

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 μm and 2.13 μm 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 μm. Thus, the retrieved cloud optical thicknesses are reduced by employing inhomogeneous ice cloud models. At a wavelength of 2.13 μm, 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

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

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

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

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  $\lambda$ , 0.65 and 2.13  $\mu\text{m}$ .  
225 The refractive indices of ice at wavelengths 0.65 and 2.13  $\mu\text{m}$  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  $\mu\text{m}$  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  $22^\circ$  and  $46^\circ$  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  $22^\circ$  and  $46^\circ$  are reduced if small air bubbles with a radius of  
238 16  $\mu\text{m}$  is embedded in the crystals. For ice crystals containing relatively large air bubbles  
239 with a radius of 34  $\mu\text{m}$ , the pronounced  $22^\circ$  and  $46^\circ$  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  $\mu\text{m}$ . 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,  $\mu$  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  $Q_{\text{vis}}$  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  $\lambda$   
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  $22^\circ$  and  $46^\circ$  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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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  $\mu\text{m}$  (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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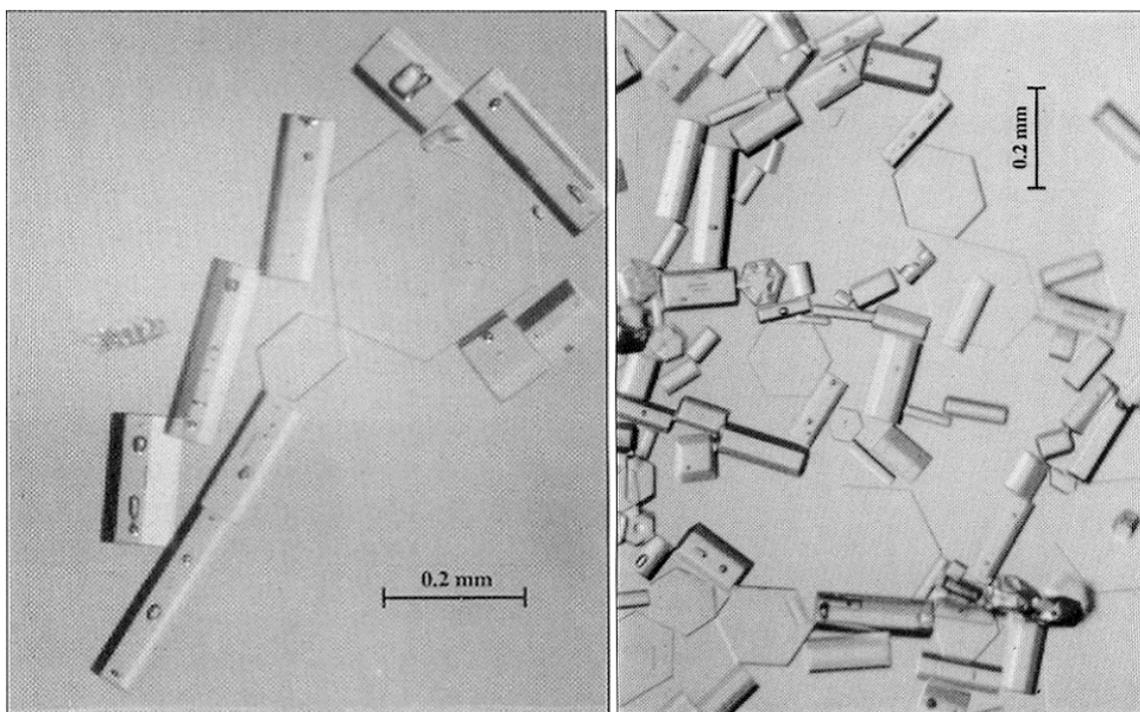
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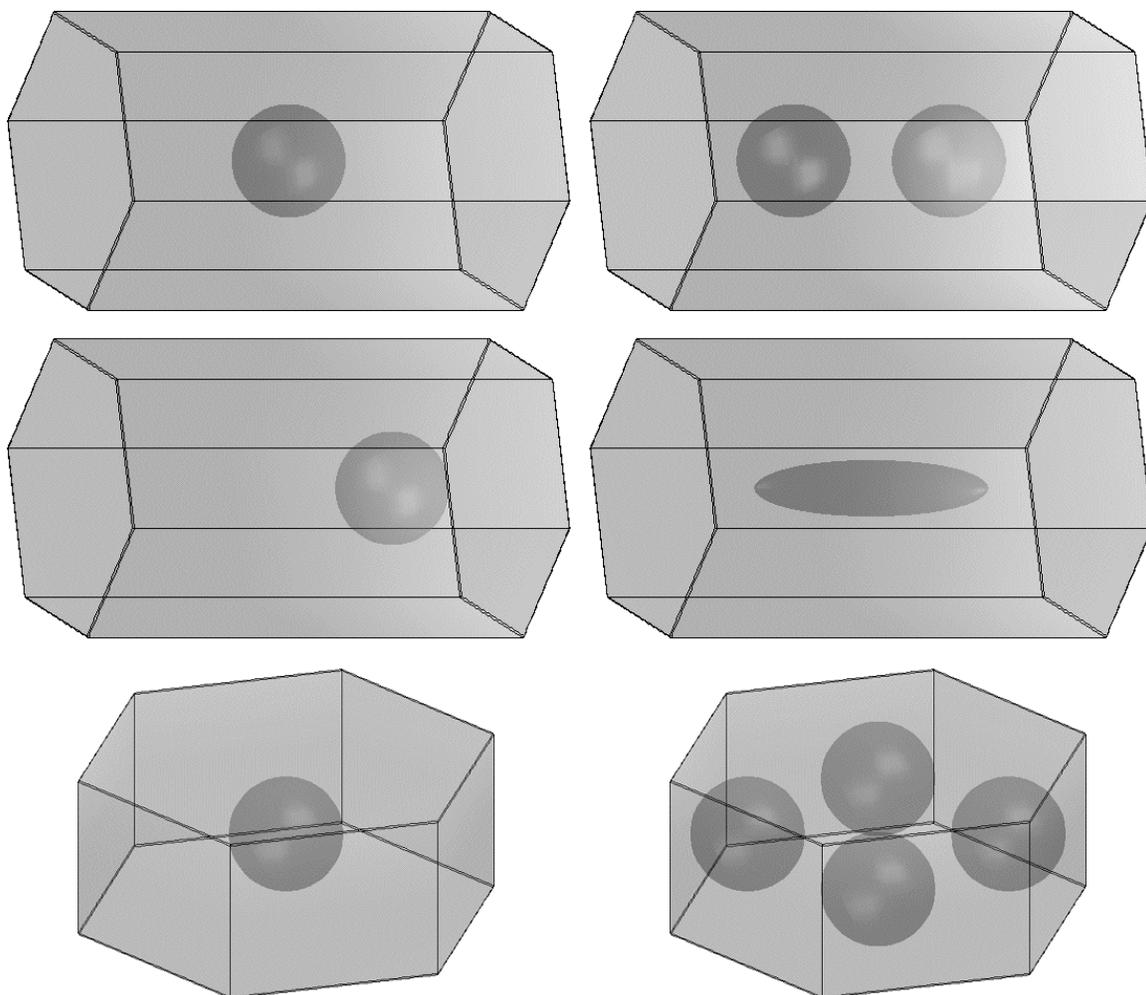
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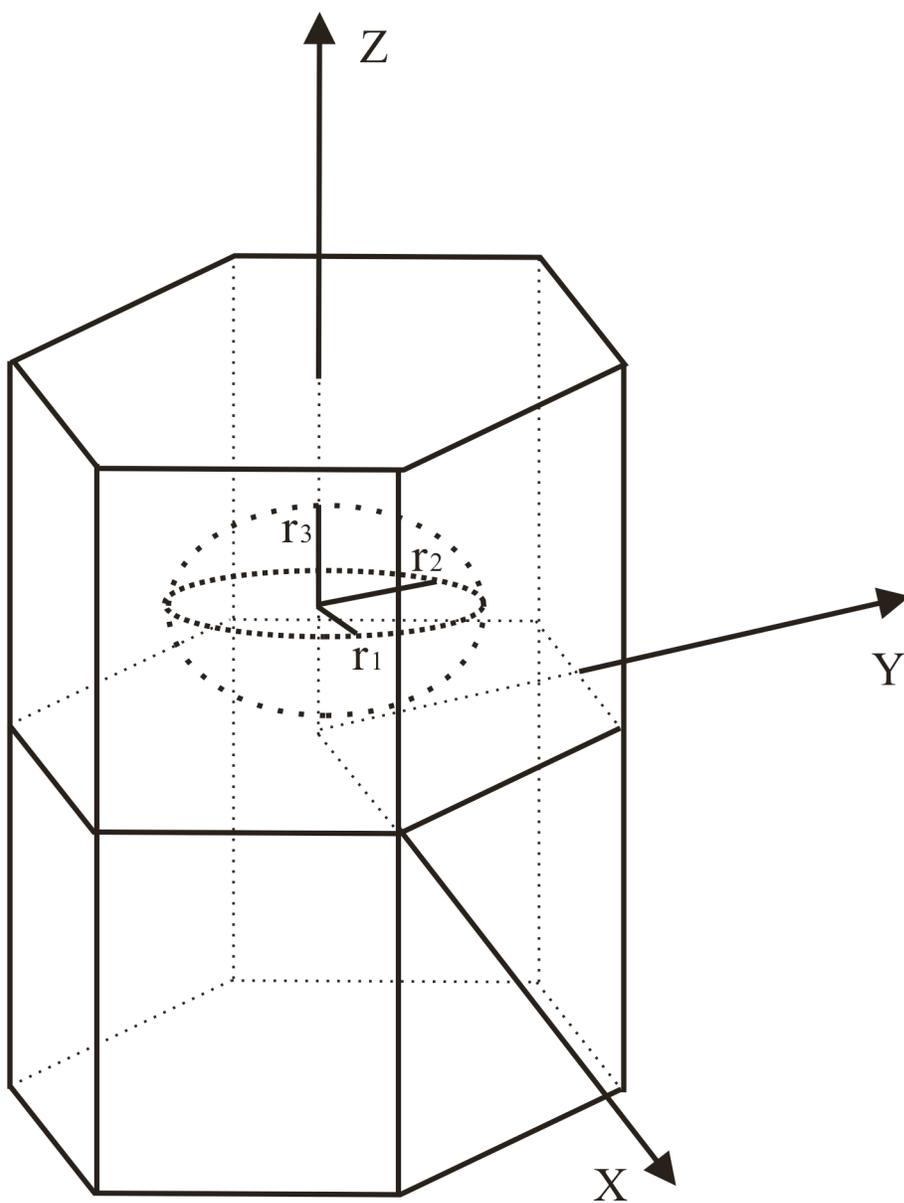
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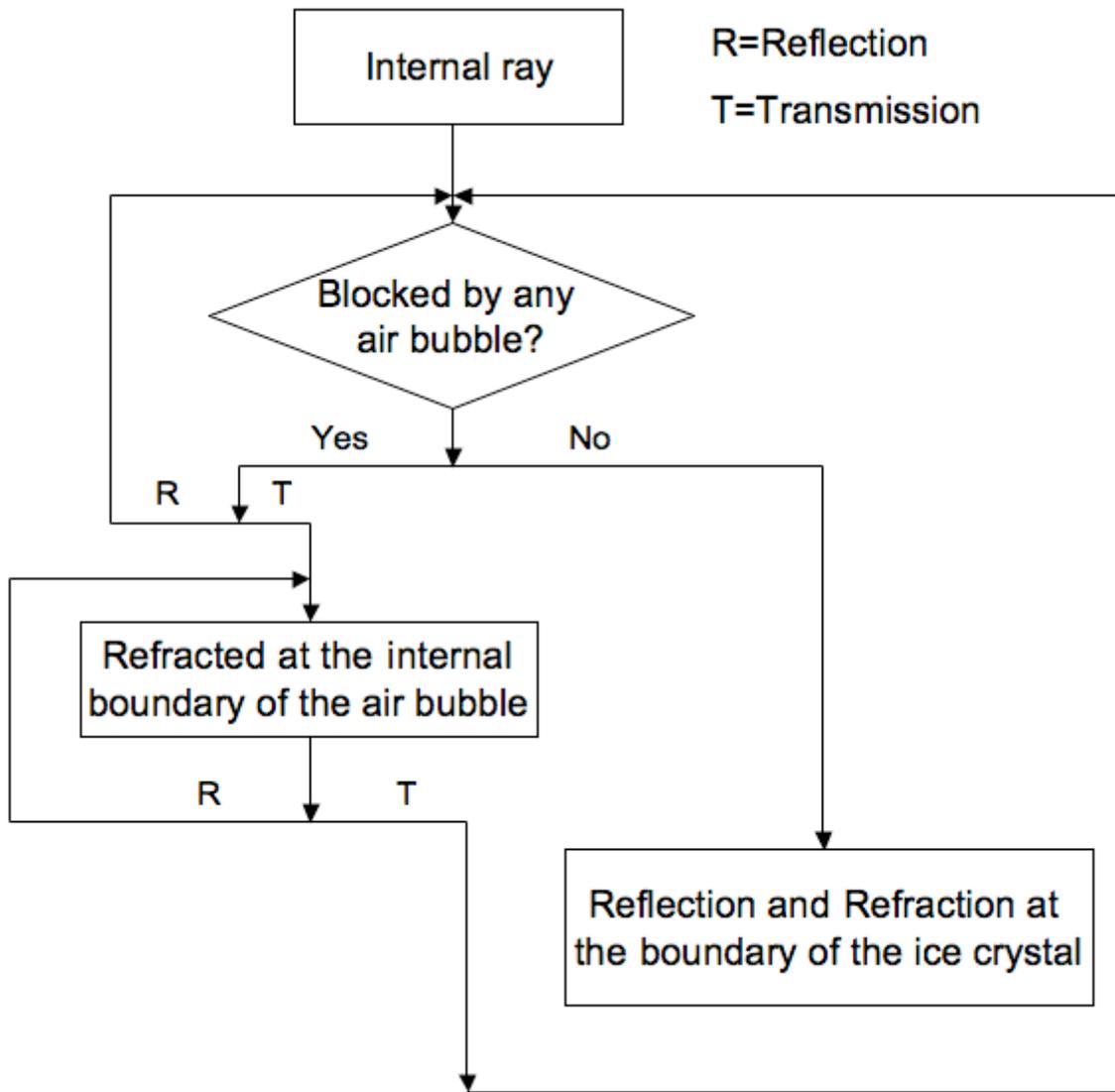
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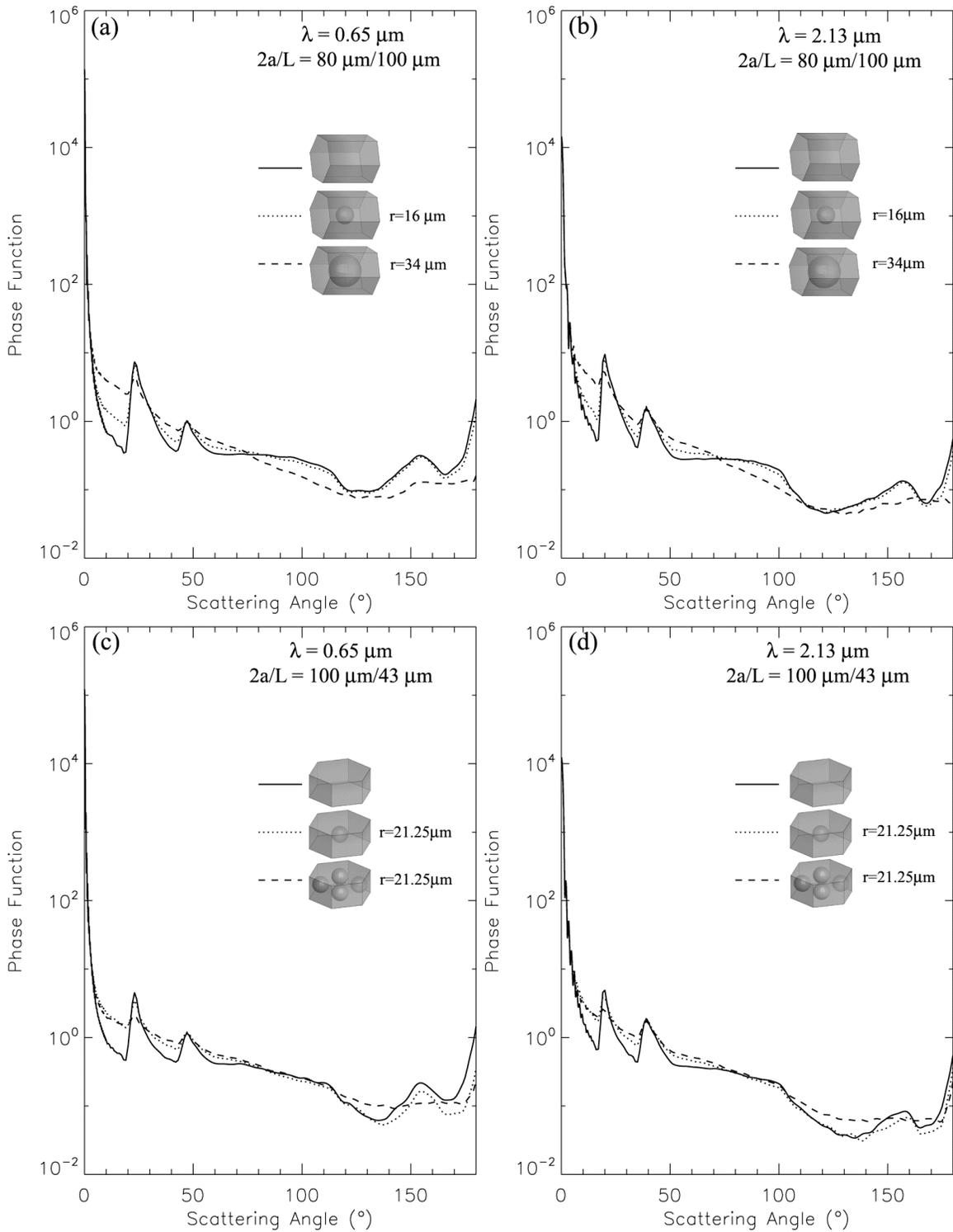
546 Fig. 3 Geometry of a hexagonal ice crystal with an air bubble inside.

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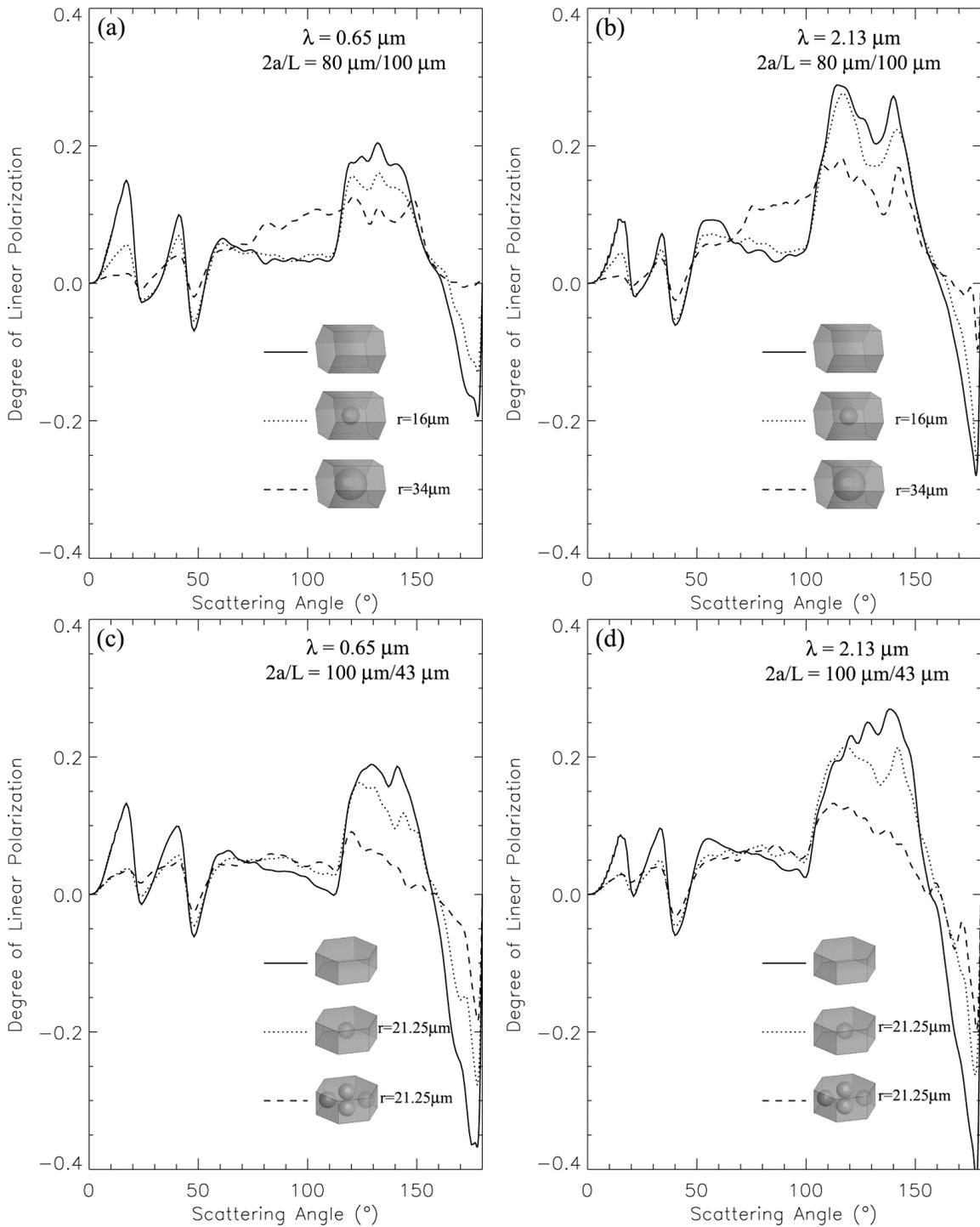
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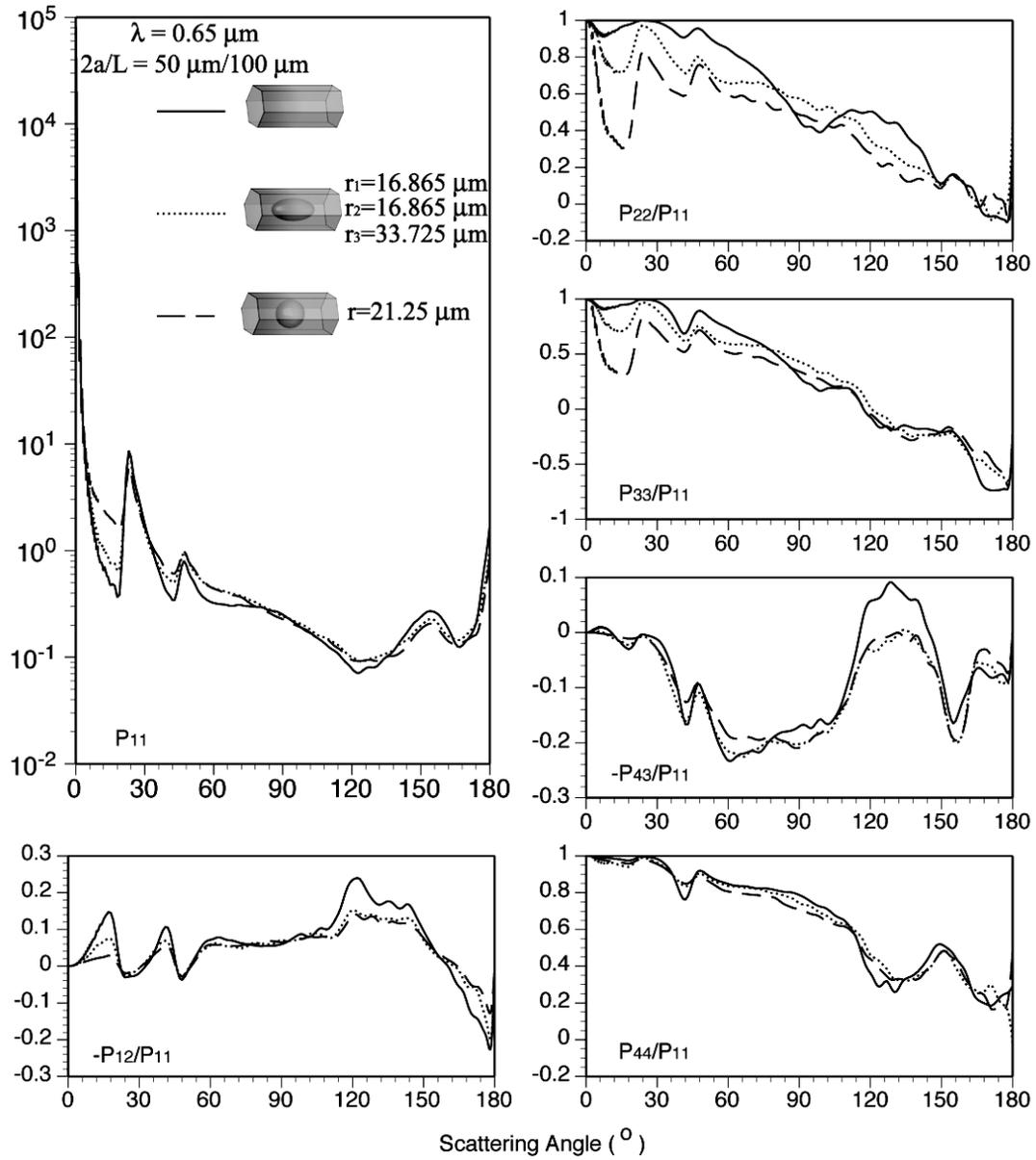


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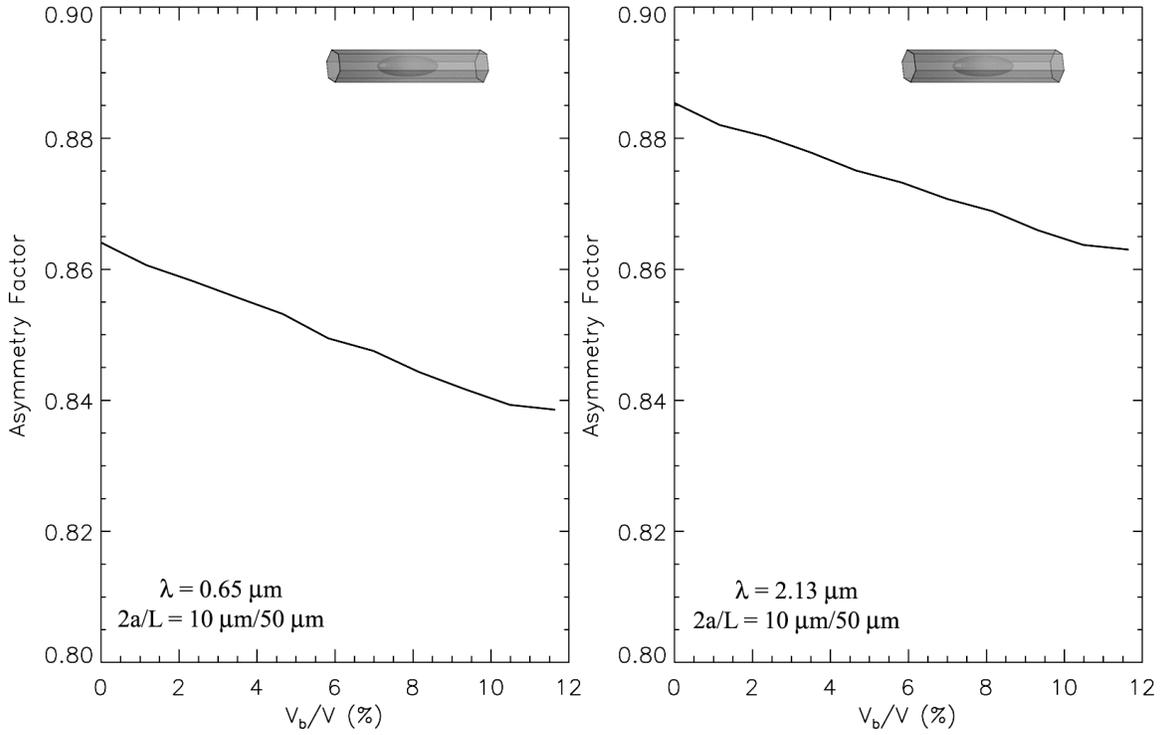
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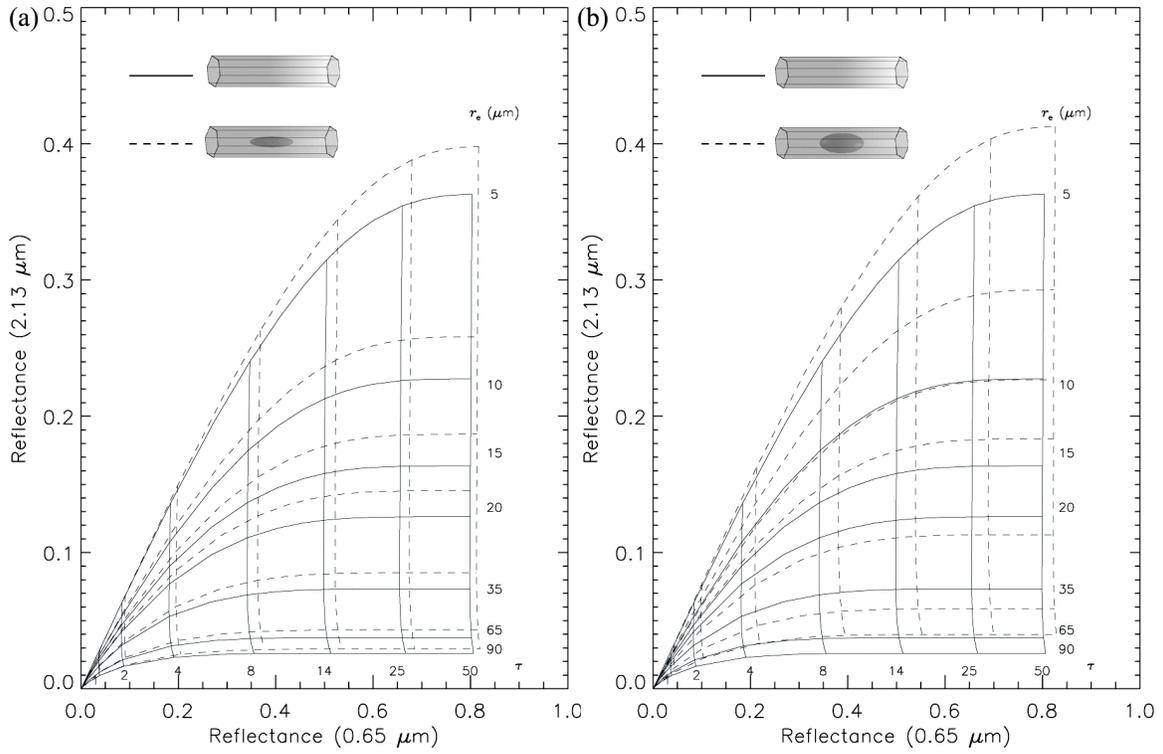
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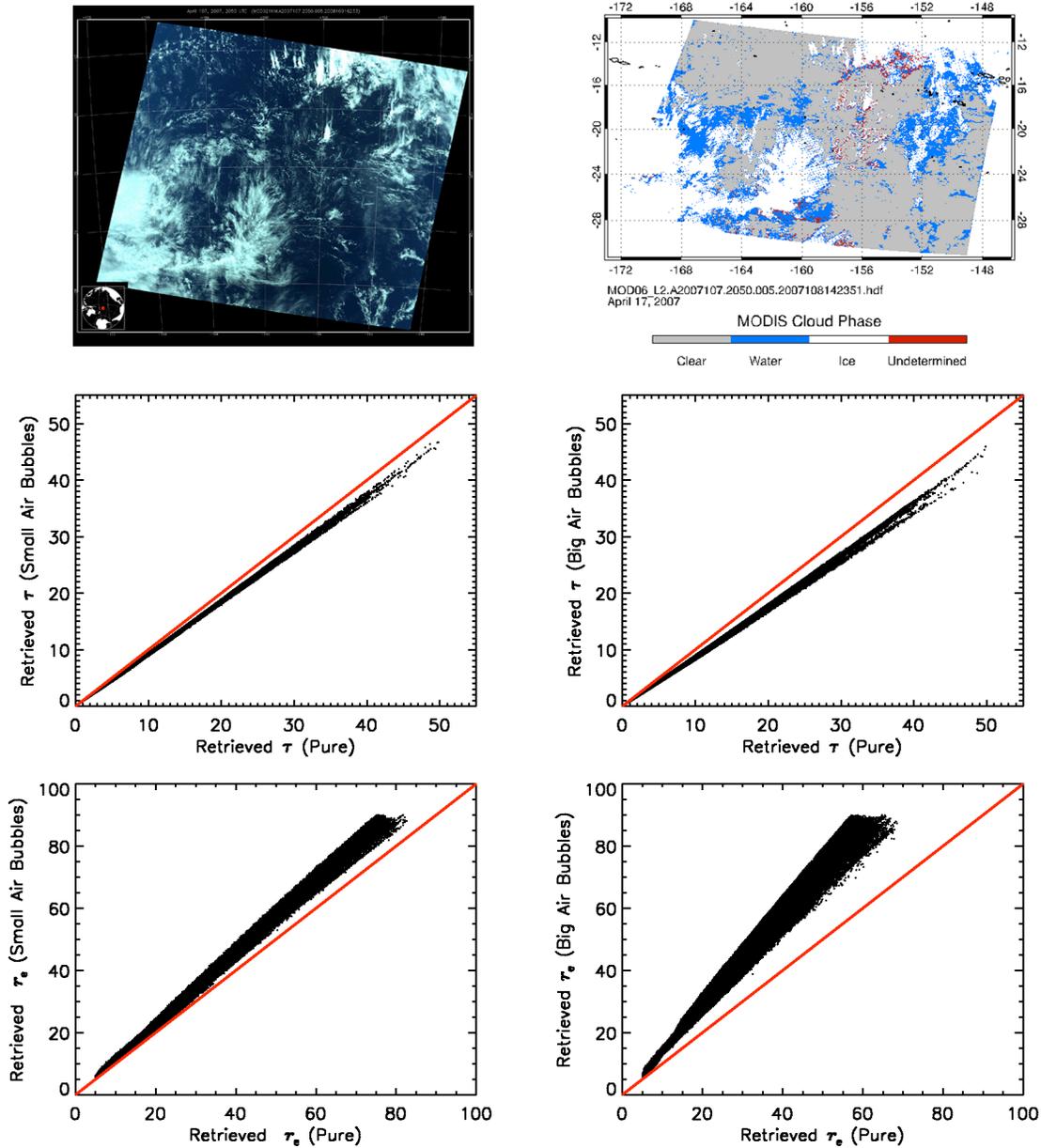
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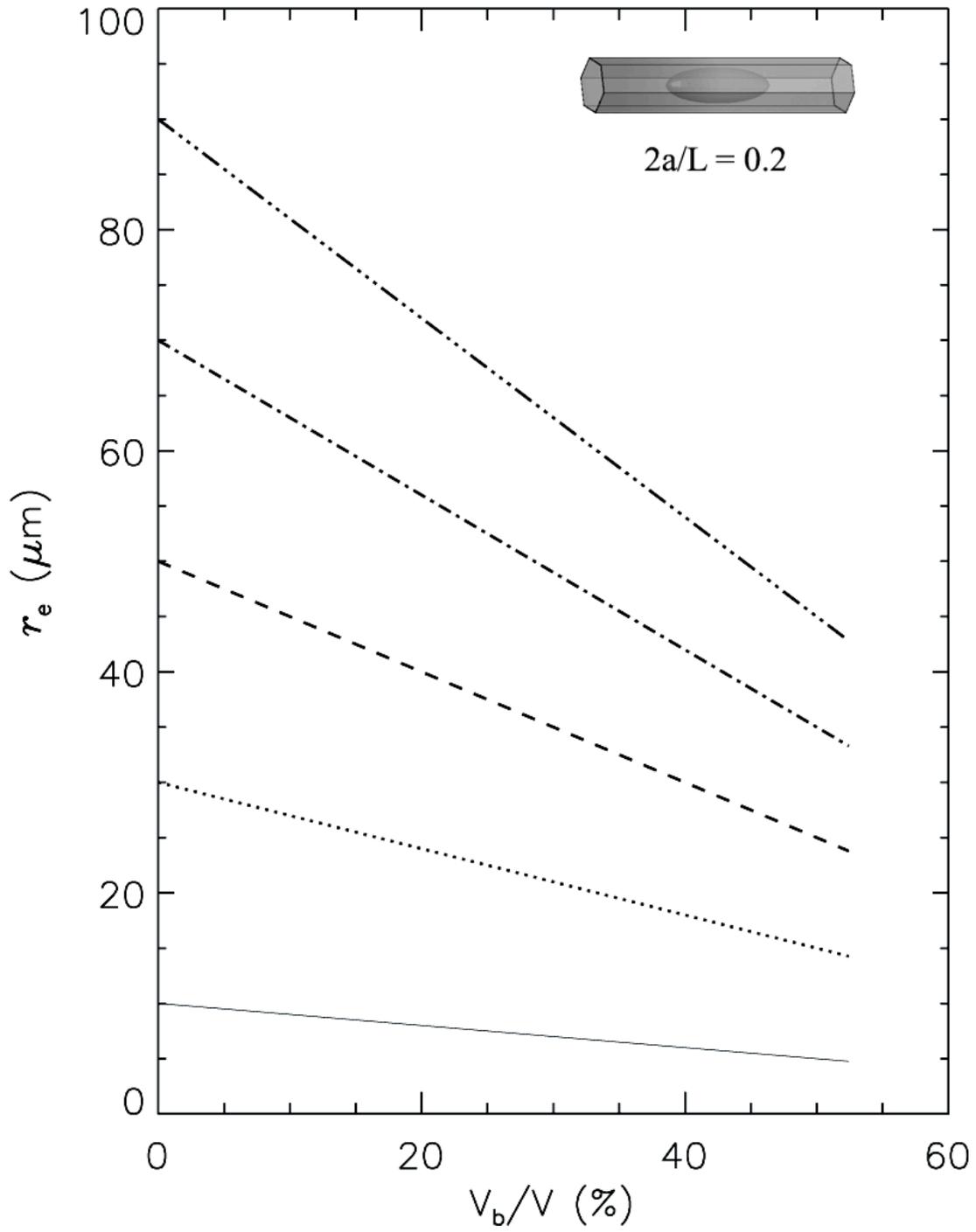
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