

1 **Effect of the Inhomogeneity of Ice Crystals on Retrieving Ice Cloud Optical**
2 **Thickness and Effective Particle Size**

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Abstract

Spherical or spheroidal air bubbles are generally trapped in the formation of rapidly growing ice crystals. In this study the single-scattering properties of inhomogeneous ice crystals containing air bubbles are investigated. Specifically, a computational model based on an improved geometric-optics method (IGOM) has been developed to simulate the scattering of light by randomly oriented hexagonal ice crystals containing spherical or spheroidal air bubbles. A combination of the ray-tracing technique and the Monte Carlo method is used. The effect of the air bubbles within ice crystals is to smooth the phase functions, diminish the 22° and 46° halo peaks, and substantially reduce the backscatter relative to bubble-free particles. These features vary with the number, sizes, locations and shapes of the air bubbles within ice crystals. Moreover, the asymmetry factors of inhomogeneous ice crystals decrease as the volume of air bubbles increases. Cloud reflectance lookup tables were generated at wavelengths $0.65\ \mu\text{m}$ and $2.13\ \mu\text{m}$ with different air-bubble conditions to examine the impact of the bubbles on retrieving ice cloud optical thickness and effective particle size. The reflectances simulated for inhomogeneous ice crystals are slightly larger than those computed for homogenous ice crystals at a wavelength of $0.65\ \mu\text{m}$. Thus, the retrieved cloud optical thicknesses are reduced by employing inhomogeneous ice cloud models. At a wavelength of $2.13\ \mu\text{m}$, including air bubbles in ice cloud models may also increase the reflectance. This effect implies that the retrieved effective particle sizes for inhomogeneous ice crystals are larger than those retrieved for homogeneous ice crystals, particularly, in the case of large air bubbles.

48 **1. Introduction**

49 An appropriate representation of cirrus clouds in radiative transfer simulations has
50 long been a subject of great interest, not only because of their importance for cloud
51 radiative forcing and energy budget of the earth, but also because of the uncertainties
52 associated with the shapes and sizes of ice crystals within these clouds [*Ramanathan et*
53 *al.*, 1983; *Liou*, 1986; Baran, 2004]. Heymsfield and Platt [1984] and Heymsfield [1977]
54 developed representative data sets for cirrus cloud particles based on in situ
55 measurements. The ice crystal samples, collected both in late winter and spring, showed
56 that hexagonal hollow columns and solid columns were predominant at temperatures
57 below 223K. Some other particle habits, such as hexagonal plates and bullet rosettes were
58 also observed at the top of ice clouds.

59 Although approximating the single-scattering properties (e.g., phase function,
60 single-scattering albedo, and asymmetry factor) of realistic ice crystals by assuming one
61 idealized geometrical shape is an oversimplification [*Foot*, 1988], it is significantly better
62 for retrieving ice cloud properties than assuming that the clouds are composed of
63 spherical ice crystals [*Minnis et al.*, 1993]. But, a more accurate representation of cirrus
64 cloud ice crystal properties is needed. For example, the use of homogeneous hexagonal
65 ice crystals [*Minnis et al.*, 1998] can yield accurate estimates of ice water path [*Mace et*
66 *al.*, 2005], but the retrieved optical depths tend to be too low [*Min et al.*, 1994] implying
67 overestimates of the effective particle size. To further improve the representation of cloud
68 ice crystals in radiative transfer calculations, steady progress has been made toward
69 single-scattering computations involving various complex particle shapes away from the
70 simple single shape. *Liou* [1972] first assumed non-spherical ice crystals as long circular

71 cylinders, who showed significant differences in the comparison of the phase functions
72 for polydisperse spheres and the counterparts for long circular cylinders. *Takano and*
73 *Liou* [1989], *Muinonen* [1989], *Brorovoi and Grishin* [2003] and many others applied the
74 traditional ray-tracing method or its modified forms to the scattering by randomly and
75 horizontally oriented hexagonal particles. The optical properties of various complicated
76 ice crystals have been simulated by *Macke* [1993], *Macke et al.* [1996], *Iaquinta et al.*
77 [1995], *Takano and Liou* [1995], *Yang and Liou* [1998], *Um and McFarquhar* [2007],
78 and *Schmitt et al.* [2006]. *Yang and Liou* [1996] employed the finite-difference time
79 domain (FDTD) method to simulate the scattering of light by small bullet-rosettes,
80 hexagonal plates, solid columns, and hollow columns. Recently, the optical properties of
81 highly complex habits, such as ice crystals with surface roughness and hollow bullet
82 rosette ice crystals, have been investigated [*Yang et al.*, 2008 a, b; *Yang et al.*, in press].
83 The results from these efforts have been used in the simulation of radiative transfer in
84 cirrus clouds.

85 Homogeneous ice crystals are extensively employed in the aforementioned
86 studies of the single-scattering properties of irregular ice particles. In the accretion and
87 aggregation of ice crystals, an ice particle may collide with supercooled water droplets or
88 other ice particles. When this happens, ice crystals can rapidly grow and form large ice
89 crystals. The collision and coalescence processes may lead to the trapping of spherical or
90 spheroidal air bubbles within ice crystals when the supercooled water droplets freeze
91 almost instantly [*Tape*, 1994]. Air bubbles may also originate when water containing
92 dissolved air freezes into ice crystals. *Hallett* [1964] showed that supercooled water turns
93 into ice by solidifying the remainder from outside but the process is quite slow. This

94 inward growth of the ice may cause the originally dissolved air to be released and
95 subsequently form small bubbles in the center of the ice particle. The size and
96 concentration of air bubbles are then influenced by the rate of freezing, amount of
97 dissolved air in water, and temperature during the freezing process [*Carte*, 1961; *Hallett*,
98 1964].

99 There are only a handful of studies reported on the optical properties of
100 inhomogeneous ice crystals because of the lack of laboratory and in situ measurements
101 and difficulties in specifying the inclusion shapes. Among these previous studies, *Macke*
102 *et al.* [1996] employed a combination of the ray-tracing and Monte Carlo techniques to
103 investigate the single-scattering properties of randomly oriented hexagonal columns
104 containing ammonium sulfate, air bubbles, and soot impurities. In their computations, the
105 scattering events at the outer boundary of the hexagonal particle are considered by using
106 the ray-tracing technique [*Macke et al.* 1993], whereas the Monte Carlo method is used to
107 account for the photon propagation directions affected by the internal inclusions. *Yang et*
108 *al.* [2000] used the FDTD technique to compute the scattering phase functions of ice
109 crystals with inclusions of soot impurities and air bubbles. *Labonnote et al.* [2001]
110 developed an Inhomogeneous Hexagonal Monocrystals (IHM) model for ice crystals
111 containing randomly located air bubbles. This single-scattering property model, based on
112 the ray-tracing and Monte Carlo techniques developed by *Macke et al.* [1996], has further
113 defined the internal air bubbles in terms of spherical voids with a size distribution.
114 Studies on the microphysical and optical properties of ice clouds were carried out by
115 *Labonnote et al.* [2001] and *Knap et al.* [2005], who used the IHM model to investigate

116 the bulk-scattering properties of ice clouds and to compare the simulations with satellite-
117 based measurements of polarized radiances.

118 The IHM model does not account for the case where an ice crystal contains only a
119 few air bubbles with specific locations. The geometries of air bubbles in the previous
120 studies are restricted to be spheres, a constraint that is not always realistic. This paper
121 reports on a new inhomogeneous ice crystal model based on the surface observations
122 reported by *Tape* [1994]. Furthermore, the effect of the air bubbles on the retrieval of
123 cloud optical thickness and effective particle sizes is investigated. This paper is organized
124 as follows. In Section 2, we describe the morphologies of ice crystals observed by *Tape*
125 [1994] and define the geometries of the inhomogeneous ice crystals for the present
126 scattering computations. Then, we introduce the single-scattering model based on an
127 improved geometrical-optics method (IGOM). In Section 3, we illustrate the effect of the
128 number, shape, size, and location of the air bubbles inside hexagonal ice crystals on the
129 single-scattering properties of these particles. In Section 4, we show ice cloud
130 bidirectional reflectance as a function of the effective particle size, optical thickness, and
131 solar and satellite viewing angles, which is computed from the inhomogeneous ice crystal
132 model. Moreover, we derive cloud microphysical and optical properties based on the
133 Moderate Resolution Imaging Spectroradiometer (MODIS) measurements and compare
134 the retrieval results from homogeneous and inhomogeneous ice crystal models. The
135 conclusions and discussions of this study are given in Section 5.

136

137 **2. Single-scattering model for inhomogeneous ice crystals**

138 Although the geometries of ice crystals in the atmosphere have been extensively
139 studied using airborne in situ observations [*Korolev and Isaac, 1999; Heymsfield and*
140 *Platt, 1984; McFarquhar and Heymsfield, 1996*], ground-based observations also provide
141 useful data for investigating ice crystal morphologies. *Tape* [1983, 1994] used Petri
142 dishes containing hexane or silicone oil and acrylic spray to collect ice crystals falling
143 near the surface and observed the ice crystal shapes using a binocular microscope. Figure
144 1 illustrates the ice crystals sampled by *Tape* [1994] at the South Pole on January 19,
145 1985 and January 17, 1986. In the photographs, the ice crystals have typical hexagonal
146 shapes and most of these particles are inhomogeneous with air bubbles inside. The
147 inhomogeneous ice crystal morphologies observed at the surface are consistent with those
148 from airborne observations. The observed inhomogeneous ice crystals spurred
149 development of the theoretical models used by *Macke et al. [1993]* and *Labonnote et al.*
150 [2001] to compute the single-scattering properties of these particles. However, unlike the
151 crystal geometries in the IHM model [*Labonnote et al. 2001*], an inhomogeneous ice
152 crystal contains a few air bubbles with visible dimensions. The sizes of the air bubbles
153 are relatively large, as the maximum dimensions of the air bubbles are comparable with
154 the width of the ice crystal. Another significant difference between the observations by
155 *Tape* [1994] and the IHM model is that the actual air bubbles are not always spheres,
156 although most of them have spherical shapes. Moreover, the air bubbles are located
157 almost exclusively along the vertical axes of the hexagons in the center of hexagonal
158 columns. However, for hexagonal plates, more than one air bubble can be horizontally
159 aligned near the surface of the particles.

160 Based on the ice particles photographed by *Tape* [1994], the geometries of
161 inhomogeneous ice crystals in this study are defined as those shown in Fig. 2. For
162 hexagonal columns, only one or two air bubbles are included within ice particles.
163 Furthermore, the air bubble inclusions in our model are all on the axes of ice crystals (see
164 the upper and middle panels in Fig. 2). For hexagonal plates, the air bubbles are aligned
165 horizontally if more than one air bubble is included (see the lower panels in Fig. 2). The
166 orientations of ice crystals for either hexagonal columns or plates are specified in the
167 OXYZ coordinate system denoted in Fig. 3. Following *Yang and Liou* [1996], the Y-axis
168 in Fig. 3 is perpendicular to one of the ice crystal's side faces, and the Z-axis is along the
169 vertical axis of the hexagon. The shape of an air bubble is defined in terms of the
170 following equation:

$$171 \quad \frac{(x - x_r)^2}{r_1^2} + \frac{(y - y_r)^2}{r_2^2} + \frac{(z - z_r)^2}{r_3^2} = 1, \quad (1)$$

172 where r_1 , r_2 , and r_3 are the three semi-axes along the X, Y and Z axes, respectively, and
173 the coordinates (x_r, y_r, z_r) specify the center of the air bubble in the OXYZ system.

174 In this study, the IGOM [*Yang and Liou*, 1996] based on the ray-tracing technique
175 is used to compute the single-scattering properties of inhomogeneous ice crystals. At the
176 outer boundary of the inhomogeneous ice crystals, the computation of reflection and
177 refraction events is the same as in the case for homogeneous hexagonal ice crystals.
178 Detailed descriptions of the IGOM method are reported in *Yang and Liou* [1996].

179 If a ray is refracted into an ice crystal, the next step is to trace the refracted ray
180 and determine if it is intersected by any air bubble within the particle. Figure 4 shows the
181 flow-chart for reflection and refraction by internal air bubbles. For an air bubble with the

182 particle shape given by Eq. (1), the coordinates of the incident point B, (x_b, y_b, z_b) , can be
 183 determined as follows:

$$184 \quad x_b = x_a + (\hat{e} \cdot \hat{x})l, \quad (2)$$

$$185 \quad y_b = y_a + (\hat{e} \cdot \hat{y})l, \quad (3)$$

$$186 \quad z_b = z_a + (\hat{e} \cdot \hat{z})l, \quad (4)$$

187 where the coordinates (x_a, y_a, z_a) indicate the position of the first incident point A at the
 188 ice crystal surface, \hat{e} is a unit vector along the incident direction, \hat{x} , \hat{y} , and \hat{z} are the
 189 unit vectors along the X, Y, and Z axes, respectively, and l is the distance between points
 190 A and B. Substituting Eqs. (2)-(4) into Eq. (1), we obtain

$$191 \quad A_1 l^2 + A_2 l + A_3 = 0, \quad (5)$$

192 where

$$193 \quad A_1 = r_2^2 r_3^2 (\hat{e} \cdot \hat{x})^2 + r_1^2 r_3^2 (\hat{e} \cdot \hat{y})^2 + r_1^2 r_2^2 (\hat{e} \cdot \hat{z})^2, \quad (6)$$

$$194 \quad A_2 = 2r_2^2 r_3^2 (x_a - x_r)(\hat{e} \cdot \hat{x}) + 2r_1^2 r_3^2 (y_a - y_r)(\hat{e} \cdot \hat{y}) + 2r_1^2 r_2^2 (z_a - z_r)(\hat{e} \cdot \hat{z}), \quad (7)$$

$$195 \quad A_3 = r_2^2 r_3^2 (x_a - x_r)^2 + r_1^2 r_3^2 (y_a - y_r)^2 + r_1^2 r_2^2 (z_a - z_r)^2 - r_1^2 r_2^2 r_3^2. \quad (8)$$

196 A ray will intercept an air bubble when A_1 , A_2 , and A_3 satisfy

$$197 \quad A_2^2 - 4A_1 A_3 > 0, \quad (9)$$

198 and

$$199 \quad \frac{-A_2 - \sqrt{A_2^2 - 4A_1 A_3}}{2A_1} > 0. \quad (10)$$

200 The directions of the reflected and refracted rays, \hat{e}_r and \hat{e}_t can be determined on the
 201 basis of Snell's law in the form of

$$202 \quad \hat{e}_r = \hat{e} - 2(\hat{e} \cdot \hat{n})\hat{n}, \quad (11)$$

$$203 \quad \hat{e}_t = N_r [\hat{e} - (\hat{e} \cdot \hat{n})\hat{n} - \sqrt{N_r^2 - 1 + (\hat{e} \cdot \hat{n})^2} \hat{n}], \quad (12)$$

204 where N_r is the real part of an adjusted refractive index that has been formularized by
 205 *Yang and Liou* [1995] and \hat{n} is the normal direction of the air-bubble surface at point B.
 206 For spheroidal air bubbles used in this study, \hat{n} can be given by

$$207 \quad \hat{n}_x = \frac{2(x_b - x_r)}{r_1^2} \bigg/ \sqrt{\left[\frac{2(x_b - x_r)}{r_1^2}\right]^2 + \left[\frac{2(y_b - y_r)}{r_2^2}\right]^2 + \left[\frac{2(z_b - z_r)}{r_3^2}\right]^2}, \quad (13)$$

$$208 \quad \hat{n}_y = \frac{2(y_b - y_r)}{r_2^2} \bigg/ \sqrt{\left[\frac{2(x_b - x_r)}{r_1^2}\right]^2 + \left[\frac{2(y_b - y_r)}{r_2^2}\right]^2 + \left[\frac{2(z_b - z_r)}{r_3^2}\right]^2} \quad (14)$$

$$209 \quad \hat{n}_z = \frac{2(z_b - z_r)}{r_3^2} \bigg/ \sqrt{\left[\frac{2(x_b - x_r)}{r_1^2}\right]^2 + \left[\frac{2(y_b - y_r)}{r_2^2}\right]^2 + \left[\frac{2(z_b - z_r)}{r_3^2}\right]^2}. \quad (15)$$

210 For the ray refracted into the air bubble, the next impinging point C, (x_c, y_c, z_c) , on the
 211 air-bubble surface can be determined as follows:

$$212 \quad x_c = x_b + (\hat{e}_t \cdot \hat{x})l', \quad (16)$$

$$213 \quad y_c = y_b + (\hat{e}_t \cdot \hat{y})l', \quad (17)$$

$$214 \quad z_c = z_b + (\hat{e}_t \cdot \hat{z})l', \quad (18)$$

215 where l' is the distance between points B and C. l' can be solved from Eqs. (5)-(8) by
 216 replacing l and \hat{e} by l' and \hat{e}_t , respectively.

217 If the conditions in Eqs. (9) and (10) are not satisfied, i.e., the incident ray does
 218 not impinge upon the air bubble centered at (x_r, y_r, z_r) , the process above will be repeated
 219 for another air bubble if more than one air bubble is embedded in the ice crystal of
 220 interest.

221

222 3. Single-scattering properties of inhomogeneous ice crystals

223 Figure 5 compares the scattering phase functions for homogeneous ice crystals
224 with their inhomogeneous ice crystal counterparts at wavelengths λ , 0.65 and 2.13 μm .
225 The refractive indices of ice at wavelengths 0.65 and 2.13 μm are $1.3080 + i1.43 \times 10^{-8}$
226 and $1.2673 + i5.57 \times 10^{-4}$, respectively. The ice crystals are assumed to be randomly
227 oriented hexagonal columns and plates with the aspect ratios, $2a/L=80 \mu\text{m}/100 \mu\text{m}$ and
228 $100 \mu\text{m}/43 \mu\text{m}$, respectively, where a is the radius of a cylinder that circumscribes the
229 hexagonal ice particle and L is the length of the ice particle. Specifically, Fig. 5a shows
230 the phase functions at $\lambda=0.65 \mu\text{m}$ for homogeneous hexagonal columns and
231 inhomogeneous columns with the same aspect ratio. For the two inhomogeneous
232 conditions, spherical air bubbles with radii of 16 or 34 μm are centered at the centers of
233 ice crystals. It is then evident from Fig. 5a that the air bubbles within ice crystals can
234 greatly affect the scattering properties of ice particles. In the homogeneous case, the
235 pronounced 22° and 46° halo peaks are quite pronounced as the typical features of the
236 phase functions for homogeneous hexagonal ice crystals. However, the magnitudes of the
237 peaks at the scattering angles 22° and 46° are reduced if small air bubbles with a radius of
238 16 μm is embedded in the crystals. For ice crystals containing relatively large air bubbles
239 with a radius of 34 μm , the pronounced 22° and 46° peaks are more significantly
240 smoothed out in the scattering phase function although they are still slightly noticeable.
241 Furthermore, the backscattering is substantially reduced in the inhomogeneous case. It
242 should be noted that a bubble embedded in ice acts as a diverging lens and affects internal
243 rays; however, the forward peaks are essentially unaffected by bubbles since diffraction
244 is the primary cause and depends primarily on the particle projected area. Figure 5b
245 shows the scattering phase functions for homogeneous and inhomogeneous hexagonal

246 columns at $\lambda=2.13 \mu\text{m}$. The effect of air bubbles at the near-infrared wavelength is
247 similar to that in the case for visible wavelengths. Figure 5c shows the scattering phase
248 functions of hexagonal plates at $\lambda=0.65 \mu\text{m}$. In this panel, the dotted line describes the
249 phase function for inhomogeneous ice crystals containing a spherical air bubble with a
250 radius of $21.25 \mu\text{m}$. For the other inhomogeneous case, four identical air bubbles are
251 aligned parallel to the basal faces of the plates. Similar to the effect in the hexagonal
252 columns, the trapped air bubbles in hexagonal plates smooth the scattering phase function
253 and reduces the back scattering. However, the scattering phase function is not sensitive to
254 the increase of the number of air bubbles in hexagonal plates. A similar effect of air
255 bubbles on the single-scattering properties is also seen in Fig. 5d for wavelength $2.13 \mu\text{m}$.

256 Figure 6 shows the degrees of linear polarization, $-p_{12}/p_{11}$, for ice crystals having
257 the same aspect ratios and inhomogeneity as in Fig. 5. Figures 6a and 6b compare the
258 degrees of linear polarization between homogeneous and inhomogeneous hexagonal
259 columns. It is seen that air bubbles embedded within ice crystals can also reduce the
260 magnitude of the degrees of linear polarization, particularly, in the case for large air
261 bubbles. The same effect can also be found for hexagonal plates, whose scattering phase
262 functions are shown in Figures 6c and 6d at $\lambda=0.65 \mu\text{m}$ and $\lambda=2.13 \mu\text{m}$, respectively.
263 However, unlike the performance of scattering phase functions in Fig. 5, increasing the
264 number of air bubbles in hexagonal plates may lead to more significant smoothing of the
265 degree of linear polarization.

266 Figure 7 shows the phase matrices for hexagonal columns at $\lambda=0.65 \mu\text{m}$. For one
267 of the inhomogeneous ice crystals, a spherical air bubble with radius of $21.25 \mu\text{m}$ is
268 included in a hexagonal column whose aspect ratio is $2a/L=50 \mu\text{m} / 100 \mu\text{m}$. To specify

269 the effect of the shapes of air bubbles on the single-scattering properties of ice crystals,
270 volume-equivalent spheroids are considered in the other type of ice crystals with the
271 same aspect ratio. It is evident from Fig. 7 that spherical air bubbles have a greater effect
272 on the phase matrix than those containing spheroidal air bubbles. This feature is
273 physically understandable since for the same volume, a spherical particle has a larger
274 cross section than a spheroid. Then the incident photon has a greater chance to be
275 intercepted by spherical air bubbles than their counterparts with other shapes. In addition
276 to the phase function and degree of linear polarization, the other elements of the phase
277 matrix are also sensitive to the presence of air bubbles.

278 To further illustrate the effect of air bubbles on the single-scattering properties of
279 ice crystals, Fig. 8 shows the asymmetry factor as a function of the volume of the air
280 bubbles at $\lambda=0.65 \mu\text{m}$ and $\lambda=2.13 \mu\text{m}$. The aspect ratio of ice crystals is $2a/L=10 \mu\text{m} /50$
281 μm . Spheroidal air bubbles are located in the center of the ice crystals where the r_1 and r_2
282 in Eq. (1) are both $4.25 \mu\text{m}$. The relative volume of the air bubble, V_b/V , can be specified
283 in terms of r_3 in Eq. (1), where V_b and V are the volumes of the air bubbles and ice
284 crystals, respectively. It is seen from Fig. 8 that the asymmetry factors decrease with
285 increasing V_b/V at both visible and near-infrared wavelengths.

286

287 **4. Effect of inhomogeneous ice crystals on ice cloud retrieval**

288 To study the effect of inhomogeneous ice crystals on retrieving ice cloud
289 properties, aspect ratios of ice crystals as well as particle size distributions in ice clouds
290 are required. In this sensitivity study, an aspect ratio of $2a/L=0.2$ is used for all ice
291 crystals, although it may not correspond well to observations [Ono, 1969]. Furthermore,

292 small ($r_1=0.45a$, $r_2=0.45a$, and $r_3=0.2L$) and relatively large ($r_1=0.85a$, $r_2=0.85a$, and
 293 $r_3=0.2L$) air bubbles are defined at the center of each inhomogeneous ice crystal. The size
 294 distribution of ice crystals is assumed to obey a Gamma distribution given by

$$295 \quad n(L) = N_0 L^\mu \exp\left(-\frac{b + \mu + 0.67}{L_m} L\right), \quad (19)$$

296 where N_0 is the intercept, μ is assumed to be 2 in this study, and L_m is the median of the
 297 distribution of L . The parameter b is taken to be 2.2. The effective particle size for a
 298 given size distribution is defined as follows [Foot, 1988]:

$$299 \quad r_e = \frac{3 \int_{L_{\min}}^{L_{\max}} V(L)n(L)dL}{4 \int_{L_{\min}}^{L_{\max}} A(L)n(L)dL}, \quad (20)$$

300 where V is particle volume, and A is projected area.

301 The ice cloud bi-directional reflectances are computed using the Discrete
 302 Ordinates Radiative Transfer (DISORT) model for $\lambda = 0.65$ and $2.13 \mu\text{m}$ at various
 303 incident-scattering configurations. The visible optical thickness at $\lambda = 0.65 \mu\text{m}$ serves as
 304 the reference optical thickness in this study. The optical thickness for a given wavelength
 305 is related to the visible optical thickness via

$$306 \quad \tau = \frac{\tau_{\text{vis}} Q}{Q_{\text{vis}}}, \quad (21)$$

307 where Q and Q_{vis} are the extinction efficiencies for $\lambda=2.13$ and $0.65 \mu\text{m}$, respectively.

308 Figure 9a shows the comparison of the lookup tables computed for the solid
 309 homogeneous ice crystals and the inhomogeneous ice crystals containing small air
 310 bubbles ($r_1= r_2=0.45a$, and $r_3=0.2L$). It is seen that the inhomogeneous ice crystals reflect
 311 slightly more than the homogeneous ice crystals at $\lambda = 0.65 \mu\text{m}$ whereas the bi-
 312 directional reflectances for the inhomogeneous ice crystals are significantly larger than

313 those for the homogeneous particles at $\lambda = 2.13 \mu\text{m}$. Figure 9b is the same as Fig. 9a
314 except that each inhomogeneous ice crystal in Fig. 9b contains bigger air bubbles with
315 radii of $r_1 = r_2 = 0.85a$, and $r_3 = 0.2L$. It is then evident that the bidirectional reflectances at λ
316 $= 0.65 \mu\text{m}$ are slightly sensitive to the air bubble size. However, large air bubbles in the
317 ice crystals can significantly increase the reflectances at $\lambda = 2.13 \mu\text{m}$.

318 The left and right panels in the top of Fig. 10 show a MODIS granule image over
319 the south Pacific Ocean on April 17, 2007 and the cloud mask from the operational
320 MODIS cloud product, respectively. The middle and bottom panels of Fig. 10 show the
321 retrieved cloud properties for the pixels that have been identified as covered by ice
322 clouds. Specifically, the middle panel on the left compares the retrieved ice cloud optical
323 thickness from homogeneous and inhomogeneous ice crystals. For the latter, small air
324 bubbles ($r_1 = r_2 = 0.45a$, and $r_3 = 0.2L$) are embedded. The middle panel on the right is the
325 same with the left panel except that the inhomogeneous ice crystals have larger air
326 bubbles ($r_1 = r_2 = 0.85a$, and $r_3 = 0.2L$). It is then evident that the cloud optical thicknesses
327 are slightly reduced by using inhomogeneous ice crystal models in ice cloud property
328 retrievals. These results are consistent with Fig. 9 where the inhomogeneous ice crystals
329 reflect more than homogeneous ice crystals at $\lambda = 0.65 \mu\text{m}$. The increase of the sizes of
330 air bubbles can further reduce the optical thickness as evident from the comparison of the
331 two middle panels in Fig. 10. Using inhomogeneous ice crystals in ice cloud models may
332 significantly increase the retrieved ice cloud effective particle sizes, as evident from the
333 bottom panels in Fig. 10. Moreover, this effect becomes more significant as sizes of the
334 air bubbles increase.

335 Figures 9 and 10 describe the sensitivities of ice cloud reflectance and cloud
336 property retrievals to optical properties of inhomogeneous ice crystals. In this study, the
337 same particle volumes and size distributions are employed for both homogeneous and
338 inhomogeneous ice crystals. However, containing air bubbles contained in ice crystals
339 will decrease the volume of ice and therefore decrease the effective particle size of ice
340 crystals in the ice cloud. Figure 11 shows the variations of effective particle sizes with the
341 volumes of the air bubbles within ice crystals. It is seen that the effective particle sizes of
342 ice clouds can be reduced to more than 50%, depending on the shape and size of the air
343 bubbles within ice crystals. Thus, the increased effective particle sizes resulting from a
344 retrieval employing inhomogeneous ice crystals in Fig. 10 can be partly compensated if
345 the volumes of the air bubbles are subtracted from the particle volumes.

346

347 **5. Summary**

348 This study reports on the single-scattering properties of inhomogeneous ice
349 crystals whose geometries are defined based on the observations made by *Tape* [1994] at
350 the South Pole. Unlike the spherical air bubbles with random locations in the IHM model
351 previously developed by *Labonnote et al.* [2001], in the present study a few spherical or
352 spheroidal air bubbles are defined at the center of hexagonal ice crystals. The sensitivity
353 of single-scattering properties to inhomogeneous ice crystals has been examined. It is
354 found that the single-scattering phase function is smoothed out and its peaks at the
355 scattering angles 22° and 46° are reduced if air bubbles are included in the ice crystals.
356 These features have been previously reported [*Labonnote et al.*, 2001; *Macke et al.*,
357 1996]. The phase function smoothing can become more pronounced by increasing the

358 number of air bubbles, enlarging the air bubbles, changing the air bubbles' shapes from
359 spheroids to spheres, or moving them from the sides to the center of an ice crystal. The
360 peaks of the degrees of linear polarization can also be reduced by considering
361 inhomogeneous ice crystals. Moreover, the asymmetry factors of inhomogeneous ice
362 crystals decrease as the relative volume of the air bubbles increases.

363 Furthermore, a lookup library of bidirectional reflectances has been developed for
364 both homogenous and homogenous ice cloud models at $\lambda = 0.65$ and $2.13 \mu\text{m}$. We
365 showed that using inhomogeneous ice cloud models can increase the bidirectional
366 reflectances at those two wavelengths. Therefore, the retrieved ice cloud optical
367 thicknesses are slightly reduced whereas the retrieved ice cloud effective particle sizes
368 can be significantly increased by including air bubbles in ice crystals, particularly, in the
369 case of large air bubbles. This effect is similar to that found when surface roughness is
370 included in the computations of ice crystal single-scattering properties [Yang *et al.*,
371 2008a,b], except that the presence of air bubbles in the crystals reduces the overall ice
372 water content compared to a solid crystal with roughened surfaces. These results
373 represent another important step in the effort to develop realistic ice crystal optical
374 properties for use in retrieving ice cloud properties from satellite imagery and
375 representing them in numerical weather and climate models. The results appear to be in
376 the right direction for decreasing the biases in retrieved ice cloud optical properties, .e.g.,
377 *Min et al.* [2004]. Additional study will be needed, however, to determine if the optical
378 properties of spheroidal bubbles, either alone or in combination with those for other ice
379 crystal formulations, can provide a more accurate representation of actual ice crystal
380 reflectance behavior.

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382

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391 **References**

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393 Baran, A. J. (2004), On the scattering and absorption properties of cirrus cloud, *J. Quant.*
394 *Spectrosc. Radiat. Transfer*, 89, 17-36.

395 Borovoi, A. G., and I. A. Grishin (2003), Scattering matrices for large ice crystal
396 particles, *J. Opt. Soc. Am. A.*, 20, 2071-2080.

397 Carte, A. E. (1961), Air bubbles in ice, *Proc. Phys. Soc.*, 77, 757-768.

398 Foot, J. S. (1988), Some observations of the optical properties of clouds. II: Cirrus, *Q. J.*
399 *R. Meteorol. Soc.*, 114, 145-169.

400 Hallett, J. (1964), Experimental studies of the crystallization of supercooled water, *J.*
401 *Atmos. Sci.*, 21, 671-682.

402 Heymsfield, A. J. (1977), Precipitation development in stratiform ice clouds: A
403 microphysical and dynamical study, *J. Atmos. Sci.*, 34, 367-381.

404 Heymsfield, A. J and C. M. R. Platt (1984), A parameterization of the particle size
405 spectrum of ice clouds in terms of the ambient temperature and the ice water
406 content, *J. Atmos. Sci.*, 41, 846-855.

407 Iaquina, J., H. Isaka, and P. Personne (1995), Scattering phase function of bullet rosette
408 ice crystals, *J. Atmos. Sci.*, 52, 1401-1413.

409 Knap, W. H., L. C. Labonnote, G. Brogniez, and P. Stammes (2005), Modeling total and
410 polarized reflectances of ice clouds: evaluation by means of POLDER and ATSR-
411 2 measurements, *Appl. Opt.*, 44, 4060-4073.

412 Korolev, A. V., G. A. Isaac, and J. Hallett (1999), Ice particle habits in Arctic clouds,
413 *Geophys. Res. L.*, 26, 1299-1302.

414 Labonnote, L. C., G. Brogniez, J. C. Buriez, and M. Doutriaux-Boucher (2001), Polarized
415 light scattering by inhomogeneous hexagonal monocrystals: validation with
416 ADEOS-POLDER measurements, *J. Geophys. Res.*, 106, 12139-12155.

417 Liou, K. N. (1972), Light scattering by ice clouds in the visible and infrared: a theoretical
418 study, *J. Atmos. Sci.*, 29, 524-536.

419 Liou, K. N. (1986), Influence of cirrus clouds on weather and climate processes: A global
420 perspective, *Mon. Wea. Rev.*, 114, 1167-1199.

421 Mace, G. G., Y. Zhang, S. Platnick, M. D. King, P. Minnis, and P. Yang (2005),
422 Evaluation of cirrus cloud properties from MODIS radiances using cloud
423 properties derived from ground-based data collected at the ARM SGP site, *J.*
424 *Appl. Meteorol.*, 44, 221-240.

425 Macke, A. (1993), Scattering of light by polyhedral ice crystals, *Appl. Opt.*, 32, 2780-
426 2788.

427 Macke, A., M. I. Mishchenko, and B. Cairns (1996), The influence of inclusions on light
428 scattering by large ice particles, *J. Geophys. Res.*, 101, 23311-23316.

429 Macke, A. J. Mueller, and E. Raschke (1996), Single scattering properties of atmospheric
430 ice crystal, *J. Atmos. Sci.*, 53, 2813-2825.

431 McFarquhar, G. M. and A. J. Heymsfield (1996), Microphysical characteristics of three
432 anvils sampled during the Central Equatorial Experiment, *J. Atmos. Sci.*, 53,
433 2401-2423.

434 Min, Q, P. Minnis, and M. M. Khaiyer (2004), Comparison of cirrus optical depths from
435 GOES-8 and surface measurements, *J. Geophys. Res.*, 109, D20119,
436 10.1029/2003JD004390.

437 Minnis, P., D. P. Garber, D. F. Young, R. F. Arduini, and Y. Takano (1998),
438 Parameterization of reflectance and effective emittance for satellite remote
439 sensing of cloud properties, *J. Atmos. Sci.*, 55, 3313-3339.

440 Minnis, P., P. W. Heck, and D. F. Young (1993), Inference of cirrus cloud properties
441 using satellite-observed visible and infrared radiances, Part II: Verification of
442 theoretical cirrus radiative properties, *J. Atmos. Sci.*, 50, 1305-1322.

443 Mishchenko, M. I., J. W. Hovenier and L. D. Travis (2000), *Light scattering by*
444 *nonspherical particles*, Academic press, San Diego, San Francisco, New York,
445 Boston, London, Sydney, Tokyo.

446 Muinonen, K. (1989), Scattering of light by crystals: a modified Kirchhoff
447 approximation, *Appl. Opt.*, 28, 3044-3050.

448 Nakajima T. and M. D. King (1990), Determination of the optical thickness and effective
449 particle radius of clouds from reflected solar radiation measurements. Part I:
450 Theory, *J. Atmos. Sci.*, 47, 1878-1893.

451 Ono, A. (1969), The shape and riming properties of ice crystals in natural clouds, *J.*
452 *Atmos. Sci.*, 26, 138-147.

453 Ramanathan, V., E. J. Pitcher, R. C. Malone, and M. L. Blackmon (1983), The response
454 of a general circulation model to refinements in radiative processes, *J. Atmos.*
455 *Sci.*, 40, 605-630.

456 Schmitt, C. G., J. Iaquinta, and A. J. Heymsfield (2006), The asymmetry parameter of
457 cirrus clouds composed of hollow bullet rosette-shaped ice crystals from ray-
458 tracing calculations. *J. Appl. Meteor. and Climat.*, 45, 973-981.

459 Takano, Y and K. N. Liou (1989), Solar radiative transfer in cirrus clouds. Part I: single-
460 scattering and optical properties of hexagonal ice crystals, *J. Atmos. Sci.*, 46, 3-19.

461 Takano, Y. and K.N. Liou (1995), Radiative transfer in cirrus clouds. III. Light scattering
462 by irregular ice crystals, *J. Atmos. Sci.*, 52, 818-837.

463 Tape, W. (1983), Some ice crystals that made halos, *J. Opt. Soc. Am.*, 73, 1641-1645.

464 Tape, W. (1994), *Atmospheric halos*, American Geophysical Union, Washington, D. C.,
465 USA.

466 Um, J., and G. M. McFarquhar (2007), Single-Scattering Properties of Aggregates of
467 Bullet Rosettes in Cirrus, *J. Appl. Meteor. and Climatology*, 46, 757-775.

468 Yang P., G. Hong, G. W. Kattawar, P. Minnis and Y. X. Hu (2008a), Uncertainties
469 associated with the surface texture of ice particles in satellite-based retrieval of
470 cirrus clouds: Part II. Effect of particle surface roughness on retrieved cloud
471 optical thickness and effective particle size, *Geo. Remote. Sens.*, 46, 1948-1957.

472 Yang, P., G. W. Kattawar, G. Hong, P. Minnis and Y. X. Hu (2008b), Uncertainties
473 associated with the surface texture of ice particles in satellite-based retrieval of
474 cirrus clouds: Part I. Single-scattering properties of ice crystals with surface
475 roughness, *Geo. Remote. Sens.*, 46, 1940-1947.

476 Yang, P and K. N. Liou (1995), Light scattering by hexagonal ice crystals: comparison of
477 finite-difference time domain and geometric optics models, *J. Opt. Soc. Am.*, A12,
478 162-176.

479 Yang, P. and K. N. Liou (1996), Finite-difference time domain method for light
480 scattering by small ice crystals in three-dimensional space, *J. Opt. Soc. Amer.*,
481 A13, 2072-2085.

482 Yang, P., and K. N. Liou (1998), Single-scattering properties of complex ice crystals in
483 terrestrial atmosphere, *Contr. Atmos. Phys.*, 71, 223-248.

484 Yang, P., K. N. Liou, M. I. Mishchenko, and B. C. Gao (2000), An efficient finite-
485 difference time domain scheme for light scattering by dielectric particles:
486 application to aerosols, *Appl. Opt.*, 39, 3727-3737.

487 Yang, P., Z. B. Zhang, G. W. Kattawar, S. G. Warren, B. A. Baum, H. L. Huang, Y. X.
488 Hu, D. Winker, and J. Iaquinta (in press), Effect of cavities on the optical
489 properties of bullet rosettes: Implications for active and passive remote sensing of
490 ice cloud properties, *J. Appl. Meteorol. Climatol.*.

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492 **Figure Captions**

493 Fig. 1. Inhomogeneous ice crystals sampled by Walter Tape [Tape, 1994] at the South
494 Pole, on January 19, 1985 (left) and January 17, 1986 (right).

495 Fig. 2. The geometries of inhomogeneous ice crystals.

496 Fig. 3 Geometry of a hexagonal ice crystal with an air bubble inside.

497 Fig. 4. Schematic flow-chart for reflection and refraction by internal air bubbles.

498 Fig. 5. Scattering phase functions for homogeneous and inhomogeneous ice crystals at
499 $\lambda=0.65 \mu\text{m}$ (panels a and c) and $2.13 \mu\text{m}$ (panels b and d).

500 Fig. 6. Degrees of linear polarization for homogeneous and inhomogeneous ice crystals at
501 $\lambda=0.65 \mu\text{m}$ (panels a and c) and $2.13 \mu\text{m}$ (panels b and d). The ice crystals' sizes
502 and morphologies in this figure are the same as those in Fig. 5.

503 Fig. 7. Scattering phase matrixes for homogeneous and inhomogeneous ice crystals at
504 $\lambda=0.65 \mu\text{m}$.

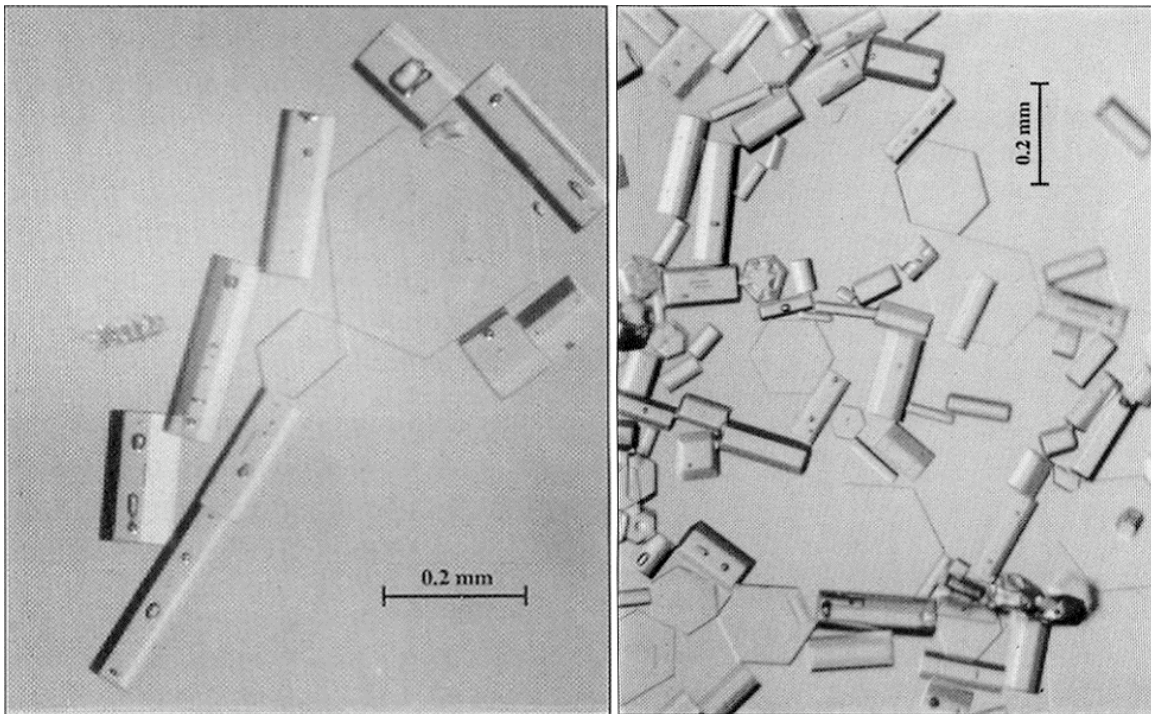
505 Fig. 8. Asymmetry factors for inhomogeneous ice crystals at $\lambda=0.65 \mu\text{m}$ (left) and 2.13
506 μm (right).

507 Fig. 9. Lookup tables using 0.65 and $2.13 \mu\text{m}$ reflectances for homogeneous and
508 inhomogeneous cloud models. $\mu_0=0.65$, $\mu=1.0$ and $\varphi - \varphi_0 = 0^\circ$.

509 Fig. 10. MODIS granule image (RGB=band 4:3:1) from Terra on April 17, 2007, and
510 MODIS cloud mask (upper panels). The comparisons of retrieved ice cloud
511 optical thicknesses from homogeneous and inhomogeneous ice crystals (middle
512 panels). The comparisons of retrieved ice cloud effective particle sizes from
513 homogeneous and inhomogeneous ice crystals (bottom panels).

514 Fig. 11. Effective particle sizes for inhomogeneous ice crystals.

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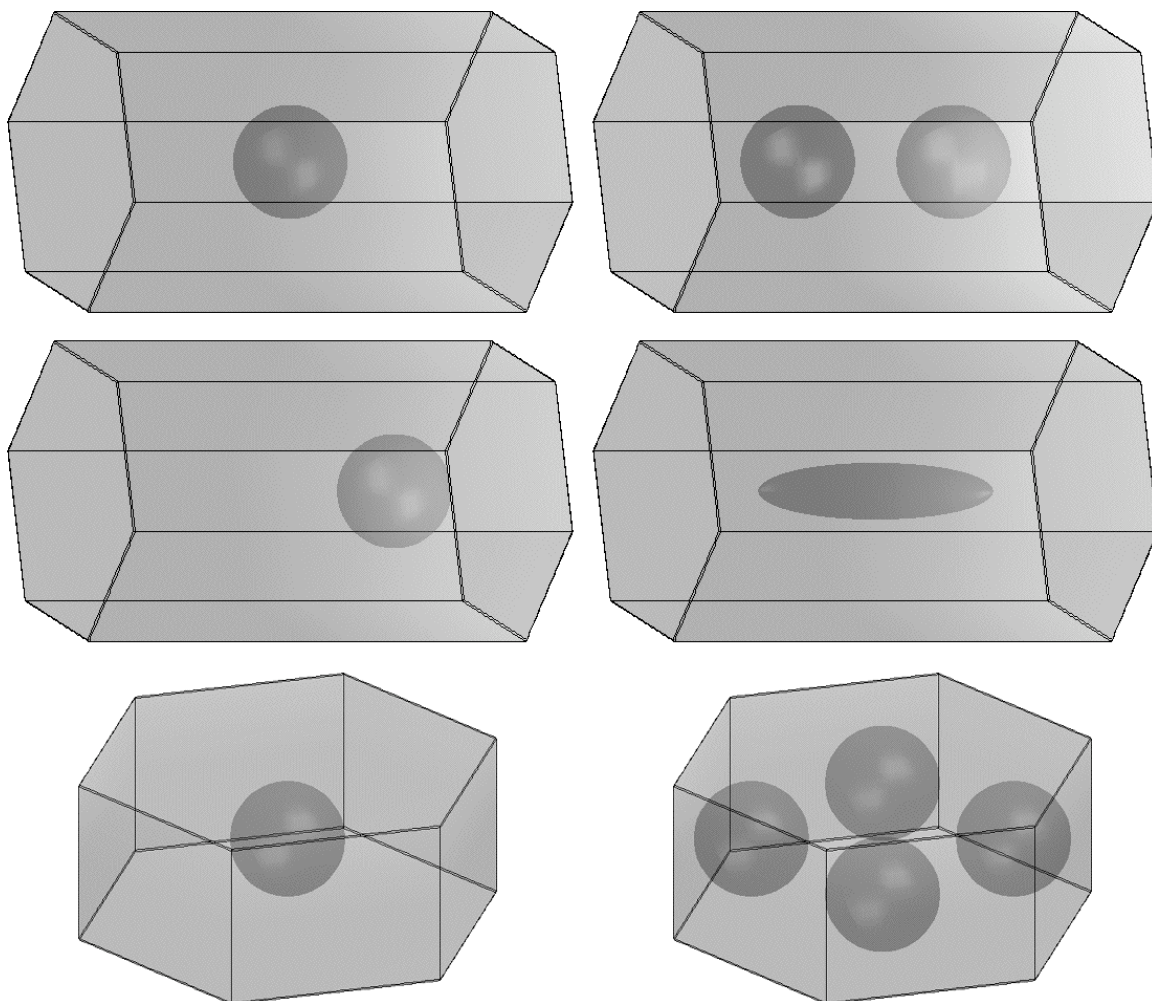
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536 Fig. 2. The geometries of inhomogenous ice crystals.

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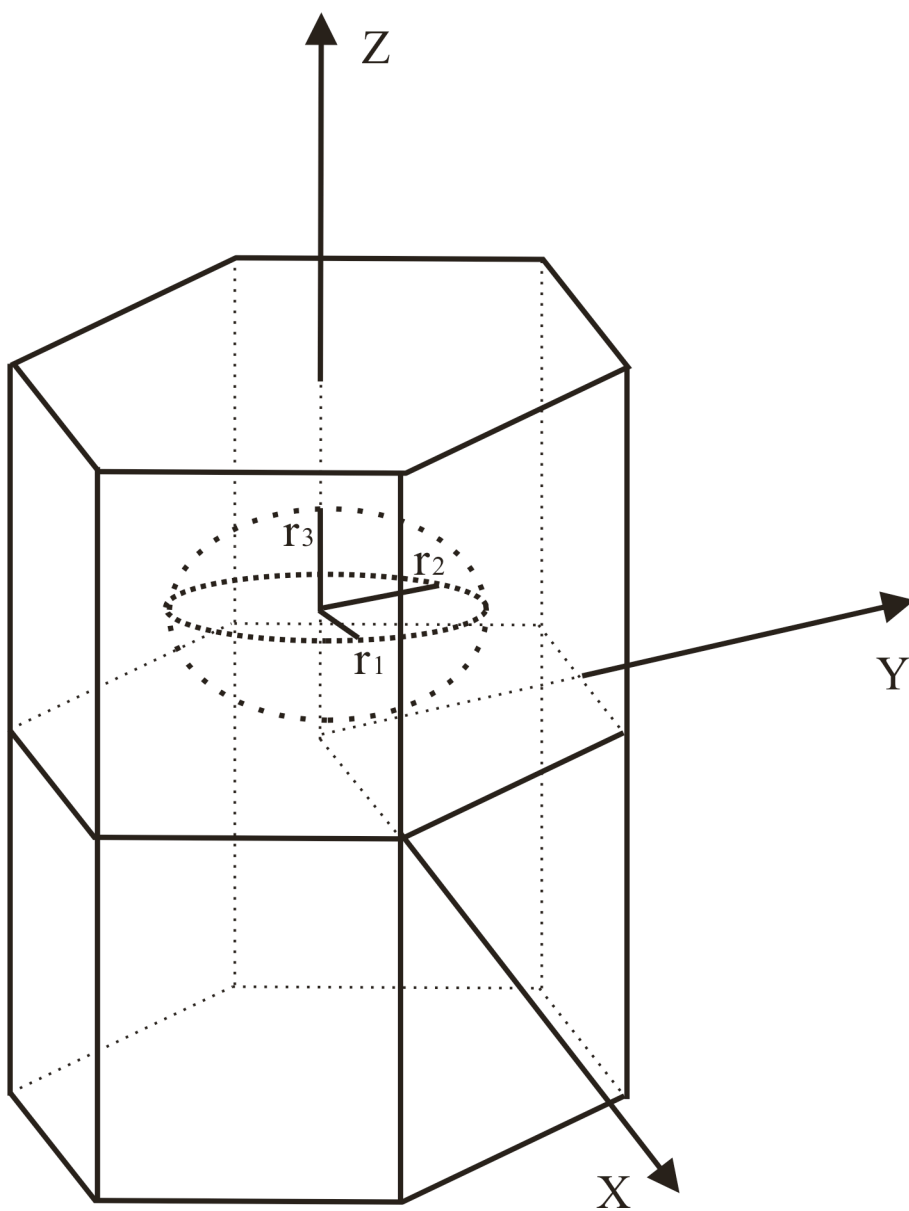
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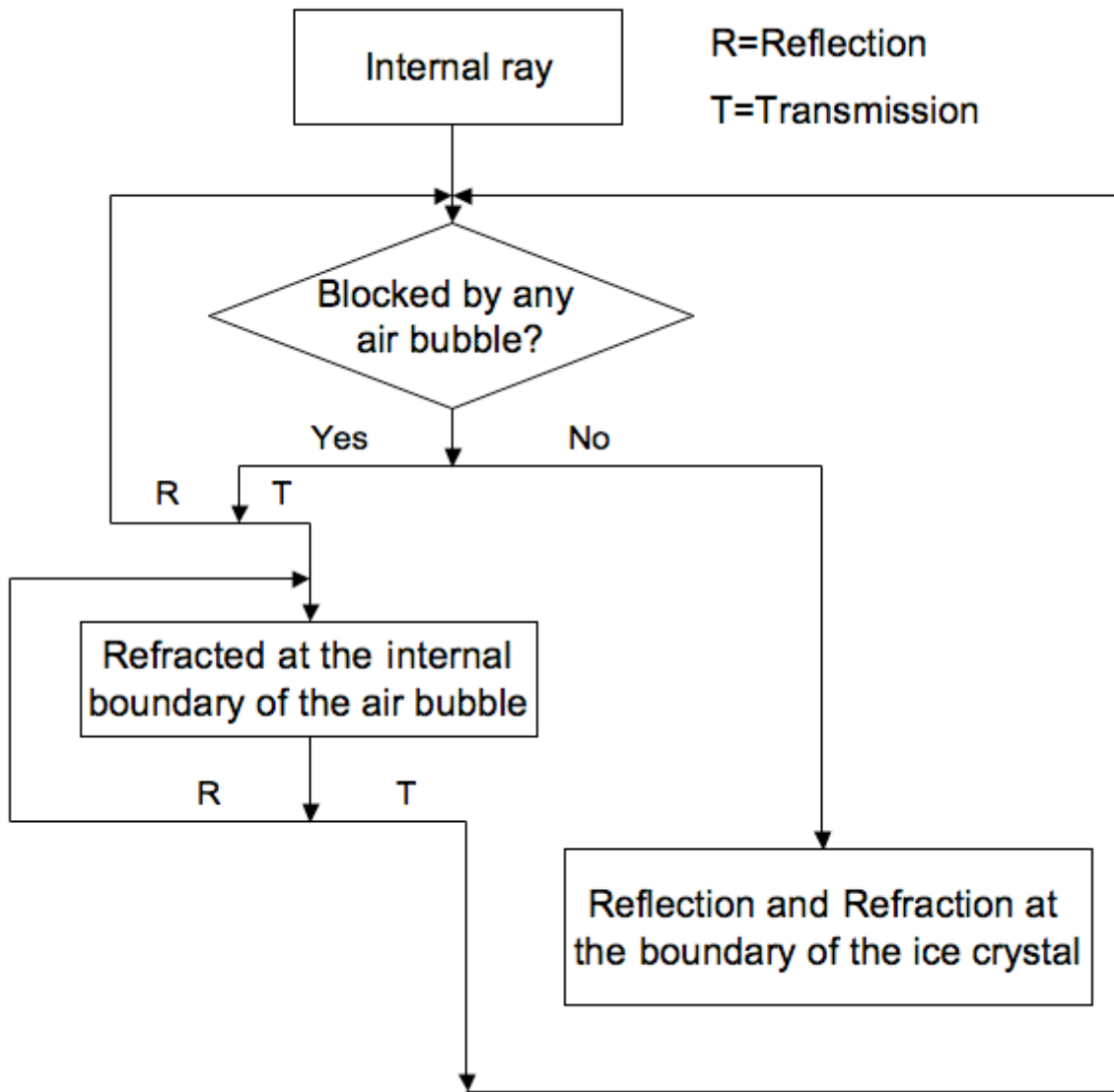
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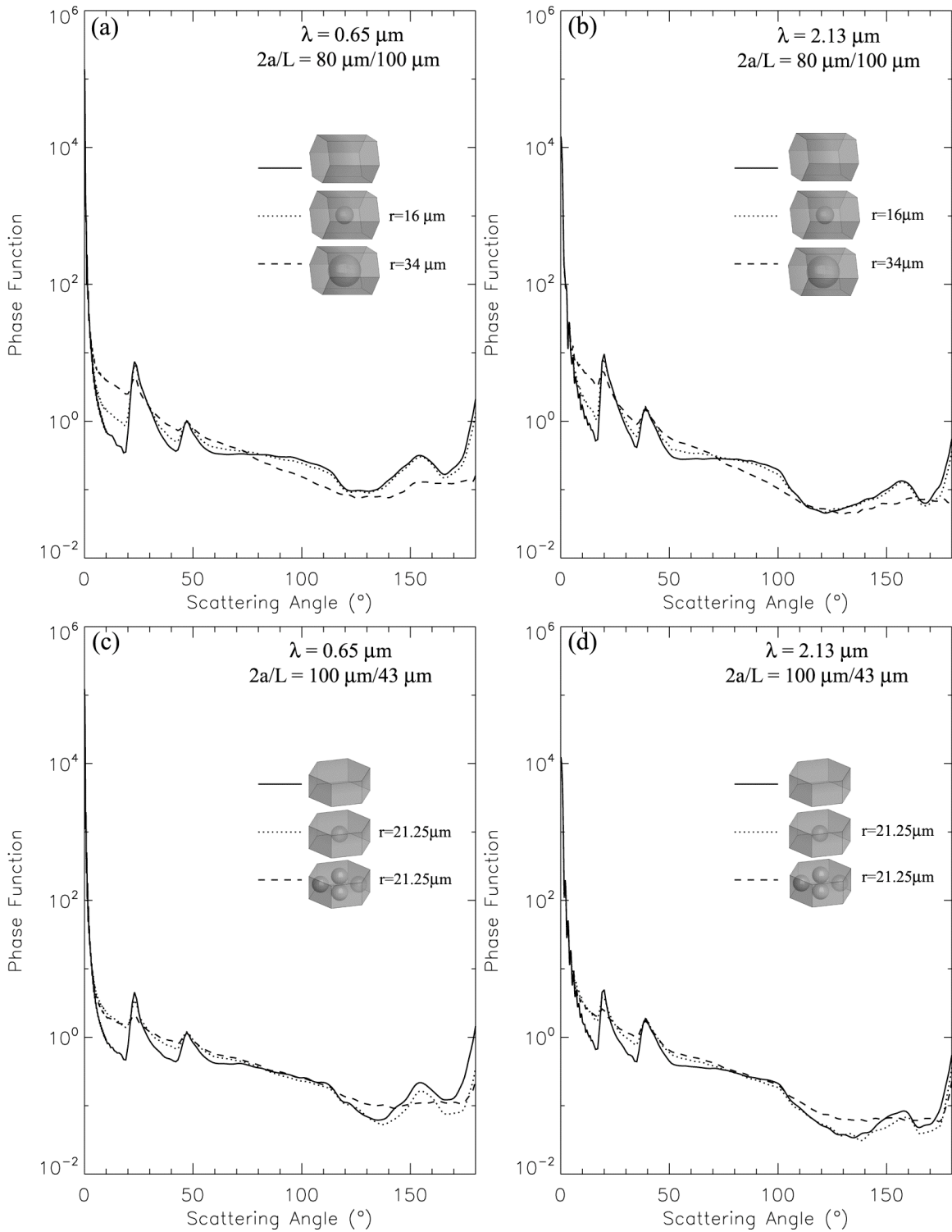
546 Fig. 3 Geometry of a hexagonal ice crystal with an air bubble inside.

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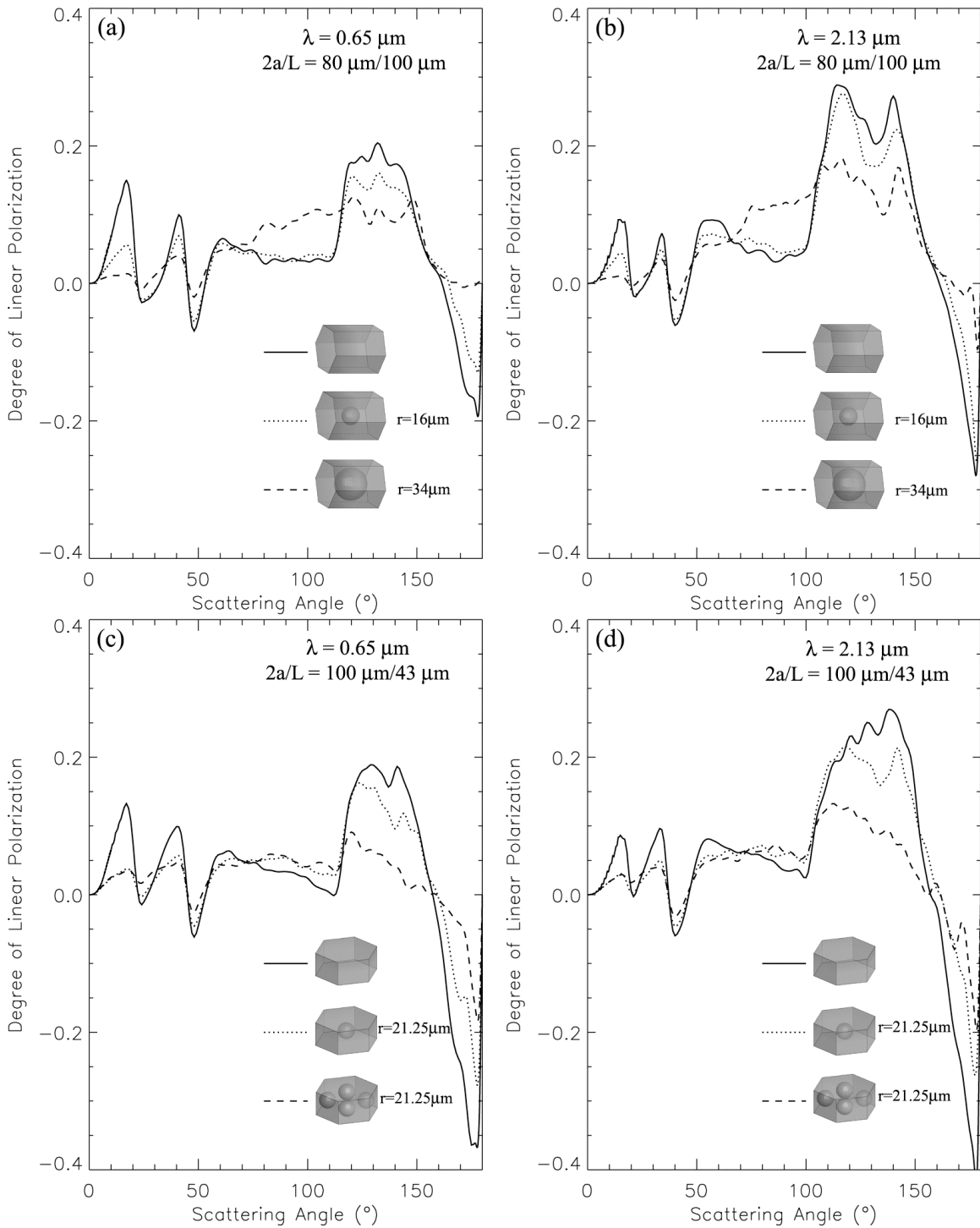
Fig. 4. Schematic flow-chart for reflection and refraction by internal air bubbles.



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557 Fig. 5. Scattering phase functions for homogeneous and inhomogeneous ice crystals at

558 $\lambda=0.65 \mu\text{m}$ (panels a and c) and $2.13 \mu\text{m}$ (panels b and d).

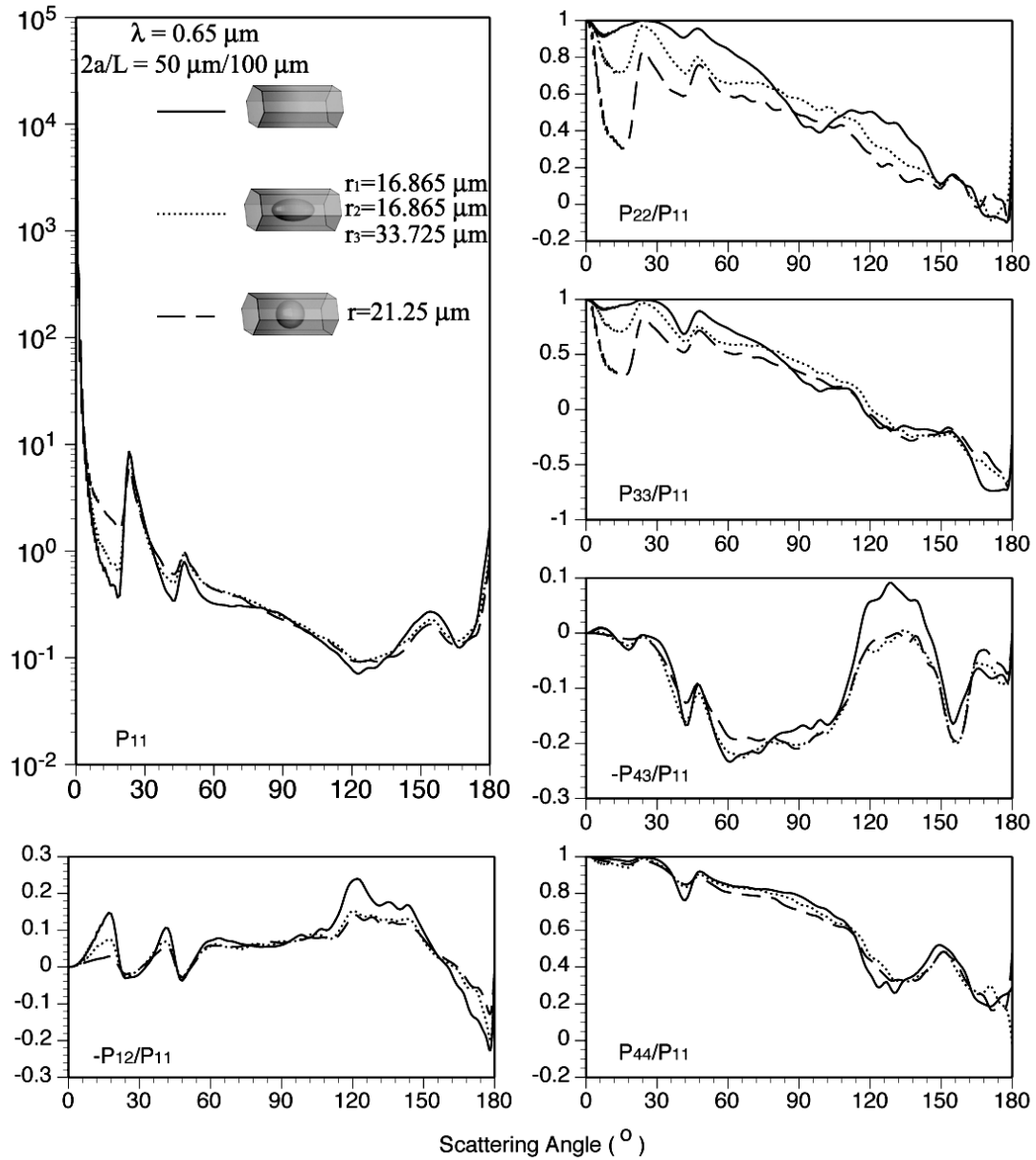


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560 Fig. 6. Degrees of linear polarization for homogeneous and inhomogeneous ice crystals at

561 $\lambda=0.65 \mu\text{m}$ (panels a and c) and $2.13 \mu\text{m}$ (panels b and d). The ice crystals' sizes and

562 morphologies in this figure are the same as those in Fig. 5.



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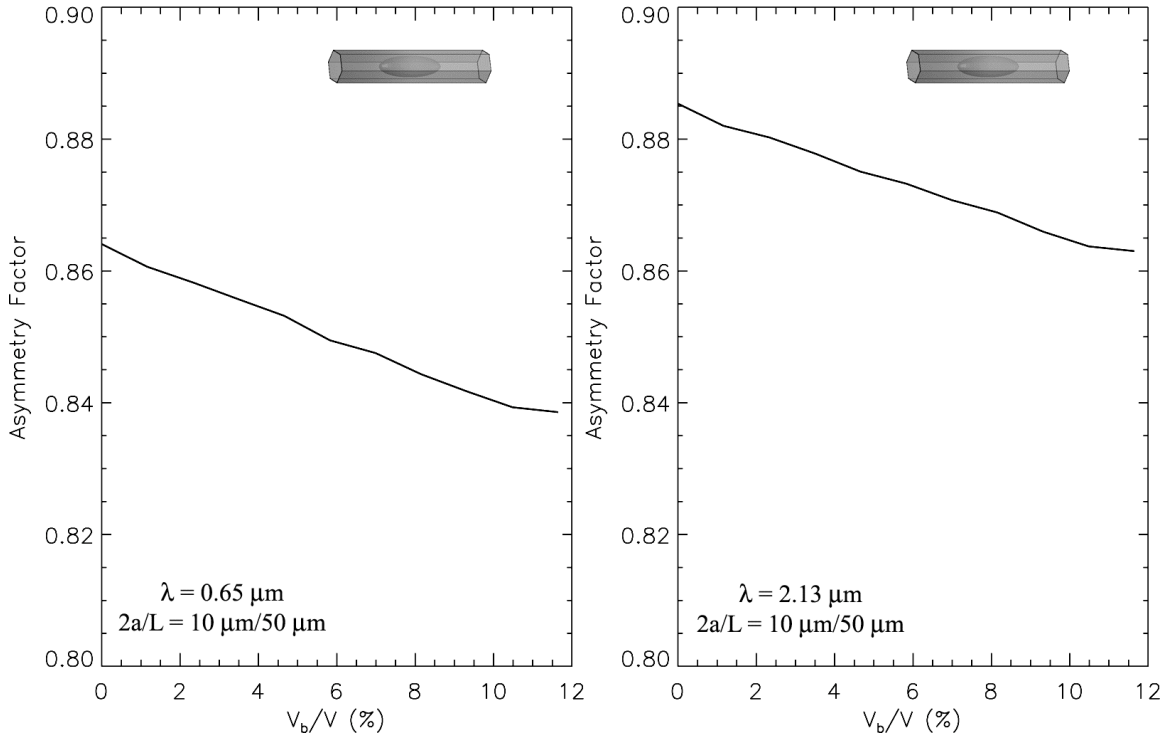
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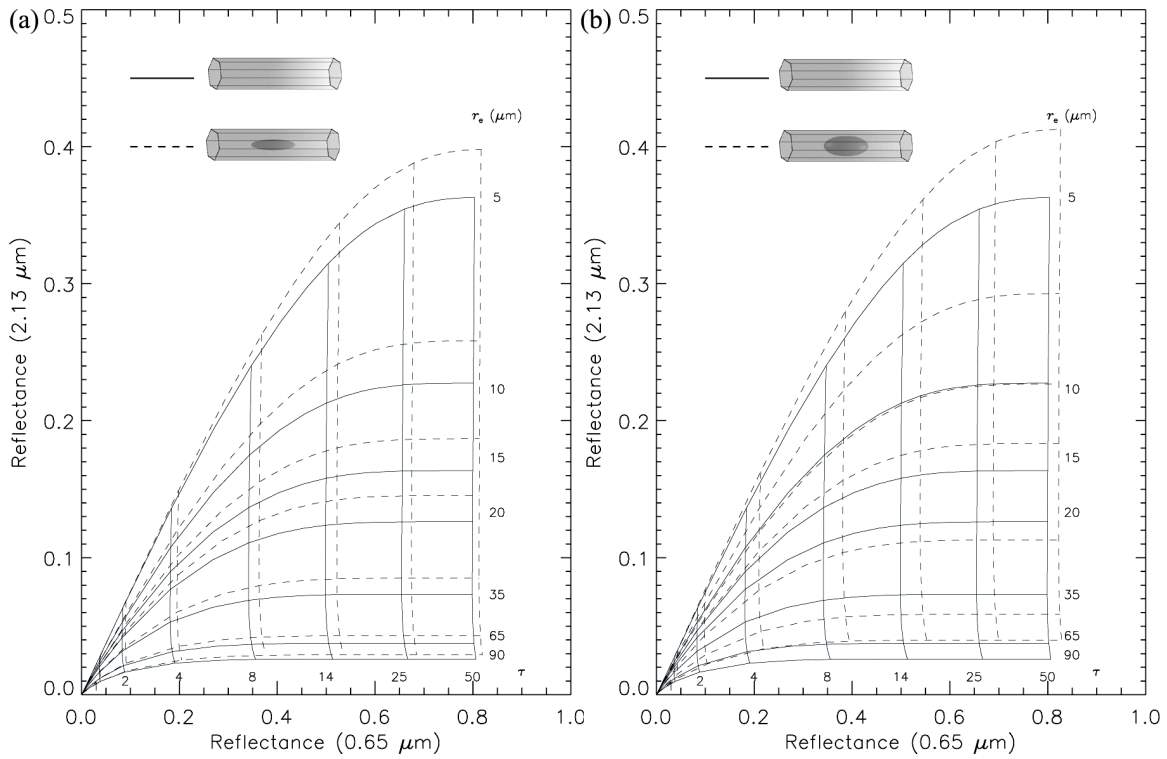
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585 Fig. 9. Lookup tables using 0.65 and 2.13 μm reflectances for homogeneous and
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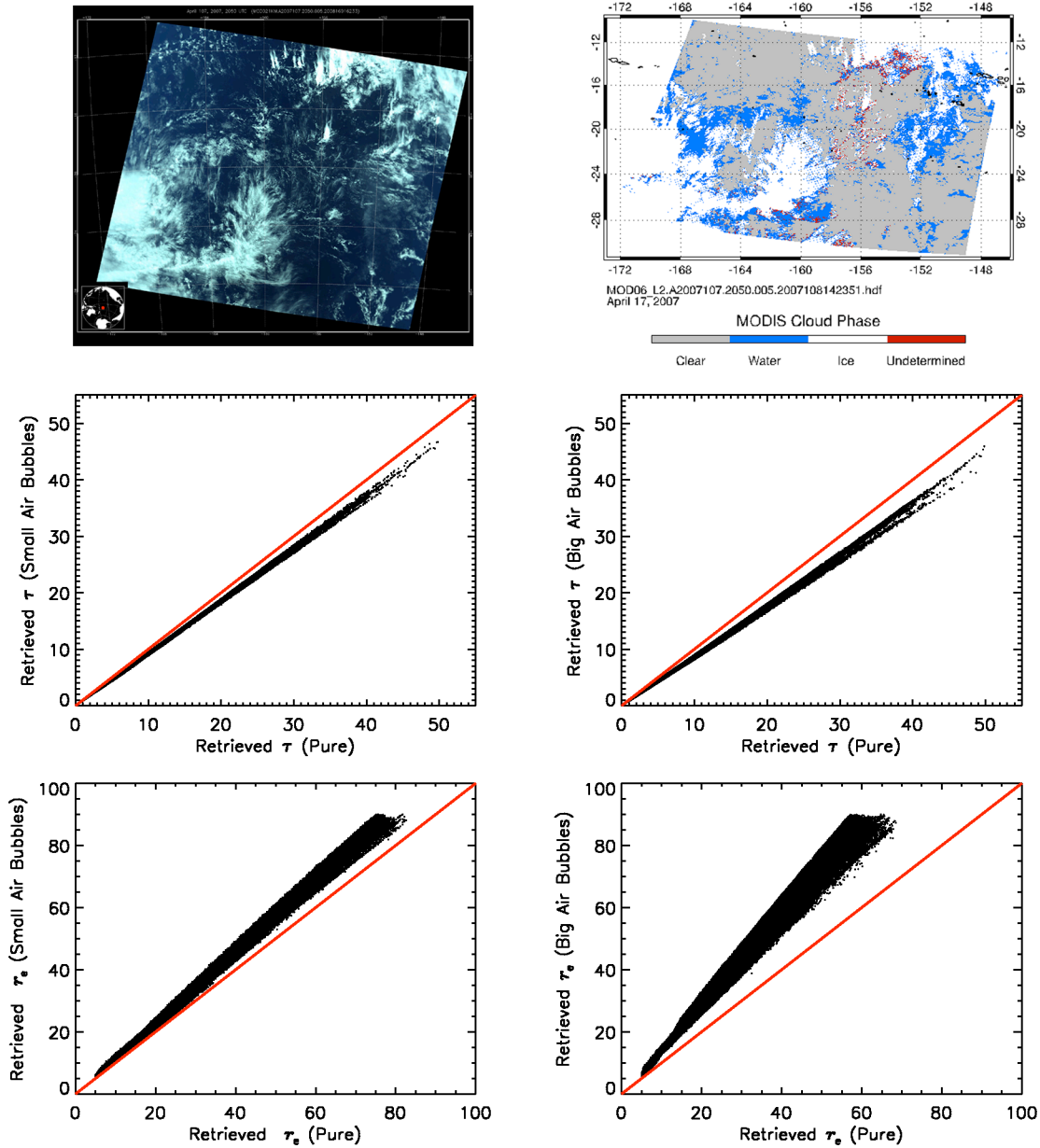
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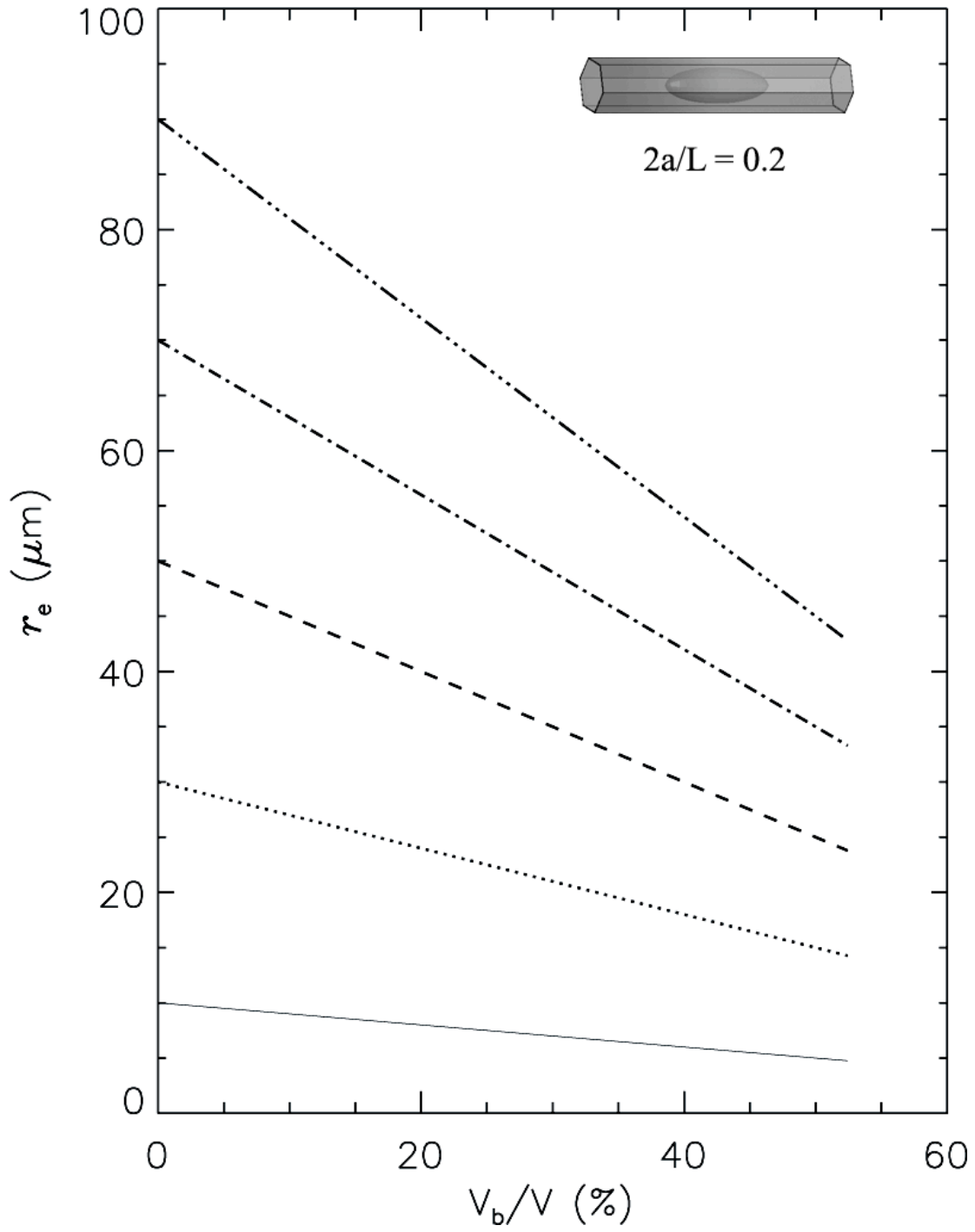
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600 thicknesses from homogeneous and inhomogeneous ice crystals (middle panels). The

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