

Response to Anonymous Referee #1

General Comments:

This study presents a novel approach in estimating ice particle properties from CPI data, and is worthy of publication in ACP after suitable revisions. It is well organized and written, with figures of good quality.

We appreciate the positive assessment and respond to comments below.

A good effort is made to compare the new ice properties with selected properties published 20 or 14 years ago, but no analytical expressions are given for the new ice properties. A table should be added to the paper, similar to Table 1, but showing the mass- and area-dimensional coefficients for the new m-D and A-D relationships (based on CPI data); like results for the new bullet rosettes, bucky-balls, and the polycrystal model. That will allow the community to compare these new results in future studies of ice properties, and promote progress in this field.

Ice properties are supplied as supplementary material (as was already noted in the appendix). Clarification now added more prominently to close of introduction: "Because the derivations here are based on crystal component geometries and do not yield continuous analytic relationships, equations are provided in Appendix A and derived ice properties are provided for download as the Supplement."

It would also be a service to the community if the recent work of Erfani and Mitchell (ACPD, 2015) were compared against the m-D and A-D results from this study. The Erfani-Mitchell expressions are not for a specific ice crystal habit, but were derived from a mixture of habits (similar to the Cotton et al. m-D results featured in this paper).

We have added Erfani and Mitchell (now ACP, 2016) fits to Figs. 5, 7, 13 and 14, and associated discussion to Sections 1, 4.1, 4.4, 5.1 and 6.

In Lawson et al. (2006, JAS), Sec. 3d, A-D power laws are given for irregulars, bullet rosettes, budding rosettes, and rimed rosettes with slide plane wafers between the branches (similar to the polycrystal model developed in this study). It would be instructive to compare these new results against those A-D expressions since they were also based on CPI data.

We have added the Lawson et al. (2006) m-A relations for budding rosettes and rosettes to Figs. 5 and 7 and associated discussion to Sections 4.1 and 5.1. We have not addressed mixed-phase clouds or riming whatsoever in this study, so we omit rimed rosettes. Because their data set obviously represents a much wider range of particle types, including mixed-phase conditions, we also omit irregulars to guarantee applicability here.

This study apparently applies a single ice crystal model over the entire ice particle size

distribution (PSD). However, this assumption is questionable based on CPI observations; see Lawson et al., 2006, JAS, Fig. 5. There you can see that, with increasing size and mass-weighted percent, the smallest crystals tend to be quasi-spherical, then small irregulars, then small irregulars and budding rosettes, then larger rosettes for a single cirrus flight. While this trend may change somewhat from flight-to-flight, it is illustrative of what is typically encountered for cirrus cloud measurements. Similar results are shown in Fig. 13 of Baker and Lawson (2006, JAS). The paper should include some discussion of this, and how such size-dependent habit variation may impact the model results.

We have expanded the discussion intended to address uncertainty in small particle shape in the conclusions (now an independent fifth paragraph of section 6), giving greater emphasis to the concept of habit evolution from quasi-spherical shapes.

Lastly, due to the large number of symbols used in this paper, an appendix for symbol definition is recommended.

We omitted this because various symbols used only in the appendix would require long definitions, and listing the appendix symbols together with those commonly used in the main text would be unnecessarily long for most readers.

Major Comments:

1. Page 8, lines 20-21: Why is arm width W twice the hexagon side length?

This choice is convenient for equations shown in the appendix.

2. Page 9, lines 5-6: Is there vapor competition between homo- and heterogeneous ice nucleation?

Clarification added to model description of homogeneous aerosol freezing: "Heterogeneous freezing is neglected." And to conclusions: "Unlike the simplified parcel simulations shown here, 3D simulations will consider competition between homogeneous and heterogeneous freezing mechanisms and results can be robustly compared with observed ice size distributions."

3. Page 9, lines 21-22: Are all ice nuclei composed of $(\text{NH}_4)_2\text{SO}_4$?

All ice crystals are formed from homogeneous aerosol freezing (see response 2).

4. Page 13, lines 14-15: How much difference is there between your D_{max} and the D_{max} that Mitchell uses? For random orientation, it seems that on average the branches would be oriented at 45 degrees relative to their maximum extension. Taking that maximum length as $L = 1.0$ (arbitrary units) and true $D_{\text{max}} = 2L$, then the percent error made by Mitchell by underestimating D_{max} as $2L \cos(45)$ (randomly

oriented) would be 29%. This seems like too small an error to account for most of the 4-fold difference in mass.

Clarification added in a follow-on sentence: “However, we are unable to quantitatively confirm that [differences in m are primarily attributable to differing approaches to defining D_{\max}] because randomly oriented maximum dimension cannot be calculated analytically for the idealized geometries derived here nor obtained from CPI images for the natural crystals.”

5. Page 17, line 3: Please add temperature information to Fig. 10 so that this sentence makes sense.

Since Fig. 10 is not intended to be statistically representative, we have changed “some rosettes falling from colder temperatures reach a plate growth regime” to “some rosettes exhibit a plate growth regime”.

6. Page 19, lines 22-24: No need to wait for future studies; this information already exists (as noted under General Comments) in Lawson et al. (2006, JAS) and Baker and Lawson (2006, JAS).

We added Lawson et al. (2006) relations for budding rosettes and rosettes to Figs. 5 and 7, as discussed above, but omitted Baker and Lawson (2006) owing to their stated conditions, i.e., “The crystal types typical of high-altitude cirrus, that is, bullet rosettes and similar spatial crystals, are not represented in this dataset.”

7. Fig. 4. Why not use log-log plots when plotting m - D and A - D since this should be quasi-linear and make the results easier to interpret?

Because of our focus on measurements made over less than one order of magnitude in maximum dimension and our concern with geometric differences, we prefer linear axes when we can use them, as in most panels of Fig. 4. We prefer log axes when emphasizing the small particle size range, where we lack Ice Crystal Ruler measurements, as in Fig. 5.

8. Fig. 15. Is there a super-position of the Mitchell and Heymsfield curves?

Clarification added to caption: “In the absence of specified α_e for some or all crystal sizes, a constant value is taken for Mitchell et al. (1996) and Heymsfield et al. (2002) ice properties (see text).”

Minor Comments:

1. Page 11, line 11: What are “cap vertices”? Please define.

Clarification added: “opposing cap vertices” replaced with “opposing edges of the hexagonal pyramids that cap each branch”, and other occurrence of “vertices” replaced with “edges” in the second sentence of Appendix A1.

2. Page 20, line 18: Should < 100 be > 200 ?

Here $<$ should have been $>$ (correction made), thank you.

3. Page 24, line 11: Does i need defining?

The i was defined in Section 4.1 (bulk density of ice).

4. Page 26, line 13: greater \Rightarrow less? This is a Christiansen band where $n_r < 1.0$ but n_i is not > 1.0 .

Indeed “greater” should have been “less” and we thank the reviewer for pointing out that this is a Christiansen band. We changed the text to read “At ~ 2.8 micron, a Christiansen band (Arnott et al. 1995) is present where a combination of strong absorption and refractive indices near or less than unity leads to a decrease in Q_e (cf. Baum et al. 2014).”

Response to Anonymous Referee #2

The research effort reported in the manuscript analyzed ice crystal images in mid-latitude cirrus clouds, towards developing internally consistent ice physical and optical properties for a size-resolved cloud microphysics model. Often reported in the literature, the parameterizations of ice cloud radiative properties and the counterparts of ice cloud microphysical properties are separately developed and thus lack internal consistency. The outcomes of this study represent an important contribution to a better understanding of ice cloud microphysical and radiative properties. Overall, the manuscript is well organized and clearly written. However, some improvements seem necessary before the manuscript is formally accepted for publication. Listed below are the reviewer's specific comments, which are mainly focused on the optical properties of ice crystals.

We appreciate the positive assessment and respond to comments below.

Several ice crystal habit models (specifically, a bucky ball model, an aggregate model, and a polycrystal model) are investigated in detail. For feasible light scattering calculation, ice crystal morphologies are highly simplified in comparison with realistic counterparts. A common justification for the simplifications is that the optical properties are realistic although ice crystal geometries are simplified and even unrealistic. An important constraint to check whether an ice crystal habit model is reasonable from the optical property perspective is to check the consistency of the corresponding optical properties between solar and infrared bands. The optical property parameterization in this study is largely based on Dr. van Diedenhoven's previous parameterizations. If the reviewer recollects correctly, Diedenhoven's previous parameterizations are developed for the solar bands, for example, van Diedenhoven et al. (2014a). Thus, it is suggested that the consistency of the present models between solar and infrared bands be validated. For the authors' information, a recent study in this regard has been reported: Holz, R.E., S. Platnick, K. Meyer, M. Vaughan, G. Wind, S. Dutcher, S. Ackerman, A. Heidinger, N. Amarasinghe, C. Wang, and P. Yang, "Resolving cirrus optical depth biases between CALIOP and MODIS using IR retrievals," *Atmos. Chem. Phys. Discuss.*, 15, 29455-29495, doi:10.5194/acpd-15-29455-2015, 2015.

As infrared radiative transfer is dominated by emission, crystal shape has relatively little influence on it. In Holz et al. (2016 now in ACP) this fact is used to evaluate the applicability of assumed habit for shortwave retrievals. Holz et al. state that "because the sensitivity of IR IOT retrievals to ice crystal habit selection is minimal, these retrievals provide an independent means to evaluate the CALIOP and MODIS solar reflectance retrievals." Thus, an evaluation as presented by Holz et al. compares retrievals of optical thickness (and/or particle size) from both shortwave and IR measurements in order to evaluate of the optical model used for the shortwave. In contrast, our investigation aims to derive optical properties consistent with the in situ observations at various levels within cloud. Representative shortwave and IR measurements for the particular clouds under investigation do not exist and an evaluation as performed by Holz et al. therefore cannot be performed for these clouds. To clarify, we added

the following text to line 28 on page 23: “Infrared radiative transfer is dominated by emission, which is affected by particle size, but its sensitivity to crystal shape is minimal (e.g., Holz et al. 2016). However, particle shape does affect the relevant shortwave optical properties substantially.”

The description of the optical property simulations requires clarification. For example, it is mentioned in the manuscript (the second paragraph on page 24) that the anomalous diffraction theory (ADT) was used to compute the extinction efficiency. However, ADT is not applicable to the phase function (thus, the asymmetry factor) computation. How is an asymmetry factor value that is consistent with the ADT simulation derived?

Since van Diedenhoven et al. (2014) is based on geometric optics calculations, it assumes extinction efficiency of 2 for all particles and wavelengths, and the anomalous diffraction was only used to partly correct this simplification for small particle sizes. Clarification added at page 24, line 13: “The van Diedenhoven et al. (2014) parameterization is based on geometric optics calculations. Accordingly, it assumes the extinction efficiency (Q_e) to be 2 for all particles and wavelengths. To partly correct this simplification for small particle sizes, here we apply anomalous diffraction ...”

On page 25 it is stated “a roughness parameter σ as defined as Mack et al. (1996) ...” (line 3) and “...we note that assuming plates with $\sigma=0.5$...”. In addition, Yang et al. (2013) and Baum et al. (2014) are cited. In Mack et al. (1996), uniformly tilting of ice crystal facets is assumed whereas the Gaussian distribution is assumed in Yang et al. (2013) and Baum et al. (2014). It is explicitly mentioned “Since Baum et al. (2014) and van Diedenhoven et al. (2014b) show that a roughness parameter of 0.5 best fit observations...”. The same roughness parameter value (0.5) cannot be applied to the aforesaid two roughness definitions. Thus, it is suggested that an explicit definition of the roughness parameter be explicitly defined (maybe, an equation should be provided here). The clarification is important because the degree of surface roughness is a critical factor in determining the radiative forcing of ice clouds as illustrated by the following paper: Yi, B., P. Yang, B. A. Baum, T. L’Ecuyer, L. Oreopoulos, E. J. Mlawer, A. J. Heymsfield, K.-N. Liou, 2013: Influence of ice particle surface roughening on the global cloud radiative effect, *J. Atmos. Sci.*, 70, 2794-2807.

Various definitions of the roughness parameters were compared by Neshyba et al. (2013) and by Geogdzhayev and van Diedenhoven (2016) and were found to be largely equivalent. This means that the same value of roughness parameter defined as by Macke et al. and that used by Baum et al. (2014) yields largely equivalent scattering properties. As demonstrated by Geogdzhayev and van Diedenhoven (2016), a roughness parameter of a given value but with different definitions represent very similar micro-structures on the crystal surfaces. As stated in the submitted manuscript (page 25, line 1): “In the van Diedenhoven et al. (2014a) parameterization, the level of surface distortion is specified by a roughness parameter δ as defined by Macke et al. (1996); differently defined roughness parameters are found to be

roughly equivalent (Neshyba et al. , 2013; Geogdzhayev and van Dienenhoven, 2016).” This text is now extended to read:

“In the van Dienenhoven et al. (2014a) parameterization, the level of surface distortion is specified by a roughness parameter δ as defined by Macke et al. (1996). The Macke et al. (1996) ray-tracing code perturbs the normal of the crystal surface from its nominal orientation by an angle that, for each interaction with a ray, is varied randomly with uniform distribution between 0 and $\delta \times 90^\circ$. Similar commonly used parameterizations of particle roughness perturb the crystal surfaces using Weibull (Shcherbakov et al. 2006) or Gaussian (Baum et al. 2014) statistics rather than uniform distributions. However, Neshyba et al. (2013) and Geogdzhayev and van Dienenhoven (2016) demonstrated that the same roughness parameter value defined through a Weibull, Gaussian or uniform distribution represents very similar crystal microscale surfaces and yields largely equivalent scattering properties.”

One page 4, acronyms SHEBA and ISDAC should be spelled out.

Now spelled out.

To resolve small sizes, it is suggested that logarithmic scale is applied to the maximum dimension in Figs. 15 and 22.

The choice of axes throughout is debatable. Here we prefer to use the same linear axis consistently across Figs. 14, 15, 22 and 23.

Derivation of physical and optical properties of midlatitude cirrus ice crystals for a size-resolved cloud microphysics model

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Abstract.

Single-crystal images collected in mid-latitude cirrus are analyzed to provide internally consistent ice physical and optical properties for a size-resolved cloud microphysics model, including single-particle mass, projected area, fall speed, capacitance, single-scattering albedo, and asymmetry parameter. Using measurements gathered during two flights through a widespread synoptic cirrus shield, bullet rosettes are found to be the dominant identifiable habit among ice crystals with maximum dimension (D_{\max}) greater than 100 μm . Properties are therefore first derived for bullet rosettes based on measurements of arm lengths and widths, then for aggregates of bullet rosettes and for unclassified (irregular) crystals. Derived bullet rosette masses are substantially greater than reported in existing literature, whereas measured projected areas are similar or lesser, resulting in factors of 1.5–2 greater fall speeds, and, in the limit of large D_{\max} , near-infrared single-scattering albedo and asymmetry parameter (g) greater by ~ 0.2 and 0.05, respectively. A model that includes commonly imaged side plane growth on bullet rosettes exhibits relatively little difference in microphysical and optical properties aside from ~ 0.05 increase in mid-visible g primarily attributable to plate aspect ratio. In parcel simulations, ice size distribution and g are sensitive to assumed ice properties.

microphysical and macrophysical properties (e.g., Stephens et al., 1990). With respect to microphysical properties, observations of cirrus cloud particle size distributions and underlying ice crystal morphology still remain subject to large uncertainties, in part owing to lack of instrumentation adequate to provide artifact-free and well-calibrated measurements of size-distributed ice particle number and mass concentrations (e.g., Baumgardner et al., 2011; Lawson, 2011; Cotton et al., 2012). With respect to single-crystal properties, the Cloud Particle Imager (CPI) instrument provides high-resolution images of crystals at 2.3 μm per pixel (Lawson et al., 2001), but to our knowledge no airborne instrumentation to date provides a direct measurement of the most fundamental quantity: single-particle mass. How important is advancement of such microphysics observations? On one hand, for instance, simulated climate sensitivity has been reported sensitive to cirrus ice fall speeds (e.g., Sanderson et al., 2008). On the other hand, statistical properties of cirrus simulated at the cloud-scale have been reported relatively insensitive to ice crystal habit assumptions (e.g., Sölch and Kärcher, 2011). Such an insensitivity to ice habit presents a contrast to mixed-phase cloud simulations, which are found sensitive to even relatively minor changes in the specification of ice microphysical properties such as habit, fall speed, and size distribution shape (Avramov and Harrington, 2010; Avramov et al., 2011; Fridlind et al., 2012; Ovchinnikov et al., 2014; Simmel et al., 2015).

It is also well known that ice crystals in the atmosphere exhibit a profound degree of diversity in morphology that impacts microphysical process rates and radiative properties (e.g., Pruppacher and Klett, 1997). Within parcel, cloud-

1 Introduction

It is well known that cirrus clouds substantially impact radiative fluxes and climate in a manner that depends upon their

60 resolving and climate model microphysics schemes, ice 115
 properties are simplified in a variety of ways, generally based
 on some degree of observational guidance. Early observa-
 tional studies using single-crystal measurement approaches
 commonly reported power-law relations between particle
 65 mass and a relevant particle dimension, such as column 120
 length or aggregate maximum dimension, generally valid
 over a relatively short range of dimensions measured for
 any particular crystal habit class (e.g., Locatelli and Hobbs,
 1974). Later work identified the importance of projected area
 70 to fall speed, reported observation-based area-dimensional 125
 power laws for a few habits, and provided estimates for a
 number of others (e.g., Mitchell, 1996). Whereas the fore-
 going studies reported mass- and area-dimensional relations
 by habit, later studies attempted to use additional measure-
 75 ments such as circumference to obtain robust relationships 130
 that do not depend upon first assigning a habit (e.g., Baker
 and Lawson, 2006a; Schmitt and Heymsfield, 2010), an ap-
 proach that is desirable in part owing to the fact that crystal
 habit is commonly irregular (e.g., Korolev et al., 2000). A
 80 convenient aspect of power-law relations, whether they are 135
 derived for one habit or a mixture, is their ease of analyti-
 cal integration in parameterized microphysics schemes (e.g.,
 Morrison et al., 2005).

In very detailed modeling studies of ice evolution, if habit
 85 geometry is well defined, precise calculations can be made 140
 for capacitance and other microphysical parameters (e.g.,
 Hashino and Tripoli, 2011). However, even in a natural cloud
 system where nearly all crystals are in the same habit class,
 that habit may be characterized by chaotically polycrystalline
 90 shapes (e.g., Bailey and Hallett, 2002), as in the case of ra- 145
 diating plates seen during the [SHEBA-Surface Heat Budget
 of the Arctic \(SHEBA\)](#) campaign (e.g., Fridlind et al., 2012),
 or may be subject to wide diversity of form, as in the case
 of dendrites ranging from plate-like to star-like seen during
 95 the [ISDAC-campaign-Indirect and Semi-Direct Aerosol 150
 Campaign \(ISDAC\)](#) (e.g., Avramov et al., 2011). The fact
 that the majority of ice crystals in natural clouds are not
 generally pristine owing at least in part to the common-
 ality of polycrystalline growth and the curving sides and
 100 edges caused by sublimation (e.g., Korolev et al., 1999) has 155
 been increasingly recognized in literature that addresses the
 consequences of morphological diversity for factors such as
 single-scattering properties (e.g., McFarquhar et al., 1999).
 Later laboratory and measurement analyses have specifically
 105 aimed to provide more generalized guidance on complex 160
 morphologies, offering revisions to earlier diagrams of habit
 as a function of temperature and supersaturation (e.g., Bai-
 ley and Hallett, 2002; Korolev and Isaac, 2003; Bailey and
 Hallett, 2009).

110 Currently, based on CPI imagery, automated identification 165
 of ice habit is relatively commonly reported (e.g., Lawson
 et al., 2006b; McFarquhar et al., 2007). However, analy-
 sis of quantitative single-crystal data on within-habit diver-
 sity to inform the representation of microphysical and radia-

tive properties of ice for modeling studies of observed case
 studies (or, by extension, cloud system classes such as cir-
 rus) remains nearly absent. The widespread occurrence of
 polycrystals and aggregates further complicates ice proper-
 ties substantially. In relatively thick mixed-phase clouds, for
 instance, cycles of riming and vapor growth may result in
 a wide variety of plate-like fin structures grown on highly
 rimed substrates (e.g., Magono and Lee, 1966, R3c habit) as
 seen during the M-PACE campaign (Fridlind et al., 2007),
 creating crystal properties so diverse that it is essentially im-
 possible to find quantitative, measurement-based guidance
 from analyses available in the literature to date.

Perhaps not yet as widely considered in models are the dif-
 ficulties of consistently assigning ice crystal component as-
 pect ratio, roundness, and microscale surface roughness for
 accurate calculation of radiative properties (e.g., van Dieden-
 hoven et al., 2014a). When using mass- and area-dimensional
 relations as a foundation for ice properties in a model, as
 most commonly done, it is possible to assign a surface rough-
 ness and aspect ratio, and to calculate optical properties
 based on columns and plates that match ice volume, pro-
 jected area and aspect ratio for any given ice class and size
 (e.g., Fu, 1996, 2007; van Diedenhoven et al., 2012). Guid-
 ance can be obtained from past studies of cirrus that quantify
 the variability of bullet arm aspect ratio, for instance (e.g.,
 Iaquinta et al., 1995; Heymsfield and Iaquinta, 2000; Um
 and McFarquhar, 2007). However, the aspect ratio of whole
 crystals and their crystalline elements are relatively scarcely
 reported and analyzed for natural ice crystals (e.g., Korolev
 and Isaac, 2003; Garrett et al., 2012; Um et al., 2015), mak-
 ing necessary some relatively poor approximations for spe-
 cific natural conditions that may be encountered in the field
 (e.g., Fridlind et al., 2012). Finally, for a size-resolved mi-
 crophysics scheme, obtaining continuity of ice particle prop-
 erties over the full size range required to represent relevant
 cloud microphysics generally requires awkward concatenation
 of aspect ratio-, mass- and area-dimensional relations
 relevant for limited size ranges (e.g., van Diedenhoven et al.,
 2012; Sölch and Kärcher, 2010), which can easily lead to
 unphysical discontinuities in derived quantities such as fall
 speed or capacitance. [Erfani and Mitchell \(2016\) recently 165
 provided polynomial mass- and area-dimensional relations
 that surmount lack of continuity and simplify to analytically
 integrable power laws that closely approximate the full
 solution over a local size range.](#)

Here we analyze single-crystal ice crystal field data with
 the primary objective of deriving physically continuous ice
 microphysical ~~and~~ optical properties over the size range
 required (1–3000 μm). As a well-defined starting place, and
 a foundation for large-eddy simulations, we focus narrowly
 on the morphological properties of a well-developed midlat-
 itude synoptic cirrus case study, taking advantage of an ex-
 isting extended analysis of single-crystal images (Um et al.,
 2015). Because the most accurate representation of cirrus
 optical properties requires consideration of polycrystal el-

170 ement aspect ratios (e.g., van Diedenhoven et al., 2014a),
 which are commonly a function of particle size in obser-
 vations, the polycrystal elements are adopted as the founda-
 tion for treating mass and projected area rather than vice
 versa (as required if area- and mass-dimensional relation- 225
 ships are instead adopted as the foundation, as most com-
 monly done); a similar approach was taken by Heyms-
 field and Iaquina (2000) for the purpose of deriving phys-
 ically based expressions for cirrus crystal terminal veloci-
 ties, such as bullet rosettes with varying numbers of arms.
 180 Parcel simulations are used to compare the ice properties
 derived in this work with ice properties available in ex-
 isting literature that have been used in large-eddy simula-
 tions of cirrus with size-resolved microphysics (Sölch and
 Kärcher, 2010). Because the derivations here are based on
crystal component geometries and do not yield continuous
analytic relationships, equations are provided in Appendix A
and derived ice properties are provided for download as the
Supplement. 235

2 Observations

190 In situ observations are analyzed from a well-sampled cir-
 rus system observed during 1 April (flight B) and 2 April
 (flight A) during the 2010 Small Particles in Cirrus (SPAR-
 TICUS) field campaign (Mace et al., 2009). Based on an ex- 245
 tensive analysis of atmospheric states during SPARTICUS,
 Muhlbauer et al. (2014) classified the 1–2 April conditions
 as ridge-crest cirrus (Fig. 1). Relative to the other non-
 convective cirrus states identified during SPARTICUS, ridge-
 crest cirrus were characterized by formation within the cold- 250
 est environments at cloud top, within considerable ice super-
 saturation, and were statistically associated with the highest
 ice crystal number concentrations and lowest ice water con-
 tents.

Previous studies using SPARTICUS data can be consid- 255
 ered in at least five general categories: characterization of the
 environmental properties observed (e.g., Muhlbauer et al.,
 205 2014), characterization of the ice crystal morphology or
 size distribution characteristics observed (e.g., Mishra et al.,
 2014; Um et al., 2015; Jackson et al., 2015), cirrus cloud pro-
 cess modeling studies (e.g., Jensen et al., 2013; Muhlbauer
 et al., 2015), evaluation of satellite retrievals (e.g., Deng
 et al., 2013), and evaluation of climate model cirrus prop-
 erties (e.g. Wang et al., 2014). The work here is in the second
 category, and is based primarily on single-crystal ice crys- 265
 tal properties using data obtained from a CPI probe on the
 Stratton Park Engineering Company (SPEC) Inc. Learjet 25
 aircraft. The ice crystals imaged by the CPI are first classified
 by habit using the scheme described by Um and McFarquhar
 (2009). Images classified as bullet rosettes and aggregates of 270
 bullet rosettes are then further analyzed using output from
 the recently developed Ice Crystal Ruler (ICR) software (Um

et al., 2015) to obtain the imaged width and length of each
 branch.

To provide context for parcel simulations, we also use
 Learjet ice particle size distributions derived from a 2D
 Stereo Probe (2DS) equipped with tips that reduce effects of
 shattering (Lawson, 2011) and analyzed as reported by Jack-
 son et al. (2015), together with in-cloud vertical wind speed
 retrievals from profiling Doppler radar measurements (Ka-
 lesse and Kollias, 2013).

3 Model description

The overall objective of this study is to use analyzed
 CPI image data to derive consistent representations of ice
 physical and optical properties for a size-resolved ice mi-
 crophysics scheme, and to compare results with existing
 literature. The target microphysics scheme is based on
 the Community Aerosol-Radiation-Microphysics Applica-
 tion (CARMA) code (Jensen et al., 1998; Ackerman et al.,
 1995). The CARMA model allows selection of an arbitrary
 number of mass bins to represent the size distributions of an
 arbitrary number of aerosol and ice classes. Within each ice
 class, the mass in each bin is a fixed multiple of the mass in
 the preceding bin.

In this work the ice crystal properties in each ice mass
 bin are represented using the approach developed by Böhm
 (1989, 1992b, c, a, 1994, 1999, 2004), as previously ap-
 plied to represent the ice crystals in mixed-phase stratus in
 Avramov et al. (2011, dendrites and their aggregates) and
 Fridlind et al. (2012, radiating plates). The Böhm scheme
 provides an integrated treatment of terminal fall speeds and
 collision efficiencies for non-spherical ice that is based not on
 specification of a particular habit but rather on four proper-
 ties that are quantitatively defined for both pristine and non-
 pristine shapes: particle mass m , a characteristic maximum
 dimension D and projected area A , and aspect ratio α . The
 foundational physical quantity of this parameterization is fall
 speed, so the characteristic quantities A , D and α are best
 defined by fall orientation, which can perhaps most simply
 be considered as the maximum projected area (which deter-
 mines the fall orientation), the maximum dimension of a cir-
 cumscribed circle around that projected area, and the aspect
 ratio of thickness normal to the fall orientation to that max-
 imum dimension (cf. Böhm, 1989). Bodily aspect ratio α is
 defined as 1 for ice crystals without a preferential fall ori-
 entation (e.g., bullet rosettes), less than 1 for oblate bodies
 (e.g., plates), and greater than 1 for prolate bodies (e.g.,
 columns). Throughout this work, α is fixed at 1 based on the geome-
 tries discussed below, and D and A by extension assumed
 equal to randomly oriented maximum dimension (D_{\max}) and
 randomly oriented projected area (A_p).

In each ice mass bin, quantities that are not considered
 in the Böhm scheme but that should ideally be specified in
 an integrated manner are capacitance and radiative scattering

and absorption coefficients. For a given crystal, the capacitance can be either specified from the literature in the case of a pristine habit or else estimated from prolate or oblate spheroids (Pruppacher and Klett, 1997, their Eqns. 13-78 and 13-79). Here we take the former approach for bullet rosettes and their aggregates and polycrystals analyzed below: given bullet arm length L and arm width W (twice the hexagon side length), we specify D_{\max} -normalized capacitance (C) as $0.4(L/W)^{0.25}$ based on the fit to calculations for six-arm rosettes by Westbrook et al. (2008).

Scattering and absorption properties assuming randomly oriented ice crystals in each mass bin are computed following van Diedenhoven et al. (2014a), which, in addition to m and A_p , also requires specification of elemental aspect ratio (α_e) and a microscale surface roughness or crystal distortion (δ), as defined by Macke et al. (1996). Here α_e required for the optical properties is identical to the bodily aspect ratio α in the case of a single-component crystal (e.g., plate or column), but for a polycrystal such as a bullet rosette α_e is the aspect ratio of constituent arms or other component crystals (cf. Fu, 2007). In this work α_e values are derived from ICR measurements where possible. Additional details are given in Section 5.4.

Parcel simulations are used to test ice properties in a simplified framework, following Ackerman et al. (2015), prior to use in computationally expensive 3D large-eddy simulations in future work. All simulations include adiabatic expansion, aerosol homogeneous freezing, diffusional growth of ice crystals, and latent heating. [Heterogeneous freezing is neglected](#). Parcels are initialized at 340 mb, 233 K, and 80% relative humidity. Saturation vapor pressures are related to water vapor mixing ratio following Murphy and Koop (2005). Each simulation is assigned a fixed updraft speed (w) of 0.01–1 m s⁻¹. Parcel expansion is treated by assuming dry adiabatic ascent and iterating three times on parcel air pressure, temperature, and density assuming hydrostatic conditions and using the ideal gas law. Latent heat is computed in accord with diffusional growth of the ice. A default time step (Δt) of 1 s is variably reduced to a minimum value of 0.1 s, which is reached when fast processes such as aerosol freezing are active, and parcel height is incremented by $w\Delta t$ each time step. Ice sedimentation, when included, assumes a vertical length scale of 100 m as in Kay and Wood (2008). Gravitational collection is neglected. We use the Koop et al. (2000) parameterization for aerosol freezing, including the Kelvin effect on surface vapor pressure, and assume that aerosol are at equilibrium with atmospheric water vapor. Aerosol are initialized with a concentration of 200 cm⁻³ lognormally distributed with geometric mean diameter 0.04 μm and geometric standard deviation 2.3 as in Lin et al. (2002), except that composition is assumed to be ammonium bisulfate. We assume a fixed ice accommodation coefficient of 1, which is within the range of recent laboratory measurements (Skrotzki et al., 2013), and account for Knudsen-number-dependent gas kinetic effects (cf. Zhang

and Harrington, 2015). Growth across mass bins is treated with the piecewise parabolic method of Colella and Woodward (1984). Simulations use 50 bins with a mass ratio of 1.65 from one bin to the next, starting with D_{\max} of 2 μm , suitable for use in 3D large-eddy simulations.

4 Derivation of ice single-crystal properties

Considering all CPI images collected during the 1–2 April flights, automated analysis places roughly half of all ice crystals in the small quasi-sphere category, and remaining crystals are primarily unclassified (Fig. 2). However, considering only ice crystals with D_{\max} greater than 100 μm , bullet rosettes emerge as the most common classified habit. Subjective examination of images suggests that bullet rosettes are the dominant habit in the coldest crystal growth regions with significant ice water content (Fig. 3), as discussed further below. We therefore begin with an analysis of ICR measurements of bullet rosette arm lengths and widths, which are suitable to describe the physical and optical properties for a cloud composed entirely of growing rosettes.

4.1 Bullet model

For each bullet rosette measured with the ICR software, Fig. 4a shows mean branch length versus measured D_{\max} . Since branches that are not aligned with the viewing plane are foreshortened, we take L as the average of all measured branch lengths minus half of randomly oriented projected end plate diameter, multiplied by a factor of $4/\pi$ to account for random orientation to first order (see Appendix A1). The relationship of L to D_{\max} is reasonably fit by a line passing through the origin.

For the same crystals, Fig. 4b and Fig. 4c show mean W , and the ratio $L/W = \alpha_e$, respectively. To account for random orientation, W is taken as the average of all measured branch widths divided by a factor of $(1 + \sqrt{3}/2)/2 = 0.933$, which is the ratio of the arithmetic mean of minimum and maximum branch projected widths to the maximum (equivalent to the ratio that would be found if measurements of projected width were made for a sufficiently large number of orientations of a bullet arm of known W). Both mean and median number of branches is six (out of four to ten measured), consistent with recent analyses from tropical and Arctic field campaigns (Um et al., 2015). Rosettes with more branches are seen to have systematically smaller W and larger α_e , consistent with competition for vapor during growth. However, a simple least squares fit of W to D_{\max} gives $W > D_{\max}$ when extrapolated to small crystal size, which is not physical; unfortunately, measurements are not available to provide guidance at such sizes.

Because we seek a continuous description of ice properties across all sizes, here we take the approach of adopting a physical model of crystal geometry to extrapolate mea-

sured properties smoothly to sizes smaller than measured. A similar approach was taken by Heymsfield and Iaquinta (2000) to improve calculated cirrus crystal fall speeds over those obtained from independently derived mass- and area-dimensional relations. We first assume branch width for rosettes consistent with the six-rosette model considered in Westbrook et al. (2008), but using a fixed angle of 44° between opposing [cap-vertices-edges of the hexagonal pyramids that cap each branch](#) (sensitivity of results to choice of cap angle is discussed at the end of Appendix A1). Selecting a fixed angle and using the linearly fit branch width at all sizes allows determination of the cap contribution to L ; L is found to consist entirely of a truncated cap at the smallest sizes and corresponding W is taken as the truncated cap base width. This model results in the line slope discontinuity seen in Fig. 4b, and resolves at least gross discrepancy of $W > D_{\max}$.

Fig. 4c shows that adopting this bullet model results in a smooth increase in branch aspect ratio $\alpha_e = L/W$ from smallest to largest sizes, suitable as a basis for calculating optical properties. L/W is constant at the smallest sizes, where only the cap contributes and both W and L are varying at the same relative rate. The range of aspect ratios measured (2–6) and the fitted trend from near-unity at the smallest sizes to roughly 5 at the largest sizes is consistent with several past studies (cf. Heymsfield and Iaquinta, 2000). As shown, the relationship of branch aspect ratio to D_{\max} also agrees with that used by Mitchell (1994) in derivation of the mass-dimensional relation for $D_{\max} > 100 \mu\text{m}$ listed in Table 1 and discussed further below.

The Westbrook et al. (2008) model assumes that all bullets are at 90° angles to one another, giving true maximum dimension of $2L$, which is $\sim 40\%$ greater than D_{\max} shown in Fig. 4a. Measured D_{\max} being a randomly oriented value can account for less than 30% discrepancy. Another source of difference is the commonly seen deviations of arm locations from 90° separations, which can only decrease D_{\max} from $2L$. Since a more quantitative explanation is beyond the scope of this initial study, we adopt the randomly oriented D_{\max} as our only defined maximum dimension, an assumption that has also been made in past studies using two-dimensional images (e.g., Heymsfield et al., 2002; Baker and Lawson, 2006a).

The bullet model described above now allows calculation of crystal surface area (A_s) and m (see Appendix A1 for details). To calculate m from the geometrical dimensions, we assume ice bulk density (ρ_i) of 0.917 g cm^{-3} ; any bullet arm hollows are neglected here owing to lack of quantitative guidance, as discussed further below. Calculated m and A_p of a six-branch rosette are seen to reasonably represent the scatter of individual crystal properties (solid lines in Fig. 4e and 4f). The ratio of measured A_p to calculated A_s is found to be about 0.11 (Fig. 4d), smaller for these concave particles than the A_p/A_s of 0.25 for convex shapes, consistent with

theoretical results (Vouk, 1948) and reasonably independent of D_{\max} across measured sizes.

In Fig. 4e derived $m(D_{\max})$ is compared with power-law relations from previous literature that have been used in similar bin microphysical schemes (Table 1). To our knowledge, only one unpublished data set has provided direct measurements of bullet rosette mass, consisting of 45 crystals with a range of 2–5 arms as reported by Heymsfield et al. (2002), but that data set is not the basis of commonly used relations. As used in Sölch and Kärcher (2010), for instance, the Heymsfield et al. (2002) relation is based on calculation of effective density from a combination of ice water content and particle size distribution measurements; coefficients in Table 1 (cf. Sölch and Kärcher, 2010) are calculated from their Equation 22, based on crystals with D_{\max} of 200–2000 μm fit in their Fig. 15. As also used in Sölch and Kärcher (2010), Mitchell (1994) combined crystal volume expressions with size-dependent bulk densities of $\sim 0.78 \text{ g cm}^{-3}$ to obtain an $m - D_{\max}$ relation for crystals with D_{\max} of 200–1000 μm (Table 1 values are taken from their equation 32); for crystals smaller than 100 μm , Mitchell et al. (1996) proposed a mass-dimensional relation using ad hoc estimates of crystal mass (Table 1 values are taken from their Table 3).

The difference between our calculated $m(D_{\max})$ and that from Mitchell (1994) is roughly a factor of four at measured crystal sizes, which results in a similar discrepancy in fall speeds and effective diameters, as shown below. We can attribute lower m in Mitchell (1994) to four factors: (i) L is substantially shorter based on the approximation $D_{\max} = 2L$ (cf. Iaquinta et al., 1995, including assumed trilateral pyramidal end following their Fig. 1), (ii) W is substantially thinner based on earlier cited literature that relates W to L and by extension $D_{\max}/2$ (see Fig. 4b), (iii) five branches are assigned instead of six found here, and (iv) ρ_i is $\sim 0.78 \text{ g cm}^{-3}$ instead of 0.917 assumed here. All else being equal, increasing their branch number and ρ_i would together increase Mitchell (1994) m by only about 40%, but m scales roughly linearly with L and geometrically with W . The trilateral pyramid ends taken from Iaquinta et al. (1995) would result in slightly greater m than ours, all else being equal. The close agreement between our arm aspect ratio L/W and that following Mitchell (1994), available for $D_{\max} > 100 \mu\text{m}$ (Fig. 4c), suggests that differences in m are primarily attributable to differing approaches to defining D_{\max} . [However, we are unable to quantitatively confirm that because randomly oriented maximum dimension cannot be calculated analytically for either the idealized geometries derived here or for CPI images of natural crystals.](#)

Our calculated $m(D_{\max})$ is also nearly a factor of two greater than that from Heymsfield et al. (2002) for ice particle ensembles (all habits, dominated by bullet rosettes) measured over the same Oklahoma location. In Heymsfield et al. (2002), D_{\max} is taken from 2DC and 2DP probe measurements and m is derived from coincident ice water content measurements from a Counterflow Virtual Impactor (CVI)

via a linear fit of effective particle density (ρ_e , the density of a sphere with diameter D_{\max}) to D_{\max} . Whereas our approach is subject to uncertainty in ICR measurements and assumed ρ_i , the Heymsfield et al. (2002) approach is subject to uncertainty in the measurement of ice particle size distribution, uncertainty in the measurement of ice water content, and the importance of any deviations of the particle ensemble from bullet rosettes. Uncertainty in CVI probe measurements are reported to be 10% for ice water contents larger than 0.2 g m^{-3} (Twohy et al., 1997), but the bin-wise uncertainty in particle size distribution measurements are generally unquantified; we consider it beyond the scope of this study to undertake the detailed analysis required to resolve such differences. Although it is not used in the $m - D_{\max}$ relationship adopted by Sölch and Kärcher (2010) and listed in Table 1, Heymsfield et al. (2002) also derive a typical ρ_i of $0.82 \pm 0.06 \text{ g cm}^{-3}$ for bullet rosettes based on independent photographic evidence for hollow bullet rosette arm ends; we make no such reduction here, as discussed above, and doing so is not a dominant cause of the differences in m .

Whereas our calculated $m(D_{\max})$ is substantially greater than that previously used in studies with size-resolved microphysics, our measured $A_p(D_{\max})$ is similar or smaller. The relationship of A_p and D_{\max} derived by Mitchell et al. (1996, their Table 1) independently from $m(D_{\max})$ for five-branched bullet rosettes in a manner similar to that here, is nearly identical to ours (cf. Fig. 4e). The less widely available $A_p - D_{\max}$ relations are surprisingly more difficult to trace, considering that they can be more directly derived from CPI images, and we are unable to identify the observational sources of the relations reported in Sölch and Kärcher (2010), which are cited from but not apparent in Heymsfield et al. (2002).

Figure 5 allows a closer examination of the extrapolation from manually measured rosette properties ($D_{\max} > 200 \mu\text{m}$) to smaller sizes using our bullet model, and shows comparisons to additional published fits. From in situ measurements of total ice water content and ice crystal size distribution and shape obtained from a 2DS probe in mid-latitude cirrus, Cotton et al. (2012) derived a mean ρ_e of 0.7 g cm^{-3} below a threshold size of $70 \mu\text{m}$ and a power law decrease of density to 0.5 g cm^{-3} at roughly $100 \mu\text{m}$ and 0.05 g cm^{-3} at roughly $1000 \mu\text{m}$. The mean ρ_e derived here happens to exhibit a similar behavior (Fig. 5a), where the discontinuity using our bullet model represents the transition to truncated branch caps. Erfani and Mitchell (2016) derived polynomial $m - D_{\max}$ relations for synoptic cirrus clouds warmer than -40°C from single-particle measurements of m , D_{\max} and A_p obtained during the 1985–1987 Sierra Cooperative Pilot Project (SCPP) (Mitchell et al., 1990), or by applying a habit-independent $m - A_p$ relation derived from the SCPP data set (Baker and Lawson, 2006a), shown in Figure 4e, to 2DS measurements obtained during 13 SPARTICUS flights (at colder temperatures). Although the SCPP data set does not contain bullet rosettes or spatial crystals

(Baker and Lawson, 2006a), Lawson et al. (2010) report that ice water content derived by applying that habit-independent $m - A_p$ relation to a combination of tropical anvil and synoptic cirrus measurements agreed with CVI measurements to within 20%. At $D_{\max} < 100 \mu\text{m}$, Erfani and Mitchell (2016) m values were calculated from CPI measurements of A_p and α assuming hexagonal column geometry (cf. Erfani and Mitchell, 2016, their Appendix B), and effective densities are similar to those derived here. At larger sizes and especially colder temperatures, Erfani and Mitchell (2016) effective densities are smaller than derived here, consistent with the (Baker and Lawson, 2006a) $m - A_p$ relation giving lower per-particle m than derived here.

Although m cannot be calculated similarly in this study for bullet rosettes that are not measurable with the ICR software or for crystals with unclassified habit, A_p is reported for all imaged crystals and can be directly compared with the bullet model. Analogous to ρ_e but dimensionless, the measured ratio of A_p to that of a sphere with diameter D_{\max} can also be compared with the bullet model. Figure 5b shows that previously applied relations are literature power law relations can become unphysical for the smallest particle sizes (projected areas greater than for a sphere of diameter D_{\max}); to correct the greatest deviations for the purposes of parcel calculations below, we adopt a constant ratio of A_p to sphere projected area where $D_{\max} < 100 \mu\text{m}$ when using Mitchell (1994) relations. When considering all rosettes automatically identified (not all of which were measurable using the Ice Crystal Ruler), the bullet model $A_p(D_{\max})$ agrees quite well with median measurements quite well and with $m - A_p$ relations for bullet rosettes and budding bullet rosettes from Lawson et al. (2006a) (Fig. 5c). However, when considering all crystals (Fig. 5d), there is a wider range of $A_p(D_{\max})$ and the bullet model underestimates median $A_p(D_{\max})$ where well sampled, as addressed further below; the Erfani and Mitchell (2016) polynomial $m - A_p$ fit from the SCPP data set (their warmest-temperature fit) and the Heymsfield et al. (2002) $m - A_p$ power law agree best with the full data set where $D_{\max} > 100 \mu\text{m}$.

4.2 Bucky ball model

To consider uncertainty in the geometry of the smallest crystals, we next consider an alternative proposed model for early bullet rosette shape: budding Bucky balls (Um and McFarquhar, 2011). The so-called budding rosette shape has been observed in laboratory grown ice and ice-analog crystals (Ulanowski et al., 2006; Bailey and Hallett, 2009), and the CPI does not have the resolution necessary to distinguish such a shape from the bullet model geometry assumed above. Here we approximate the Bucky ball core as a sphere of diameter $10 \mu\text{m}$ and then assume that arms emerge with initial width $4 \mu\text{m}$. If we assume that L falls linearly to zero at D_{\max} equal to the core dimension (Fig. 6a) and W correspond-

ingly falls linearly to its minimum initial width (Fig. 6b), then branch α_e is relatively constant near the mean observed (Fig. 6c). A_s and m can now be calculated using this Bucky ball model, except that A_p of the smallest crystals must be interpolated to bridge the geometry of a sphere ($D_{\max} < \text{core diameter}$) and that of a rosette; to do this, we calculate a mass-weighted sum of A_p obtained from the linear relation in Fig. 4d and that of a sphere (see Appendix A2). Thus, as m converges to that of a sphere, so does A_p . Results are similar to those of the bullet model at larger particle sizes (Fig. 6d-f), with $m(D_{\max})$ still larger than previous estimates and $A_p(D_{\max})$ still similar or smaller.

However, using this simplified Bucky ball model, a developing six-arm rosette has a systematically smaller ρ_e and A_p than it did with the bullet model (Fig. 7). Although this particular version of a Bucky ball model, with only six arms even at small sizes, gives substantially smaller $A_p(D_{\max})$ than measured for automatically classified rosettes at small D_{\max} (Fig. 7c), it does serve to provide a quite close match to the minimum area relative to that of a sphere over the full particle data set (Fig. 7d), and is therefore included in parcel calculations below. In reality it seems likely that not all budding arms grow evenly. For instance, Um and McFarquhar (2011) propose a Bucky ball model with 32 regular and irregular hexagonal arms, one growing from each of the ball's 20 hexagonal and 12 pentagonal planes. From this study, it is apparent that only up to about 12 arms commonly reach substantial lengths, and most commonly only six such arms are seen. Faced with the problem of how to introduce geometry that smoothly transitions from an unknown larger number of sub-100- μm arms to roughly six arms at larger sizes with no quantitative basis for how to introduce such added complexity here, we have simply assumed six arms throughout.

4.3 Aggregate model

We return now to the distribution of habits during the April 1–2 flights, and consider the properties of crystals in the observed cirrus deck that are not identified as bullet rosettes. The rosettes are most common in the upper cloud regions at temperatures colder than -40°C (Fig. 8), consistent with previous findings that rosette shapes in the temperature range -40 to -55°C are mostly pristine (Lawson et al., 2006a). In this case, at slightly warmer temperatures, aggregates of bullet rosettes become most common. Using ICR measurements for aggregates of bullet rosettes, it is straightforward to extend the bullet model to rosette aggregates (Fig. 9, see Appendix A3), where the mean and median branch numbers are found to be 12 per aggregate, consistent with aggregation of two typical bullet rosettes. Compared to single rosettes, aggregate properties are generally similar to those of single rosettes except shifted in size to a larger maximum dimension. We do not dwell here on the properties at the smallest sizes since aggregates are born from fully

formed bullet rosettes and this study is focused on crystal growth (neglecting sublimation).

However, aggregates of pristine rosettes also represent a small fraction of ice crystals observed in this case, at least on a number basis. CPI images show that ~~rosettes falling from colder temperatures~~ some rosettes reach a plate growth regime (Fig. 10), a phenomenon well documented in previous cirrus field observations and laboratory measurements (Bailey and Hallett, 2009). In the lower cloud regions at temperatures warmer than -40°C , modified bullets have been described as mostly “platelike polycrystals, mixed-habit rosettes, and rosettes with side planes” (Lawson et al., 2006a), where side plane growth on columns may be attributable to facet instability on prism faces (Bacon et al., 2003).

4.4 Polycrystal model

For the purposes of considering how plate-like growth impacts rosette single-crystal properties, it is notable from the SPARTICUS images in this case that radiating side plane elements appear to increasingly fill the space between the arms of rosettes and rosette aggregates, giving the impression of cobwebs that lead to blocky ice particle shapes (e.g., Fig. 10). In such a process, particle m could increase without rapid expansion of particle D_{\max} . Such a tendency for crystals to become less florid may be related to the finding of side plane growth on rosettes in the laboratory exclusively originating from the rosette center, consistent with an important role for defect and dislocation sites (Baker and Lawson, 2006b). Toward cloud base, sublimation then increasingly rounds crystal edges (Fig. 11). Rosettes that did not enter a side plane growth stage appear now with rounded arms that can still be counted, whereas rosettes that did experience substantial side plane growth emerge from sublimation zones as relatively large quasi-spheres, which appear as a non-negligible percentage of large particle habit; the existence of such large quasi-spheres would be otherwise difficult to explain. The smallest sublimated crystals appear occasionally as sintered chains.

We next consider an approximate model for the physical and optical properties of these more common, irregular crystals. In the data set examined here, we are unable to find a consistent increase in projected area ratio with increasing temperature that would be expected if rosettes are modified by side plane growth during sedimentation from colder to warmer temperatures, but we do find that unclassified crystals at all temperatures exhibit consistently larger area ratios than rosette crystals (Fig. 12). To account for rosette shape evolution in a manner amenable to calculation of radiative and microphysical properties at least for growing crystals, we attempt to coarsely estimate the side plane mass added to pristine rosettes and its associated elemental aspect ratio as follows.

We first calculate the additional projected area that can be attributed to side plane growth. Considering all unclassified crystals, a fit of measured A_p to calculated bullet surface area (based on measured maximum dimension and assuming a bullet model rosette with six arms) yields a slope of 0.15 (Fig. 13a), which is larger than the slope of 0.11 found using the bullet model for measured rosettes, consistent with greater area ratios for unclassified crystals. If we make the ad hoc assumption that the relationship of surface area to projected area is close to that for bullet rosettes, we can attribute the surface area beyond that of the bullet model to plates. If we make the ad hoc assumption that a plate-like side plane grows on each of six arms and neglect plate thickness, the plate or side plane surface area can be considered as the sum of hexagonal faces of the six plates, and the plate diameter can be calculated. If we further relate plate thickness to plate diameter as described in Appendix A4, then mass can now be calculated as the sum of bullet and plate contributions for a typical particle (e.g., Fig. 13c, solid line). For this crude representation of plate-like growth on the bullet model, the calculated crystal properties agree reasonably well with Cotton et al. (2012) effective density in the limit of small D_{\max} (Fig. 13e) and with the area ratio as a function of D_{\max} over all unclassified crystals (Fig. 13f) if the following choices are made: the cap angle β is increased to 25° , plates are assumed present only where branches extend beyond truncated caps, and the plate surface area is assumed to increase inverse exponentially to its terminal value with a length scale equal to the diameter at L greater than L_c (see Appendix A4 for details). Where $D_{\max} > 100 \mu\text{m}$, the resulting polycrystal model A_p agrees closely with the Erfani and Mitchell (2016) fit for warmest-temperature synoptic cirrus, but resulting m is now also correspondingly greater than and further from Erfani and Mitchell (2016) than in the bullet model (cf. Fig. 5a).

The foregoing results for this polycrystal model are dependent upon the underlying bullet model assumed, the assumed ratio of A_s to A_p , and the assumed plate or side plane geometry, for which no quantitative guidance exists in the current data set. This polycrystal model is intended only as a relatively simple example of ice properties that is guided by available observations and allows calculation of internally consistent physical and radiative properties in a continuous fashion over all crystal sizes that need to be represented in our microphysics model. In order to evaluate the need for further consideration of ice properties in greater detail, we next consider parcel simulations to evaluate the influence of ice models on predicted size distributions and optical properties.

5 Model results

5.1 Fall speed and capacitance

Figure 14 shows a point calculation of fall speeds (v_f) at 350 mb and 233 K for comparison with Sölch and Kärcher (2010, their Fig. A1). Our bullet model gives $v_f(D_{\max})$ values that are more than a factor of 1.5–2 greater than derived from Mitchell and Heymsfield ice properties and used by Sölch and Kärcher (2010) in a microphysics model similar to ours. Increasing crystal m calculated from the literature by a factor of 0.917/0.78 can account for relatively little of the difference (not shown), indicating that the main differences are attributable to crystal geometries. In the case of Mitchell properties, as discussed above, the main difference ~~can be traced~~ may be traceable to differing D_{\max} definition used to calculate m , whereas $A_p(D_{\max})$ is very similar. In the case of Heymsfield properties, $m(D_{\max})$ is closer to ours but $A_p(D_{\max})$ is also larger, a factor that should be relatively more easily resolved in future studies since both A_p and D_{\max} can be directly measured. As shown in Fig. 5c, for instance, an $m - A_p$ relation from earlier midlatitude cirrus measurements (Lawson et al., 2006a) agrees well with SPARTICUS rosette measurements and with our bullet model. At the warmest temperatures considered by Erfani and Mitchell (2016), $A_p(D_{\max})$ is similar to or even greater than the bullet or polycrystal models but substantially lower $m(D_{\max})$ leads to substantially lower $v_f(D_{\max})$; the Erfani and Mitchell (2016) trend toward greater decrease in m than A_p with decreasing temperature leads to increasing divergence between the models derived here and their results.

Given literature ice properties, using our model to calculate crystal fall speed as detailed in Avramov et al. (2011) results in v_f values that appear similar to those of Sölch and Kärcher (2010) and are also within roughly 10% of those calculated using the method described in Heymsfield and Westbrook (2010) (not shown). However, our aggregate model gives fall speeds roughly one-third reduced from similar-sized bullet model ice, which is a substantially larger difference than that using Heymsfield ice properties for aggregates and their rosettes shown in Sölch and Kärcher (2010). We can trace this greater difference in part to substantially larger $m(D_{\max})$ derived here, as shown above. Overall, we conclude from comparison of our results with those of Sölch and Kärcher (2010) that the precise method of calculating v_f as a function of m and A_p appears to be responsible for relatively little spread, but differences in ice properties themselves (m and A_p) introduce $v_f(D_{\max})$ differences that are substantially larger than expected, as discussed further below.

Owing to the dependence of parameterized capacitance on bullet arm aspect ratio alone (see Section 3), capacitance differences are nearly negligible for crystals larger than $\sim 400 \mu\text{m}$ across all bullet models derived here, in

800 sharp contrast to factor of two differences in fall speed at
 such sizes. Because assumed or derived bullet arm aspect
 ratios vary most where D_{\max} is less than 300 μm , capaci-
 tance differences up to roughly 25% are most pronounced 855
 at those sizes. Although aspect ratios used in derivation
 of the Mitchell ice properties are similar to ours where
 805 $D_{\max} < 100 \mu\text{m}$ (see Fig. 6), no such aspect ratios
 are provided for smaller Mitchell crystals or for Heyms-
 field ice properties. For parcel calculations, we therefore 860
 adopt a C value of 0.25 derived for aggregates (Westbrook
 et al., 2008), taken here as representative of polycrystals with
 810 unspecified aspect ratios. A similar assumption would be
 required for Erfani and Mitchell (2016) ice properties; since
 parcel simulations are also not configured for changes in
 ice crystal properties during a single simulation, we omit
 815 Erfani and Mitchell (2016) ice properties from the remaining
 calculations.

5.2 Parcel simulations without sedimentation

To grossly evaluate the potential effect of different model
 ice properties on ice crystal nucleation and growth, we first
 820 consider parcel simulations without the complication of sed-
 imentation. Since aggregation is neglected, aggregate ice
 properties are not considered. As described in Section 3,
 parcels begin at 233 K (-40°C), 340 mb, and 80% relative
 humidity. Vertical wind speed (w) is fixed at 0.01, 0.1 or
 825 1 m s^{-1} , within the range of millimeter cloud radar retrievals
 of in-cloud w from the beginning of the first flight examined
 here to the end of the second flight (Fig. 16). Aside we note
 that a parcel simulation is not a realistic rendition of natural
 cirrus cloud evolution, which is characterized by extensive
 830 growth and sublimation during particle sedimentation. But
 a similar framework has been used to test cirrus models (Lin
 et al., 2002), and in this case it allows a simple comparison of
 particle growth to sizes that span the range observed during
 835 SPARTICUS, as discussed further below.

Figure 17 shows the ice particle size distribution (PSD) for
 each simulation at -55°C , which is close to the average of
 in-cloud temperatures observed during the 1–2 April flights.
 Figs. 18–21 show the ice crystal number concentration (N_i), 840
 number-weighted mean diameter (D_i), total projected area
 (A_i), and relative dispersion (ν), respectively, as a function
 of parcel ascent distance. In the absence of sedimentation, ice
 mass is essentially distributed across differing crystal sizes
 depending upon w and ρ_e . The magnitude of w primarily de- 845
 termines N_i : when w is strongest, vapor growth competes
 least with nucleation, resulting in greatest N_i (Fig. 18a). Nu-
 cleated number concentrations range from several per liter
 when w is 0.01 m s^{-1} to several per cubic centimeter when
 850 w is 1 m s^{-1} , consistent with past studies (e.g., Lin et al., 2002).
 The Heymsfield ice properties give roughly a doubling of N_i
 relative to other ice properties, owing to the densest small ice
 accompanied by a fixed capacitance for non-spheres (in the

absence of obvious means of transitioning capacitance from
 spheres to non-spheres).

The strongest w and associated fastest aerosol freezing,
 which leads to largest N_i , leads to the correspondingly small-
 est D_i (Fig. 19a) and the greatest A_i (Fig. 20a). Where N_i is
 insensitive to ice properties, the sensitivity of D_i and A_i to
 ice properties at a given w can be seen as simply scaling in-
 versely with effective density and area per unit mass, respec-
 tively. Ice properties assumptions lead to roughly a factor of
 2 range of D_i at lowest w and nearly a factor of 4 range of
 D_i at highest w . Whereas D_i is variable with ice properties,
 A_i at all w falls into two groups: Mitchell and Heymsfield
 properties, with relatively large A_i , and all other ice proper-
 ties including spheres and our models derived here, with A_i
 systematically smaller by roughly a factor of 3 at all w . Dis-
 870 persion (ν) exhibits up to factors of 2–3 difference (Fig. 21e).
 The ice properties associated with the lowest effective den-
 sity (Mitchell) have greatest D_i and ν . However, at lowest
 w the Bucky ball model exhibits substantially greater D_i but
 similar ν as spheres, which can be attributed to a weak depen-
 dence of ρ_e on D_{\max} at $D_{\max} > 100 \mu\text{m}$ that is more similar
 to spheres than other ice models (Fig. 7).

In summary, in the simple case of a non-sedimenting par-
 cel, differing ice property assumptions lead to a factor of 2
 difference in N_i and factor of 3 in A_i . Up to a factor of 2
 increase in ν is also induced by ice properties that exhibit
 a trend in ρ_e across the relevant size distribution relative to
 ice properties with constant ρ_e . Differences in ρ_e across ice
 properties considered here (regardless of trend) also lead to
 factors of 2–3 difference in D_i .

5.3 Parcel simulations with sedimentation

When sedimentation is included with an assumed parcel
 depth of 100 m following Kay and Wood (2008), results
 are largely unchanged at the strongest w since $v_f \ll w$ (cf.
 Fig. 14); the only notable change is roughly a factor of 2
 reduction in N_i by -55°C , seen primarily as a uni-
 form downward shift of the PSDs between Fig. 17a and
 Fig. 17b. However, at $w \ll 1 \text{ m s}^{-1}$, the parcel behavior
 changes rather dramatically because sedimentation reduces
 surface area sufficiently to allow aerosol freezing events
 repeatedly as the parcel ascends, every 250–500 m when
 855 $w = 0.1 \text{ m s}^{-1}$ (Fig. 18d) and at least ten times more fre-
 quently when $w = 0.01 \text{ m s}^{-1}$ (Fig. 18f). At intermediate w ,
 nucleation occurs roughly 50% less frequently for the slow-
 est falling ice (Mitchell, Heymsfield) than for other ice prop-
 erties. At the greatest w , nucleation does occur eventually if
 parcel ascent is continued for several kilometers (not shown).
 Thus, the frequency of nucleation events is impacted by the
 differing assumptions about ice properties and capacitance,
 and the spread in N_i seen for $w = 1 \text{ m s}^{-1}$ can be viewed as a
 frequency difference with a very long period. With sedimen-
 tation at -55°C , Fig. 17 shows that some size distributions

happen to be in a period with small crystals present whereas others do not.

Although sedimentation only reduces parcel m and A_i , maximum parcel N_i may be increased over their values without sedimentation by more than a factor of two owing at least in part to faster aerosol freezing at colder temperatures. Nonetheless, in parcels subject to repeated nucleation events, sedimentation reduces time-averaged N_i by nearly an order of magnitude and time-averaged A_i by even more. D_i and ν_i experience briefer discontinuities associated with nucleation events, D_i dropping and ν_i increasing each time new crystals appear. The bullet and polycrystal models derived here exhibit a lagged transition in ν_i after each nucleation event compared with the other ice properties, which can be attributed to evolution between ρ_e varying not all with $D_{\max} < 100 \mu\text{m}$ (giving minimum ν_i) to ρ_e decreasing with $D_{\max} > 100 \mu\text{m}$ (increasing ν_i only when new crystals grow past $100 \mu\text{m}$).

At the greatest w , sedimentation results in D_i and ν similar in magnitude to that without sedimentation (e.g., Fig. 19f versus Fig. 19c), but the addition of sensitivity to fall speed increases the spread across N_i and A_i . Thus, we conclude that with or without sedimentation, a chief effect of varying ice properties is on the size distribution of ice owing to differing ρ_e , leading to roughly factor of 2–3 differences in D_i and ν_i in this parcel framework. We note that these parcel simulations with and without sedimentation generate results that span the range of N_i , D_i , A_i , and ν_i observed in situ during SPARTICUS, but we do not attempt any direct comparisons owing to the lack of realism of this simulation framework.

5.4 Optical properties

Extinction cross sections, scattering asymmetry parameters, and single-scattering albedos that are consistent with the derived crystal geometries are needed for interactive radiative calculations in cloud-resolving simulations and for calculation of diagnostic fluxes and radiances to be compared with measurements (e.g., van Diedenhoven et al., 2012). Infrared radiative transfer is dominated by emission, which is affected by particle size, but its sensitivity to crystal shape is minimal (e.g., Holz et al., 2016). However, particle shape does affect the relevant shortwave optical properties substantially. Detailed, accurate calculations of optical properties of non-spherical ice particles are generally computationally expensive. Existing databases and calculations of optical properties (e.g., Um and McFarquhar, 2007, 2011; Yang et al., 2013) assume crystal geometries based on sparse measurements and ad hoc assumptions that generally do not match the geometries derived here. As an alternative, approaches such as those of Fu (1996, 2007) and van Diedenhoven et al. (2014a) can be used to approximate the optical properties of complex crystals based on those of hexagonal prisms that serve as radiative proxies. Here we adopt the van Diedenhoven et al. (2014a) parameterization to approximate the optical

properties of our derived crystal geometries. This parameterization provides the extinction cross section, asymmetry parameter, and single-scattering albedo at any shortwave wavelength for ice particles with any combination of crystal volume ($V = m/\rho_i$), A_p , and α_e and roughness of crystal components. Ice refractive indices are taken from Warren and Brandt (2008).

~~In the van Diedenhoven et al. (2014a) parameterization, the van Diedenhoven et al. (2014a) parameterization is based on geometric optics calculations. Accordingly, it assumes the extinction efficiency (Q_e) is assumed to be 2 for all particles and wavelengths. Here To partly correct this simplification for small particle sizes, here we apply anomalous diffraction theory to adjust Q_e at wavelength λ for particles with effective size parameter $P = 2\pi V(m_r - 1)/(\lambda A_p)$ less than $\pi/2$, where m_r is the real part of the ice refractive index (Bryant and Latimer, 1969). We also apply the edge effect adjustment given by Nussenzveig and Wiscombe (1980). Both adjustments depend on V and A_p .~~

The single-scattering albedo (w_s) is parameterized as a function of V , A_p , and α_e of the crystal components. All models use α_e of bullet arms for this calculation. In the case of the bullet and aggregate models, the arm length is taken to include the cap, and the width is taken as the cap base width where arms comprise only caps. In case of the polycrystal model, we use only bullet arm α_e , neglecting the slight increase of w_s owing to the thinness of the plates between arms. For the Bucky ball model, the α_e of the arms as given in Fig. 6c is limited to values of unity or greater to roughly account for the influence of the compact core where budding arms remain shorter than they are wide.

The asymmetry parameter (g) depends on particle V , A_p , and α_e values, as well as the crystal surface roughness, which may substantially lower g (e.g., Macke et al., 1996; van Diedenhoven et al., 2014a). In the van Diedenhoven et al. (2014a) parameterization, the level of surface distortion is specified by a roughness parameter δ as defined by Macke et al. (1996); ~~differently defined roughness parameters are found to be roughly equivalent (Neshyba et al., 2013; Geogdzhayev and van Diedenhoven, 2016).~~ The Macke et al. (1996) ray-tracing code perturbs the normal of the crystal surface from its nominal orientation by an angle that, for each interaction with a ray, is varied randomly with uniform distribution between 0 and δ times 90° . Similar commonly used parameterizations of particle roughness perturb the crystal surfaces using Weibull (Shcherbakov et al., 2006) or Gaussian (Baum et al., 2014) statistics rather than uniform distributions. However, Neshyba et al. (2013) and Geogdzhayev and van Diedenhoven (2016) demonstrated that the same roughness parameter value defined through a Weibull, Gaussian or uniform distribution represents very similar crystal microscale surfaces and yields largely equivalent scattering properties. Unfortunately, the roughness parameter cannot be constrained by the

CPI data used here. Laboratory studies demonstrate that the microscopic structure of ice crystals is dependent on the environmental conditions in which they grow (Neshyba et al., 2013; Magee et al., 2014) (Neshyba et al., 2013; For the same reason, forecasts with sedimentation is generally lower. Since Baum et al. (2014) and van Diedenhoven et al. (2014b) show that a roughness parameter of 0.5 best fit observations, that is the default value we adopt here. For the Bucky ball model, we average core and arm g values, weighted by their relative contributions to total A_p (cf. Fu, 2007; van Diedenhoven et al., 2015). For the polycrystal model, the arm and plate g values are averaged in the same way. Since the plate-like structures on the polycrystals shown in Fig. 10 appear relatively transparent, we assume smooth surfaces for the plates (i.e., $\delta = 0$).

Calculated Q_e , g , and w_e are shown in Fig. 22 as a function of crystal D_{\max} . Also shown are the optical properties of the six-branch bullet rosette model calculated by Yang et al. (2013). The geometry of the bullet rosettes assumed by Yang et al. (2013) is taken from Mitchell and Arnott (1994) and is similar to that of Mitchell et al. (1996) shown in Fig. 4. Yang et al. (2013) calculate the optical properties using a combination of improved geometric optics and other methods, which reveals resonances in the extinction efficiencies that are not seen in our results. However, such resonances largely cancel out when integrated over size distributions (Baum et al., 2014). The calculated g values generally increase with size because of increasing α_e with size (cf. Fig 4c). At visible wavelengths, g of the bullet, Bucky ball, and aggregate models, as well as the Yang et al. (2013) bullet rosettes, converge at about 0.81 at large sizes. Because of the addition of thin smooth plates to the polycrystal model, its g is generally greater. Aside we note that assuming plates with $\delta = 0.5$ reduces $0.5\text{-}\mu\text{m}$ g by only about 0.01 in the limit of large D_{\max} (not shown), indicating that plate aspect ratio is the main cause of g increase. At $2.1\text{ }\mu\text{m}$, g values increase owing to ice absorption (e.g., van Diedenhoven et al., 2014a). The $2.1\text{-}\mu\text{m}$ g from Yang et al. (2013) is generally lower than our results because w_s is generally greater. At a given λ , w_s of an ice crystal is mostly determined by the particle effective diameter $D_{\text{eff}} = 3V/(2A_p)$ (van Diedenhoven et al., 2014a). Figure 23 shows D_{eff} as a function of D_{\max} . For the Yang et al. (2013) bullet rosettes, $D_{\text{eff}}(D_{\max})$ is generally substantially smaller than for our models, which is consistent with our generally greater V and smaller A_p compared to the Mitchell et al. (1996) bullet rosettes (see Fig. 4).

Figure 24 shows the shortwave optical properties integrated over the model size distributions at -55°C shown in Fig. 17. Extinction efficiencies generally increase slightly with λ as P decreases and are therefore generally greater for the cases with sedimentation owing to the smaller crystal sizes. At $\lambda \sim 2.8\text{ }\mu\text{m}$, a [Christiansen band \(Arnott et al., 1995\)](#) is present where a combination of strong absorption and refractive indices greater near or less than unity leads to a decrease in Q_e (cf. Baum et al., 2014). The w_s is generally greater for cases with sedimentation since

these simulations lead to small $D_{\text{eff}} \sim 20\text{ }\mu\text{m}$, whereas D_{eff} produced by simulations without sedimentation range from about 50 to 500 μm , primarily depending on w (cf. Fig. 17). For the same reason, forecasts with sedimentation is generally lower.

6 Discussion and conclusions

In preparation for large-eddy simulations with size-resolved microphysics for a case study of mid-latitude synoptic cirrus observed on 1–2 April 2010 during the SPARTICUS campaign (Muhlbauer et al., 2015), here we use CPI image analysis to develop ice crystal geometries that are physically continuous over the required crystal size range and suitable to calculate internally consistent physical and optical properties. The model to be used employs the Böhm (1999, 2004) approach to calculate fall speeds and pairwise collision rates (based on crystal mass m , maximum projected area A , corresponding maximum dimension D , and bodily aspect ratio α) and the van Diedenhoven et al. (2014a) approach to calculate radiative properties (based on crystal mass m , maximum projected area A , and crystal or polycrystal element aspect ratios α_e). Assuming bullet rosettes as a typical geometry, we approximate α as unity (no preferred fall orientation), consistent with adoption of measured (randomly oriented) maximum dimension D_{\max} and projected area A_p for physical and optical properties. We then take an approach to estimating mass from CPI image data that begins with derivation of geometric crystal components suitable for calculation of optical properties, based on available ICR measurements. We also use derived α_e values in calculation of capacitance for vapor growth. This approach to ice crystal properties offers an advance over our past, ad hoc approach of using piecewise mass- and area-dimensional relations as a foundation, and then separately assigning aspect ratios based on sparse literature sources (e.g., Avramov et al., 2011; Fridlind et al., 2012; van Diedenhoven et al., 2012).

Our results using a typical bullet model of rosettes give $m(D_{\max})$ systematically larger than literature values used in similar past size-resolved microphysics simulations (Sölch and Kärcher, 2010), and $A_p(D_{\max})$ systematically smaller or similar. Taken together, these differences lead to v_f greater by a factor of 1.5–2, and w_s and g respectively greater by about 0.2 and 0.05 in the limit of large D_{\max} at near-infrared λ . A polycrystal model that estimates side plane growth on bullet rosettes increases v_f by only about 15%, indicating that the effect of increased m outweighs that of increased A_p given the relatively ad hoc assumptions made here. In the polycrystal model, side plane growth also increases g by about 0.05, primarily owing to plate aspect ratio.

In parcel simulations with and without sedimentation, differing ice properties lead to factors of 2–4 difference in crystal number concentration N_i , number-weighted mean diameter D_i , total projected area A_i , and size distribution relative

dispersion ν_i . When crystal effective density ρ_e is smaller, D_i is larger; when ρ_e varies with size, ν_i is larger. When $v_f \gg w$, faster falling crystals are associated with more frequent nucleation events, by roughly 50% at $w = 0.1 \text{ m s}^{-1}$.

Overall, it appears that the main differences between our models and past literature arises from differences in bullet rosette geometry (i.e., ~~bullet arm properties~~ single-particle mass) or its representation (i.e., definition of D_{\max}). Where available, ~~derived arm α_e values appear~~ $A_p(D_{\max})$ and arm $\alpha_e(D_{\max})$ appear more similar, by contrast. Based on ad hoc assumptions made here, the chief potential impact of side plane growth could be an increase in g by ~ 0.05 in the mid-visible. More detailed observational analysis would be needed to confirm side plane properties assumed here. However, differences between our polycrystal and bullet properties are surprisingly substantially less than the differences between our bullet properties and those in past literature, which may prioritize better establishing the baseline bullet rosette model over working out details of irregular crystal properties. ~~With respect to the bullet rosettes,~~

Evolution of newly nucleated ice crystals may proceed from amorphous shapes to more defined habits (e.g., Baker and Lawson, 2006b; Schnaiter et al., 2016) in a manner that may depend in part on nucleation mode (e.g., Bacon et al., 2003; Schnaiter et al., 2016), but observations considered here are inadequate to derive a robust geometric model for D_{\max} smaller than roughly $100 \mu\text{m}$ for the case study considered here, as in other recent work (e.g., Erfani and Mitchell, 2016). However, it appears we find that growth from a budding Bucky ball shape versus an idealized bullet rosette shape could lead to non-negligible differences in normalized capacitance of nearly 0.1 (cf. Fig. 15). If such geometry is important at the tops of cirrus to predicted PSD evolution, deriving a statistically decreasing number of arms with increasing size could be needed to simultaneously represent the evolution of crystal m , A_p , and α_e . Or more accurate geometries could be established (e.g., Nousiainen et al., 2011; Schnaiter et al., 2016) and relevant physical and optical properties made appropriately consistent and continuous for modeling purposes. Evident diversity of both small and large crystal properties at a given D_{\max} , even when most rigorously defined, could also be relevant.

It may be the case that uncertainties in ice crystal m and its relationship to morphological properties, which together determine factors such as v_f and radiative properties, are not sufficiently considered in current literature. Single-crystal mass measurements that were made laboriously in studies decades ago (e.g., Kajikawa, 1972) (e.g., Kajikawa, 1972; Mitchell et al., 1990) not been replaced by improved measurements or substantially augmented since that time. In the case of bullet rosettes, for instance, we are aware of only one unpublished data set comprising 45 crystals, we are aware of no such measurements made at cirrus elevations, and it appears

that those ground-level measurements may be biased to fewer branches, as discussed above. From analysis of the SPARTICUS data here, we can see that such a bias in branch number could likely be correlated with a bias in α_e and m . The degree to which a single habit-independent $m - A_p$ power law applied to 2DS PSDs leads to accurate calculation of $m(D_{\max})$ for both anvil and synoptic cirrus crystal conditions may also warrant additional investigation (cf. Fig. 4e). As discussed by Baker and Lawson (2006a), for instance, particles are not entirely randomly oriented in the petri dish measurement approach used in the SCPP data set; to the extent that non-random orientation favors a higher ratio of A_p/m on a petri dish, the derived $m(A_p)$ could be biased correspondingly low when applied to randomly oriented crystal images.

With respect to classification of morphological properties, it also appears to be the case that classification algorithms may give substantially differing results. For instance, whereas here roughly 80% of crystals with D_{\max} greater than $100 \mu\text{m}$ are unclassified (irregular), the algorithm reported by Lindqvist et al. (2012) classifies more than 50% of crystals as rosettes in a similar mid-latitude cloud. The fact that their study places fewer than 20% of crystals in an irregular class across tropical, Arctic, and mid-latitude conditions suggests that it is fundamentally different from the algorithm applied here. The fact that unclassified crystals here differ relatively little in derived properties from bullet rosettes (with the possible exception of g , given some relatively ad hoc assumptions) suggests that algorithms may currently differ in the allowable degree of deviation from a pristine state. It may be useful to establish comparable statistics from differing algorithms to allow comparison of circumference or other non-habit-dependent measures.

Overall, the results obtained here motivate the use of our derived ice properties in comparison with more widely used values in 3D simulations of the April 1–2 SPARTICUS conditions, which can in turn be compared with in situ ice size distribution observations.

Appendix A: Ice crystal models

A fundamental geometric element of all ice models considered below is the regular hexagonal column with length L and width W , defined here as twice hexagon side length. In all cases the true mean branch width W is taken as the mean of the measured widths of all branches divided by a factor of $(1 + \sqrt{3}/2)/2$ to correct for random orientation. Thus, true mean branch width W is about 7% wider than measured branch width.

Since branch length measurements extend from crystal center to the outermost edge of projected randomly oriented branches, the true mean total branch length (including cap or core contributions, depending on the model) is taken as the mean of the measured lengths less one-half of the mean of

the measured widths times $\pi/4$ (the contribution of randomly oriented projected base to measured length), all multiplied by a factor of $4/\pi$ to account for branch foreshortening by random orientation. Thus, true mean branch length is about 30% longer than measured mean branch length corrected for the contribution of column base projection.

All non-aggregate models (bullet, Bucky ball, and polycrystal) assume six branches, consistent with mean and median number found over all bullet rosettes measurable with the ICR software.

Derived ice properties are supplied as [supplementary material](#) [the Supplement](#).

A1 Bullet model

The bullet model assumes that each hexagonal column has a single cap, and that the six caps meet at a point in the center of the crystal. If the cap is a hexagonal pyramid with a fixed angle β between [hexagonal-base-vertices](#) [pyramid edges](#) and the line defining pyramid height, then cap length L_c scales with column width according to

$$L_c = \frac{W}{2 \tan \beta}. \quad (\text{A1})$$

Thus, wider branches have longer caps. Here we assume fixed β , and assign a value of 22° ; generally wider angles have been assumed in previous work, as discussed further at the end of this section.

For the bullet model, total true branch length includes both hexagonal column length L and hexagonal pyramid cap length (L_c). A least squares linear fit of total mean branch length $L+L_c$ to measured maximum dimension (uncorrected for random orientation throughout, see Section 3) gives a line nearly through the origin. Adopting only the slope (cf. Fig. 4a) gives

$$L + L_c = 0.691 D_{\max}. \quad (\text{A2})$$

A least squares fit of mean branch width W to D_{\max} gives (cf. Fig. 4b, μm units)

$$W = 0.139 D_{\max} + 40.6. \quad (\text{A3})$$

In the limit of zero D_{\max} , Equation A3 would give a non-zero branch width. As a simple physical solution, we assume that branch width is equal to cap base width wherever predicted cap length per Equations A1 and A3 is greater than total branch length per Equation A2, designated as $D_{\max,L0}$ (where branch length L is zero). Thus, for the bullet model, crystals with mean branch width less than $D_{\max,L0} \sim 50 \mu\text{m}$ comprise hexagonal pyramids without developed hexagonal columns extending from them; we note that no ICR measurements were possible at such small sizes.

With crystal geometry now defined using the bullet model, it is straightforward to calculate the aspect ratio $W/(L+L_c)$,

surface area A_s , and mass m for each measured crystal (symbols in Fig. 4c–e). The crystal model derived and used in simulations is that for a corresponding typical crystal with number of branches fixed to six, equal to both mean and median of measured branch numbers (solid lines in Fig. 4c–e), with mass therefore defined as

$$m = 6\rho_i \frac{\sqrt{3}}{8} W^2 (3L + L_c), \quad (\text{A4})$$

where ρ_i is the bulk density of ice, taken here as 0.917 g cm^3 , and surface area defined as

$$A_s = 6 \left(3LW + \frac{3\sqrt{3}}{8} W^2 + \frac{3}{4} W \sqrt{\frac{3}{4} W^2 + 4L_c^2} \right). \quad (\text{A5})$$

However, the randomly oriented projected area A_p is not analytically defined. A least squares fit of measured crystal projected areas to bullet model crystal surface areas results in a line nearly through the origin (cf. Fig. 4d), and this is used with Equation A5 to define model projected area

$$A_p = \lambda_b A_s, \quad (\text{A6})$$

where $\lambda_b = 0.107$. The linear relationship and slope < 0.25 are consistent with theory for convex particles (Vouk, 1948), as discussed above.

If the cap angle β is increased, effective density and projected area ratio increase for ice crystals with D_{\max} smaller than about $90 \mu\text{m}$. In the limit of small D_{\max} , a β of 22° is selected to give effective density and projected area ratio no larger than that calculated for any measured rosettes (cf. Fig. 5a, b). Calculated fall speeds are not strongly sensitive to changes in β because effective density and projected area increase or decrease together. Regarding choice of β , Iaquinta et al. (1995) have noted that a bullet rosette with a six-faced pyramidal end and a 56° angle between opposing faces has been assumed in past work but cannot fit to form a multibranch bullet rosette, leading to their adoption of a trilateral pyramidal end as “only an idealized form of the sharp end of natural ice crystals”; we select β values here in the same spirit.

A2 Bucky ball model

The Bucky ball model assumes that each hexagonal column grows initially from a Bucky ball face. The core is approximated as a sphere with diameter D_c of $10 \mu\text{m}$, and budding columns are assigned an initial width W_{\min} of $4 \mu\text{m}$. In order to insure a branch length of zero when D_{\max} is equal to that of a sphere with core diameter D_c , a slope is fit to L as a function of $D_{\max} - D_c$ (Fig. 6a), giving

$$L = 0.684(D_{\max} - D_c). \quad (\text{A7})$$

Similarly, in order to insure a branch width of W_{\min} when the maximum dimension is equal to the core diameter D_c , a

slope is fit to $W - W_{min}$ as a function of $D_{max} - D_c$ (Fig. 6b), giving

$$W - W_{min} = 0.216(D_{max} - D_c). \quad (\text{A8})$$

With crystal geometry now defined using the Bucky ball model, it is straightforward to calculate the branch aspect ratio (L/W) and mass for each measured crystal. For the canonical crystal with six arms, where $D_{max} < D_c$, then values are those of a sphere with diameter D_{max} and density ρ_i . Otherwise, using W and L from Equations A7 and A8,

$$m = \rho_i \left(\frac{\pi}{6} D_c^3 + 6 \frac{3\sqrt{3}}{8} W^2 L \right). \quad (\text{A9})$$

Rigorous calculation of A_s for the model crystal with typical six branches is less straightforward. Here we take the ad hoc approach of first estimating the surface area of measured crystals as the total of branches with one end each, neglecting the inner end faces (Equation A5 without the third term that represents cap surface area). A fit of A_p measured to $A_{s,est}$ so estimated gives a slope 0.0921 (Fig. 6d). The model crystal with six arms is then assigned $A_s(D_{max})$ as a weighted average of estimated A_s and that of a sphere with diameter D_{max} ,

$$A_s = f_s 4\pi D_{max}^2 + f_b A_{s,est}, \quad (\text{A10})$$

where m_r is the ratio of m to that of a sphere with diameter D_{max} , $m_{r,max}$ is the value in the limit of large D_{max} (roughly 0.24),

$$f_s = 1 - \frac{1 - m_r}{1 - m_{r,max}}, \quad (\text{A11})$$

and

$$f_b = 1 - f_s. \quad (\text{A12})$$

Relative to the bullet model, the Bucky ball model exhibits a stronger increase of W with increasing D_{max} (cf. Fig. 4b and 6b) and a nearly constant aspect ratio at D_{max} greater than 100 μm (cf. Fig. 4c and 6c). Smooth variation of radiative properties and capacitance in the limit of small D_{max} is achieved with

$$\alpha_e = \max(1, L/W) \quad (\text{A13})$$

and (Fig. 15)

$$\frac{C}{D_{max}} = \min \left(0.5, 0.4 \left(\frac{L}{W} \right)^{0.25} \right). \quad (\text{A14})$$

A3 Aggregate model

The ‘aggregate model’ is an extension of the bullet model. A least squares fit of mean branch length $L + L_c$ to D_{max} gives a

line again nearly through the origin. Adopting only the slope as in the bullet model (cf. Fig. 9a) gives

$$L + L_c = 0.461 D_{max}. \quad (\text{A15})$$

The mean and median measured number of arms is twelve, consistent with aggregates primarily of two typical single rosettes. The roughly 30% reduction in slope compared with single rosettes can be attributed to the overlap of aggregate arms, compounded by random orientation when two crystals create a linearly aligned pair that will be rarely normal to the viewing angle. A least squares fit of mean branch width W to D_{max} gives (cf. Fig. 9b, μm units)

$$W = 0.0886 D_{max} + 44.9. \quad (\text{A16})$$

To handle unphysical branch widths in the limit of zero D_{max} , we again assume that branch width is equal to cap base width wherever predicted cap length per Equations A1 and A16 would be greater than total branch length per Equation A15.

Using this model for aggregates, mass and projected area are simply twice that of bullet rosettes,

$$m = 12\rho_i \frac{\sqrt{3}}{8} W^2 (3L + L_c), \quad (\text{A17})$$

where ρ_i is the bulk density of ice, taken here as 0.917 g cm^3 , and surface area defined as

$$A_s = 12 \left(3LW + \frac{3\sqrt{3}}{8} W^2 + \frac{3}{4} W \sqrt{\frac{3}{4} W^2 + 4L_c^2} \right). \quad (\text{A18})$$

Using Equation A18 with A1, A15 and A16 to calculate A_s , measured A_p is found to be 10% of calculated A_s (cf. Fig. 9d), roughly 1% lower than found for single rosettes using the bullet model, consistent with branch entanglement that reduces A_p but not A_s relative to a pair of single rosettes.

A4 Polycrystal model

The polycrystal model is derived for unclassified crystals using plate growth on the bullet model as a basis. When the unclassified crystals are initially assumed to follow the bullet model, and measured A_p is regressed against calculated bullet surface area ($A_{s,b}$) following Equation A5, assuming six arms per crystals, the slope is greater than found for the bullet model, consistent with systematically greater projected area than rosettes demonstrated in Fig. 12. We adopt the ad hoc assumption that additional projected area can be attributed to side plane growth, represented here for simplicity as growth of hexagonal plates. Continuing with the six-arm bullet model as a basis, we further make the ad hoc assumption that a single plate is grown on each arm with sufficient total plate surface area ($A_{s,p}$) to restore $A_p/(A_{s,b} + A_{s,p})$ to a value near λ_b in the limit of large D_{max} .

Based on trial and error, taking the foregoing assumptions as a recipe, the following prescription was found to match effective density from Cotton et al. (2012) in the limit of small D_{\max} (Fig. 13e) and median measured $A_p(D_{\max})$ for unclassified crystals at all sizes (Fig. 13f) to the extent possible without exceeding the effective diameter of equivalent-sized spheres. First, to increase effective density relative to the bullet model where crystals are entirely truncated caps ($D_{\max} < D_{\max,L0}$), β is increased to 25° . If $A_{s,b}$ is then calculated following Equation A5 for measured crystals, a slope $\lambda_i = 0.147$ is found (Fig. 13a), larger than $\lambda_b = 0.107$ found for rosettes using the bullet model for measured rosettes (Equation A6). Next A_p/A_s is matched to allow zero plate contribution to surface area where $D_{\max} < D_{\max,L0}$ (noting that increased β slightly reduces $D_{\max,L0}$ relative to that for the bullet model) and maximum contribution to surface area within an ad hoc scale length of $2D_{\max,L0}$ using

$$\Delta D = D_{\max} - D_{\max,L0} \quad (\text{A19})$$

and, taking $\lambda_p = 0.1$ as the ratio of A_p to A_s for polycrystals (reduced by an ad hoc amount from that for bullets on the basis that A_s has increased relatively more than A_p , but not so much that effective density exceeds that of equivalent-sized spheres),

$$\lambda = \lambda_p + (\lambda_i - \lambda_p) \left(1 - \exp\left(-\frac{\Delta D}{2D_{\max,L0}}\right) \right). \quad (\text{A20})$$

For the model crystal with six arms, the plate contribution to surface area is then

$$A_{s,p} = 6 \left(\frac{\lambda}{\lambda_p} - 1 \right) A_{s,b}. \quad (\text{A21})$$

If plate surface area is approximated as twice the face areas (neglecting edge contributions), then per-plate diameter (D_p), defined for $D_{\max} > D_{\max,L0}$, can be calculated from

$$6D_p = \sqrt{\frac{2A_{s,p}}{9\sqrt{3}}}. \quad (\text{A22})$$

Plate thickness L_p , which is neglected in the addition of plate surface area to A_s but is included in calculation of plate contribution to crystal mass, is taken as (Pruppacher and Klett, 1997, their Table 2.2a, cm units):

$$L_p = \min(0.1D_p, 0.0141D_p^{0.474}). \quad (\text{A23})$$

In the limit of zero plate size, L_p is not permitted to exceed $0.1D_p$. Thus, for radiative calculations, the maximum plate aspect ratio $\alpha_{e,p} = L_p/D_p = 0.1$ and the bullet arm aspect ratio remains as $\alpha_{e,b} = (L + L_c)/W$. Normalized capacitance is calculated as for a bullet rosette with $L(\beta)$ and $W(\beta)$, neglecting the presence of plates, for lack of another obvious strategy.

Plate contribution to polycrystal mass can be calculated as

$$m_p = \frac{A_{s,p}}{2} L_p. \quad (\text{A24})$$

Total mass is then m_p plus bullet mass following Equation A4 with $\beta = 25^\circ$, and total projected area $A_p = \lambda(A_{s,p} + A_{s,b})$.

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Table 1. Mass- and area-dimensional power law coefficients for $m = aD_{\max}^b$ and $A = cD_{\max}^d$ in cgs units.

Habit	D_{\max}	a	b	c	d	Source [†]
Small bullet rosettes	<0.01	0.1	2.997	0.629535 [‡]	2.0 [†]	Mitchell (1994)
Large bullet rosettes	0.02–1	0.00308	2.26	0.08687	1.568	Mitchell (1996)
Large bullet rosettes	0.02–2	0.0139	2.54	0.2148 [‡]	1.7956 [‡]	Heymsfield et al. (2002)
Bullet rosette aggregates	0.04–2	0.00183	2.04	0.0803	1.45	Heymsfield et al. (2002)

[†] Calculated as described in text.
[‡] As cited in Sölch and Kärcher (2010), see text.

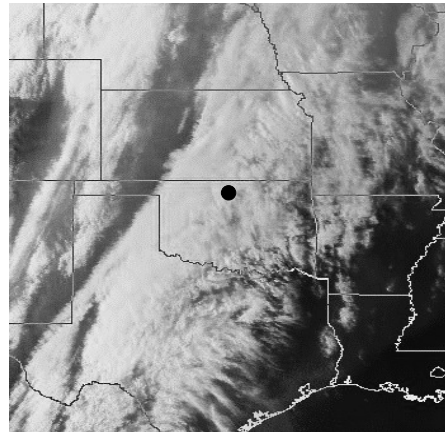


Figure 1. GOES composite image at 23:39 UTC on 1 April 2010. Black circle indicates the Southern Great Plains long-term measurement site in Oklahoma.

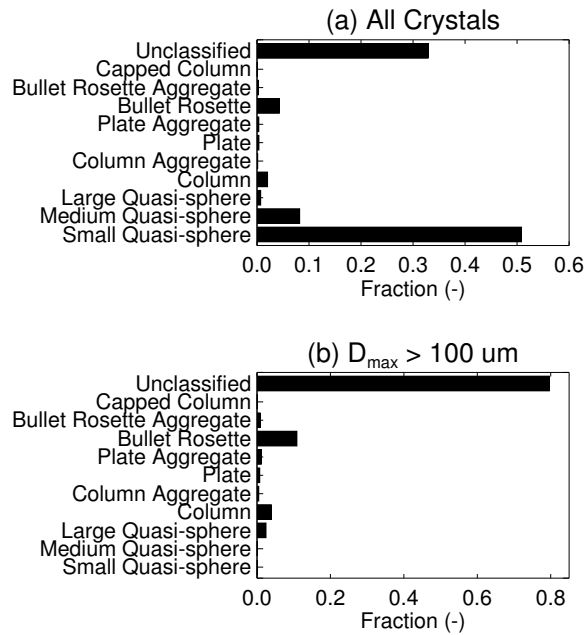


Figure 2. Fraction of ice by habit class for all crystals imaged (a) and for those with D_{\max} greater than 100 μm (b).

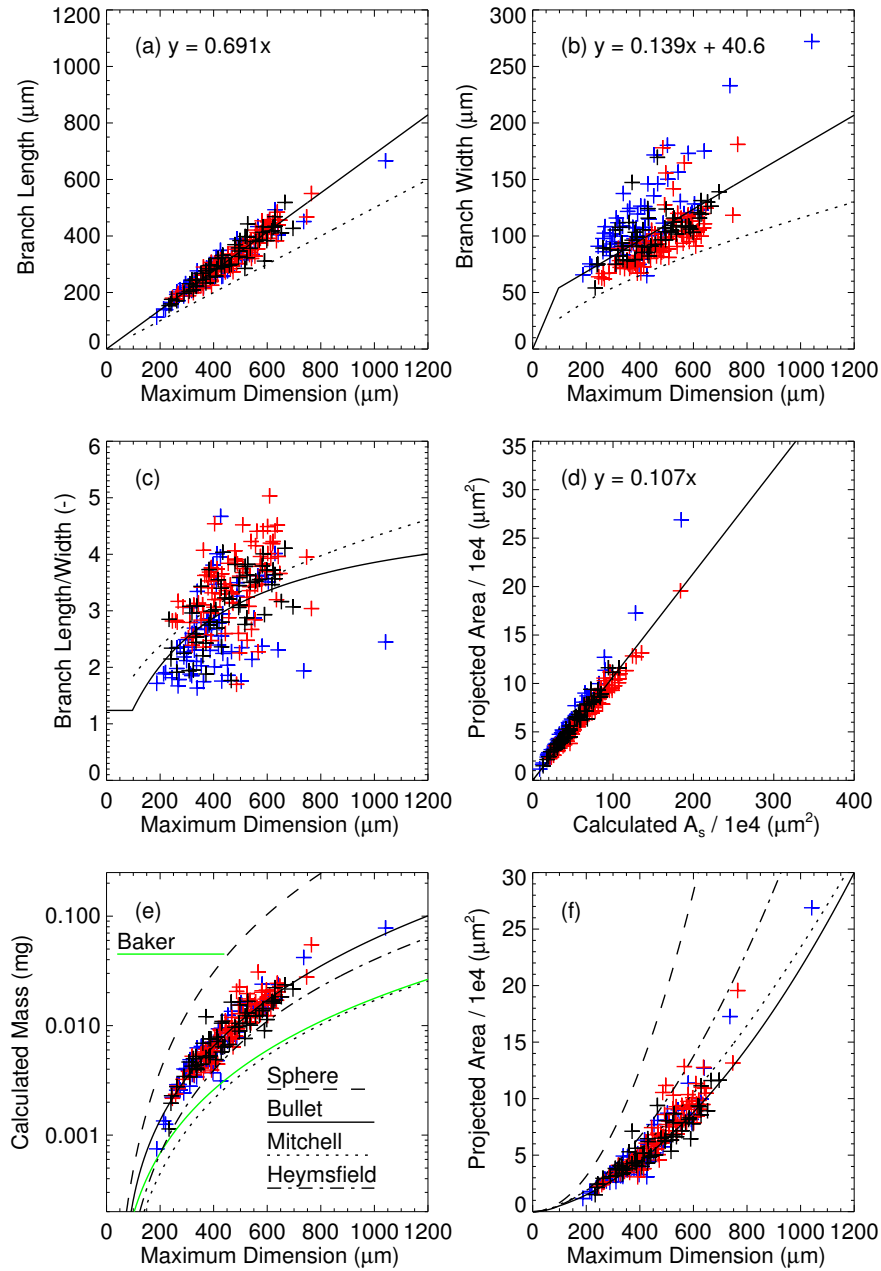


Figure 4. Bullet model: measured and calculated properties of imaged bullet rosettes with six arms (black symbols), fewer than six arms (blue symbols), and more than six arms (red symbols). Line types indicate derived ice properties as follows (see legend in panel e): a sphere, a six-arm bullet rosette per the bullet model (see Section 4.1 and Appendix A1), five-arm rosettes from Mitchell et al. (1996), and cirrus crystals from Heymsfield et al. (2002). Also shown is the habit-independent $m - A_p$ relation derived by Baker and Lawson (2006a).

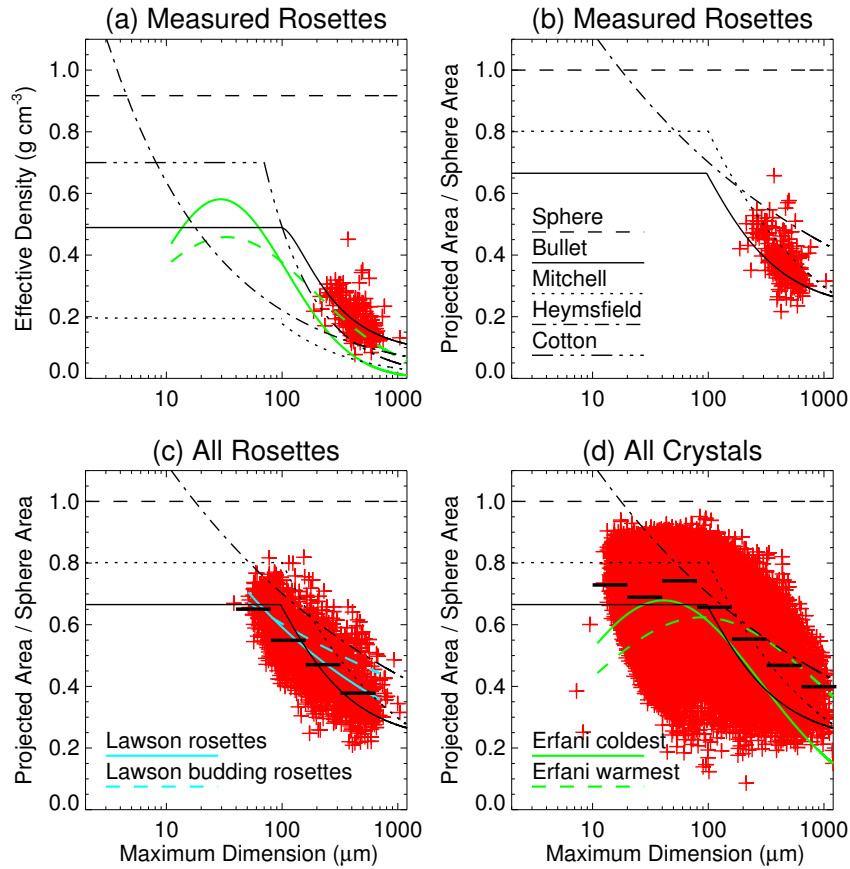


Figure 5. Bullet model: measured and calculated properties of imaged ice crystals (red symbols) emphasizing the transition to the smallest sizes. Effective density and projected area for bullet rosettes with ICR measurements (a, b), and projected area for all bullet rosettes identified (c, including those not measurable with the ICR software), and for all crystals imaged during the April 1–2 flights (d). Within D_{max} doubling bins, the median of measurements is shown where a bin contains more than 100 measurements (thick solid line segments, c and d only). Other line types indicate derived ice properties as follows (see legend in panel b): a sphere, a six-arm bullet rosette per the bullet model (see Section 4.1 and Appendix A1), five-arm rosettes from Mitchell et al. (1996), and cirrus crystals from Heymsfield et al. (2002) and Cotton et al. (2012). Also shown (see legends in c and d): fits to measured areas of bullet rosettes and budding bullet rosettes from Lawson et al. (2006a), and polynomial fits from Erfani and Mitchell (2016) for synoptic cirrus crystals at -55 to -65°C (coldest range fitted) and -20 to -40°C (warmest, see text).

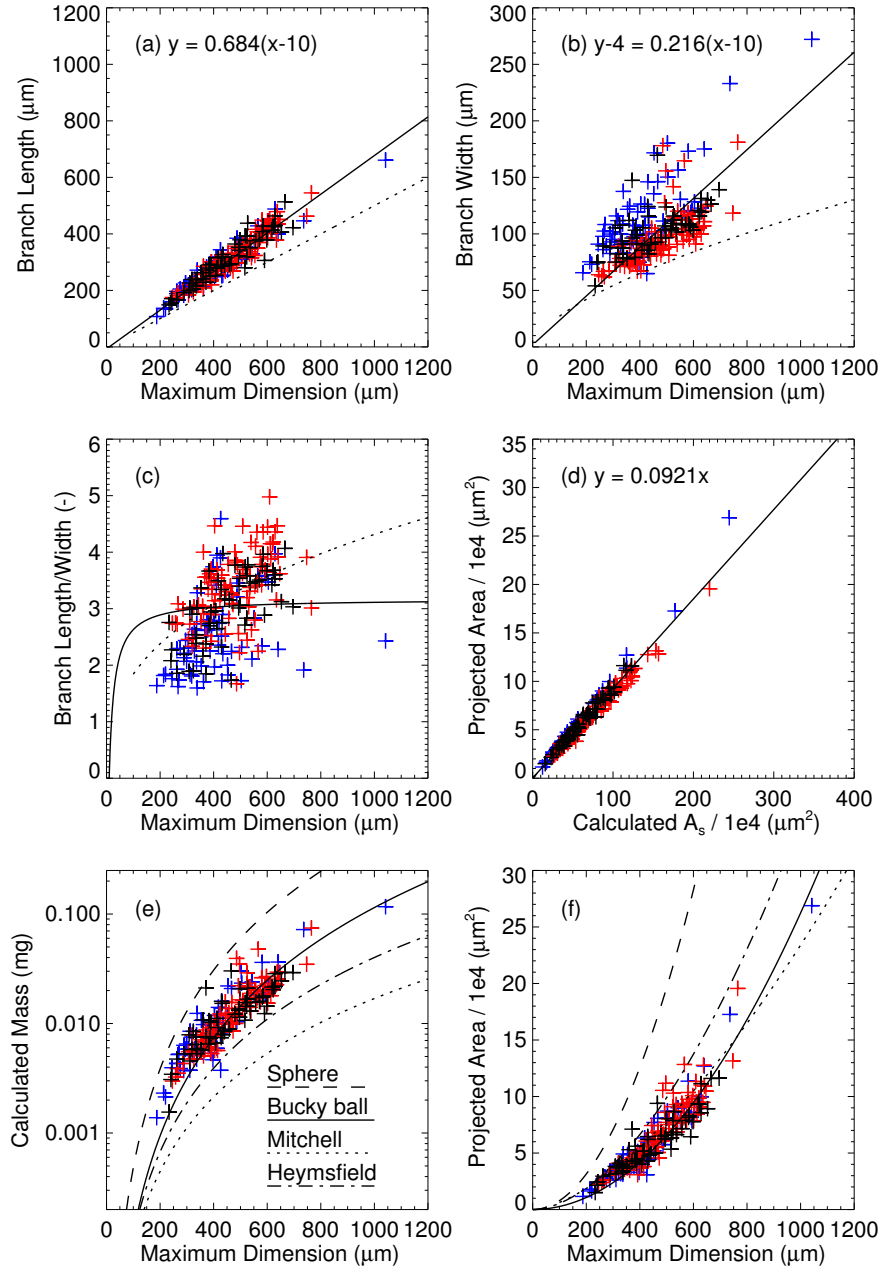


Figure 6. As in Fig. 4 except for Bucky ball model (see Section 4.2 and Appendix A2).

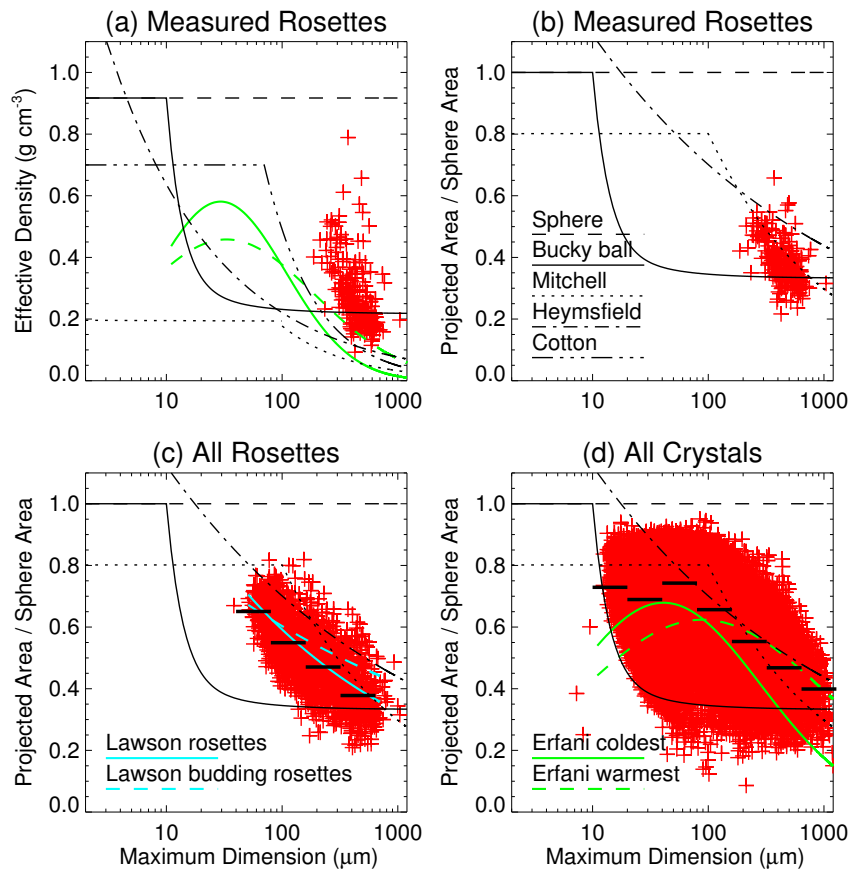


Figure 7. As in Fig. 5 except for Bucky ball model.

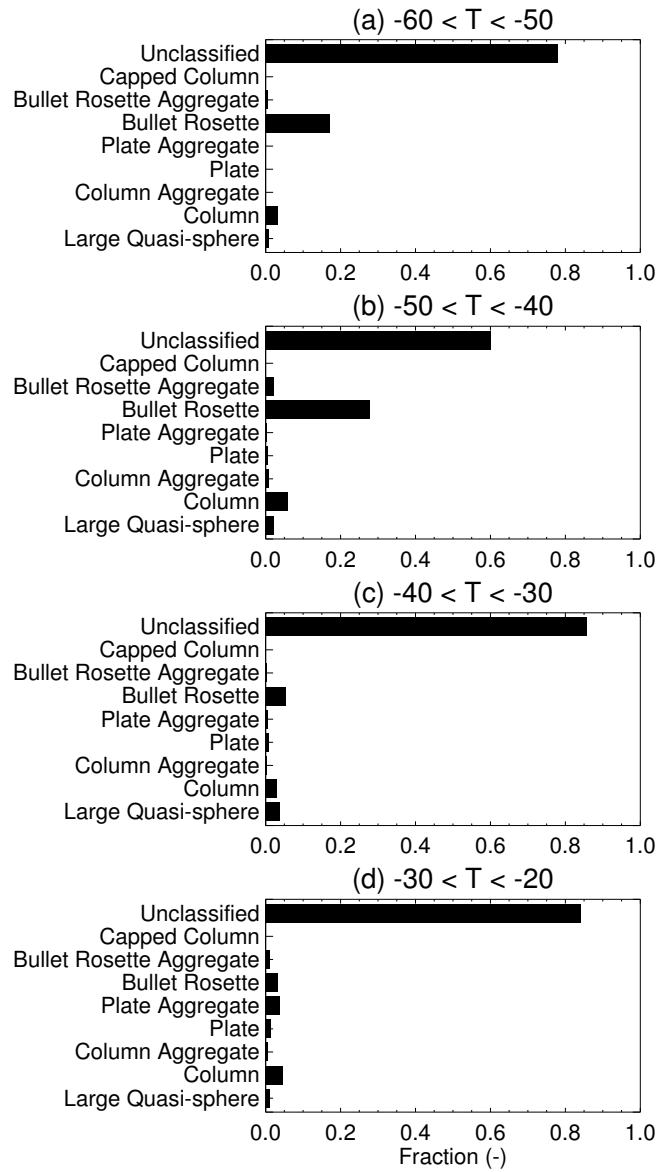


Figure 8. Fraction of imaged ice crystals with D_{\max} greater than $100 \mu\text{m}$ in four temperature ranges in degrees Celsius.

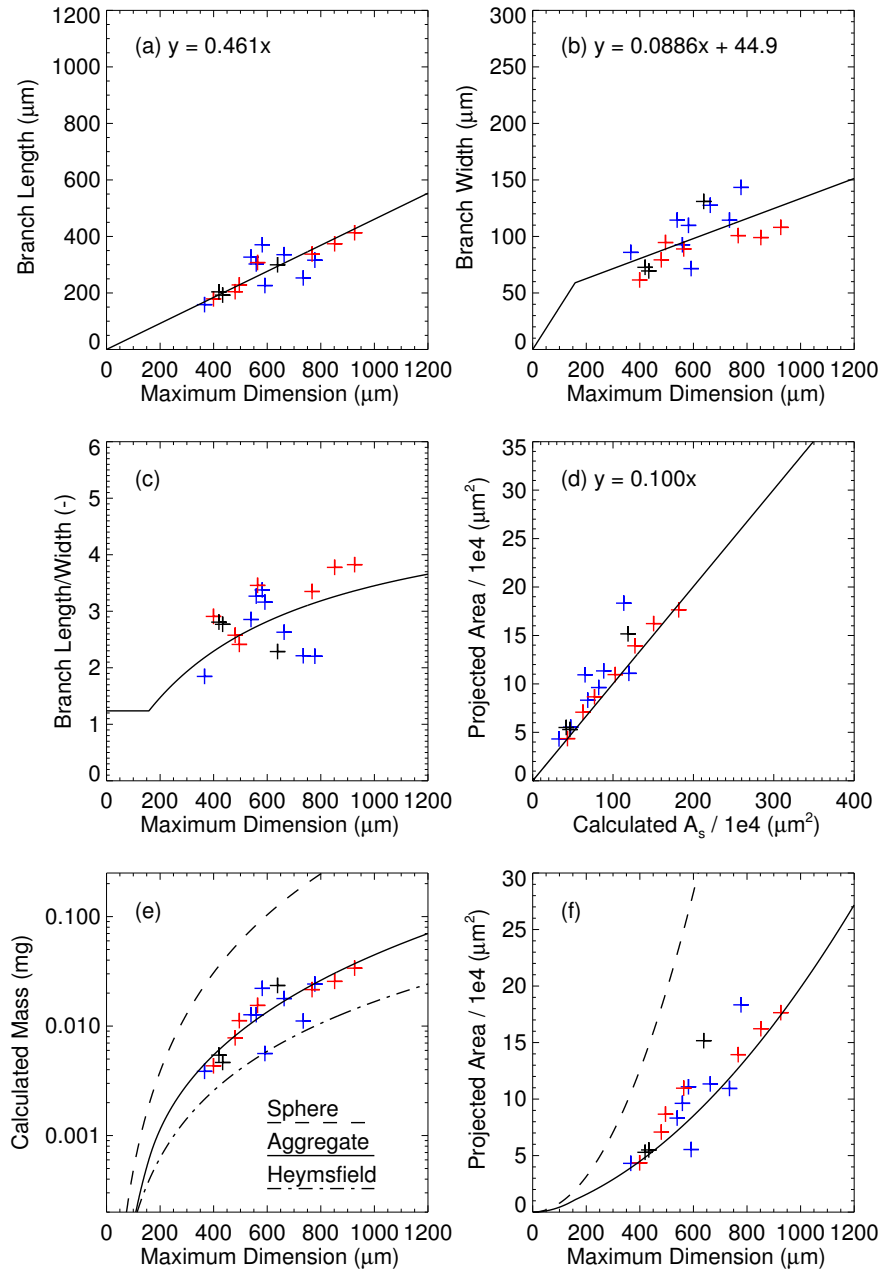


Figure 9. Aggregate model: measured and calculated properties of aggregates of bullet rosettes with twelve arms (black symbols), fewer than twelve arms (blue symbols), and more than twelve arms (red symbols). Line types indicate derived ice properties as follows (see legend in panel e): a sphere, a twelve-arm bullet rosette aggregate (see Section 4.3 and Appendix A3), and aggregates of bullet rosettes from Heymsfield et al. (2002).



Figure 10. Bullet rosettes and unclassified crystals with radiating growth imaged on 1 April.

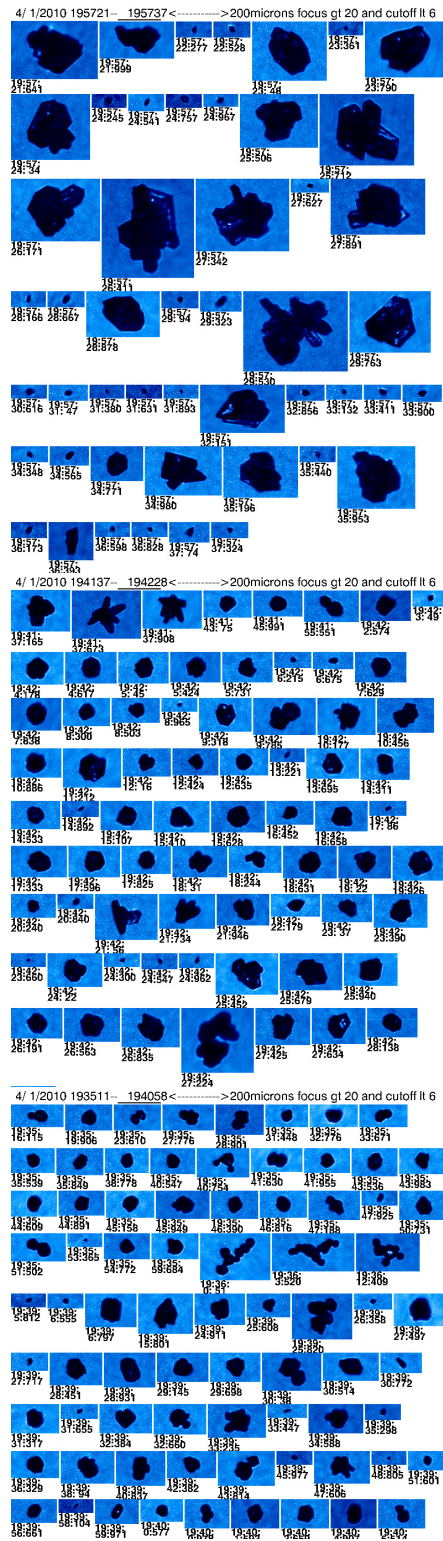


Figure 11. Unclassified ice crystals with sublimated edges imaged on 1 April.

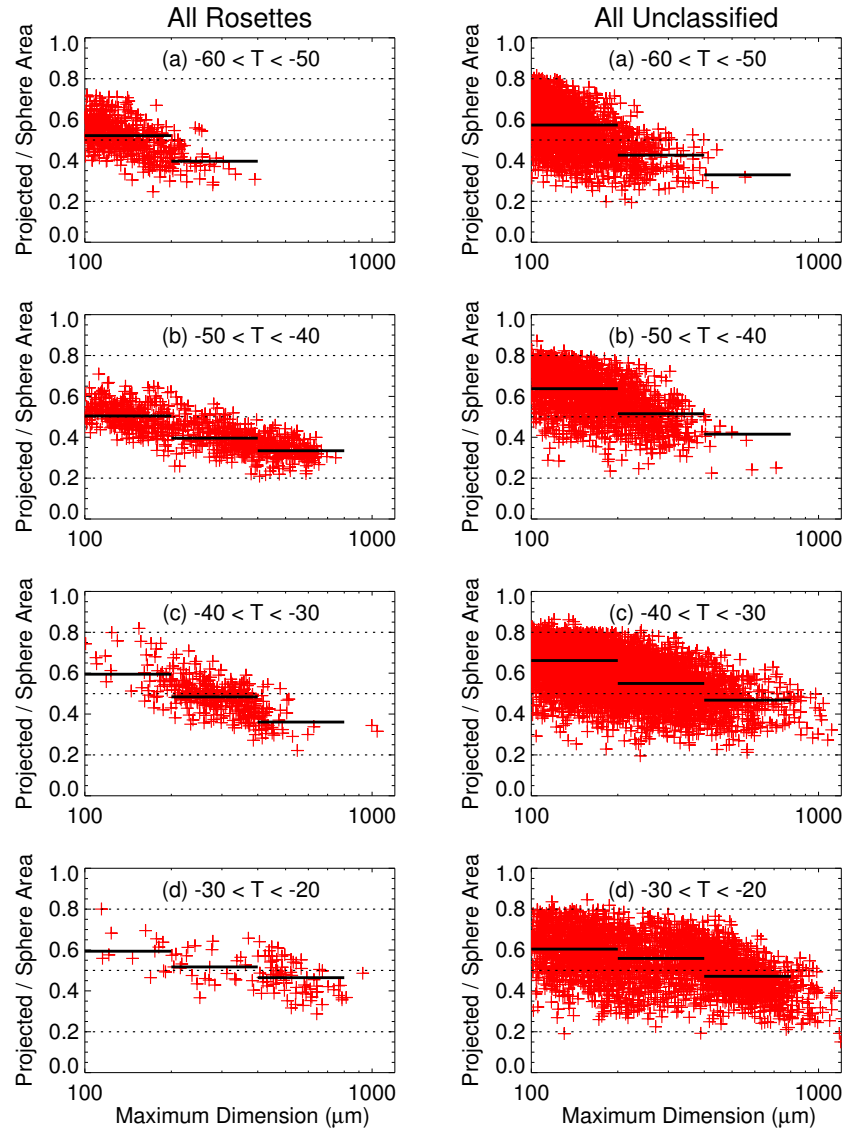


Figure 12. Ratio of measured A_p to area of a sphere with diameter D_{max} in four temperature ranges for all bullet rosettes (left column) and all unclassified crystals (right column). Overplotted solid line segments indicate median value over D_{max} -doubling bins.

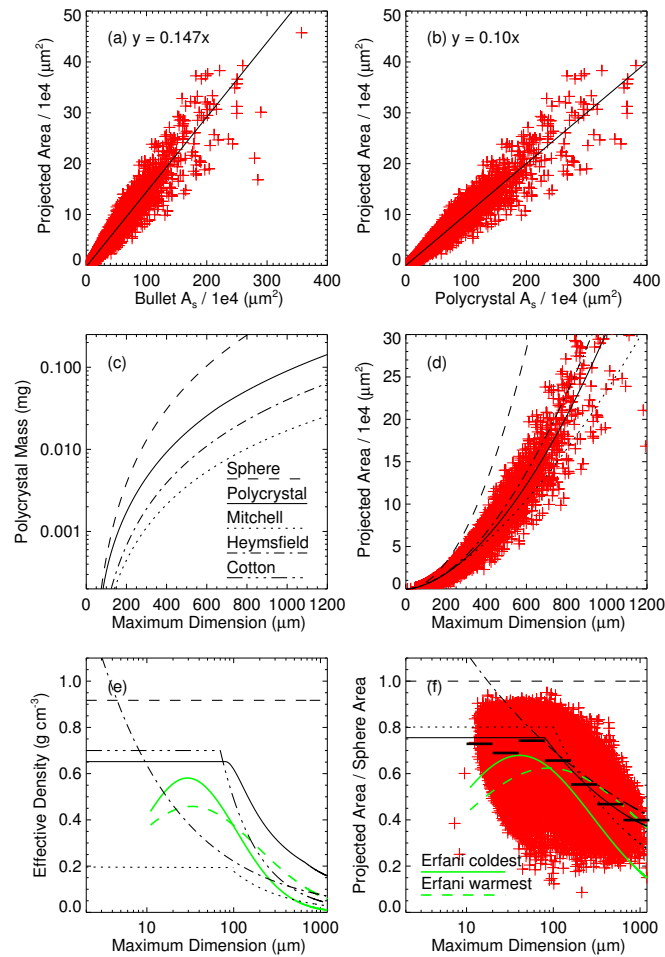


Figure 13. Polycrystal model: measured and calculated properties of unclassified ice crystals (red symbols). Line types indicate derived ice properties as follows (see [legend legends](#) in [panel panels c and f](#)): a sphere, a polycrystal based on a six-arm bullet rosette (see Section 4.4 and Appendix A4), five-arm rosettes from Mitchell et al. (1996), and cirrus crystals from Heymsfield et al. (2002) and Cotton et al. (2012), and Erfani and Mitchell (2016).

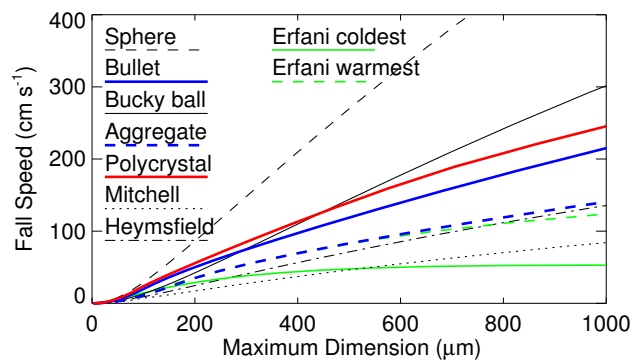


Figure 14. Ice crystal fall speeds at 350 mb and 233 K for derived ice properties as follows (see legend): a sphere, six-arm rosettes following the bullet and Bucky ball models, twelve-arm aggregates following the bullet model, the polycrystal model, five-arm rosettes from Mitchell et al. (1996), and cirrus crystals from Heymsfield et al. (2002) and from Erfani and Mitchell (2016) assuming ice crystal properties at -55 to -65°C (coldest range fitted) and -20 to -40°C (warmest, see text).

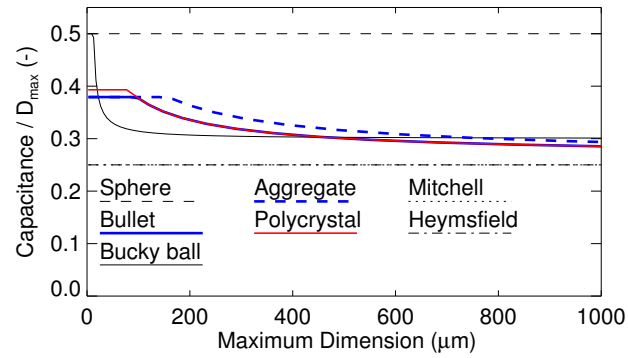


Figure 15. Ice crystal capacitance normalized by maximum dimension at 350 mb and 233 K for derived ice properties as in Fig. 14. [In the absence of specified \$\alpha_c\$ for some or all crystal sizes, a constant value is taken for Mitchell et al. \(1996\) and Heymsfield et al. \(2002\) ice properties \(see text\).](#)

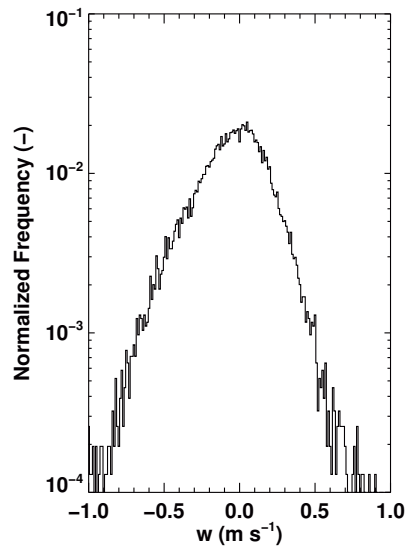


Figure 16. Vertical wind speeds retrieved from 19:06 UTC on 1 April to 2:23 UTC on 2 April at elevations of 6.1–12.0 km, from a sample size of 123,469 retrievals obtained in 47 layers at 10-s resolution.

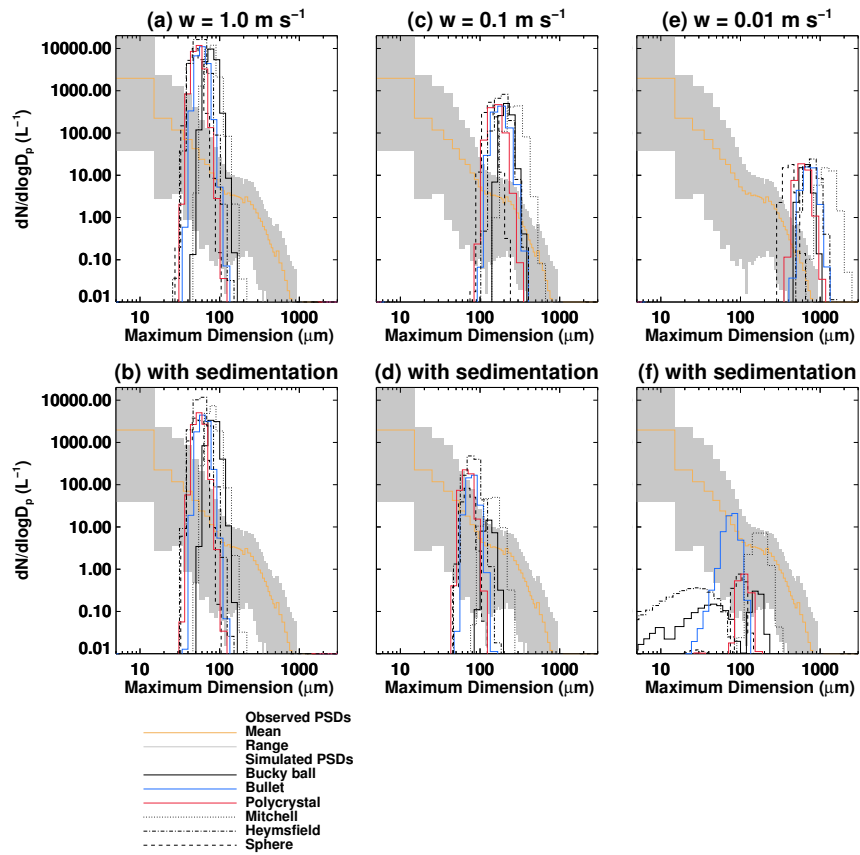


Figure 17. Ice particle size distributions (PSDs) simulated at -55°C with different ice properties (see legend) and different updraft speeds without sedimentation (top row) and with sedimentation (bottom row). For context are shown also the mean and range of all PSDs observed over the 1–2 April flights using an in-cloud ice water content threshold of 0.001 g m^{-3} following Jackson et al. (2015).

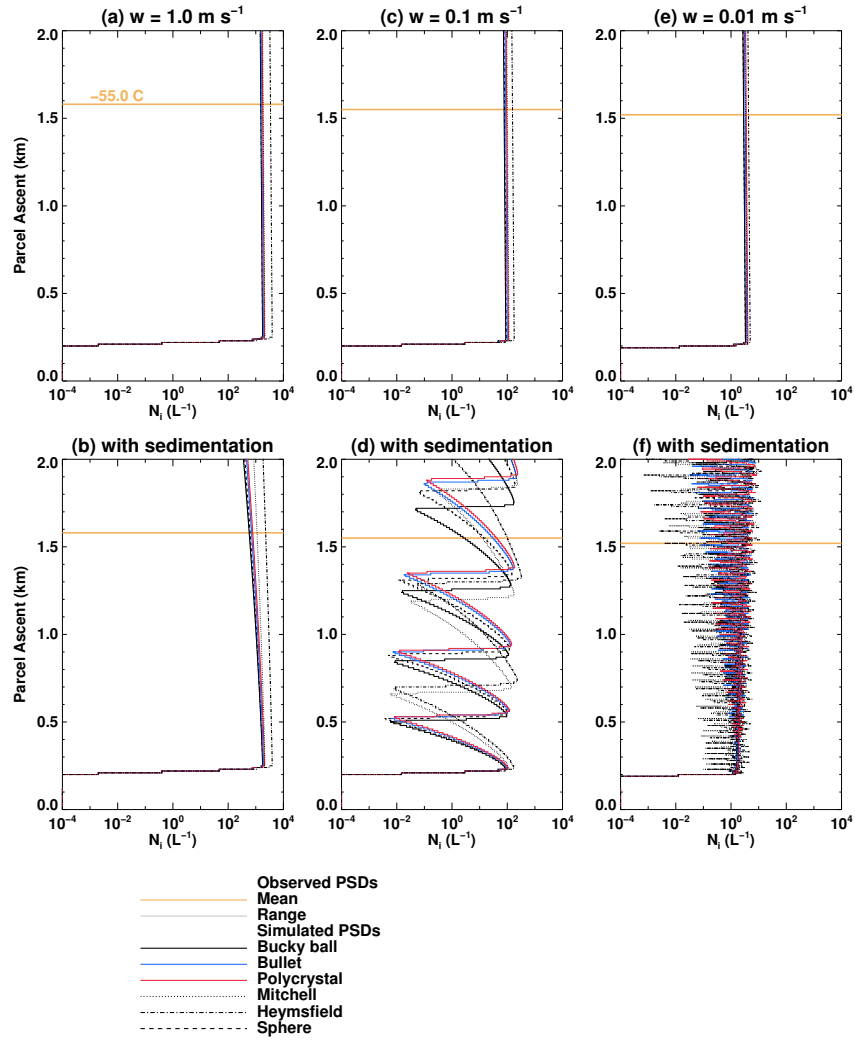


Figure 18. Simulated ice crystal number concentration as a function of parcel distance from initiation at -40°C , with ice properties as in Fig. 14 (see legend) and updraft speeds of 1, 0.1, and 0.01 m s^{-1} without sedimentation (top row) and with sedimentation (bottom row). Parcel level corresponding to -55°C corresponding to size distributions in Fig. 17 is shown as dotted yellow line.

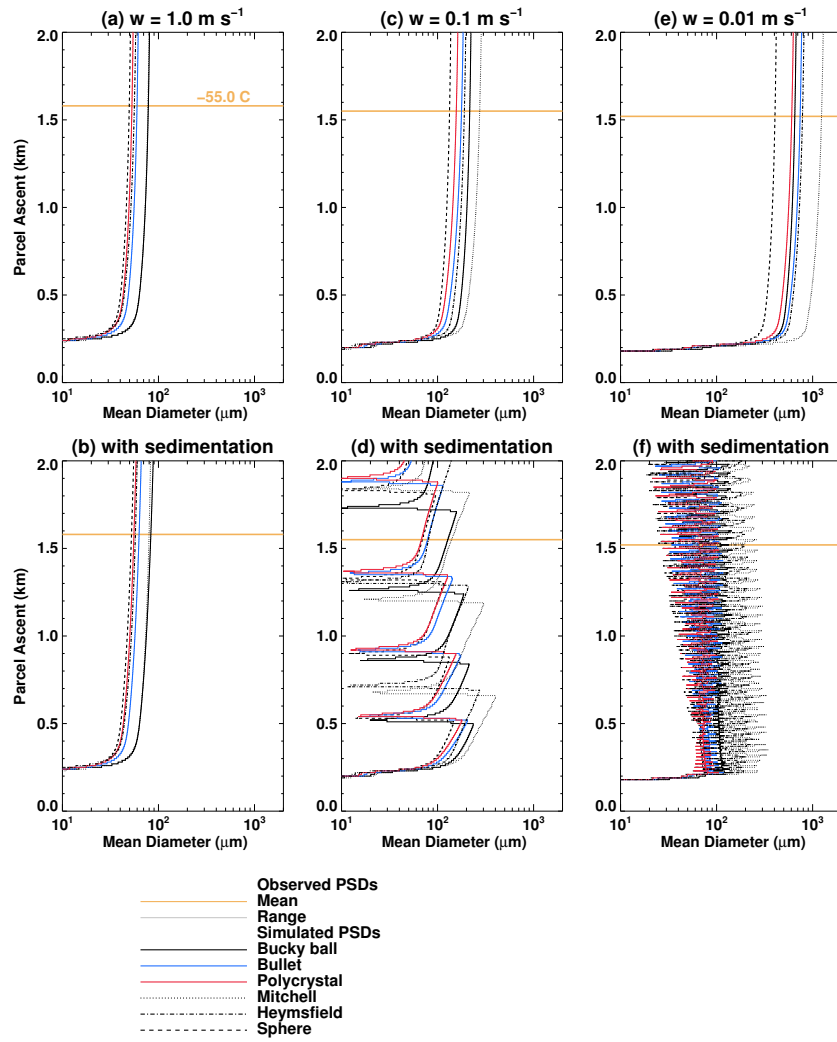


Figure 19. As in Fig. 18 except number-weighted mean ice crystal diameter.

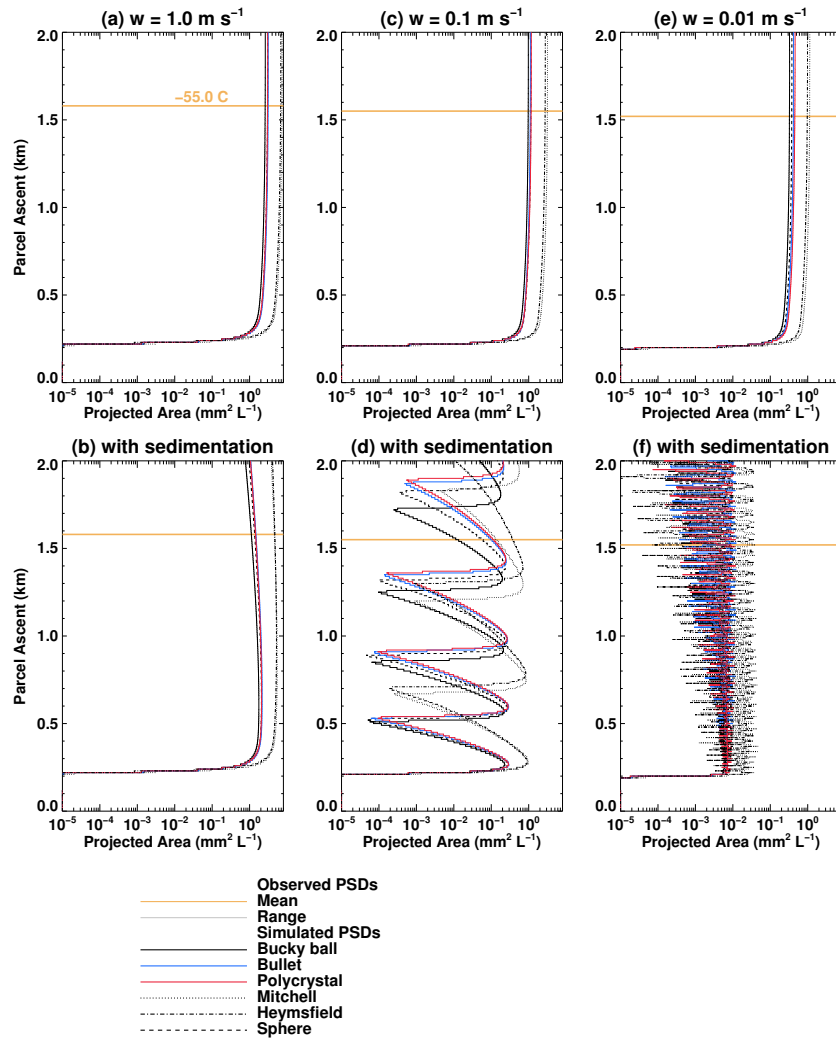


Figure 20. As in Fig. 18 except total ice crystal projected area.

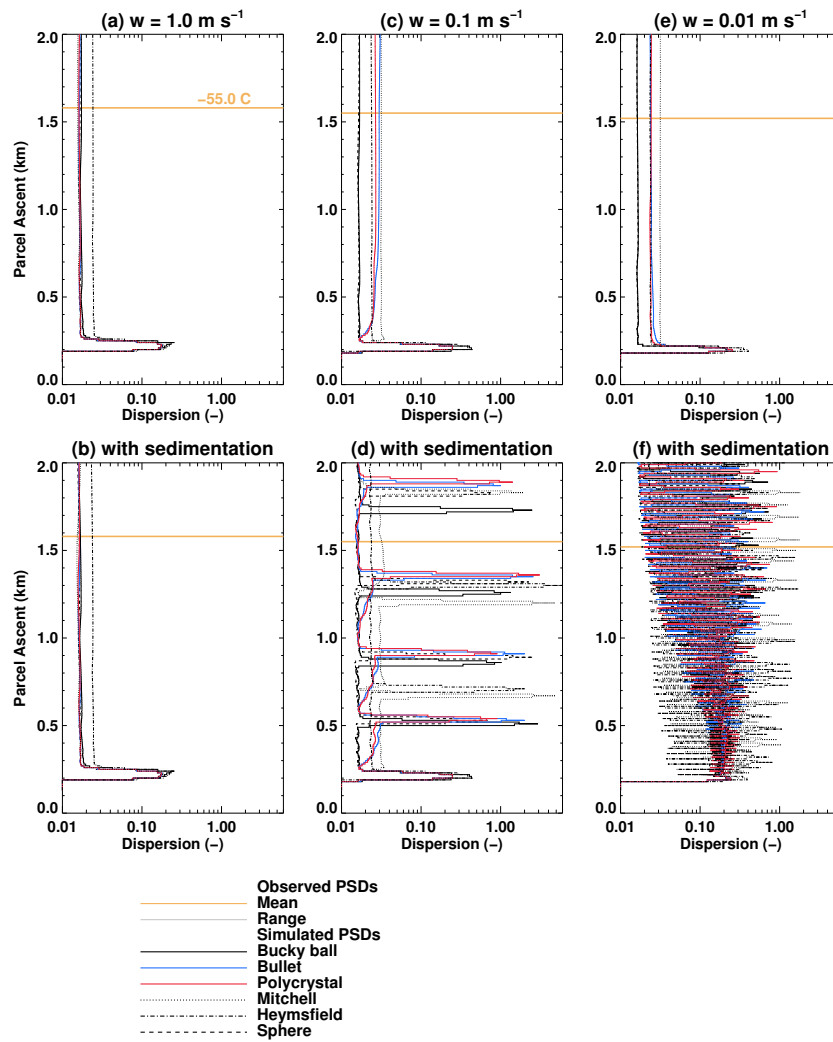


Figure 21. As in Fig. 18 except relative dispersion of the ice crystal size distribution.

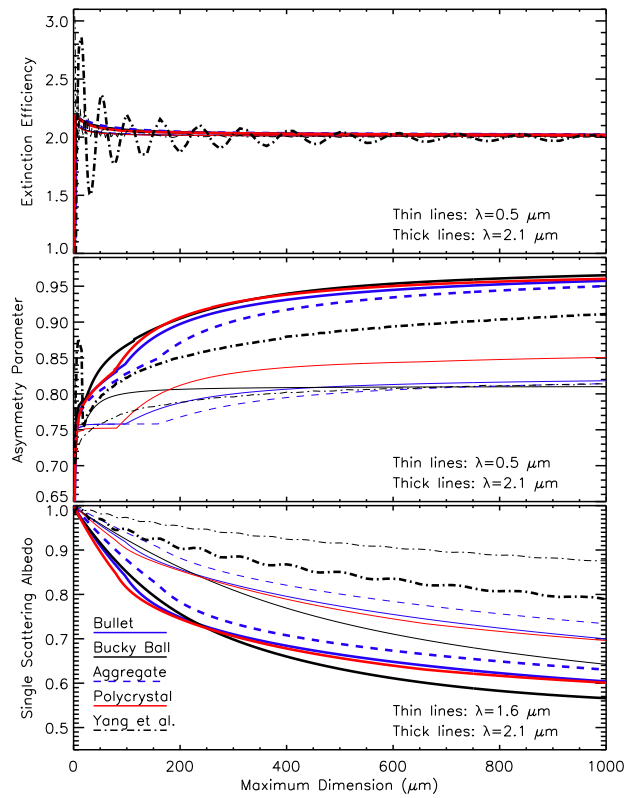


Figure 22. Ice single-crystal optical properties as a function of maximum dimension for the bullet, Bucky ball, aggregate, and polycrystal models, and for Yang et al. (2013) bullet rosettes (see line types in legend) at scattering and absorbing wavelengths (thin and thick lines in panels a and b) or two absorbing wavelengths (thin and thick lines in panel c).

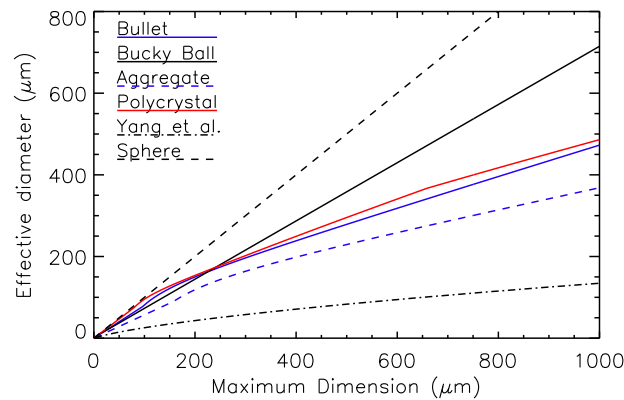


Figure 23. Ice single-crystal effective diameter as a function of maximum dimension for the bullet, Bucky ball, aggregate, and polycrystal models, Yang et al. (2013) bullet rosettes, and spheres (see legend).

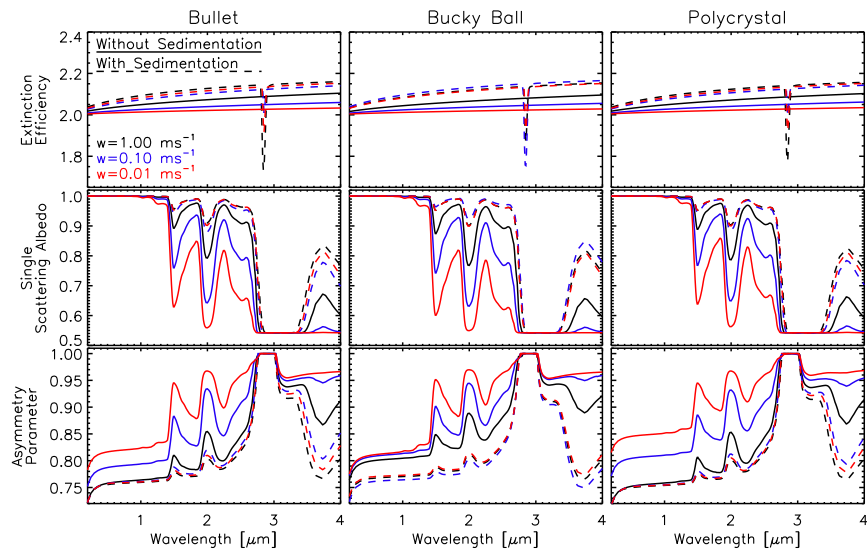


Figure 24. Optical properties of ice crystal size distributions shown in Fig. 17, as simulated at -55°C with varying ice properties (bullet, Bucky ball and polycrystal: left to right) and varying updraft speeds (line colors per legend), with and without sedimentation (solid and dashed lines per legend).