Effective Radius of Ice Particles in Cirrus and Contrails

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(Manuscript received 25 May 2010, in final form 8 September 2010)

ABSTRACT

This paper discusses the ratio $C$ between the volume mean radius and the effective radius of ice particles in cirrus and contrails. The volume mean radius is proportional to the third root of the ratio between ice water content and number of ice particles, and the effective radius measures the ratio between ice particle volume and projected cross-sectional area. For given ice water content and number concentration of ice particles, the optical depth scales linearly with $C$. Hence, $C$ is an important input parameter for radiative forcing estimates. The ratio $C$ in general depends strongly on the particle size distribution (PSD) and on the particle habits. For constant habits, $C$ can be factored into a PSD and a habit factor. The PSD factor is generally less than one, while the habit factor is larger than one for convex or concave ice particles with random orientation. The value of $C$ may get very small for power-law PSDs with exponent $n$ between $-4$ and $0$, which is often observed. For such PSDs, most of the particle volume is controlled by a few large particles, while most of the cross-sectional area is controlled by the many small particles. A new particle habit mix for contrail cirrus including small droxtal-shape particles is suggested. For measured cirrus and contrails, the dependence of $C$ on volume mean particle radius, ambient humidity, and contrail age is determined. For cirrus, $C$ varies typically between 0.4 and 1.1. In contrails, $C = 0.7 \pm 0.3$, with uncertainty ranges increasing with the volume radius and contrail age. For the small particles in young contrails, the extinction efficiency in the solar range deviates considerably from the geometric optics limit.

1. Introduction

This study addresses the relationship between the optical effective radius $r_{\text{eff}}$ and the volume mean radius $r_{\text{vol}}$ of ice particles in upper tropospheric ice clouds (cirrus) and aircraft condensation trails (contrails). The effective particle radius is defined such that the extinction coefficient (optical depth) is proportional to the ice water content (IWC) [ice water path (IWP)] divided by the effective radius (Hansen and Travis 1974; Garrett et al. 2003). While the volume mean radius can be computed for given IWC, ice bulk density, and number of ice particles, the effective radius depends on details of the particle habits and the particle size distribution (PSD) (McFarquhar and Heymsfield 1998), which currently cannot be predicted from first principles (Bailey and Hallett 2009). The radius relationship is of importance for models that study the influence of the number concentration of ice particles and the IWC on the optical properties of cirrus (e.g., Lohmann et al. 2008; Liu et al. 2009; Penner et al. 2009; Spichtinger and Gierens 2009). It is also important for sedimentation because of volume-dependent weight and area-dependent drag. Since many climate effects, such as radiative forcing, scale about linearly with the optical depth, it is important to know the ratio

$$C = \frac{r_{\text{vol}}}{r_{\text{eff}}}$$

between the volume specific and the effective radius (for definitions see section 2) and possible parameterizations.
This study has been performed in the course of development of a contrail cirrus prediction tool (CoCiP) (Schumann 2009). CoCiP predicts the IWC and the number of ice particles and the area covered by contrail cirrus. The radiative forcing by the contrail cirrus is parameterized as a function of the effective particle radius and the optical depth (Schumann et al. 2009). For this, we need to know the factor $C$. Knowledge about $C$ for liquid clouds (droplets) has been derived from PSD statistics and measurements (Pontikis and Hicks 1992; Martin et al. 1994; Korolev et al. 1999). For example, Martin et al. (1994) suggested $C = 0.67$ for continental and 0.9 for maritime warm stratocumulus clouds. For ice clouds, some $C$ parameterizations have been introduced for two-moment cirrus models with little theoretical or experimental support so far (Lohmann 2002; Liu et al. 2007; Ström and Gierens 2002).

The concept of the effective radius was introduced first for liquid (spherical) cloud particles by Hansen (1971) as the ratio of the third moment of the PSD to the second moment of the PSD. He used the fact that a cloud particle scatters an amount of light approximately in proportion to its area (van de Hulst 1957). Hansen and Travis (1974) discussed this radius in comparison to alternative definitions. Stephens (1978) parameterized the optical thickness of liquid clouds as the ratio between the liquid water path and the effective radius. These studies noted that the effective radius concept is valid only for short wavelength $\lambda$ or large particles (i.e., for size parameters $x = 2\pi r_{\text{eff}}/\lambda \gg 1$), for which the extinction efficiency $Q_{\text{ext}}$ is close to 2, the geometric optics limit.

Cirrus cloud ice particles are composed of various types of nonspherical crystals (Weickmann 1945; Heymsfield and Iaquinta 2000; Bailey and Hallett 2009). For modeling, one often assumes prescribed particle habits (Baum et al. 2005a; Baran 2009). Recent laboratory data reveal that most ice particles at low temperatures ($-40^\circ$ to $-70^\circ$C) have columnar shapes (with variable aspect ratio length/diameter) and that the very small crystals growing at low ice supersaturation are compact faceted polycrystals (Bailey and Hallett 2009). Often ice particles are hollow (Weickmann 1945; Schmitt and Heymsfield 2007). Ice particles may result from homogeneous freezing of initially liquid (water or solution) droplets, which then grow by deposition of water vapor. Ice particles also originate from heterogeneous freezing on preexisting ice nuclei. The different freezing processes may impact the particle habits. Once entering a subsaturated environment, ice particles will sublime and may become more spherical again (Nelson 1998). Also, particle orientation may play a role for cloud optical properties, in particular for large ice particles (Reichardt et al. 2008; Westbrook et al. 2010).

Therefore, the concept of effective radius has been generalized to account for the nonspherical shapes (Foot 1988). Most studies now define the effective radius as proportional to the ratio between mean particle volume and mean particle projected cross-sectional area (mean shadow area) (Foot 1988; Francis et al. 1994; Fu 1996; McFarquhar and Heymsfield 1998; Wyser 1998; Hong et al. 2009). Some earlier alternative definitions (e.g., Ebert and Curry 1992) are still in use (e.g., Fusina et al. 2007).

Parameterizations of the bulk radiative properties of ice clouds for solar and terrestrial radiation are beyond the scope of this paper, but we note that they often use the concept of effective radius, both for applications in climate models and remote sensing (e.g., Ebert and Curry 1992; Fu 1996; Fu et al. 1998; Stubenrauch et al. 2004; Hong et al. 2009; Minnis et al. 1998, 2010, submitted to IEEE Trans. Geosci. Remote Sens., hereafter MIN). Various papers have discussed the usefulness and limitations of this concept (Fu 1996; Wyser 1998; Mitchell 2002; McFarquhar et al. 2003; Baran and Havemann 2004; Baran 2005; Yang et al. 2005; De Leon and Haigh 2007; Edwards et al. 2007). In many studies the optical properties are modeled as a function of ice water content, temperature, and the effective radius. Such one-moment models are independent of the volume mean radius and hence not a function of $C$. In principle, the effective radius can be avoided and the optical and physical properties of the clouds can be parameterized directly as a function of ice water content and temperature if crystal sizes and habits are known or can be suitably prescribed (Mitchell et al. 2008; Baran et al. 2009). In fact, many global circulation models use a prescribed effective radius, which is either given as a constant or as a function of ice water content and temperature (Liou et al. 2008). For large size parameters, the parameterizations depend also on the details of the assumed size and habit distributions. Early parameterizations were based on size distributions derived from a few field experiments. For example, Yang et al. (2000) used 30 particle size distributions as collected by Fu (1996) and Mitchell and Arnott (1994). Minnis et al. (1998) used about 10 size distributions partly from the same measurements. More recent studies (Hong et al. 2009) added further measurement cases.

Measurements of the number, area, and volume size distributions of cirrus particles as a function of particle sizes are difficult and prone to uncertainties for many reasons. Ice particles occur at sizes of about 1–8000 $\mu$m (Dowling and Radke 1990); in this paper we mainly consider the lower part of this size range. Many studies detect and size small ice particles by forward scattering particle counters (Knollenberg 1972; Baumgardner et al. 2001). Such forward scattering spectrometer probe (FSSP)
instruments measure the light scattered by the particles. The signal depends on many parameters including the size and shape of the particles (Baumgardner et al. 1992). To invert the measured signals into actual particle sizes (and areas and volumes), Mie calculations or T-matrix calculations for spheres and spheroids have been used (Borrmann et al. 2000). More recent studies use the FSSP-300 for small particles, which allows identifying ice particles larger than about 0.4 μm. Early instruments underestimated the number of small ice particles. However, cirrus may contain significant concentrations of ice particles smaller than 30 μm (Baumgardner et al. 1992; Noone et al. 1993; Gayet et al. 2002; Febvre et al. 2009; Hong et al. 2009). This is the case in spite of the fact that shattering of large crystals on the probe inlet may cause an overestimate of the concentration of small ice crystals in cirrus that also contain many large ice particles (Field et al. 2003; Korolev and Isaac 2005; McFarquhar et al. 2007; Heymsfield 2007; Westbrook and Illingworth 2009). Two-dimensional optical probes are used to directly depict the shadow area of particles larger than about 25 μm (Baumgardner et al. 2001; Lawson et al. 2001). New instrument versions now allow detection of shattering by measuring the interarrival times of particles (Field et al. 2003, 2006). The particle volume for given particle sizes is estimated using empirical relationships (e.g., Baker and Lawson 2006). As an alternative to measuring the cross-sectional area of particles, one may measure the extinction coefficient β, which is directly related to the cross section of the ensemble of particles for constant extinction efficiency (Garrett et al. 2003). The polar nephelometer (Gayet et al. 1997) measures the scattering phase function of an ensemble of cloud particles for particle diameters below about 800-μm diameter. From the scattering phase function one can evaluate, among other properties, the optical extinction coefficient (Gayet et al. 2002, 2004).

Particles in contrails differ from cirrus in that they are smaller and more numerous (Schumann 1996; Gayet et al. 1996a; Baumgardner and Gandrud 1998; Sassen 1997), at least for some time. Atmospheric observations in young contrails (ages between seconds and 0.5 h) with impactors (first by Weickmann 1945) and particle size measurements (Knollenberg 1972) show ice particles composed of nearly spherical ice crystals with mean diameters in the range of 1–10 μm and narrow size distributions (Petzold et al. 1997; Poellot et al. 1999; Schröder et al. 2000), at least for cold ambient conditions (below −50°C). Contrail particles are small inside the contrail and may get larger by uptake of humidity from ambient air at the outer contrail edge (Petzold et al. 1997; Heymsfield et al. 1998; Lawson et al. 1998). The smallness of the particles causes more variability of the extinction and absorption efficiencies (Poellot et al. 1999). Febvre et al. (2009) observed optical properties of young and slightly aged (15 min) contrails and ambient cirrus. The measurements confirmed that quasispherical ice particles with diameters smaller than 5 μm control the optical properties of the plume shortly after formation. In the slightly aged contrails the optical properties are governed by larger nonspherical ice crystals. The particles with sizes smaller than 10 μm contribute about 80% to the total extinction and therefore dominate the optical properties. The shape of most of the observed ice particles of sizes smaller than 10 μm in the aged contrail strongly deviates from spherical. In aged contrail cirrus (age of 35–90 min), in particular in highly supersaturated air masses, more complex ice habits have been observed (Gayet et al. 1996a; Baumgardner and Gandrud 1998; Heymsfield et al. 1998; Lawson et al. 1998; Baumgardner et al. 2005). Heymsfield et al. (1998) and Lawson et al. (1998) find many small ice particles (diameters of 1–20 μm) but also a few large ice particles. Some of the latter could originate from mixing with ambient cirrus. Fall streaks of large ice crystals have been observed in more than 2-h aged contrail cirrus at temperatures above −40°C (Atlas et al. 2006).

Numerical simulations with the contrail–cirrus model described in Unterstrasser and Gierens (2010) show that the mean diameter evaluated over the total contrail area increases from 5 to 15–40 μm within the first 2–3 h for a large variety of ambient conditions. Temperature has the largest impact on the crystal sizes for two reasons: the water vapor concentrations increase with temperature and more ice crystals are lost during the vortex phase in a warmer environment (Unterstrasser et al. 2008). The ambient relative humidity affects the crystal sizes to a much lesser extent, since for increasing relative humidity the two effects of increasing water vapor concentrations and of a larger number of ice crystals surviving the vortex phase nearly cancel out each other.

Often contrails show higher lidar depolarization than cirrus, indicating possibly large deviations from sphericity of the ice particles, with details depending on particle sizes, contrail age, and temperature (Freudenthaler et al. 1996; Mishchenko and Sassen 1998; Sassen and Hsueh 1998; Del Guasta and Niranjan 2001; Langford et al. 2005). Some papers list measured values of the volume mean and effective radius or diameter of cirrus and contrails (e.g., Gayet et al. 1996a,b; Schröder et al. 2000), but without discussing the importance of the ratio between the two particle scales.

So far, little is known about the transition of the habit mix from young contrails to natural cirrus. Contrail ice particles originating from freezing droplets first take a droxtal shape (Thuman and Robinson 1954; Ohtake...
1970; Roth and Frohn 1998) and later may become more aspherical according to laboratory measurements (Gonda and Yamazaki 1984). The optical properties of droxtals are systematically different from those of spheres, in particular in the solar range, with less forward scattering (Yang et al. 2003; Zhang et al. 2004; Wendisch et al. 2007). After some time, contrail particles become more similar to cirrus particles. From the abovementioned observations in aged contrail cirrus, Liou et al. (1998) derived an ice crystal shape model for contrails consisting of bullet rosettes, hollow columns, and plates—hence, without near-spherical particles.

In this paper we will show that $C$ may be larger or smaller than one and in general depends strongly on the PSD and on the particle habit. In particular, we identify upper and lower bounds for $C$ for special cases. The value of $C$ is deduced from measured size distributions with parameterized volume and projected area values for ice particles of given habit mixes. For contrail cirrus, a habit mix is suggested that reflects the few existing observations. Moreover, we collect measured data on number density, ice water content, and projected area or extinction coefficient from experiments in cirrus and contrails to derive a set of $C$ values. This includes published measurements as well as new data from the recent Contraill and Cirrus Experiment (CONCERT; Voigt et al. 2010). Based on these data, the dependence of $C$ on cirrus particle habits, volume mean radius, ambient humidity, and the age of contrails is discussed. For contrails and cirrus, a simple parameterization of the factor $C$ suitable for two-moment models is suggested. For uncertainty estimates, approximate upper and lower bounds are provided.

2. Definitions

We consider cirrus clouds containing a large number of different ice particles with random (uniformly distributed) orientations, with different sizes and different habits (see, e.g., Fig. 1). We assume that we know the PSD for this ensemble of particles (see Fig. 2). The PSD defines the number of ice particles $n_p(D)$ per unit volume of air in the size interval $D$ to $D + dD$ as a function of a measurable particle scale $D$. Here $D$ may be the maximum dimension $d_{\text{max}}$, or the equivalent spherical diameter $d$, or the radius $r = d/2$. In addition we need to know $V_p(D)$ and $A_p(D)$, that is, the mean particle volume and mean particle projected cross-sectional area in the cirrus as a function of particle size. Then, the volume density $V$, projected area density $A$, and number density $N$ on average over all particles and all orientations per unit volume are defined by

$$V = \int V_p(D)n_p(D)\,dD,$$

$$A = \int A_p(D)n_p(D)\,dD, \quad \text{and}$$

$$N = \int n_p(D)\,dD.$$

The effective radius $r_{\text{eff}}$ and the area and volume mean radii $r_{\text{area}}$ and $r_{\text{vol}}$ are commonly defined by

$$r_{\text{eff}} = (3V)/(4A),$$

$$r_{\text{vol}} = [3V/(4\pi N)]^{1/3}, \quad \text{and}$$

$$r_{\text{area}} = [A/(\pi N)]^{1/2}.$$

We note that these definitions apply to both spherical and nonspherical particles. The factor $3/4$ in Eq. (5) is introduced so that

$$r_{\text{eff}} = \frac{\int r^3 n_p(r)\,dr}{\int r^2 n_p(r)\,dr}$$

for spherical particles. The factors with $\pi$ in Eqs. (6) and (7) are introduced so that $r_{\text{vol}} = r_{\text{area}} = r_{\text{eff}}$ for spheres in a monodisperse size distribution with $n_p(r) = \delta(r, r_{\text{eff}})N$.

The definitions in Eqs. (1) and (5)–(7) imply a useful relationship between the three radius definitions:

$$C = r_{\text{vol}}/r_{\text{eff}} = r_{\text{area}}^2/r_{\text{vol}}^2.$$

For $C \leq 1$, this implies
r \text{ area} \leq r \text{ vol} \leq r \text{ eff}, \quad (10)

but the opposite is true for $C \geq 1$, and both variants can be found in the literature (Korolev et al. 1999; Yang et al. 2000). The definitions also imply

$$C = \left( \frac{16}{9\pi} \right)^{1/3} \frac{A}{N^{1/3} V^{2/3}} = \left( \frac{16}{9\pi} \right)^{1/3} \frac{(A/N)}{(V/N)^{2/3}}. \quad (11)$$

Hence, $C$ increases linearly with the cross-sectional area and decreases more weakly with volume and number concentration of particles. The second version is useful for a single ice particle ($N = 1$).

For constant extinction efficiency $Q_{\text{ext}}$, the extinction coefficient is

$$\beta = \int Q_{\text{ext}} A_p(D) n_p(D) \, dD = Q_{\text{ext}} A. \quad (12)$$

The exact value of $Q_{\text{ext}}$ is size and wavelength dependent and oscillates strongly around 2 for size parameters $x < 30$ (van de Hulst 1957). Small size parameters may apply for contrail ice particles. Any broadband mean value of $Q_{\text{ext}}$ depends on the spectrum of radiation (Baum et al. 2005b). Broadband mean values of $Q_{\text{ext}}$ are of course less variable than narrowband values. The effective extinction efficiency $\langle Q_{\text{ext}} \rangle$ is the average over the area distribution (and the spectral band):

$$\langle Q_{\text{ext}} \rangle = \frac{\int Q_{\text{ext}}(D) A_p(D) n_p(D) \, dD}{A_p(D) n_p(D) \, dD}. \quad (13)$$

For illustration, Fig. 3 shows $\langle Q_{\text{ext}} \rangle$ in the visible range. The data represent Mie scattering by spherical ice particles (Wiscombe 1980; Warren 1984), weighted with the solar Planck function, for cirrus PSDs as in Key et al. (2002) and for contrail PSDs with smaller mean radius (Voigt et al. 2010). For small spheroidal particles, similar values result from the T-matrix method (Mishchenko and Travis 1994). The parameterization of Hong et al. (2009) is shown as derived for this spectral range, as an average over various PSDs, for the habit mix of Baum et al. (2005a), with small particles represented by droxtals. Also plotted is an often used simple algebraic approximation of the Mie theory derived for refraction index near 1 by van de Hulst (1957), slightly underestimating $Q_{\text{ext}}$ for ice at small $x$. For $r_{\text{eff}} > 1 \mu m$, $\langle Q_{\text{ext}} \rangle$ in the solar range varies between 1.9 and 2.5.

With the above definitions, the IWC, IWP, and the optical depth follow from vertical integrals over the cloud layer depth $H$:

$$\text{IWC} = \int \rho_{\text{ice}} V_p(D) n_p(D) \, dD = \rho_{\text{ice}} V, \quad (14)$$

$$\text{IWP} = \int_0^H \text{IWC}(z) \, dz, \quad \text{and} \quad \tau = \int_0^H \beta(z) \, dz \quad (15)$$

with bulk ice density $\rho_{\text{ice}}$ (typically $\rho_{\text{ice}} = 917$ kg m$^{-3}$ for ice without hollows). The optical depth $\tau$ could be
computed from any of the following three equivalent expressions:

\[
\tau = \langle Q_{\text{ext}} \rangle A H = \frac{\langle Q_{\text{ext}} \rangle A \text{IWP}}{\rho_{\text{ice}} V} = \frac{3 \langle Q_{\text{ext}} \rangle \text{IWP}}{4 \rho_{\text{ice}} r_{\text{eff}}}. \quad (16)
\]

Hence, for \( \langle Q_{\text{ext}} \rangle \) = constant, and given IWP, \( \tau \) is determined by \( r_{\text{eff}} \) regardless of the details of the particle size and habit distributions. For small particles, we need to know \( \rho_{\text{ice}} \) in addition.

For given IWC and \( N \), it is easy to compute the volume mean radius:

\[
r_{\text{vol}} = \left[ 3 \text{IWC}/(4\pi \rho_{\text{ice}} N) \right]^{1/3}. \quad (17)
\]

However, in order to derive \( r_{\text{eff}} \) from IWC and \( N \), we need to know \( C = r_{\text{vol}}/r_{\text{eff}} \). In fact, \( C \) enters the relationship linearly:

\[
\tau = C \frac{3 \langle Q_{\text{ext}} \rangle \text{IWP}}{4 \rho_{\text{ice}} r_{\text{vol}}}. \quad (18)
\]

This shows the importance of \( C \): any uncertainty in \( C \) has direct impact on the accuracy of the computed optical depth.

Parameterizations that relate \( \beta = \tau/\text{IWP} \) directly to the IWC and \( r_{\text{eff}} \) (Hong et al. 2009) leave the relation between \( r_{\text{eff}} \) and IWC, and hence \( N \) of ice particles, open.

### 3. Split into habit and size distribution factors

In this section we will split the factor \( C \) into two factors that are independent of each other, at least for an ensemble of particles with size-independent habits. This splitting will help us to understand the dependence of the value of \( C \) on the PSD and particle habits.

We introduce the volume equivalent radius

\[
r_{\text{vol},p} = \left[ 3 V_p/(4\pi) \right]^{1/3} \quad (19)
\]

and the equivalent projected spherical area

\[
A_{s,p} = \pi r_{\text{vol},p}^2 \quad (20)
\]

for individual particles with given particle volume \( V_p \). For sake of a simpler notation we define \( r = r_{\text{vol},p} \).

Without loss of generality,

\[
r_{\text{eff}} = \frac{\int r^3 n_p(r) \, dr}{\int C_{\text{habit}}(r) r^2 n_p(r) \, dr} \quad (21)
\]

follows as identity from Eqs. (2), (3), and (5). Here, \( C_{\text{habit}}(r) \) is the ratio of the projected area of the particles of size \( r \) relative to the projected area of a sphere of equal volume:

\[
C_{\text{habit}}(r) = A_p(r)/A_{s,p}(r). \quad (22)
\]

For constant (size independent) particle habits,

\[
C_{\text{habit}} = \frac{A_p}{A_{s,p}} = \frac{A_p}{(3\pi/4)^{2/3}} \quad (23)
\]

is constant; otherwise, we consider the weighted mean value

\[
\overline{C}_{\text{habit}} = \frac{\int C_{\text{habit}}(r) r^2 n_p(r) \, dr}{\int r^2 n_p(r) \, dr}. \quad (24)
\]

For the given \( r \) and arbitrary particle habit, Eq. (6) implies

\[
r_{\text{vol}}^3 = \frac{\int r^3 n_p(r) \, dr}{\int n_p(r) \, dr}. \quad (25)
\]

With the \( k \)th moment of the particle size distribution \( n_p(r) \), defined as

\[
M_k = \int_0^\infty r^k n_p(r) \, dr, \quad (26)
\]

this can be written as

\[
r_{\text{vol}}^3 = M_3/M_0, \quad (27)
\]

and

\[
r_{\text{eff}} = M_3/(M_2 \overline{C}_{\text{habit}}). \quad (28)
\]

Hence, \( C = r_{\text{vol}}/r_{\text{eff}} \) can be factored into

\[
C = \overline{C}_{\text{habit}} C_{\text{PSD}}, \quad (29)
\]

with

\[
C_{\text{PSD}} = (M_3/M_0)^{1/3} (M_2/M_3)^{2/3} = N_2/N_3^{2/3}. \quad (30)
\]

Here

\[
N_k = M_k/M_0 \quad (31)
\]

is the normalized \( k \)th moment of the PSD.
For constant particle habits and aspect ratio, the two factors are independent; $C_{PSD}$ accounts for the PSD, while $C_{habit}$ or $C_{habit}^{r}$ accounts for the particle habits. In this form, we next can discuss the magnitude of these factors separately.

4. The particle size distribution factor $C_{PSD}$

In this section we discuss the particle size distribution factor $C_{PSD}$. We first show that $0 < C_{PSD} \leq 1$ in general and then discuss the influence of the PSD on the value of $C$.

a. Lyapunov limit for $C_{PSD}$ for constant habits

For constant habits, see Eq. (30):

$$C_{PSD} = N_{2}^{2/3}.$$  (32)

This ratio is obviously positive for any nonzero PSD. Moreover, it is smaller than or equal to one for any PSD; this can be shown as follows: After taking the square root of $C_{PSD}$, we have

$$C_{PSD}^{1/2} = N_{2}^{1/2}\langle N_{3}^{1/3}. \quad (33)$$

Hence, the Lyapunov inequality (Ljapunoff 1900; the name is sometimes romanized as Ljapunov or Ljapunoff), as proven in Fisz (1970),

$$N_{n}^{1/n} \leq N_{m}^{1/m} \quad \text{for} \quad n < m,$$  (34)

implies

$$0 < C_{PSD} \leq 1.$$  (35)

b. $C_{PSD}$ for a lognormal distribution

Here we discuss the sensitivity of $C_{PSD}$ to the shape of the PSD $n_{p}(r)$ for the first of three model distributions as plotted in Fig. 4, for which we can compute $C_{PSD}$ analytically.

Obviously, $C_{PSD} = 1$ for a monodisperse PSD but is smaller than one otherwise. Measured size distributions $n_{p}(r)$ often exhibit a lognormal distribution (Hansen and Travis 1974; Spichtinger and Gierens 2009; Tian et al. 2010):

$$n_{p}(r) = \frac{N}{\sqrt{2\pi \ln(\sigma_{r}/r_{m})}} \exp\left\{-\frac{1}{2} \left[ \ln\left(\frac{r}{r_{m}}\right) \right]^{2}\right\}, \quad (36)$$

where $r_{m}$ is near the modal radius [for which $n_{p}(r)$ takes its maximum; the actual modal radius is smaller because of the factor $1/r$ in the premultiplier], and $\sigma_{r}$ is the spectral dispersion (i.e., the standard deviation of the logarithm of the volumetric particle radius $r$) (Hansen and Travis 1974). The lognormal distribution is completely specified once its zeroth, first, and second moment are given. Here we have input for the zeroth and third moment ($N$ and IWC), leaving at least one free parameter. Some modelers prescribe the standard deviation $\sigma_{r}/r_{m}$ as a constant value, typically between 1.2 and 2.1 (Spichtinger and Gierens 2009). This means that the spectral dispersion is proportional to the mean radius. As we will see, this implies a specific value of the ratio of $C_{PSD}$.

The order-$k$ moments of the size distribution are

$$M_{k} = \int_{0}^{\infty} r^{k} n_{p}(r) \, dr = N r_{m}^{k} \exp\left\{\frac{1}{2} k^{2} \left[ \ln\left(\frac{r_{m}/r}{1/r_{m}}\right) \right]^{2}\right\}. \quad (37)$$

Hence, the zeroth, second, and third moments are

$$M_{0} = N,$$  (38)

$$M_{2} = N r_{m}^{2} \exp\left\{\frac{1}{2} \left[ \ln(\sigma_{r}/r_{m}) \right]^{2}\right\}, \quad \text{and} \quad (39)$$

$$M_{3} = N r_{m}^{3} \exp\left\{\frac{1}{2} \left[ \ln(\sigma_{r}/r_{m}) \right]^{2}\right\}. \quad (40)$$

As a consequence,

$$C_{PSD} = \exp\{-[\ln(\sigma_{r}/r_{m})]^{2}\}. \quad (41)$$

Obviously, $C$ satisfies Eq. (35) and is constant for fixed dispersion (e.g., for $\sigma_{r}/r_{m} = 2$, $C_{PSD} = 0.615$). For other values, see Fig. 5.

In agreement with Pontikis and Hicks (1992), $C_{PSD}$ is directly related to the dispersion or the variance of the size distribution for the lognormal distribution. But this
is not the case for other PSDs in general. Hansen and Travis (1974) and Mishchenko and Travis (1994) discuss various variance definitions, which partly scale with the fourth moment of the PSD and hence cannot generally be related to $C_{PSD}$, which depends on the second and third moment of the PSD only. For general PSDs, Martin et al. (1994) find that $C_{PSD}$ depends on the skewness of the size distribution; $C_{PSD}$ gets smaller for large positive skewness (long tail at large radius). However, Martin’s result $C_{PSD} > 1$ for large negative skewness contradicts the Lyapunov limit.

c. $C_{PSD}$ for a power-law distribution

Often, measured size distributions follow a power law (e.g., see Fig. 2). As we will see, knowledge of the steepness of the PSD allows for rough estimate of $C$. Therefore, we compute the PSD factor assuming that $n_p(r)$ follows a power law in a finite interval $[R_1, R_2]$ and is zero outside, as indicated by the long-dashed curve in Fig. 4:

$$n_p(r) = n_0 (r/R_1)^n, \quad R_1 \leq r \leq R_2, \quad n < 1.$$  (42)

Unfortunately, there are no general physical constraints on the value of $n$. However, this model serves for understanding of the large range of $C_{PSD}$ values that we find in observations (see below).

The moments $M_k$ of these distributions can be evaluated by analytical integration:

$$M_k = \frac{n_0}{n+k+1} (R^{n+k+1} - 1), \quad \text{for } n+k+1 \neq 0,$$

$$M_k = n_0 \ln R, \quad \text{for } n+k+1 = 0,$$  (43)

$$M_{-1} = -\frac{n_0}{r_m} \ln \frac{R_2}{R_1}, \quad \text{for } n+k+1 = 0.$$  (44)

Figure 5. $C_{PSD}$ ($r_{vol}/r_{eff}$ for spheres) as a function of the spectral dispersion $\sigma_r$ or standard deviation of the volumetric particle radius for a lognormal size distribution with $r_m$.

with $R = R_2/R_1$. With these moments, $C_{PSD}$ follows from Eq. (30):

$$C_{PSD} = \frac{1 + n R^{3+n} - 1}{3 + n R^{3+n} - 1} \left(\frac{4 + n R^{1+n} - 1}{1 + n R^{4+n} - 1}\right)^{2/3}.$$  (45)

The expression is well behaved near $n = -1, -3, -4$, where the above expression is indeterminate: for $n = -1$, $C_{PSD} = (\frac{1}{2})(R^3 - 1)(3 \ln R)^{1/3}(R^2 - 1)/(R^2 - 1)$, with $C_{PSD} \to (\frac{1}{2})(3 \ln R)^{-1/3}$, for $R \to \infty$. For $n = -3$, the rule of d’Hospital implies $C_{PSD} = \ln R (2R)^{-2/3}$. For $n = -4$, $C_{PSD} = (3 - 1)/(3 \ln R)^{2/3}(R^3 - 1)^{1/3}$, with $C_{PSD} \to 3(3 \ln R)^{-2/3}$, for $R \to \infty$.

Figure 6 shows that $C_{PSD}(n, R)$ is a smooth function of $n$, $C_{PSD}(n, R) = 1$ for $R = 1$, and $C_{PSD}(n, R) < 1$ otherwise, decreasing with growing $R = R_2/R_1$, consistent with Eq. (35). For $n \leq -5$ or $n \geq 0$, and $R \to \infty$, $C_{PSD}$ stays finite; $C_{PSD} = (1 - n)^{1/3} (-4 - n)^{2/3}(-3 - n)^{1/3}$, for $R \to \infty$.

Between $-4 < n < -1$, $C_{PSD}$ goes to zero for large $R$ with a flat minimum at $n = -3$.

For $n > -3$, the large particles contribute most to volume; hence $r_{vol} \to R_2$. For $n < -2$, the small particles contribute most to area; hence $r_{area} \to R_1$. As a consequence, there exists a subrange $-3 < n < -2$ where the ratio $r_{area}/r_{vol}$ is less than one and $C_{PSD}$ decreases strongly with $R$. Outside this range, in the larger subrange $-4 < n < -1$, the decrease is slower. The asymptotic case $C_{PSD} \to 0$ implies $r_{eff} \to 0$, $\tau \to 0$ for fixed IWC. Hence, a few very large particles may dominate the mass while contributing little to the optical properties.

Obviously, all values $0 < C_{PSD} < 1$ are possible. However, for the limited range $R < 10$, $C_{PSD}$ varies between about 0.7 and 1.

d. $C_{PSD}$ for bimodal peaks

Perhaps the most simple distribution in this connection is a bimodal distribution, consisting of two peaks,
i = 1, 2, where each has a monodisperse distribution represented by Dirac functions δ(Ri, ni) and n2/n1 = Rn, R = R2/R1. Hence, the peaks at r = R1 and R2 have amplitudes that differ by a factor similar to the previous power law. The size distribution n_p(r) is zero outside the peaks (i.e., also between the two peaks; see the full lines in Fig. 4).

The moments M_k of these distributions are

\[ M_k = R_1^k n_1 + R_2^k n_2, \quad n_2/n_1 = R^n, \quad R = R_2/R_1. \]  

(46)

With these moments, \( C_{PSD} \) follows from Eq. (30):

\[ C_{PSD} = \frac{1 + R_2^{2+n}}{(1 + R^n)^{1/2} (1 + R_1^{3+n})^{2/3}}. \]  

(47)

As shown in Fig. 7, this expression implies that \( C_{PSD} = 1 \) for \( R = 1 \), and \( C_{PSD} < 1 \) for \( R > 1 \), consistent with Eq. (35). Here \( C_{PSD} \) takes its minimum for \( n = -2 \) and \( C_{PSD} \to 0 \) for \( n = -2 \) and \( R \to \infty \). For large \( R \), \( C_{PSD} \) is small for \(-3 < n < 0\), with a flat minimum at \( n = -2 \).

Hence, this distribution is similar to that for a power law, but with \( n \) replaced by \( n + 1 \). Obviously, for \( n > -2 \), the large particles contribute most to volume; hence \( r_{vol} \to R_2 \). For \( n < -2 \), the small particles contribute most to area; hence \( r_{area} \to R_1 \). As a consequence, there exists a subrange \(-3 < n < 0\) where the ratio \( r_{area}/r_{vol} \) is less than one and \( C_{PSD} \) decreases with \( R \). Outside this range, \( C_{PSD} \) first decreases with \( R \) and then returns to 1 for very large values of \( R \).

Again, all values \( 0 < C_{PSD} \leq 1 \) are possible. However, for the realistic range limitation \( R < 10 \), \( C_{PSD} \) varies between about 0.4 and 1. Here, the lower limit is smaller than in the power-law case.

e. \( C_{PSD} \) values for averaged PSDs

The factor \( C_{PSD} \) is nonlinear in the size distributions. Hence, the value of \( C \) for the average of a set of measured PSDs is different from the average value of \( C \) for the individual PSDs. The same is true for \( r_{eff} \) and \( r_{vol} \).

Accordingly, the mean optical properties of a statistically inhomogeneous cloud depend on the ensemble over which the mean value is defined. Moreover, the following shows that a larger cloud domain in most cases has smaller \( C \) values than a smaller domain.

For illustration, we computed \( C \) for the individual PSDs plotted in Fig. 2 and for the arithmetic average of all possible combinations of two PSDs out of this set of 30 PSDs. In none of these combinations did the PSD resulting from combining two PSDs have a larger \( C \) than the maximum of the individual \( C \) values for the two PSDs. We conjecture that this statement is generally true, although we were not able to find a formal proof. On the other hand, in nearly 40% of all combinations, the averaged \( C \) value over two PSDs is less than the minimum of the \( C \) values of the two individual PSDs. Strongest reduction of \( C \) for the average PSD is obtained in the cases where the two individual PSDs had largest \( C \) (i.e., were close to monodisperse).

In fact, monodisperse PSDs (with \( C_{PSD} = 1 \)) occur rarely. Even when all particles have the same size at a specific time and position in space but these sizes vary with time and position, its mean PSD is nonmonodisperse. The bimodal spectrum discussed above may be interpreted as the mean spectrum over two subensembles with mono-
disperse PSDs. The results show that the \( C_{PSD} < 1 \) for an ensemble of clouds even when subensembles may be monodisperse.

5. The habit factor \( C_{habit} \)

This section shows that \( C_{habit} \) (i.e., the ratio of mean projected areas of particles and of volume equivalent spheres) is larger than or equal to one

\[ C_{habit} = (A_p/A_{sp}) \geq 1 \]  

(48)

for convex and concave particles of random orientation. The same is true for its weighted mean value \( T_{habit} \). Obviously, \( C_{habit} = A_p/A_{sp} = 1 \) for spherical particles. Otherwise, \( C_{habit} \geq 1 \) can easily be proven for convex particles using two well-known facts. First, the surface \( S_p \) of a particle with fixed volume \( V_p = V_s = (4/3)\pi r^3 \) takes its minimum \( S_s = 4\pi r^2 \) when the particle is spherical; otherwise,

\[ S_p > S_s. \]  

(49)

This so-called isoperimetric inequality of differential geometry is valid for both convex and concave particles (Schmidt 1939). Second, the ratio between the mean projected surface \( A_p \) and the total surface \( S_p \) is
\[ A_p/S_p = 1/4, \] (50)

on average, for an ensemble of convex particles with random orientation (Cauchy 1908; Vouk 1948). Since Eq. (50) is true also for spheres, we have \( A_p/S_p = A_{s,p}/S_p = 1/4 \). Multiplication with \( S_p/A_{s,p} \) gives \( A_p/A_{s,p} = S_p/S_s \). From Eq. (49), \( S_p/S_s \approx 1 \), and hence \( A_p/A_{s,p} \geq 1 \) for all convex particles of the same volume with random orientation.

As an illustrative example we consider the well-known case of a cylinder with diameter \( d \) and length \( L \) (Martin et al. 1988; Mishchenko et al. 1996). The projected area \( A_p \) of such cylinders is

\[ A_p = \frac{\pi}{4} d^2 \cos \theta + Ld \sin \theta, \] (51)

where \( \theta \) is the angle between the normal to the top plane surface and the projection direction. For random orientation in three dimensions, the mean values of the cosine and the sine of the angle \( \theta \) are

\[ \cos(\theta) = \int_0^{90} \cos(\theta) \sin(\theta) 2\pi d\theta/\pi = 1/2 \quad \text{and} \] (52)

\[ \sin(\theta) = \int_{-90}^{90} \sin(\theta)^2 2\pi d\theta/\pi = \pi/4. \] (53)

Hence, the mean projected surface is \( A_p = \pi d^2/8 + \pi Ld/4 \). Obviously, the result satisfies Eq. (50). With \( V_p = (\pi/4)d^2L \), it follows from Eq. (23) that

\[ C_{\text{habit}} = \frac{1/2 + L/d}{[(3/2)(L/d)]^{2/3}}. \] (54)

The function \( C_{\text{habit}}(L/d) \) takes its minimum \((1/2)^{1/3}\) or about 1.145 for \( L/d = 1 \) and approaches large values both for large and small ratios \( L/d \) (see Fig. 8).

The figure also contains \( C_{\text{habit}} \) for randomly oriented hexagons and spheroids as a function of the aspect ratio \( L/d \). For hexagonal cylinders (Takano and Liou 1989; Ebert and Curry 1992; Fu and Liou 1993), the volume and the area are \( V_p = A_hL \) and \( A_p = (2A_h + 3Ld)/4 \), with the hexagon area \( A_h = (3)^{1/2}d^2/8 \). Here, \( d \) is the basal diameter (i.e., the largest diagonal or twice the side length of the hexagon). Hence,

\[ C_{\text{habit}} = \frac{(3)^{1/2}/4 + L/d}{[(\pi)^{1/2}(3/4)(L/d)]^{2/3}}. \] (55)

Because of larger surface to volume ratio, \( C \) is slightly larger for hexagons than that for cylinders (1.185 instead of 1.145 for \( L/d = 1 \)). For very small aspect ratios, both hexagons and cylinders would have the same asymptote if plotted versus \( L/D \) instead of \( L/d \), where \( D \) follows from \( \pi D^2/4 = A_h \).

Spheroids are rotationally symmetrical ellipsoids. The aspect ratio \( L/d \) is defined here as the ratio between the rotational semiaxis \( b = L/2 \) and the semiaxis \( a = d/2 \) perpendicular to this. Depending on \( L/d \), the shape of spheroids ranges from spheres to needles (prolates) and plates (oblates). The volume of a spheroid is \( V_p = (\pi/6)\pi d^2L \), and the area \( A_p \) follows from known relationships (Mishchenko and Travis 1994; Krotkov et al. 1999). Spheroids are used to approximate real ice particle habits (e.g., in analysis of optical particle measurements; Borrmann et al. 2000). For spheroids, the value of \( C \) is one for \( L/d = 1 \) (sphere) and larger than one otherwise. Because of smaller surface to volume ratio, \( C \) for spheroids is less than that for cylinders except for extreme aspect ratios.

The previous examples were convex bodies. Chylek (1977) notes that Eq. (48) is enforced even more for concave particles of random orientation, since for any concave body there exists a convex body with smaller projected area but the same volume. From this it follows that

\[ \frac{A_p(\text{concave body})}{A_{s,p}} > \frac{A_p(\text{convex body})}{A_{s,p}} \geq 1. \] (56)

### 6. Evaluation of \( C \) for given PSD and habits

#### a. Habit model

This section provides parameterizations to compute the habit factor \( C_{\text{habit},h} \) of ice crystals of given size for seven different habits \( (h = 0–6; \text{see Table 1}) \), including convex and concave ones.
For the habits specified in Table 1, Yang et al. (2000), with input from earlier studies, derived algebraic parameterizations for the mean volume and projected cross-sectional area, which can be used to compute the volume mean radius \( r \) and the effective radius \( r_e \) of individual particles as a function of its maximum size \( D_{\text{max}} \). Note that the quantities \( r \) and \( r_e \) refer to an individual particle, whereas \( r_{\text{vol}} \) and \( r_{\text{eff}} \) describe a particle size distribution. For an individual particle \( C_{\text{PSD}} = 1 \). From this it follows that

\[
C_{\text{habit}} = C = r/r_e. \tag{57}
\]

Models usually provide input for \( r_{\text{vol}} \) or \( r \), not for \( D_{\text{max}} \). Therefore, we set up the inverse function \( r_e(r) \), first individually for each habit class, by fitting data for \( r \) and \( r_e \) versus \( D_{\text{max}} \). The result is given in Table 1. For variable aspect ratios (such as for solid columns), the results depend on the size of the particles. The table lists also the maximum volume mean radius (between 800 and 4500 \( \mu m \)) for which these data were provided. The standard error of the fits is at maximum 0.0078 \( \mu m \) for \( r_e \) of rosettes.

The results imply habit factors \( C_{\text{habit}} \) as a function of \( r \), as plotted in Fig. 9. As expected, \( C_{\text{habit}} \) equals one for spheres and is slightly larger than one for droxtals, and the largest habit factors are computed for rosettes and for plates.

b. \( C_{\text{habit}} \) for habit mixes

To account for cirrus particles of different habits, Baum et al. (2005a) suggested a habit mixture that prescribes the mass fractions of various habits as a function of maximum dimension \( D_{\text{max}} \). Their mixture assumed that all particles with \( D_{\text{max}} < 60 \mu m \) are droxtals while larger particles have mixtures of more complex habits. (We note that the mix in the size range \( 1000 < D_{\text{max}} < 2000 \mu m \) implies smaller habit factors for this size range than for the neighboring size ranges. But this is of minor importance for the present study, which concentrates on the size range below 1000 \( \mu m \).) For contrails, the observations cited in the introduction are consistent with droxtals for small particles but imply a transition to complex particles already at smaller dimensions. For larger dimensions, contrail particles seem to become similar to other cirrus crystals. Therefore, we suggest the contrail–cirrus habit mix defined in Table 2. Since the models predict the volume mean radius, we express the mixture weights as a function of both \( r \) and \( D_{\text{max}} \) (see Table 2).

Next we determine the effective radius and the habit factor \( C_{\text{habit}} \) for this contrail–cirrus habit mixture, with...
fractions $G_h(r) = \sum_{p,h} n_{p,h}(r)$, with $n_{p,h}$ being the number of particles of habit class $h$.

The equivalent sphere cross section for each habit class is $A_{p,h} = \pi r_e^2$ [cf. Eq. (20)]. The individual cross section $A_{p,h}$ is determined from Eq. (5) (applied to a PSD with a single particle)

$$A_{p,h} = \frac{3}{4} V_{p,h} \pi r_{e,h}^3,$$

with the effective radius $r_{e,h}$ of a single particle of habit $h$. Then $C_{\text{habit}}$ of the mixture is given by

$$C_{\text{habit}}(r) = \frac{\sum_{h=0}^{6} G_h(r) \cdot C_{\text{habit},h}(r)}{\sum_{h=0}^{6} G_h(r) \cdot C_{\text{habit},h}(r)},$$

From this it follows that

$$C_{\text{habit}}(r) = \sum_{h=0}^{6} G_h(r) C_{\text{habit},h}(r),$$

with $C_{\text{habit},h} = r/r_{e,h}$. Hence, $C_{\text{habit}}$ of the mixture is solely the weighted mean of the $C_{\text{habit},h}$ of each habit class. However, this statement is not generally true and is only valid if $G_h(r)$ exists or $G_h(D_{\text{max}})$ can be reasonably well recast into the form $G_h(r)$.

The weights $G_h$ are dependent on $D_{\text{max}}$. Hence, we need to know $D_{\text{max}}$ for given $r$. For the given habit mix one may compute the mean values of $D_{\text{max}}$ and $r$ by weighting the individual habit contributions. The result can be approximated as

$$D_{\text{max}} = 2[r_0 + a \ln(r/r_0) + b \ln(r/r_0)^2],$$

with $r_0 = 1 \mu m$ (see Fig. 10). Here, $y_0 = 1.402$, $a = -0.4555$, and $b = 0.1390$. (For the Baum cirrus mixture the fit is very similar: $y_0 = 1.395$, $a = -0.4851$, and $b = 0.1439$.)

Figure 11 shows the resultant factor $C_{\text{habit}}(r)$ (thick black curve). We see that $C_{\text{habit}}(r)$ grows with volume mean radius from 1.06 for small particles (droxtals) to values of order 1.6 for large particles (mainly rosettes). The $C_{\text{habit}}$ steps at $r = 5, 9.5,$ and 23 $\mu m$ are consequences of the assumed stepwise transition from pure droxtals to more complex habit mixes. For larger radius, the results are a consequence of the Baum cirrus mixture.

The step curve $C_{\text{habit}}(r)$ can be roughly approximated as (see Fig. 11)

$$C_{\text{habit}} = 2.2 + 0.0013 \frac{r}{r_0} - 1.121 \exp\left(-0.011 \frac{r}{r_0}\right).$$

For other habit mixes, the results may differ considerably. For example, Fig. 12 shows, among others, this

Table 2. Habit mixture weights $G$ (%) as a function of $D_{\text{max}}$ or $r$ for contrail cirrus.

<table>
<thead>
<tr>
<th>$D_{\text{max}}$ (µm)</th>
<th>$r$ (µm)</th>
<th>col</th>
<th>hol</th>
<th>agg</th>
<th>ro6</th>
<th>pla</th>
<th>dro</th>
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<td>0</td>
<td>3</td>
<td>97</td>
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<td>0</td>
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</table>

FIG. 10. $D_{\text{max}}$ for contrail–cirrus habits vs $r$ with data from Yang et al. (2000).

FIG. 11. $C = C_{\text{habit}}C_{\text{PSD}} = r_{\text{vol}}/r_{\text{eff}}$ vs $r_{\text{vol}}$ or sphere-equivalent particle radius $r$ for 30 different measured size distributions as provided by Key et al. (2002), assuming various habits, as listed. The blue symbols denote the result for the contrail cirrus habit mix of Table 2. The solid curve is $C_{\text{habit}}(r)$ for this mixture; the dashed curve depicts the approximation, Eq. (62).
approximation together with the habit factor that would result for the habit mix of Liou et al. (1998). The Liou mix implies about a factor of 2 larger habit factor than suggested in this study. It contains 50% bullet rosettes, 30% hollow columns, and 20% plates, that is, particles with larger habit factors than for the near-spherical droxtals. For the reasons given in the introduction, and in agreement with Baum et al. (2005a), droxtals appear to be more representative for the smallest ice particles.

c. Empirical habit factors

The habit factor for the cirrus habit mixture can be compared with empirical relationships. Several authors have derived simple power laws relating the mass \( M \) (or diameter \( D_{eq} \)) of the droplet after melting the ice particle to the projected area \( A \) of individual ice particles with certain habits, \( M/M_0 = \alpha (A/A_0)^{\beta} \), with empirical coefficients \( \alpha \) and \( \beta \) (depending on the assumed particle habit) and reference dimensions \( M_0 \) and \( A_0 \) (Cunningham 1978; Moss and Johnson 1994; Baker and Lawson 2006). These relationships imply a volume–area relationship and hence a habit factor. Relationships between mass and maximum particle dimension can be used for this purpose only when accompanied by area–dimension relationships.

For example, Baker and Lawson (2006) derived \( M/M_0 = \alpha (A/A_0)^{\beta} \) with \( \alpha = 0.115, \beta = 1.218, M_0 = 1 \) mg, and \( A_0 = 1 \) mm² for measured projected areas and volumes of ice particles. The data were derived from images of a set of ice particles collected on a dish at ground and of the resultant droplets after melting. This implies \( C_{\text{habit,BL}} = 5.67(r/r_0)^{0.45} \), with \( r_0 = 1 \) mm. Figure 12 shows that this habit factor increases with \( r_{\text{vol}} \) as expected because of growing complexity of the large ice particles and may reach values up to an order of 10 for millimeter-sized ice particles. Obviously, this relationships should be applied only for sufficiently large \( r > 25 \) μm, for which the habit factor is larger than one. Gayet et al. (1996a) use a relationship \( D_{eq} = D_0(A/A_0)^{\gamma} \), derived by Cunningham (1978) for bullet-rosettes with \( A < 0.21 \) mm², \( \gamma = 0.39, D_0 = 1 \) mm, and \( A_0 = 1 \) mm². This implies \( C_{\text{habit,C}} = 10.3(r/r_0)^{0.564} \) (for \( 0.016 < r/r_0 < 0.14 \)). Gayet et al. (2004) use similar formulas with other coefficients. The empirical relationships derived from imaging particles provide larger habit factors than the habit mixtures of Baum et al. (2005a). Possibly the image method overestimates the average projected area because the crystals most likely come to rest on the dish in their most stable orientation with largest projected area. The large differences between the approximations from various sources suggest large uncertainties in these factors. In any case, large \( C \) values are to be expected for large \( r_{\text{vol}} \) unless small \( C_{\text{PSD}} \) values counteract this trend, which is probable because large particles often imply a wide size distribution with small \( C_{\text{PSD}} \).

d. Habit influence on \( C \) for given cirrus ice particle size distributions

Key et al. (2002) set up tables with size distributions from 30 field experiments (see Fig. 2). These size distributions define the discrete concentrations \( n_i \) for size bins with index \( i \), of ice particles with mean (i.e., averaged over the cirrus particle ensemble) maximum particle sizes \( D \) in size intervals \( D_i \leq D < D_{i+1} \) of width \( \Delta D_i = D_{i+1} - D_i \). In addition we use the computed volume and projected area of ice particles in such intervals, \( V_{\text{eff}} \) and \( A_{\text{eff}} \), for each size interval \( \Delta D_i \) and given habit type using input from Yang et al. (2000). With these data, we evaluate the integrals, Eqs. (2)–(4), for \( V, A, \) and \( N \) by numerical integration:

\[
r_{\text{eff}} = \frac{3}{4} \sum_i V_p(i) n_p(i) \Delta D_i, \quad N = \sum_i n_p(i) \Delta D_i, \quad \text{and} \quad C_{\text{eff}} = \frac{3}{4} \sum_i A_p(i) n_p(i) \Delta D_i \tag{63}
\]

\[
r_{\text{vol}} = \left[ \frac{3V}{(4\pi N)^{1/3}} \right] \frac{1}{\Delta D_i}, \quad V = \sum_i V_p(i) n_p(i) \Delta D_i \tag{64}
\]

The results for \( C \) for individual habit types (see various black symbols in Fig. 11) scatter between 0.14 and 1.52. This large scatter is no longer unexpected because of highly different size distributions and particle habits. The largest values \( C \) are computed for the size distribution number 25, which stays flat until a kink near a particle size \( d \) of 40 μm and then decreases rapidly (see Fig. 13). In this case, both \( r_{\text{vol}} \) and \( r_{\text{eff}} \) are close to...
20 \mu m, and C is near 1 therefore. The smallest C value is computed for the size distribution number 1, which decreases smoothly following closely a power law with exponent $-3$. In view of our results for power laws (Fig. 7), the small value of C was to be expected for this case.

Far less scatter results when evaluating the factor C for a given habit mixture. The blue symbols in Fig. 11 show the C result for the individual PSDs using the contrail cirrus habit mix. The C values show a decreasing trend with particle size for small particles (because of increasing width of the PSDs) and then a slight increase, presumably due to growing contributions from large irregular ice particles. These results will be used below to relate C to $r_{vol}$.

7. C from experimental data

As a guide for proper C parameterizations, this section presents measured values. We first discuss results for cirrus without obvious anthropogenic influences, and then for contrails. The experimental data used are described in the appendix.

a. Experimental data for C in cirrus

The results derived from measurements in cirrus are plotted in Fig. 14. The data labeled Baum et al. (2005a) were derived similarly to those shown in Fig. 11: instead of using the contrail–cirrus habit mix, these data were computed with habit factors of Yang et al. (2000) and PSDs of Key et al. (2002) as before, but using the habit mix of Baum et al. (2005a). The differences of the results for the two habit mixes (cirrus and contrail cirrus) are small because of small number concentrations in the size range below 60 \mu m.

The results show C values scattering mostly between 0.4 and 1.1. Most of the data support the scatter and trends derived from cirrus mix of Baum et al. (2005a). These data show mean (min–max) values for $C = 0.7$ (0.4–1.1) for $r_{vol}/\mu m = 22(5–55)$. An exception is the data of Gayet et al. (2006) for large radius, which show smaller values down to 0.25. PSDs with large dispersion would explain these low values, but underestimates of habit factors and particle shattering may have also contributed to these small values. In fact, the value of C gets smaller when the number N of small particles increases [see Eq. (11)]. However, even for constant volume, an increase of N may be partly balanced by an increase in $A$, so shattering does not always imply smaller values of C. However, shattering is most important in the presence of many large ice particles, and such cases are likely connected with small negative power-law PSD exponents $n$. Therefore, we expect that shattering causes a steeper PSD with smaller $n$. For small negative $n$, C gets smaller for decreasing $n$ (see Fig. 6)—hence shattering of large particles is likely to cause a decrease of C. In the cirrus dataset of de Reus et al. (2009), incidences of shattering from the CIP instrument were removed based on particle interarrival times (Field et al. 2006), and this dataset gives results for C consistent with the other datasets. Although we cannot exclude particle shattering, we do not expect that shattering has large effects on the values of C on average, at least for the range of $r_{vol}$ discussed in this paper.
b. Experimental data for $C$ in contrails

Because contrail ice crystals are of different origin, have small mean size with narrow PSD, and are nearly spherical, the value of $C$ may be different for contrail cirrus compared to other cirrus, particularly at small ages. Only a few measurements are available to evaluate $C$ for contrails. In the appendix we collect the existing information and add new results from recent in situ measurements in contrails during CONCERT (Voigt et al. 2010). The results are plotted versus $r_{\text{vol}}$ in Fig. 15 and versus plume age in Fig. 16. Both figures use logarithmic abscissa to cover the highly variable sizes and ages.

As a whole, Fig. 15 shows $C$ values with considerable scatter from 0.3 to 1.2. The values are closer to one for small particles and the data generally indicate a decrease of $C$ with $r_{\text{vol}}$, similar to the cirrus cases. The smallest volume mean radius in these data is near 0.33–0.5 μm (Petzold et al. 1997; Schröder et al. 2000), causing significant deviations from $Q_{\text{ext}} = 2$. For small contrail ice particles, the $C$ values are close to one because of narrow size distributions (Petzold et al. 1997). Smaller $C$ values arise because of widening size distributions. The $C$ values of Schröder et al. (2000) and Febvre et al. (2009) in contrails are smaller than those obtained by the same teams in ambient cirrus, presumably because of larger habit factors for the larger cirrus particles. For larger sizes, some of the large $C$ values in contrails may be caused by nearly monodisperse size distributions and strongly nonspherical particles (e.g., in warm and humid atmospheres) (Gayet et al. 1996a). However, large $C$ values may be also a consequence of a large habit factor assumed for large particles (see Fig. 12). The $C$ values show no systematic dependence on the aircraft type (Voigt et al. 2010) or the fuel sulfur content (Petzold et al. 1997). In general, the $C$ values are within the range found in Fig. 14 for cirrus.

Figure 16 shows the dependency on plume age. For fresh contrails (<10 s), $C$ is in between 0.7 and 0.9. Such rather large values are expected for small spectral dispersion and near-spherical particle shape (droxtals). For larger plume ages, the data exhibit large scatter (around $0.7 \pm 0.4$) without obvious systematic trends. With growing plume age, some ice particles grow in size and the spectral dispersion and the habits may approach common cirrus-cloud values (Schröder et al. 2000). Some of the contrails were reported as “evaporating” (Schröder et al. 2000). In these cases, sublimation may cause a rounding of ice particles (Nelson 1998) and hence a reduction of $C$.

c. Variation of $C$ in cirrus and contrails with relative humidity

For a few cases (Schröder et al. 2000; de Reus et al. 2009; Voigt et al. 2010) we have simultaneous measurements of PSDs together with relative humidity over ice $\text{RH}_i$. $\text{RH}_i$ was measured in situ with different frost point hygrometers (Schröder et al. 2000; de Reus et al. 2009) or a fluorescence hygrometer combined with temperature measurements (Voigt et al. 2010). The cited publications report estimated uncertainties of $\text{RH}_i$ of about 15% (Voigt et al. 2010; for details, see the references cited). Hence, we can investigate the impact of humidity on $C$ (see Fig. 17). Of course, the measured $\text{RH}_i$ is the humidity at the time of ice particle measurement, not the humidity during cloud formation. This may contribute to the scatter of the data. The data of de Reus et al. (2009) were obtained mostly at very low temperatures and therefore may be well comparable to contrail cases.
Contrails form by condensing water mainly emitted by the aircraft engines; they later take up water from ambient air in ice supersaturated air masses. In subsaturated air masses the particles sublimate. Contrails formed by heavy four-engine aircrafts can survive for several minutes even for considerable subsaturation since large amounts of water vapor are emitted. Contrails of smaller two-engine aircrafts can evaporate after a few seconds (Sussmann and Gierens 2001). For supersaturated air masses, the results indicate a wide range of $C$, without clear indications for a dependence on humidity. For subsaturated cases, the CONCERT $C$ values seem to increase with decreasing humidity. The humidity measurements by Schroeder et al. (2000) show rather low humidity (down to 50% for contrail ages up to 120 s, and 60% in ambient cirrus) and large scatter for $C$.

The data suggest that for high ambient humidity, the PSD widens (causing reduced PSD factors). In subsaturated air masses the measured PSD contains less particle volume in the larger size bins, causing smaller volume mean radius. The effective radius gets also reduced, perhaps even more quickly, because of more efficient sublimation of particles with large surface. In addition we expect a rounding of the shape of the sublimating particles, although this cannot be detected with the instruments used. The variability of humidity obviously explains a large part of the scatter of the data seen in Figs. 14 and 15.

### 8. Parameterization of $C$

#### a. Parameterizations used in previous models

The need for parameterization of $C$ has been acknowledged by all teams who use two-moment cirrus models. Models that assume a lognormal PSD with fixed dispersion (e.g., Spichtinger and Gierens 2009) effectively prescribe the value of $C$ with this assumption [see Eq. (41)]. Other models use parameterizations as plotted in Fig. 18.

Lohmann (2002) includes a prognostic equation for the ice particle number concentration in a global climate model and prescribes $C$ as a function of a given temperature-dependent effective radius. Lohmann et al. (2008) essentially use the same relationship but with $C$ as a function of the volume mean radius that follows from the computed IWC and ice particle number concentration (see Fig. 18). This function implies a monotonic decrease of $C$ with $r_{vol}$ that may be reasonable for spherical particles but does not account for more complex particle habits. Hence, this model may underestimate the optical depth of thick natural cirrus clouds. The underestimate could be even stronger when shattering causes an underestimate of $C$ in the data.

Ström and Gierens (2002) study the impact of aircraft particle emissions on the radiative properties of contrails. They use a prognostic model for IWC and the number density of ice crystals and relate the effective radius to the mean ice particle mass, which can be expressed as a function of $r_{vol}$. For large $r_{vol}$, their result is similar to the Lohmann model. At small $r_{vol}$, the tendency to large $C$ values is unrealistic, as we now know. It overemphasizes the climate impact of very small ice particles.

Liu et al. (2007, 2009) and Penner et al. (2009) parameterize $C$ as a function of temperature and IWC based on data derived by Wyser (1998). We have plotted their $C$ value [with the coefficients as given in Penner et al. (2009)] for a median midlatitude IWC–temperature correlation (Schiller et al. 2008). Observed IWC values show large scatter relative to this median. Therefore, we also show curves for the same IWC multiplied with 0.1 or 10. The $C$ parameterization gives reasonable values in the range as observed but tends to overshoot above one for small IWC values and low temperatures. This could cause an overestimate of the radiative impact of thin and subvisible cirrus and an underestimate of the impact of thick cirrus. In any case, these models do not account for changes in the particle habits.

#### b. New parameterization

In view of the data and understanding gained so far, we are looking for a simple parameterization that reflects the deduced dependencies on the particle habit, the volume mean radius, and the relative humidity. The above data suggest some coherent relationships between $C$ and $r_{vol}$ and between $C$ and RH. No separate function is needed to account for contrail age. The contrail age is
Fig. 18. (top) $C$ vs $r_{vol}$ for the parameterizations of Lohmann et al. (2008) and Ström and Gierens (2002). (bottom) $C$ vs temperature, as used in Liu et al. (2007, 2009) and Penner et al. (2009), for a median IWC–temperature correlation (factor 1) and its multiples (factors 0.1 and 10).

Reflected in the particle sizes, which grow with age in supersaturated air or shrink otherwise. Therefore, we suggest the approximation

$$C(r_{vol}, RH_i) = 1 + (C_r - 1) \frac{[1 - \exp(-RH_i)]}{[1 - \exp(-1)]},$$

with

$$C_r(r_{vol}) = 0.9 + (C_{habit} - 1.7)[1 - \exp(-r_{vol}/r_1)]$$

and with $C_{habit}(r_{vol})$ as given in Eq. (62), and $r_1 = 20 \mu$m. For $RH_i = 1$, $C = C_r$. This approximation is plotted for $RH_i = 1$ in Figs. 14 and 15. In this approximation, $C$ assumes that the value $C_{habit}$ for a narrow PSD with aspherical particles (about 0.9 for spherical particles) goes to one in very dry air ($RH_i \to 0$) and decreases slightly in ice supersaturated air ($RH_i > 1$). This approximation uses input as available in two-moment models.

For uncertainty analysis, one may vary $C$ in the limits

$$C_{min} < C < C_{max},$$

where $C_{min}(r_{vol}) = 0.6 \ C_r(r_{vol})$ and $C_{max}(r_{vol}) = 0.9 \ C_{habit}(r_{vol})$. Of course, these are not strict bounds but appear to be reasonable limits for the data shown in Figs. 14 and 15.

9. Conclusions

We have discussed the relationship $C = (r_{area}/r_{vol})^2 = r_{col}/r_{eff}$ between the volume-specific radius and the effective radius. For ice particles with constant extinction efficiency, the optical depth is determined when knowing the IWP, the number concentration of ice particles, and the factor $C$. For small ice particles, as in young contrails, or for thermal wavelengths, the deviation of the effective extinction efficiency from the geometric optics limit has to be taken into account (Key et al. 2002; Yang et al. 2005; Hong et al. 2009). The ratio $C$ is a purely geometrical parameter and depends in general strongly on the PSD and on the particle habits; $C$ can be expressed as the product of a habit and a PSD-related factor $C_{PSD}C_{habit}$. We have shown strictly that $0 < C_{PSD} \approx 1$ for arbitrary PSD in general and equal to 1 for a monodisperse PSD. Moreover, $C_{habit} \approx 1$ for convex and concave particles with random orientation in general and is equal to 1 for spherical particles.

In general, $C$ may get very small for PSDs that decay with size similar to a power law with exponent $n$ between $-4$ and $-1$ ($-3$ and $0$ for bimodal spectra). This has been derived analytically for simple model PSDs. For such wide and slowly decaying PSDs, most of the particle volume (and ice water content) is controlled by a few large particles, while most of the cross-sectional area (and optical depth) is controlled by the many small particles. The same is true for lognormal size distributions with large dispersion. Since $C$ is a nonlinear function of the size distributions, the mean optical properties of a statistically inhomogeneous cloud depend also on the ensemble over which the mean value is defined. A larger cloud domain tends to have smaller average $C$ values than a smaller domain.

We provide an analysis of PSD and habit data of Yang et al. (2000) and Key et al. (2002). The analysis provides specific values of $C$ as a function of particle sizes and habits. In addition, we analyze the value of $C$ from previous and recent in situ measurements. In the various measured cases, for $r_{vol}$ below about 100 $\mu$m, $C_{PSD}$ varies typically between values of order 0.4 and 1, and $C_{habit}$ ranges between 1 and about 2. Because of growing aspect ratios, $C_{habit}$ grows with the particle size. The modeled habit factor is somewhat smaller than assumed in data analysis of particle measurements.

For contrail cirrus, a new particle habit mix is suggested. This habit mix includes near-spherical (droxtal)-shaped particles for the very small particles and allows for more complex shapes already at rather small maximum dimension. In the size range of contrails crystals, the habit factor for this mix is about a factor of 2 smaller than for the habit mix of Liou et al. (1998).
For contrails and cirrus we suggest a new parameterization to estimate \( C \) in two-moment models as a function of IWC, number of ice particles, and possibly relative humidity. For parameter studies reasonable upper and lower bounds are estimated. For fresh contrails, \( C = 0.7 \pm 0.3 \) because of near-spherical particles and PSDs with small dispersion. Any uncertainty in \( C \) has direct impact on the accuracy of the computed optical depth of the cloud.

In the new parameterization, \( C \) is decreasing less quickly with \( r_{\text{vol}} \) than in older parameterizations (Lohmann et al. 2008; Penner et al. 2009). This increases the relative contribution of large particles to optical depth. As a consequence, the climate impact of freshly nucleated small ice crystals may be smaller than estimated before.

For further progress, one would need more basic understanding of the processes and parameters controlling the ice habits and particle size distributions.

Acknowledgments. This work was performed within the DLR project “Climate-Compatible Air Transport System” (CATS). The CONCERT campaign was organized by the HGF junior research group “Impact of Aircraft Emissions on the Heterogeneous Chemistry of the Tropopause Region” (AEROTROP). For computation of the extinction efficiency of spheroids, T-matrix results from Josef Gasteiger were used. Philip Kotter helped in the formulation of the mathematical properties of convex and concave bodies. Stephan Borrmann made the data from de Reus et al. (2009) available to us. Discussions with Pat Minnis and Joyce Penner during the “Aviation Climate Change Research Initiative” (ACCRI) helped our understanding.

APPENDIX

Experimental Data Used to Derive \( C \) in Cirrus and Contrails

Fu and Liou (1993) report data for IWC, \( N \), and \( r_{\text{eff}} \) for a series of 11 size distributions measured in various cirrus clouds. They assumed the ice particles to be hexagonal solid columns. Since their definition of \( D_{\text{eff}} \) is based on \( V/A \), and not on \( 3V/(2A) \), we multiply their effective diameter values by a factor \( \sqrt{3} \) to be consistent with the present definitions. With these data we compute the equivalent spherical volume mean radius [Eq. (17)] and \( C \). The results are shown in Fig. 14. They scatter between 0.5 and 1, presumably because of different PSD widths and different aspect ratios.

Minnis et al. (1998) used basically the same size distributions and a case representing a large-particle cirrus cloud. For the mean volume \( V/N \) and the mean projected area \( A/N \) from MIN, we compute \( r_{\text{vol}} \) and \( C \). The results (see Fig. 14) are, as expected, similar to the Fu and Liou results. The results scatter between 0.74 and 1.06.

Gayet et al. (2002, 2004, 2006) measured ice particle properties in cirrus at southern midlatitudes westward of Punta Arena, Argentina. Using the published mean values of ice water content and extinction coefficients \( \beta \), we compute the volume mean and effective radius as explained above, resulting in the \( C \) values shown in Fig. 14. The \( C \) values generally decrease systematically with \( r_{\text{vol}} \), indicating widening PSDs. The scatter of the individual values is rather small when one considers the large statistical uncertainties (on the order of 30%–100%) of IWC, \( N \), and \( \beta \) from the high-frequency (5 s) data (Gayet et al. 2002) and possible deviations from \( Q_{\text{ext}} = 2 \) for small particles. This shows that these data contain consistent physical information. The \( C \) values are significantly smaller than the values reported by Fu and Liou (1993) and Minnis et al. (1998). This may be a consequence of the different particle measurement instruments. The older data were based on measurements with particle sizes larger than 25 \( \mu \)m, while Gayet et al. (1996b) included FSSP-300 data for particles in the size range \( 0.3 < D < 25 \mu \)m. Inclusion of the smaller crystals measured with the FSSP reduces the effective radius.

De Reus et al. (2009) performed in situ measurements of ice crystal PSDs in the tropical troposphere and lower stratosphere using a high-altitude research aircraft during a campaign in Darwin, Australia. The measurements were performed using a combination of a Forward Scattering Spectrometer Probe (FSSP-100) and a Cloud Imaging Probe (CIP) with removal of shattering incidences (Field et al. 2006; de Reus et al. 2009). In total 89 valid PSDs at temperatures between \(-89^\circ \) and \(-35^\circ \)C (mostly below \(-60^\circ \)C) were available to us, together with relative humidity data for about half of the cases. Volume and area of the particles were computed assuming that the FSSP measured the scattering cross section of ice spheres while the CIP measured the projected area of complex ice particles with a mass–area relationship as in Baker and Lawson (2006) (see Fig. 12). Significantly smaller \( C \) and larger \( r_{\text{vol}} \) would result if the CIP data were assumed to represent spheres.

For contrails, Petzold et al. (1997) report seven data points for IWC, ice particle number concentration \( N \), and effective diameter or radius \( r_{\text{eff}} \) for spheres [Eq. (8)], from in situ measurements in young contrails behind two aircraft (ATTAS and A310–300), in a very young (250-m distance, <2-s plume age), and in slightly aged contrails (1800 m, 10 s). The measurements were taken in contrails behind engines burning either high or low fuel sulfur contents. The data reflect only a slight impact of the variations in fuel sulfur content on the ice particle...
size spectra. The data are used to compute the volume mean radius [Eq. (17)]. The results for C (see Fig. 15) scatter between 0.8 and 0.9.

Four data points are available from Gayet et al. (1996a,b) for contrails, partly embedded into thin natural cirrus, at rather warm temperatures (−37°C) and apparently high ambient humidity, at contrail ages of about 300–400 s. The measurements took place at 7-km altitude over the North Sea. Gayet et al. (1996a) reported analysis of the data for particles >25 μm; later Gayet et al. (1996b) also evaluated the FSSP probe measuring in the size range 2–25 μm. Inclusion of the smaller particles reduces the value of C. The results show quite large C values, partially larger than one possibly because of a large area–mass relationship assumed for the larger particles.

The data point from Poellot et al. (1999) corresponds to the contrail case (CON) discussed by Minnis et al. (1998). The CON case is based on First International Satellite Cloud Climatology Project (ISCCP) Regional Experiment (FIRE) Cirrus Intensive Field Observation (IFO)-II aircraft measurements taken in a contrail embedded within a cirrus cloud during a 22 November 1991 flight, with plume age of about 11.5 min (Poellot et al. 1999). Minnis et al. (1998) also presented a young contrail case (NCON) that was constructed to represent an “extreme” case with ice crystal sizes below 20 μm. The values of C computed from the given volume V/N and area A/N per ice particle (MIN) are C = 0.8 (CON) and 1.2 (NCON).

Schröder et al. (2000) report measurements of ice particles in contrails of various ages. They tabulate the volume mean diameter and the effective diameter, from which we can evaluate C without further assumptions. The young contrail particles were close to spherical as evidenced by impactor measurements. Some of the contrails were observed at low ambient humidity and reported as “evaporating.”

Febvre et al. (2009) present data for a young contrail (2.5 min), for a 20-min-aged persistent contrail, and for frontal cirrus clouds without obvious aircraft signatures (such as enhanced nitrogen oxide). They report ice water content and particle concentrations as needed to compute the volume mean radius. In addition, they report measured β values, from which [see Eqs. (15) and (16)] the effective radius results:

\[ r_{\text{eff}} = \left[ \frac{Q_{\text{ext}}}{2\rho_{\text{ice}}} \right] \frac{\text{IWC}}{\beta}. \]  

(A1)

The data (see Fig. 14) show small volume mean radii (3 and 4.5 μm) for 2.5- and 20-min contrail ages, respectively, and rather small C values of about 0.5, decreasing with contrail age, because of widening size distributions.

Recently, we performed particle and humidity measurements in contrails behind seven different airliners, at contrail ages between 1 and 10 min, in super- and subsaturated air masses, within and outside other cirrus clouds (CONCERT; Voigt et al. 2010). Contrail and cirrus were discriminated based on nitrogen oxides and small particle concentration measurements. The number density of ice particles at sizes 0.45–25 μm was measured with an FSSP-300; larger particles were detected with various two-dimensional optical probes but were unimportant for this analysis because the ambient cirrus had far lower crystal concentrations than the contrails (Voigt et al. 2010). Hence the large concentrations in the contrails are not shattering artifacts. The bin boundaries of the FSSP were derived assuming prolate spheroids (aspect ratio = 2:1) as in Bormann et al. (2000). The same analysis assuming spherical ice particles instead of spheroids results in about 10% smaller C values on average because of about 7% smaller habit factor and different attribution of the counted particles to the individual size bins. The results show values C = 0.7(0.46 – 0.93) for \( r_{\text{eff}} = 2.7(0.5 - 3.3) \) μm.

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