and an estimate of $D_1$ at any height.

Given Eqs. (19) and (20), the change in $\lambda$ due to diffusional growth and aggregation as a function of height can be computed using Mitchell’s (1988) solution for a steady-state cloud

$$\frac{d\lambda}{dz} = c_1 \frac{\lambda}{\phi} \frac{d\phi}{dz} - c_2 E_\phi \lambda^{\gamma - 2},$$

where $E_\phi$ is an aggregation efficiency and
Fig. 14. TRMM flight on 11 Sep 1999: $P$–$D_1$ plot. Plus symbols are observations; solid line is model results. The dotted line represents contours of constant radar reflectivity.

Fig. 15. $N_0$–$\lambda$ plot of model–data intercomparison for the 9 Mar 2000 ARM flight. The data are plotted as plus symbols; the model is the solid line.

Fig. 16. ARM flight on 9 Mar 2000. (top) Parameter $\lambda$ vs height plot. Thin black line is the observations; thick gray line is model results. (bottom) Parameter $N_0$ vs height plot. Thin black line is the observations; thick gray line is the model results.

$$c_1 = \frac{1}{\beta} \left[ 1 - \frac{2\Gamma(\beta + \delta + 1)\Gamma(\beta + b + 1)}{\Gamma(\delta + 1)\Gamma(2\beta + b + 1)} \right],$$

$$c_2 = \frac{\pi\eta}{4\beta \eta \Gamma(\beta + b + 1)\Gamma(2\beta + b + 1)},$$

$$L = \int_0^\infty \int_0^\infty x^\delta y^b (x + y)^{\alpha} \left| x^b - y^b \right|$$

$$\times \exp(-x - y) \, dx \, dy,$$

where $\delta$ is a diffusional growth coefficient ($\delta = 1$, following Mitchell 1988), and $x$ and $y$ are normalized particle sizes: $x = D_x / D_1$; $y = D_y / D_1$. The first term on the right-hand side of Eq. (22) is the change in $\lambda$ due to diffusional growth and the second term is the change due to aggregation. We note here that the collection kernel $K(D_x, D_y)$ for the aggregation is assumed to be for simple hydrodynamic capture:

$$K(D_x, D_y) = E_a \pi(D_x + D_y)^2 |v_x - v_y| / 4,$$

where $v_x$ and $v_y$ are the terminal velocities of the ice crystals of sizes $D_x$ and $D_y$. Any shortcomings of this simple kernel will be accommodated in the value of $E_a$.

8. Model results

When applying the model to real cases, the only unknown is the aggregation efficiency that can be varied to optimize results for each situation. Here, we present steady-state cloud solutions to flights carried out during the ARM, TRMM, and FIRE I campaigns. For each case the results are displayed in $\lambda$–$N_0$ space, $\lambda$ altitude, $N_0$ altitude, and $P$–$D_1$ space.

For the TRMM case (Figs. 12–14), $E_a = 0.6$, that is, roughly one sticking event for every two collisions. There is good agreement with the $\lambda$–$N_0$ trajectory with a tendency for the trajectory to lie along a locus of
constant $\phi$ until the cloud begins to sublimate below 4500 m. The $\lambda-N_0$ versus altitude plots indicate that the steady-state cloud reproduces the observed vertical structure well. Finally, the $P-D_1$ plot for the TRMM case graphically shows that $P$ is approximately constant implying that aggregation is dominating the PSD evolution until the sublimation region is reached. The results for the ARM (Figs. 15–17) and FIRE I (Figs. 18–20) case use $E_a = 0.7$ and $E_a = 0.3$, respectively. The FIRE case shows that there are problems with the model when the ice is sublimating. In reality breakup is probably also occurring, which would work to decrease the value of $D_1$ (see Fig. 20). This exercise suggests that the biggest uncertainty will lie with the aggregation ef-
size distributions. Consequently, these two variables are required to make a good estimate of the radar reflectivity. Similarly the inverse problem of obtaining precipitation rate from radar measurements requires an estimate of $D_i$ from dual wavelength retrievals, for example, as well as reflectivity.

The application of the parameterized $\theta$ and $\eta$ values, together with the assumption of a steady-state cloud, appear to accurately reproduce the structure of clouds observed in ARM, TRMM, and FIRE1.

The generalized relations given in Table 1 apply to a wide range of cloud types and environmental conditions. The reason for this generality is related to the universality of the aggregation process, which operates in ice clouds.

9. Summary and conclusions

Similar to other nonmicrophysical aggregating systems it is seen that PSDs evolved through ice crystal aggregation are scalable when it is assumed that ice mass flux density is conserved during aggregation. This scalability occurs in spite of the fact that the numbers of component crystals in the aggregates are only of order 10 compared to other colloidal systems where aggregates can be composed of many orders of magnitude more component crystals. The form of the scaled PSD is exponential out to at least nine times the number-weighted mean diameter $D_1$.

The scaling exponent $\theta$ is correlated with precipitation rate and approaches a value of 3 for large precipitation rates. Consideration of theoretical arguments allows the quartet of parameters necessary to state the mass–diameter and fall speed–diameter power-law relations explicitly to be determined. This implies that the mass exponent $\beta$ approaches 2 for large precipitation rates.

It is also clear that two variables, precipitation rate (or IWC, given the power-law relations) and $D_1$, are required to provide an accurate estimate of ice particle aggregation efficiency $E_a$ that may be affected by habit, humidity, temperature, electrical effects, inadequacies in the estimation of $\theta$ from $P$, and assumptions about the nature of the collection kernel.

The choice of $E_a$ is subjective so a likely random error of 0.10 is to be borne in mind. Values for $E_a$ were determined for the other flights considered and are given in Table 2. There is no obvious temperature dependence, although this may be masked by the wide range of temperatures covered by the spirals and the selection of only one value of $E_a$ for each spiral. The values tend to be around 0.3. For one of the spirals [1 Nov 1986 (b)] Kajikawa and Heymsfield (1989) estimated a value of $E_a$ of 0.1. When we consider the same depth of cloud we find that $E_a = 0.1$ provides good agreement between the model and observations. Mitchell’s (1991) estimates of $E_a$ also tend to be of the order 0.1.

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