

## Responses to Reviewers (ACP MS No.: acp-2014-441)

We would like to thank the reviewers for their valuable comments and constructive suggestions. In the revised manuscript, we have accommodated all the suggested changes.

The reviewers' comments/recommendations are copied here as texts in blue; the authors' responses are texts in red.

Note, a marked-up version of the revised manuscript is attached to this response.

### Anonymous Referee #1

#### General comments

The paper presents a two-habit mixture model to represent the general microphysical, optical and radiative properties of cirrus. The authors base their two-habit mixture model on recent observations that show ice crystals evolving from simple compact shapes to more spatially aggregated shapes with increasing ice crystal maximum dimension (observations that are generally shown throughout the literature as well as aggregation simulation studies). The habit mixture can be changed continuously across the PSD without discontinuities. The authors use the latest light scattering methods to compute the single scattering properties of their model and include both surface roughness and hollow hexagonal cavities. They make extensive use of in situ, laboratory, and satellite remote sensing measurements, inclusive of polarization, to show that some form of habit mixture is required to consistently simulate the observations from across the spectrum and as a function of scattering angle. A single hexagonal column model is shown not to simulate the same observations to the same degree as the habit mixture model. The results presented are worthy of publication and the paper is thorough and understandable. Although their model consists of only two habits, the second habit is composed of 20 hexagonal monomers, this leads to the following questions and points that the authors need to address before the paper can progress to ACP.

The points and questions below are not considered major but need to be addressed to improve the paper.

#### Points to consider

1. The aggregate model consists of 20 monomers, why 20? Why not 10, 15, 18? Please justify why 20 has to be used. Is it the case that to satisfy the measurements of Dmm and IWC this many monomers is required? From light scattering calculations it is shown in Figures 5 and 6 of Baran (2009) that adding hexagonal monomers beyond 3 components does not significantly change the phase function (asymmetry parameter) due to the aggregates being spatial, i.e., multiple scattering between monomers is not significant. Indeed, the g-values are shown to asymptote. The results contained in Baran (2009) are based on an ice aggregation model developed by Westbrook et al. (2004). In the case of the aggregate model proposed by the authors how many monomers are required for the phase function (asymmetry parameter) to asymptote?

**Response:** Although Baran (2009) demonstrated that adding hexagonal monomers with the element number beyond 3 does not significantly alter the asymmetry factor, in this study we select 20 monomers for three reasons: 1) as an appropriate particle geometry is sought to mimic the complicated morphologies of realistic aggregates within ice clouds and it seems to be an oversimplification if only a few monomers are used; 2) an aggregate geometry corresponding to a potentially lowest value of the asymmetry factor is desired, and it is found

1 that the asymmetry factor slightly decreases as the number of monomers increases; 3) with the  
2 trial and error method, the use of 20 monomers is optimal in terms of the balance between the  
3 computational effort in light scattering simulation and the performance of the particle habit  
4 model in fitting the measured microphysical properties (specifically,  $IWC$  and  $D_{mm}$ ).

5  
6 2. In the construction of the aggregate model are intersecting planes avoided? This should be  
7 stated.

8  
9 **Response:** The criteria used by Xie et al. (2011) to detect overlapping of two hexagonal  
10 particles are used to avoid intersecting particle faces in this study, as explained in the revised  
11 manuscript.

12  
13 3. Please could the authors state the orientation-averaged area ratio and fractal dimension of  
14 their aggregate model in the paper and how do these compare to observation?

15  
16 **Response:** We did not consider the orientation-averaged area ratio or fractal dimension of the  
17 model. However, the two parameters are very important for determining particle geometries,  
18 and have been investigated by other researchers using available observations.

19  
20 To give more information about the geometry of the aggregate used in this study, we added  
21 the geometric parameters, volume and projected area of the aggregate in the revised  
22 manuscript (please see Appendix A and Table A1).

23  
24 4. In the appendix please also include the full co-ordinate (x,y,z) geometry of the hexagonal  
25 aggregate model and are the aspect ratios of each monomer kept constant at a value of 1?

26  
27 **Response:** The parameters used to fully determine the aggregate as well as its volume and  
28 projected area are added in the appendix. As we explain in the manuscript, the aspect ratios  
29 are randomly chosen between 0.8 and 1.

30  
31 5. The definitions of maximum dimension between the SCM and ice aggregate are not the  
32 same. If definitions were the same what effect would this have on your calculations when  
33 comparing properties of the same maximum dimension? This will not fundamentally alter  
34 their conclusions but will impact their calculations to some degree, the question is how  
35 important is it? Is the definition of maximum dimension applied to the aggregate robust  
36 under different viewing geometries?

37  
38 **Response:** It is true that the definition of maximum dimension does not fundamentally affect  
39 our conclusions. For the aggregate, the maximum dimension is defined as the maximum  
40 distance of two points on the aggregate element surfaces, and it is independent of particle  
41 orientation.

42  
43 6. I dispute the use of the term “spectral consistency”. What is shown in the paper is that the  
44 two component model is more consistent between 5 wavelengths, and the wavelengths are  
45 composite band-averages rather than monochromatic differences. To be truly spectrally  
46 consistent the authors need to show that the model is monochromatically consistent across  
47 high-resolution radiance spectra spanning the visible, near-ir and long-wave regions as  
48 demonstrated by Baran and Francis (2004). At the moment, the authors may only state that  
49 their model fits composite band-averaged measurements comprising of five wavelengths.

**Response:** The use of the term “spectral consistency” may overstate the advantage of the two habit model. However, we clearly explained the term in the manuscript, and a reader should understand the meaning of “spectral consistency” within context. So we prefer to keep the term.

7. Figure 4. Could the authors be more quantitative? especially when measurements of IWC are over many orders of magnitude. I suggest plotting PDFs of measurements and model results over intervals of  $D_{mm}$  and IWC and using a statistical method to quantify the goodness of fit?

**Response:** The histograms of the distributions of the measured and calculated  $D_{mm}$  and IWC are given (see the lower panels of Fig. 4), and, as expected, both the  $D_{mm}$  and IWC distributions given by the THM closely agree with those of the measurements. Furthermore, we illustrate the mean relative differences and their standard deviations of the theoretical microphysical properties at different bins of  $D_{mm}$  and IWC. More detailed discussions of the comparison between the theoretical and measured microphysical properties are added in the revised manuscript. In addition, a new table (Table 1) and a new figure (Fig. 5) have been added.

8. Figure 6. These are bulk comparisons. The authors employ a number of different light scattering methods to compute the scalar optical properties as a function of  $D$ . I would also like to see a figure showing a plot of the scalar optical properties as a function of maximum dimension to show that there are no discontinuities occurring between the different light scattering methods.

**Response:** Following the suggestion, we added a new figure (Fig. 7) for the extinction efficiency, single-scattering albedo and asymmetry factor of the SCM and THM as functions of  $D$ . It can be seen that there are no noticeable discontinuities in the results.

9. The paper does not at all discuss how cloud vertically inhomogeneity and 3D cloud effects may impact their results. Some discussion of these effects is warranted, especially with regard to the more recent study by Fauchez et al. (2014), found here <http://www.atmos-chem-phys.net/14/5599/2014/acp-14-5599-2014.pdf>

**Response:** This study does not address issues related cloud vertical inhomogeneity and 3-dimensional radiative effects. This point is clearly stated and the recommended reference (Fauchez et al., 2014) is cited in the revised manuscript.

Minor points and typos

1. Page 19547 line 10, “an ensemble habits”-> “an ensemble of habits..”

**Response:** corrected. Thanks for pointing out the typo.

2. Page 19547 line 15 consider adding this citation Baran et al. (2014), as the paper demonstrates the importance of constraining habit mixture models and PSDs, assumptions regarding the former and latter are shown to significantly affect climate model calculations of SW and LW fluxes at TOA (Baran, A., P. Hill, K. Furtado, P. Field, and J. Manners, 2014: A Coupled Cloud Physics-Radiation Parameterization of the Bulk Optical Properties of Cirrus and its Impact on the Met Office Unified Model Global Atmosphere

5.0 Configuration. J. Climate. doi:10.1175/JCLI-D-13-00700.1, in press.)

**Response:** This is a relevant paper, and is now cited in the revised manuscript.

3. Page 19548. In the discussion of surface roughness a citation to Ulanowski et al. (2014) should also be added, which can be found here <http://www.atmos-chemphys.net/14/1649/2014/acp-14-1649-2014.pdf>

**Response:** The paper is cited.

4. Page 19548, the word “numerous” is in my opinion not justified as Figure 7 shows one example of a laboratory measured phase function and one example of an in situ measured phase function. Please re-write accordingly.

**Response:** Indeed, only two cases are compared between the theoretical calculations and laboratory measurements. However, we state that “the ice cloud optical properties were obtained in numerous laboratories and field campaigns” by other researchers.

5. Page 19548, when discussing the PN I believe you are missing a number of Gayet et al. citations. Please include some of those citations in your manuscript.

**Response:** Two more papers by Gayet et al. on the subject are cited in the revised manuscript.

6. Page 19549, line 1. All the citations are somewhat biased towards particular groups, what about work that has used the dual-viewing ATSR-2 instrument and multiple viewing MISR, for instance McFarlane et al citation should also be added here. Here are some suggestions.

<http://onlinelibrary.wiley.com/doi/10.1029/2007JD009191/abstract>

<http://onlinelibrary.wiley.com/doi/10.1029/1999JD900842/abstract>

<http://www.opticsinfobase.org/ao/abstract.cfm?uri=AO-44-19-4060>

**Response:** These three papers are cited.

7. Page 19552. Lines 13-19. This argument is only true if the monomers making up the aggregate are sufficiently separated from each other so that multiple scattering between monomers is negligible. There might be instances where the constructions are such that the phase functions could be different between different realizations due to multiple scattering between monomers.

**Response:** We agree with the reviewer, and modified the relevant statement accordingly.

8. Page 19551, line 4. The authors state “..seldom does an cloud model...” This statement has been addressed by Baran et al. (2014), whom show that an ensemble model can indeed be consistently applied across the spectrum to simulate different measurements from the UV to radar frequencies, see <http://onlinelibrary.wiley.com/doi/10.1002/qj.2193/abstract>

**Response:** This paper is now cited in the revised manuscript. .

9. Page 19553, line 16. Please add in the discussion of surface roughness the Ulanowski et



al. (2014) citation and also Ulanowski et al. (2006), which is found here [http://homepages.see.leeds.ac.uk/~lecsjed/huiyi/habit/habit\\_Aug\\_2011/papers/sdarticle%5B1%5D.pdf](http://homepages.see.leeds.ac.uk/~lecsjed/huiyi/habit/habit_Aug_2011/papers/sdarticle%5B1%5D.pdf)

**Response:** Both papers are cited now.

10. Page 19553, line 20, the application of surface roughness to the two component model is this applied to all sizes? If so, please state this or the size range over which it is applied.

**Response:** The surface roughness is applied for particles over the entire size range, and we clarify this point in the revised manuscript.

11. Page 19554, is the determination of in situ IWC based on the PSD integration assuming some mass-D relationship or bulk measurements of IWC? In the former case, is the exponent assumed in the mass-D relationship the same as the fractal dimension of your aggregate model?

**Response:** Instead of the mass-D relationship, we gave the volume-D relationship in the revised manuscript (see Eq. 4).

12. Following equation (1)  $D_{mm}$  should follow?

**Response:** Based on the definition of  $D_{mm}$ , it is only used as the upper/lower limits of the integrals in Eq. (2). It is unlikely to obtain an analytical expression in the form " $D_{mm} = \text{something}$ ". However,  $D_{mm}$  included in Eq. (2) can be obtained numerically.

13. Page 19555, line 3, "...is the density of ice" -> "...is the density of solid ice.." and please state the density of solid ice assumed.

**Response:** The definition is specified, and the value used is given.

14. Page 19555, line 8, the 11 field campaigns, the PSD measurements, what was the range of particle size measured? Was the maximum particle size < 1 mm? This is important as it has implications for the effects of shattering on their dataset, see Korolev et al. (2013). Indeed, in this section the authors should state whether their datasets are affected by shattering or how shattering was minimized in their case.

**Response:** The details of the microphysical observations are not given in the manuscript. We use results from 11 field campaigns, and the information on them can be easily found in the relevant reference. Indeed, there are observations with small particles. In the revised manuscript, we discussed the different performance of the THM in modeling the microphysical properties at different bins of  $D_{mm}$  and IWC.

15. Page 19555, line 12 suggest replacing "under" by "colder than"

**Response:** Modified.

16. Page 19556, line 23, they use the term "solved", their "solution" is not unique as a combination of microphysical models could be used to give similar results, I believe the word "solved" is not warranted. Please re-write this sentence accordingly. Indeed, the

1 above paragraph lines 12-21 directly contradict the statement contained in section 4.

2  
3 **Response:** The sentence is rewritten.

4  
5 17. Page 19557 line 5, do they mean “increases” rather than “decreases” as particle size  
6 increases? (Auer and Veal, 1970).

7  
8 **Response:** The model we used is the same as that used by Yang et al. (2013) and Bi et al.  
9 (2014), and the details can be found in the cited references.

10  
11 18. Page 19557, section 4.1. I assume all single-scattering calculations are for random  
12 orientation? If so please state this.

13  
14 **Response:** Random orientation condition is stated in the revised manuscript.

15  
16 19. Page 19558, line 5, suggest “repeated” rather than “recaptured”.

17  
18 **Response:** the suggestion is taken and revision is made accordingly.

19  
20 20. Page 19558, line 29, how well does the value of the asymmetry parameter of the habit  
21 mixture model compare against observations?

22  
23 **Response:** The values of the modeled and observed asymmetry parameters of the two cases  
24 are given in the revised manuscript.

25  
26 21. Title section 4.2, no need for the word “the” in the section title.

27  
28 **Response:** “The” has been deleted in the revised manuscript.

29  
30 22. Page 19559, line 15, there are exceptions to featureless phase functions at backscattering  
31 angles such as the cases discussed by Gayet et al. (2012) and Baran et al. (2012).  
32 <http://www.atmos-chem-phys.net/12/9355/2012/acp-12-9355-2012.pdf> and references  
33 therein and other studies.

34  
35 **Response:** Actually, both papers were cited in the manuscript. In the revised manuscript, it is  
36 further clarified that the unusual scattering phase functions were observed from in situ  
37 measurements, while they are not considered for building the THM.

38  
39 23. Page 19559, in the discussion of number concentration measured by the PN on line 21, the  
40 likely effect of shattering on this instrument should be discussed.

41  
42 **Response:** For the case we use, Febvre et al. (2009) articulated that the effects of ice crystal  
43 shattering on the measurement is not very important, and, thus, they will not be considered in  
44 our study. We added the relevant discussion in the revised manuscript.

45  
46 24. From Figure 8, what is the value of the phase function at 180° given by the two  
47 component model? And how does this value compare against the CALIOP observations  
48 given in Baum et al. (2011)?  
49

**Response:** Considering the difficulty associated with accurate simulation of the phase function at  $180^\circ$  (especially for the geometric optics method), we didn't discuss the back scattering in this paper.

25. Page 19561, line 9, do you mean the far-infrared? In which case you should cite Cox et al. (2010) located here <http://onlinelibrary.wiley.com/doi/10.1002/qj.596/abstract;jsessionid=AA7EBE2992CB5F4DD67A9062F9BB574B.f01t03>

**Response:** The paper is cited.

**Anonymous Referee #2**

**Major comments:**

1) While this paper does address ice cloud microphysics, it is only addressed to the extent necessary for obtaining optical properties from a given ice particle size distribution (PSD). This point needs to be made more clearly in the paper. Thus the content of the paper may be more appropriately expressed using a title like “A two-habit model for the optical properties of ice clouds”. After all, the model conserves two microphysical properties but does not predict them.

**Response:** The motivation of building a new ice cloud model is to represent both its microphysical and optical properties, and this is also the most important and unique highlight of the paper. We have a full section, i.e. Section 3, to discuss the microphysical properties of the THM. For a given PSD, it is straightforward to use the THM to calculate the ice water content and medium-mass diameter for a given PSD. Furthermore, in the revised manuscript we have added an expression of particle volume as a function of particle maximum dimension. Thus, we prefer not to change the title of the paper.

2) Page 19555, lines 6-7: Since  $D$  = ice particle maximum dimension, it is not possible for both  $V_c(D)$  and  $V_a(D)$  to be proportional to  $D^3$ . Numerous papers show  $V_a(D)$  to be roughly proportional to  $D^2$ , and the exponent on  $D$  for  $V_c(D)$  lies typically between 2.5 and 3. So what expressions were used to represent  $V_c(D)$  and  $V_a(D)$ ?

**Response:** The V-D relationship is given in Eq. (4). For particle sizes between 100  $\mu\text{m}$  and 1500  $\mu\text{m}$ ,  $V$  is proportional to the  $D^2$ . In smaller or larger size regions,  $V$  is proportional to  $D^3$ .

3) Page 19555, lines 13-15: Here it states that Eqns. 1 and 2, using the THM habit fractions, can be used to calculate IWC and Dmm from the observed PSD. A research paper should provide the necessary information that allows other investigators to test the study's findings. This is not possible for this study since the relationships for  $V_c(D)$  and  $V_a(D)$  are not reported, but evidently these volumes are calculated as described in Appendix A. It could be very useful to the cloud physics community if these volumes could be related to their maximum dimension  $D$  in log-log space, with V-D power laws given. For example, this may allow other investigators to generalize the results reported in Fig. 4 to other cloud physics applications.

**Response:** In the revised manuscript, detailed information about the aggregate particle geometry used in the two habit model is presented (see Table A1). In addition, the V-D relation is also given (see Eq. 4).

4) Page 19555, lines 13-15: Do these calculations use the gamma PSD fits noted above in lines 10-11? One might assume that they do, or else why are the gamma fits mentioned? Nonetheless, this point should be clarified.

**Response:** Yes, the dataset provides two parameters of each fitted gamma distribution, and we clarified this point in revised manuscript.

5) Page 19556, lines 3-11: The agreement between measured and computed IWC in Fig. 4 is remarkable; so remarkable that it is hard to believe if taken at face value since. That is, direct in situ measurements of IWC (e.g. CVI or CSI probes) compared against IWC calculated from collocated PSD measurements, using either ice particle mass-dimension (m-D) or mass-area

(m-A) power law relationships, show a great deal of scatter; see for example the comparison in Fig. 6 of Lawson et al. (2010, JGR). This is the best agreement I have seen between direct measurements of IWC and IWC calculated from measured PSD & m-A expressions, and still there are differences of a factor of 2 or even 3. Questions that naturally arise when inspecting Fig. 4 are:

a. What are these measured IWCs? Are they direct measurements from probes like the CVI, or are they calculated from PSD measurements assuming some m-D or m-A expression(s)?

b. If they are calculated from PSD measurements, what m-D or m-A expressions were used?

c. What are the expressions for  $V_c(D)$  and  $V_a(D)$ , used to calculate IWC in the THM?

If the measured IWC was calculated from observed PSD, then what is actually being compared in Fig. 4 are two calculations; one based on observed PSD and some undisclosed m-D or m-A expression(s) and the other based on Eq. 1 in the THM. If this is the case, then the agreement observed in Fig. 4 is plausible since much of the natural variability will be removed by invoking the m-D or m-A expression(s).

The same concerns noted above for IWC also apply to the Dmm comparisons.

**Response:** Yes, these IWCs are all direct measurements from probes of different kinds, and data collected from 11 field campaigns are used. Actually, the TC-4 discussed by Lawson et al. (2010) is one of the 11 field campaigns. Many more details about the in situ measurements can be found from the references we cited. To clarify, we added the final format of the relationship we found between  $V$  and  $D$ , which leads to close agreement between the theoretical and observed microphysical properties.

6) Page 19557, line 5: The convention in cloud physics for aspect ratio is to define it as more than or equal to 1.0 for columnar ice crystals and less than or equal to 1.0 for planar ice crystals (e.g. Lamb & Verlinde, 2011: Physics and Chemistry of Clouds, Cambridge; see Ch. 8).

**Response:** The definition of the aspect ratio is clearly given in the manuscript. Many references cited in this manuscript use the same definition as ours. So we prefer to keep this definition.

7) Page 19559, end of Sec. 4.1: Please comment on the importance of the random distribution of aspect ratio and size regarding the aggregate components. For example, to what extent do the optical properties change when a realistic fixed aspect ratio/monomer size assumption is imposed?

**Response:** The effect of the aspect ratio of a hexagonal column on its scattering properties has been well studied (e.g., Yang, P., and Q. Fu, 2009: Dependence of ice crystal optical properties on particle aspect ratio, *J. Quant. Spectrosc. Radiat. Transfer*, 110, 1604-1614), and we will not repeat those in this paper. For this study, we try to build a particle with relatively small asymmetry factor (at visible and near infrared wavelengths), and, thus, the aspect ratios of hexagonal monomers are kept close to 1. In other words, it is important to have monomers with aspect ratios close to 1, and the asymmetry factor will become larger as the aspect ratio deviates from unity. However, the random distribution is implemented by assuming that the monomers of a realistic aggregate do not have the same aspect ratio.

8) Page 19560, line 9: Wang et al. (2014) is not referenced. Wang et al. (2013a) and (2013b) are referenced, but are not cited in this paper apparently. The Wang et al. 2013 papers do not retrieve the scattering phase function of ice clouds from satellite observations (which the

Wang et al. 2014 paper allegedly does).

**Response:** We have the references of Wang et al. 2013a, 2013b and 2014. The citations about the three papers are checked and corrected. Thanks for pointing out the typo.

9) Page 19560, line 20: How is  $D_{\text{eff}}$  defined in this study? Some investigators use extinction to define it, others use PSD projected area, and so on.

**Response:** We add the definition of  $D_{\text{eff}}$  in the text. In addition, the paper by Foot (1988) has been cited, which introduces the definition.

10) Page 19560, lines 17-20: Mishra et al. (2014, JGR) show that  $D_{\text{eff}} = 100\mu\text{m}$  is also common for cirrus clouds. Please consider adding a THM bulk phase function for  $D_{\text{eff}} = 100\mu\text{m}$  to Fig. 8, showing how insensitive the phase function is to  $D_{\text{eff}}$ .

**Response:** One of the most significant features of the THM is that its phase functions (as well as the asymmetry factor) at visible and near infrared channels are not sensitive to particle size, as can be seen from Fig. 7. The phase function with  $D_{\text{eff}} = 50\mu\text{m}$  is very close to that with  $D_{\text{eff}} = 100\mu\text{m}$  (except for the forward diffraction peaks that are proportional to particle geometrical cross section). To better illustrate this, we added the phase function with  $D_{\text{eff}} = 100\mu\text{m}$  in the figure for comparison.

9) Page 19561, lines 1-3: Could another explanation be that  $D_{\text{eff}}$  is smaller over land (relative to the oceans)?

**Response:** As we have demonstrated in the paper, the phase function of the THM at visible/near infrared bands does not vary significantly for different  $D_{\text{eff}}$  (except for the forward peak) and this should not be the reason for the relatively poor agreement for data over land.

Technical comments:

1) Page 19547, line 10: ensemble habits => ensemble of habits?

**Response:** Corrected.

2) Page 19547, line 29: growth, => diffusion growth, ?

**Response:** Modified. Thanks for the suggestion.

3) Page 19548, line 9-10: ice crystal particle => ice particle?

**Response:** Corrected

4) Page 19567, line 14: ranage => range?

**Response:** Corrected.

5) Page 19568, line 21: dada => data?

**Response:** Corrected.

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6) Figure 7: The text within Fig. 7b refers to a wavelength of 0.804  $\mu\text{m}$ , but the caption refers to a wavelength of 0.86  $\mu\text{m}$ .

**Response:** Corrected.

1 **Short Comment** by Dr. Bastiaan van Dienenhoven

2  
3 It is not my intent to provide a full review to the manuscript submitted to ACPD. There was  
4 one important comment that I missed in the previous reviews and I would like to address that  
5 in this writing.

6  
7 The manuscript by Liu et al. presents a two-habit model (THM) for the microphysical and  
8 optical properties of ice crystals in ice clouds. The authors show that this model represents the  
9 microphysical and remote sensing data rather well in general. This model could be useful for  
10 modeling and remote sensing applications. As they state in the manuscript, to better illustrate  
11 the advantages of the THM, they also consider a single hexagonal column model (SCM) for  
12 comparison. Based on the comparisons of the optical properties of the THM and SCM to  
13 measurements, the authors seem to suggest that a single column model cannot adequately  
14 represent the optical properties of ice clouds. However, that conclusion is not supported by the  
15 work in this paper.

16  
17 The SCM used by the authors has aspect ratios that increasingly deviate from 1 with  
18 increasing size and does not assume any surface roughness, based on choices made many  
19 years ago. These choices of aspect ratio and roughness are largely determining the optical  
20 properties. Especially the difference in roughness is determining the differences in optical  
21 properties between the SCM and THM. The paper does not show that there are no other  
22 choices of aspect ratio and roughness possible that would lead to a similar agreement with the  
23 measurements as is reached with the THM. Indeed, Wang et al. (2014) show that the phase  
24 function data shown in Fig. 8 is sufficiently well fit by a rough solid column model, at least  
25 over ocean. Furthermore, Cole et al. (2013) show the POLDER data shown in Fig. 10 is well  
26 fit using a single rough hollow column model.

27  
28 Thus, in my opinion, the authors should at least make clear throughout that their choice of  
29 SCM is a very particular one. Basically all recent literature on ice scattering models agrees  
30 that crystal surface roughness is prevalent in natural ice crystals. This paper once again shows  
31 that a pristine crystal model does not fit the data, which is not very relevant anymore.  
32 However, it does not show that a SCM with adjusted roughness and aspect ratio would not fit  
33 the data.

34  
35 **Response:** Dr. van Dienenhoven's suggestions were followed and relevant revisions were  
36 made in the revised manuscript. To be more specific, in the conclusion we have added "It  
37 should be noticed that the SCM we used for this study is based on pristine particles with  
38 smooth surfaces, and the conclusions obtained with the present SCM should not be  
39 generalized to other single column models. Furthermore, models based on single column or  
40 plates are still widely used for radiative flux calculation and remote sensing implementations  
41 (e.g., Fu, 2007; van Dienenhoven et al., 2014), which are articulated to be rational with  
42 demonstrated success for some specific applications."

43  
44 In addition, in the revised conclusion, we further emphasized the point that "Furthermore, we  
45 would like to emphasize that the SCM we used for comparison is based on pristine ice crystals  
46 with smooth surfaces and certain aspect ratio values, and the findings based on the assessment  
47 of the performance of SCM in remote sensing applications may not necessarily applicable to a  
48 different single column/plate model, particularly, when particle surface roughness is  
49 considered."



1 The microphysical data shown in Fig. 4 will likely not be fit as well using a single column  
2 model and this is an advantage of using a multi-habit model. However, for remote sensing this  
3 is not a concern and if a single particle model with adjusted aspect ratios and roughness  
4 produces correct optical properties, it would be adequate for remote sensing purposes.

5  
6 **Response:** The advantage of the THM to consider both the microphysical and optical  
7 properties for the same model. This is one of our motivations to build the new model. The  
8 SCM we used in the study can not represent the microphysical properties of ice cloud  
9 regardless its performance in the optical property calculation.

10  
11 I suggest removing the SCM results and any statements about the SCM from the paper.  
12 Alternatively, the SCM could be renamed “the single pristine column model” (SPCM) and the  
13 THM then should be renamed the “roughened two-habit model” (RTHM). It should then be  
14 clearly explained in the text that the differences in optical properties are largely due to the  
15 differences in roughness and aspect ratio choices and not because one is a two-habit model  
16 and the other is a single habit model. The authors should acknowledge that this work does not  
17 prove that there does not exist any SCM (or single plate models) with adjusted aspect ratio  
18 and roughness such that it would fit all optical data presented here.

19  
20 **Response:** We have clearly stated that smooth surface is used for the SCM, whereas the THM  
21 assumes rough surfaces. However, surface structure is only one factor of the model (it is  
22 definitely an important one), the aspect ratio, hollow structure, and aggregation configurations  
23 are all essential properties determining the model. We do not think there is any special reason  
24 to emphasize the importance of surface roughness and ignore the others. Historically, the  
25 SCM based on smooth surface and the aspect ratios that are the same as those used in our  
26 study were applied to remote sensing and radiative transfer simulations. However, the  
27 performance of the SCM has not been systematically evaluated. As a result, we prefer to keep  
28 the comparison of SCM and THM as they were used in the previous form of the manuscript.  
29 However, we added more detailed discussions in the revised manuscript to demonstrate the  
30 importance of surface roughness, and the limitation of the SCM used in this study.

31  
32 Furthermore, at two places in the revised manuscript, we explicitly stated that the findings  
33 associated with the current SCM should not be generalized to other single habit models,  
34 particularly, those including the surface roughness.

35  
36 **Minor comment:** Please change the x-axis label of Figure 6 to “effective particle diameter”.

37  
38 **Response:** Modified. Thanks for the suggestions.

# A two-habit model for the microphysical and optical properties of ice clouds

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## Abstract

To provide a better representation of natural ice clouds, a novel ice cloud model is developed by assuming an ice cloud to consist of an ensemble of hexagonal columns and twenty-element aggregates with specific habit fractions at each particle size bin. The microphysical and optical properties of this two-habit model (THM) are compared with both laboratory and in situ measurements, and its performance in downstream satellite remote sensing applications is assessed. The ice water contents and median mass diameters calculated based on the THM closely agree with in situ measurements made during 11 field campaigns. In this study, the scattering, absorption, and polarization properties of ice crystals are calculated with a combination of the invariant imbedding T-matrix, pseudo-spectral time domain, and improved geometric-optics methods over an entire practical range of particle sizes. The phase functions, calculated based on the THM, show close agreement with counterparts from laboratory and in situ measurements and from satellite-based retrievals. When the THM is applied to the retrievals of cloud microphysical and optical properties from MODIS observations, excellent spectral consistency is achieved; specifically, the retrieved cloud optical thicknesses based on the visible/near infrared bands and the thermal infrared bands agree quite well. Furthermore, a comparison between the polarized reflectivities observed by the PARASOL satellite and from theoretical simulations illustrates that the THM can be used to represent ice cloud polarization properties.

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# 1 Introduction

Ice clouds, i.e., high clouds containing [ice crystals with](#) various sizes and shapes, on average cover over 20% of the Earth and up to 60-70% of the tropical areas (Lynch et al., 2002; Nazaryan et al., 2008; Baran, 2009). Not surprisingly, ice clouds significantly influence both the climate system radiation budget and large-scale circulations in the atmosphere (Herman et al., 1980; Liou, 1986; Minnis et al., [1993a, 1993b](#); Sassen and Comstock, 2001; Stephens [et al., 1990](#); Stephens, 2005; Ramanathan, [et al., 2007](#); Loeb et al., 2009). However, owing to uncertainties in the ice microphysical properties (particle habit and size distribution) and optical properties (extinction coefficient, single-scattering albedo, and scattering phase matrix), ice clouds are still one of the least understood atmospheric components from the perspective of remote sensing and radiative transfer simulations involved in General Circulation Models (GCMs). Thus, a realistic and robust ice cloud model is being sought and of vital importance to atmospheric research.

A numerical model of ice clouds normally assumes either a single particle habit (i.e., particle shape) or an ensemble [of](#) habits (Baran and Labonnote, 2007; Baran et al., 2009; Baran, 2009, 2012; Yang et al., 2013; Baum et al., 2014). The optical properties determined based on the particle habit/habits are fundamental to the downstream applications in remote sensing, radiative transfer and GCMs (Ebert and Curry, 1992; Fu and Liou, 1993; Fu, 1996, 2007; Minnis et al., 1998; Katagiri et al., 2010; Heymsfield and Miloshevich, 2003; Baum et al., 2011; Edwards et al., 2007; Yi et al., 2013; [Baran et al., 2014a](#)). Thus, in order to reduce the uncertainties in downstream applications, an improved representation of ice cloud particle habits and optical properties is needed for the construction of a robust ice cloud model. Numerous laboratory and in situ measurements have been made to improve our knowledge [of](#) ice clouds, and various satellite observations have also played important roles in determining their microphysical and optical properties (Minnis et al., 1993b; Heymsfield and Miloshevich, 1995; Gayet et al., 2006, 2012; Lawson et al., 2008; Febvre et al., 2009; Heymsfield et al., 2013). The observations from different perspectives serve as the most practical and insightful standards from which to develop an ice cloud model. This study considers the currently available data in an attempt to improve the representation of ice clouds with a theoretical model based on two particle geometries.

As one of nature's greatest artworks, ice crystals show a myriad of variations in shape/habit for different meteorological conditions. Ice cloud habit study begins with an understanding of the microphysical processes necessary for nucleation, [diffusion](#) growth, collision and

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1 aggregation within the atmosphere. Both laboratory and in situ observations have contributed  
2 meaningful information about ice crystal shapes (Magono and Lee, 1966; Heymsfield et al.,  
3 2002, 2005; Lawson et al., 2006). The studies indicate that ice crystals occur with geometries  
4 having various degrees of complexity, e.g., pristine hexagonal columns, plates, and bullets,  
5 rosettes of different forms, and complicated and irregular aggregates. Furthermore, some  
6 detailed structures, such as surface roughness, hollow structure, and inhomogeneity (with air  
7 bubbles or ice nuclei inside), have been widely noted in the observations and considered for  
8 numerical studies (Ulanowski et al., 2012, [2014](#); Schmitt and Heymsfield, 2007; Labonnote et  
9 al., 2001). Both the ice, particle overall geometry and detailed structure have a significant  
10 effect on the optical properties. Thus, constructing an idealized model both geometrically and  
11 optically representative of natural particles is quite challenging.

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12 In addition to observations of particle geometries, various measurements have been attempted  
13 to obtain reliable information on the microphysical and optical properties of ice clouds (Curry,  
14 [et al., 2000](#); Heymsfield et al., 2013). A series of field campaigns were [conducted at](#) a variety  
15 of midlatitude and tropical locations in both hemispheres over the period between 1999 and  
16 2006 to study the microphysical properties of ice clouds (Heymsfield et al., 2013). The  
17 microphysical data collected includes the particle size distribution (PSD), ice water content  
18 (*IWC*), and median mass diameter ( $D_{mm}$ ). The  $D_{mm}$  is defined as the diameter at which the  
19 mass in the PSD with smaller particles equals that with larger ones. [Moreover](#), the ice cloud  
20 optical properties were obtained in numerous laboratories and field campaigns. The polar  
21 nephelometer (PN) was widely used to measure the scattering phase function of an ensemble  
22 of cloud particles (water droplets, ice crystals, or mixture of both) (Sassen and Liou, 1979;  
23 [Gayet et al., 1998, 2004](#); Barkey and Liou, 2001; Auriol et al., 2001; Febvre et al., 2009).  
24 Although limited spatially and temporally, the measurements [have](#) played an essential role in  
25 the numerical studies of ice clouds, and will be fully considered in this study.

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26 Satellite observations are used to infer cloud properties by [comparing](#) sensor measurements  
27 and radiative transfer simulations for a set of known cloud and atmospheric conditions  
28 (Wielicki et al., 1998; [Chepfer et al., 2002](#); Winker et al., 2003; Platnick et al., 2003; [Knap et](#)  
29 [al., 2005](#); [McFarlane and Marchand, 2008](#); Minnis et al., 2011; [Baran et al., 2012b](#)). The  
30 satellite measurements may be at either visible/near-infrared solar bands or thermal infrared  
31 (IR) bands, and may also include polarization. Sensors on board satellites flying as part of the  
32 NASA Earth Observing System A-Train constellation simultaneously provide measurements  
33 encompassing all of these characteristics ([L'Ecuyer and Jiang, 2010](#)). The satellite-based

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1 retrieval of ice cloud properties, e.g., the effective particle diameter ( $D_{eff}$ ) and optical  
2 thickness ( $\tau$ ), relies on the use of accurate and efficient radiative transfer models to simulate  
3 the cloud radiances at the top of atmosphere, and the optical properties of a given ice cloud  
4 model are required for those simulations (Minnis et al., 1993a, 1993b, 2011; Platnick et al.,  
5 2003). However, when applied to satellite remote sensing (e.g., based on the Moderate  
6 Resolution Imaging Spectroradiometer (MODIS) observations), most ice cloud models  
7 encounter a challenge known as spectral inconsistency, i.e., significant differences occur in  
8 cloud optical thicknesses retrieved with different spectral bands (e.g., solar or thermal IR  
9 bands) for the same cloud model (Baum et al., 2014). Thus, another goal of this study is to  
10 construct an ice cloud model that can infer consistent optical properties in solar- and thermal  
11 IR-band retrievals.

12 Using the ice cloud polarization properties (e.g., polarized reflectivity) has increasingly  
13 gained attention as a means to infer the microphysical and optical properties (van  
14 Diedenhoven et al., 2012, 2013; Labonnote et al., 2001), and such applications can be widely  
15 found with available observations from the PARASOL (Polarization and Anisotropy of  
16 Reflectances for Atmospheric Sciences coupled with Observations from a Lidar) satellite  
17 (Labonnote et al., 2001; Baran and Labonnote, 2007; Baran, 2009; Cole et al., 2013). The  
18 radiometer/polarimeter on board POLDER (POLarization and Directionality of the Earth's  
19 Reflectances) measures the  $I$ ,  $Q$ , and  $U$  components of the Stokes vector at three wavelengths  
20 with up to 16 viewing angles for each pixel (Deschamps et al., 1994). Previous studies have  
21 indicated that the polarized reflectivities simulated based on several ice cloud models, using  
22 either an individual habit or a mixture of multiple habits, can approximately match those of  
23 PARASOL observations (Doutriaux-Boucher et al., 2000; Baran, 2009; Cole et al., 2013). The  
24 polarized reflectivity data from the satellite have also been used to retrieve the ice particle  
25 habit (Sun et al., 2006) and degree of surface roughness (Cole et al., 2014). Thus, as another  
26 unique perspective of ice cloud, the polarization properties of a numerical model must be  
27 carefully checked.

28 With the variety of laboratory experiments, field campaigns, and satellite sensors to measure  
29 the microphysical and optical properties, constructing an ice cloud model that can consistently  
30 represent a wide range of perspectives is extremely challenging. This study strives to develop  
31 a robust ice cloud model based on two particle geometries, the two-habit model (THM), and  
32 to verify its performance in modeling the microphysical and optical properties of natural ice  
33 clouds. Section 2 reviews some of the previous ice cloud models and introduces the novel

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1 THM. Section 3 discusses the THM microphysical properties. Section 4 shows the optical  
2 properties of the THM, and comparisons with measurements. The THM performance in  
3 satellite remote sensing applications is presented in Section 5. Section 6 contains the  
4 conclusions.

5

## 6 **2 Two-habit Model**

7 Ice cloud particle geometries show significant variations for different meteorological  
8 conditions, especially temperature and relative humidity (Magono and Lee, 1966; Heymsfield  
9 and Miloshevich, 1995). A tremendous amount of effort has been devoted to the detailed  
10 study of ice crystal geometries from both laboratory and in situ observations, and to classify  
11 ice crystals into multiple categories based on the general geometries. The most widely  
12 observed ice crystal types include hexagonal columns, hexagonal plates, bullet rosettes, and  
13 aggregates of various pristine particles (Magono and Lee, 1966; Korolev et al., 1999;  
14 Heymsfield et al., 2002, 2005; Evans et al., 2005; Lawson et al., 2006). A number of studies  
15 have been reported on building numerical models for ice clouds with idealized geometries,  
16 and in using the corresponding microphysical and optical properties to represent natural  
17 clouds. For different applications, either the microphysical or the optical properties at certain  
18 wavelengths are normally considered in the models, and seldom does an ice cloud model  
19 consistently represent all ice cloud properties for different applications, (Baran et al., 2014b).

20 Due to the limitation in numerical simulations of light scattering by non-spherical particles,  
21 the single hexagonal column model for ice clouds was introduced to atmospheric applications  
22 in the 1970s and 1980s (Wendling et al., 1979; Cai and Liou, 1982; Takano and Liou, 1989a,  
23 1989b), but additional particle habits have since been developed and applied (Macke, 1993;  
24 Takano and Liou, 1995; Macke et al., 1996; Yang and Liou, 1998; Um and McFarquhar,  
25 2007; 2009). Various commonly occurring ice cloud habits are now widely used in radiative  
26 transfer and remote sensing, and popular examples include hexagonal columns and plates of  
27 various aspect ratios, droxtals, polycrystals, solid or hollow bullet rosettes, and aggregates of  
28 columns, plates or rosettes (e.g., Baran, 2009; Yang et al., 2013; and references cited therein).  
29 The models use either an individual particle habit or an ensemble of habits. When multiple  
30 habits are used, the habit fractions normally vary for different particle sizes. The optical  
31 database and parameterization based on the numerical models are normally made for further  
32 applications in remote sensing, radiative transfer and GCMs (Fu, 1996, 2007; Minnis et al.,

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1 | 1998; Edwards et al., 2007; Letu et al., 2012; [van Diedenhoven et al., 2014](#)). Ice cloud models  
2 | with a single habit have been found useful for some special applications as the computational  
3 | burden for single-scattering simulations is minimized. However, the single habit models are  
4 | [limited in the aspect of consistently representing](#) multiple ice cloud properties (Baum et al.,  
5 | 2014). Natural ice clouds show unclear variation of particle habits and have different habit  
6 | preferences at different size ranges. Multiple ice habits definitely introduce much more  
7 | freedom to accurately represent the microphysical and optical properties. However, without  
8 | explicit theoretical or observational data, the choice of the habits and habit fractions is  
9 | arbitrary. Furthermore, the accurate calculation of the scattering properties of different non-  
10 | spherical habits is very time-consuming. Thus, we will attempt to construct an ice cloud  
11 | model that can capture and represent all major properties by using as few particle habits as  
12 | possible, which will simplify the model and minimize the computational burden for  
13 | [computing](#) the single-scattering properties.

14 | A study by Schmitt and Heymsfield (2014) [suggests](#) that atmospheric ice particles can be  
15 | separated into two categories in terms of particle complexity (i.e., simple and complex) by  
16 | using particle imagery data from high-resolution aircraft particle imaging probes. A  
17 | dimensionless parameter representing the particle ‘complexity’ is defined based on particle  
18 | projected area, area ratio and perimeter, and a cutoff value is chosen to identify pristine and  
19 | complex ice particles imaged by the Cloud Particle Imager (CPI) probe with a resolution of a  
20 | few microns. The results of two example data sets from in situ measurements indicate that  
21 | complex particle habit fraction increases as the particle maximum dimension increases. The  
22 | idea of separating ice crystals into two general categories and the conclusions obtained from  
23 | the [study of Schmitt and Heymsfield \(2014\)](#) are of great importance in the simplification of  
24 | ice cloud models. [Moreover](#), numerical studies indicate that the optical properties of particles  
25 | of the same kind are not strongly affected by the [number and orientations of monomers of](#),  
26 | e.g. complex aggregates of hexagonal plates (Xie et al., 2011) or bullet-rosettes (Um and  
27 | McFarquhar, 2007), [particularly if the monomers are sufficiently separated that the multiple](#)  
28 | [scattering among the monomers is negligible. Therefore, as a quite accurate approximation, it](#)  
29 | [is possible to use a relatively simple particle morphology to represent a group of more](#)  
30 | [complicated counterparts in the computation of the particle optical properties.](#)

31 | [Based on the preceding physical rationale and](#) the observations and classifications given by  
32 | Schmitt and Heymsfield (2014), [in addition to the consideration of](#) the computational burden,  
33 | [this study explores the feasibility of using a simple habit and a complex habit to represent](#) ice

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1 | clouds. To construct the model, we need to determine the most representative particle habits.  
2 | A hexagonal column, considered as the simple/pristine particle, is the primary candidate, and  
3 | a type of complex aggregate is the second most widely observed and studied particle habit.  
4 | We use hexagonal columns as the aggregate monomers. By changing the geometric  
5 | parameters related to the aspect ratio, number of the monomers, and aggregation  
6 | configuration, the optical properties of the particles are optimized to match those of natural ice  
7 | clouds. The aspect ratio of a hexagonal column is defined as  $2a/L$ , where  $a$  is the semi-width  
8 | of the hexagonal cross section and  $L$  is the column length. A hexagonal column with an aspect  
9 | ratio equal to one, and an aggregate with 20 hexagonal column monomers are used in this  
10 | study. The relative sizes and aspect ratios of the 20 monomers are randomly generated, and  
11 | they are point-attached to form the aggregate. The particle maximum dimension  $D$  is used to  
12 | specify the particle size (i.e.,  $L$  for the hexagonal column, and the maximum distance between  
13 | two points on the particle for the aggregate), and the size parameter, which is an important  
14 | parameter for light scattering simulations, is defined as  $\pi D/\lambda$ .

15 | In addition to the overall geometries of the two habits, the detailed structures of natural  
16 | crystals are considered. In situ measurements have indicated that ice crystals have  
17 | predominantly hollow structures (Walden et al., 2003; Schmitt and Heymsfield, 2007) and  
18 | irregular geometries, and, for consideration of these facts in the THM, the hexagonal  
19 | monomer of the aggregate is assumed to have hollow structures similar to those used by Yang  
20 | et al. (2013). Figure 1 illustrates the geometry of a hollow hexagonal element. The depth of  
21 | the hollow structure is specified by  $d$  and  $d/L=0.25$  in this study. From observations, particle  
22 | surface roughness is widely noted as an important ice crystal feature (Cross, 1969; Ulanowski  
23 | et al., 2006, 2012, 2014; Neshyba et al., 2013), and numerical studies indicate that the surface  
24 | roughness has significant influence on the particle optical properties and cloud radiative effect  
25 | (Yi et al., 2013), especially the angular-dependent scattering phase matrix elements  
26 | (Peltoniemi et al., 1989; Macke et al., 1996; Shcherbakov et al., 2006; Yang et al., 2008). The  
27 | hexagonal columns and the aggregates over the entire size range considered will be treated as  
28 | roughened particles, with the same degree of surface roughness. The technical details of the  
29 | roughened surface definition can be found in Liu et al. (2013). In this study, severely  
30 | roughened particles (Yang et al., 2013) are used.

31 | Figure 2 shows the two particle geometries used for the THM, and both the hollow structure  
32 | and surface roughness are illustrated in the figure. The column is clearly a single but  
33 | 'compact' particle, whereas the aggregate is very complex and loose in the space. The two

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habits represent the simple and complex ice crystals classified by Schmitt and Heymsfield (2014). The processes necessary to form the hexagonal aggregate and its geometric parameters are detailed in the Appendix A.

### 3 Microphysical properties

With the explicit geometries of the two particle habits defined, the habit fraction, as a function of particle maximum dimension, becomes a key parameter to determine the microphysical properties of the THM. This section introduces the habit fraction used for the THM and compares the simulated microphysical properties, i.e.,  $IWC$  and  $D_{mm}$ , with those from in situ measurements.

As discussed in Section 2, Schmitt and Heymsfield (2014) separate ice crystals into simple and complex categories by analyzing CPI images, and show that the complex habit fraction increases as the maximum dimension increases. In the THM, the hexagonal column and aggregate correspond, respectively, to the simple and complex particles. Although that the exact percentages of the simple and complex particles may differ from case to case, the first role in qualitatively determining the habit fractions is to increase the aggregate fraction as the particle maximum dimension increases, which ensures that the geometric model represents natural crystals.

A more quantitative way to determine the habit fraction is to consider the microphysical data sets from the in situ measurements. The  $IWC$  and  $D_{mm}$  of ice clouds are highly related to the ice particle volume, which is a strong function of the particle maximum dimension. With a given PSD and the two particle habits in the THM, the  $IWC$  and  $D_{mm}$  are determined by:

$$IWC = \rho_{ice} \int_{D_{min}}^{D_{max}} [V_c(D)f_c(D) + V_a(D)f_a(D)]n(D)dD, \quad (1)$$

and

$$\begin{aligned} & \rho_{ice} \int_{D_{mm}}^{D_{max}} [V_c(D)f_c(D) + V_a(D)f_a(D)]n(D)dD \\ &= \rho_{ice} \int_{D_{min}}^{D_{mm}} [V_c(D)f_c(D) + V_a(D)f_a(D)]n(D)dD = IWC/2, \end{aligned} \quad (2)$$

where  $\rho_{ice}$  is the density of solid ice (a value of  $0.917 \text{ g cm}^{-3}$  is used in this study),  $D_{min}$  and  $D_{max}$  are the minimum and maximum particle sizes in the distribution, and  $n(D)$  is the number concentration of particles with a maximum dimension of  $D$ .  $f_c(D)$  and  $f_a(D)$  are the

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1 habit fractions of the column and aggregates in the THM, and, for any size,  $f_c(D) + f_a(D) =$   
2 1.  $V_c(D)$  and  $V_a(D)$  indicate particle volume for the two particle habits.

3 We use the microphysical data collected from 11 field campaigns, and a detailed summary of  
4 these data can be found in Heymsfield et al. (2013) and Baum et al. (2014). Coefficients for  
5 the gamma size distribution are fitted to the datasets of the particle number concentration  
6 versus size and provided for each individual PSD, and a total of over 14,000 PSDs with cloud  
7 temperatures colder than -40°C are used to ensure the measured clouds are indeed ice clouds.  
8 With certain habit fractions, the  $IWC$  and  $D_{mm}$  based on the THM can be computed for each  
9 PSD by the integrals given by Eqs. (1) and (2), and may then be compared with the  
10 observations. Fitted gamma size distributions from the data are used for the aforementioned  
11 integral. After we tested different habit fractions to minimize the differences between the  
12 simulated and observed  $IWC$  and  $D_{mm}$ , we chose a continuous habit fraction for the hexagonal  
13 column that leads to close agreement of the microphysical properties, which is given by:

$$f_c(D) = \begin{cases} 0.81 & D < 100\mu m \\ \frac{85}{D} - 0.04 & 100\mu m \leq D < 1500\mu m \\ 0.017 & D \geq 1500\mu m \end{cases} \quad (3)$$

15 and the fraction of aggregate is given by  $f_a(D) = 1 - f_c(D)$ . Figure 3 shows the THM habit  
16 fractions obeying Eq. (3). The fraction of the aggregate, i.e., the complex particle, smoothly  
17 increases with increasing particle diameter, and the trend is the same as that obtained from ice  
18 crystal image analysis. For small particles with maximum dimensions less than 100  $\mu m$ , we  
19 assume over 80% of the ice crystals to be hexagonal columns, but the fraction drops to only  
20 1.7% for particles larger than 1500  $\mu m$ .

21 Note that, considering the uncertainties in the observations, the final habit fraction we use for  
22 the THM does not necessarily give the best fit to all in situ data, but we find that all habit  
23 fractions with similar trends lead to similar agreement in the microphysical properties.  
24 Furthermore, considering the significant variation of ice clouds under different meteorological  
25 conditions, no single ‘best’ exists for all ice clouds, because the best for one condition may  
26 not represent the ice cloud properties under another condition. For applications of ice clouds  
27 having very different microphysical properties, the fractions of the two habits can be easily  
28 modified to match the specific properties. Thus, the continuous habit fraction given by Eq. (3)  
29 is used in the THM and the following simulations.

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$$\begin{cases} 0.8 & D < 100\mu m \\ \frac{80}{D} & 100\mu m \leq D < 2500\mu m \\ 0.032 & D \geq 2500\mu m \end{cases}$$

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Moved down [2]: Slight differences are noticed for  $D_{mm}$  at values larger than 500  $\mu m$ . The largest differences in the  $IWC$  are shown for data from the CRYSTAL-FACE campaign.

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Moved down [1]: the measured and calculated  $IWC$  and  $D_{mm}$  values for each of the PSDs from the 11 field campaigns. The names of the field campaigns are listed in the figure and differentiated by both colors and symbols. The values, for both  $IWC$  and  $D_{mm}$ , calculated with the THM are in

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With the habit fractions given, the upper panels of Fig. 4 compare the measured and calculated  $IWC$  and  $D_{mm}$  values for each of the PSDs from the 11 field campaigns. The names of the field campaigns are listed in the figure and differentiated by both colors and symbols. The values, for both  $IWC$  and  $D_{mm}$ , calculated with the THM are in close agreement with the observations. Slight differences are noticed for  $D_{mm}$  at values larger than 500  $\mu\text{m}$ . The largest differences in the  $IWC$  are shown for data from the CRYSTAL-FACE campaign. Overall, Fig. 4 indicates that, with two particle habits with a habit fraction given by Eq. (3), the THM can reasonably represent the microphysical properties of ice clouds. The lower panels of Fig. 4 show the histograms of the distributions of the measured and calculated  $IWC$  and  $D_{mm}$ . As expected, the THM-based distributions are essentially the same as the measured counterparts.

The relative differences (RDs) between the theoretical microphysical properties based on the THM and the in situ measurements for each of the 11 field campaigns are listed in Table 1. The names of the field campaign and the related numbers of PSDs with temperatures colder than  $-40^\circ\text{C}$  are given. For both  $D_{mm}$  and  $IWC$ , the means and standard deviations (STD) of the RDs are also listed. We can see that the mean RDs for  $D_{mm}$  are generally less than 5%. The only exception is the case of the Stratospheric-Climatic links with emphasis On the Upper Troposphere and lower stratosphere (SCOUT) field campaign with an average RD of 17% because relatively small  $D_{mm}$  (less than 50  $\mu\text{m}$ ) values were observed during this field campaign and the measurements are less reliable with such small particle sizes. Averaged for all the field campaign data, the model shows a mean RD of -0.27% with a standard deviation of 5.2% for  $D_{mm}$ . The RDs for  $IWC$  are almost one order larger in magnitude compared with the case of  $D_{mm}$ , because the values of measured  $IWC$  span more than 6 orders in magnitude. Furthermore, the mean RDs can be as large as 148% with a standard deviation of 50% for the CRYSTAL-FACE campaign, although the model works well in the case of data obtained during other campaigns such as the TRMM, ARM-IOP and MPACE. Overall, the present model overestimates the  $IWC$  by approximately 13% with a standard deviation of 24%.

To further quantify the performance of the THM for modeling the microphysical properties of ice clouds, Figure 5 illustrates the mean RDs and standard deviations for different bins of  $D_{mm}$  and  $IWC$ . The solid dot symbols in Fig. 5 indicate mean RDs, and the error bars indicate the corresponding standard deviations. For different bins of  $D_{mm}$ , the mean RDs and the corresponding standard deviations for both  $D_{mm}$  and  $IWC$  approach to zero as  $D_{mm}$  increases (see the left panels of Fig. 5). We call special attention to the fact that there are significant uncertainties related to the measurements of small particles, and the RDs at small  $D_{mm}$  bins

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show quite large standard deviations. The performance of the model is also sensitive to the *IWC*. Both the standard deviations of the RDs for  $D_{mm}$  and *IWC* tend to decrease as *IWC* increases, particularly, when *IWC* is larger than  $10^{-2} \text{ g m}^{-3}$ .

Furthermore, the relationship between particle volume (*V*) and the particle maximum dimension (*D*) determines *IWC* and  $D_{mm}$  for a given PSD. The V-D relationship based on the THM is given by

$$V(D) = \begin{cases} 0.53D^3 & D < 100\mu\text{m} \\ 53D^2 & 100\mu\text{m} \leq D < 1500\mu\text{m} \\ 0.036D^3 & D \geq 1500\mu\text{m}. \end{cases} \quad (4)$$

In the above expression, *D* is specified in units of  $\mu\text{m}$ , and *V* is in units of  $\mu\text{m}^3$ . Figure 6 illustrates the *V-D* relationship given by Eq. (4). Given the large amount of in situ measurements we used in this study and the quite reasonable agreement between the model results and measurements, the preceding *V-D* relationship can be used as a reasonably accurate expression to estimate the variation of ice crystal volume as a function of particle maximum dimension for other relevant applications.

#### 4 Optical properties

With the geometrical and microphysical model (i.e. the two particle habits as well as their habit fractions) discussed above, we turn to the optical properties of the THM. First, we give a brief introduction of the numerical algorithms used to obtain the optical properties, and illustrate the single-scattering properties of the THM. The second subsection compares the modeled phase functions with the results from both measurements and satellite retrievals.

To better illustrate the advantages of the THM, here we also consider a single hexagonal column model for comparison. This single column model (SCM) is based on a smooth surface, and the aspect ratio decreases as the particle size increases. The details of the single column model as well as its microphysical and optical properties can be found in Yang et al. (2013) and Bi et al. (2014). It should be noticed that the SCM we used for this study is based on pristine particles with smooth surfaces, and the conclusions obtained with the present SCM should not be generalized to other single column models. Furthermore, models based on single columns or plates are still widely used for radiative flux calculation and remote sensing implementations (e.g., Fu, 2007; van Deidenhoven et al., 2014), which are articulated to be rational with demonstrated success for some specific applications.

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#### 1 4.1 Single-scattering simulations

2 Numerical simulations of light scattering by [randomly oriented](#) non-spherical particles are a  
3 major challenge limiting the development of ice cloud models. The conventional geometric-  
4 optics method (CGOM), which is relatively simple and computationally efficient, is one of the  
5 most popular methods for the solution of light scattering by ice crystals ([Cai and Liou, 1982](#);  
6 [Takano and Liou, 1989a](#); [Macke et al., 1996](#); [Baran, 2009](#)), although its accuracy in the [cases](#)  
7 of small and moderate size parameters is questionable due to the inherent shortcomings of the  
8 ray-tracing technique. [Bi et al. \(2014\)](#) elaborate on the uncertainties with the CGOM in  
9 remote sensing applications and radiative transfer simulations by comparing with results from  
10 a benchmark scattering dataset obtained with a combination of the Invariant Imbedding T-  
11 matrix method (II-TM) ([Bi and Yang, 2014](#)) and the Improved Geometric-Optics Method  
12 (IGOM) ([Yang and Liou, 1996](#); [Bi et al., 2009](#)). The study indicates that the CGOM errors in  
13 inferring the optical thickness and effective diameters from the MODIS observations can be  
14 up to 20%, and on the order of  $10 \text{ Wm}^{-2}$  in ice cloud radiative forcing calculations.

15 The single-scattering properties of ice crystals given by the II-TM ([Bi and Yang, 2014](#)) can be  
16 considered as a benchmark, because the II-TM solves Maxwell's equations from first  
17 principles. Note, the II-TM is applicable to moderately large size parameters for which the  
18 IGOM has reasonable accuracy. However, due to the loose structure of the hexagonal  
19 aggregate considered in the THM, the computational memory used by the II-TM simulations  
20 increases significantly as the particle size increases. To optimize numerical computations, in  
21 this study the pseudo-spectral time domain method (PSTD) that is a numerically accurate  
22 technique ([Liu, 1997](#); [Liu et al., 2012a, 2012b](#)) is employed for the size parameters in the  
23 regime between the II-TM and IGOM simulations. The applicability and accuracy of these  
24 three methods have been extensively studied in previous studies, and, thus, are not [repeated](#)  
25 here. Without discussing technical details, we use a synergic combination of those three  
26 numerical models to minimize the bias introduced by light scattering simulations, and [the](#)  
27 single-scattering properties of the two particles habits with maximum diameters from 2 to  
28  $10000 \mu\text{m}$  at interested wavelengths are simulated. [Furthermore, the scattering properties](#)  
29 [involved in this study are associated with particles with random orientations.](#)

30 [Figure 7 shows the THM and the SCM extinction efficiencies, single-scattering albedos](#)  
31 [and asymmetry factors as functions of the particle maximum dimension. The single-](#)  
32 [scattering properties at three wavelengths, 0.67, 2.13 and  \$12.0 \mu\text{m}\$ , are illustrated. The](#)  
33 [SCM data are obtained from a combination of the II-TM and IGOM as shown by \[Bi et al.\]\(#\)](#)

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(2014). The II-TM, PSTD and IGOM are used to cover the entire practical size range that we consider for the THM. With the edge effect included in the results following the approach used in Yang et al. (2003), we can see that smooth curves are obtained for the extinction efficiency and the single-scattering albedo. The SCM and the THM show quite similar patterns for both the extinction efficiency and single-scattering albedo, whereas differences are evident in their asymmetry factors. At visible wavelengths, i.e. 0.67  $\mu\text{m}$ , the THM exhibits an almost constant asymmetry factor with a value of approximately 0.76, whereas the SCM values increase to almost 0.9 as the particle maximum dimension increases. Based on a climatic feedback sensitivity study, Stephens et al. (1990) suggest that a reduction of the Mie-theory-based asymmetry factor ( $\sim 0.87$ ) to a lower value of 0.7 may be necessary to achieve broad agreement between theory and observation. Thus, reduction of the asymmetry factor from its SCM value (as large as  $\sim 0.9$ ) to the THM ( $\sim 0.76$ ) is in alignment with the previous speculation.

For remote sensing applications, the bulk scattering properties of an ensemble of ice particles with specified size distributions are normally used. We assume a Gamma size distribution (Hansen and Travis, 1974) to integrate the bulk scattering properties of the THM. The dimensionless effective variance is assumed to be 0.1, and the effective diameter values increase from 10 to 180  $\mu\text{m}$  in steps of 10  $\mu\text{m}$  (McFarquhar and Heymsfield, 1998). The effective diameter of the particle is defined to be  $1.5 \times V/A$  following Foot (1988), where  $V$  and  $A$  are the volume and projected area of the particles.

Figure 8 shows the bulk non-zero phase matrix elements of the THM and SCM at the wavelengths of 0.67, 2.13 and 12.0  $\mu\text{m}$  in the case of an effective particle diameter of 30  $\mu\text{m}$  for both models. With the surface roughness considered in the THM, the halo peaks observed in the case of pristine hexagonal columns are smoothed out, and featureless phase matrix elements are obtained with THM. In Fig. 8, the bulk extinction efficiency ( $Q_{\text{ext}}$ ), single-scattering albedo (SSA), and asymmetry factor ( $g$ ) are also given for both the THM and SCM. We call special attention to the fact that the SCM has larger asymmetry factors at all three wavelengths. Although the oscillations of the phase matrix elements of the SCM consisting of pristine ice crystals can be smoothed out by surface roughness, the effects of surface structure on the values of the integral scattering properties (e.g., the extinction efficiency and the asymmetry factor) are relatively small.

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1 The bulk extinction efficiency, single-scattering albedo and asymmetry factor of the THM and  
 2 SCM are shown in Fig. 9 as functions of the effective particle diameter. As expected, the  
 3 extinction efficiency of the THM converges to 2 for larger particles, and the single-scattering  
 4 albedo given by the two models are quite similar. It should be noticed that, at the 0.67- $\mu\text{m}$   
 5 visible wavelength, the THM results give an almost constant asymmetry factor with a value of  
 6 approximately 0.76, whereas the values for the SCM increase as the effective particle  
 7 diameter increases (from 0.78 to almost 0.84). Larger asymmetry factors are also obtained  
 8 with SCM at the other two wavelengths.

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## 9 4.2 Comparison with observations

10 Compared with the large number of datasets on the microphysical properties of ice clouds and  
 11 images of their geometries, our observations and understanding of the optical properties are  
 12 relatively limited. The measured ice cloud phase functions have been widely used to construct  
 13 and verify the numerical models (Baran et al., 2001, 2012a), and results from the THM are  
 14 compared with those from laboratory and in situ measurements as well as satellite retrievals.

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15 The PN probe has been used in various laboratories and field campaigns to measure the  
 16 scattering phase function of ice clouds simultaneously with the size distribution. The PN  
 17 measurements suggest that ice clouds show featureless phase functions with a relatively flat  
 18 trend at backscattering angles (Barkey and Liou, 2001; Gayet et al. 1998, 2004; Febvre et al.,  
 19 2009). It should be noticed that unusual scattering phase functions with certain features were  
 20 also observed from some in situ measurements (Gayet et al., 2012; Baran et al., 2012), and we  
 21 will not consider these special cases when building our THM. However, we compare the  
 22 phase functions simulated based on the THM with measurements from laboratory and in situ  
 23 measurements at a visible and a near infrared wavelength. Barkey and Liou (2001) reported  
 24 the light scattering measurements of small ice crystals generated in a cloud chamber at a  
 25 wavelength of 0.67  $\mu\text{m}$ . In situ measurements of light scattering and microphysical  
 26 characteristics presented by Febvre et al. (2009) show the phase function of ice clouds at  
 27 0.804  $\mu\text{m}$ . In addition, both studies measured the ice crystal number concentrations. For the  
 28 case we use, Febvre et al. (2009) articulated that the effects of ice crystal shattering on the in  
 29 situ measurement are probably not very important, and, thus, they will not be considered in  
 30 our study. Figure 10 shows comparisons of the bulk phase functions between the THM and  
 31 the observations (left panels), and the corresponding number concentrations are given in the  
 32 right panels. The effective diameters of the two cases are approximately 5  $\mu\text{m}$  and 35  $\mu\text{m}$ ,  
 33 respectively. The THM exhibits a reasonable agreement in both cases. Note that the phase

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1 function of the in situ observations is normalized to the values at 30°. Both the modeled and  
 2 measured phase functions show similar and relatively smooth overall trends. The absence of  
 3 halo phenomena, i.e. scattering peaks commonly seen at 22° and 46°, is an indication of the  
 4 irregularity or surface roughness of ice crystals, and demonstrates the necessity and  
 5 importance of including the hollow structure and surface roughness in the THM. For the  
 6 laboratory results (upper panel), the THM slightly overestimates the phase function values  
 7 with scattering angles between 60° and 90°, but underestimates the values at scattering angles  
 8 larger than 100°. The modeled phase function shows larger values at scattering angles  
 9 between 60° and 120° compared with the in situ observations (lower panel). The asymmetry  
 10 factors of the laboratory and in situ measurement is approximately 0.76 and 0.79, respectively,  
 11 and the corresponding modeled values of the THM are 0.77 and 0.78.

12 Wang et al. (2014) retrieve the scattering phase function of ice clouds from satellite  
 13 observations. To reduce the impact from surface reflection and highlight thin ice clouds in the  
 14 upper troposphere, the reflectance at MODIS 1.38 µm channel is used to statistically derive  
 15 the scattering phase function, and the phase function values at 30 scattering angles between  
 16 90° and 180° are obtained for ice clouds over ocean and land. Figure 11 illustrates the  
 17 modeled (both single-column and two-habit models) and retrieved phase functions of ice  
 18 clouds, and the upper and lower panels are for the retrieved results respectively over ocean  
 19 and land. The red circles in the figure represent the averaged phase functions, and the error  
 20 bars indicate the standard deviations. Because the variation of a phase function with a change  
 21 of effective diameter for the THM can be ignored compared with the standard deviations of  
 22 the retrieved phase functions, especially for the backward scattering, the THM bulk phase  
 23 functions with  $D_{eff}$  of 50 and 100 µm are used for comparison. The phase functions of the  
 24 THM at the two sizes are almost indistinguishable except for the forward peaks, illustrating  
 25 that the THM-based phase function in the side and backward scattering directions are not  
 26 sensitive to particle effective sizes at visible and near-infrared wavelengths. The phase  
 27 function of the SCM at a single effective particle diameter of 50 µm is used. The phase  
 28 functions given by the THM at two sizes almost perfectly match the retrieved values over  
 29 ocean, which was also achieved by Wang et al. (2014) using three particle habits, whereas the  
 30 one based on the SCM shows significant oscillation in the region. For ice cloud over land, the  
 31 agreement between the satellite retrieval and numerical result is relatively limited, although  
 32 the modeled results are within the standard deviations of the retrieval over the entire backward  
 33 direction. The THM underestimates the phase function values for scattering angles larger than

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1 | 120°. Note that, even considering [the](#) phase functions of ice habits with three different degrees  
2 | of surface roughness given by Yang et al. (2013), Wang et al. (2014) cannot accurately match  
3 | the inferred results over land, and this may be due to the larger uncertainties associated with  
4 | the inferred phase function over land.

5 | Overall, considering the comparisons between the phase functions calculated based on the  
6 | THM and those from measurements or retrievals, the THM does show excellent  
7 | representation of the optical properties of ice clouds at visible and near infrared wavelengths.  
8 | However, we are far from claiming the THM to be an optimal ice cloud model, and the optical  
9 | properties of the THM over longer wavelengths are not verified because of a lack of  
10 | observations with which to compare [\(Cox et al. 2010\)](#).

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## 12 | **5 Satellite remote sensing applications**

13 | Both the microphysical and optical properties of the THM match the measurements closely,  
14 | and another important goal in the development of the THM is to improve the consistency in  
15 | the downstream remote sensing of ice cloud properties. One issue is the significant difference  
16 | between ice cloud optical thicknesses retrieved from solar and infrared [bands](#) (Wang et al.,  
17 | 2013b; Baum et al., 2014). The polarization properties observed from the PARASOL satellite  
18 | are an important aspect widely used to test ice cloud models (Baran, 2009; Cole et al., 2013).

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19 | [Note, the plane-parallel radiative transfer model with single cloud layer is assumed in this](#)  
20 | [study, and the vertical inhomogeneity and 3-dimensional effects of clouds \(Yang et al., 2001;](#)  
21 | [Fauchez et al., 2014\) are not considered in this study.](#)

### 22 | **5.1 Comparison between the solar- and IR-band retrieved optical thicknesses**

23 | Two popular methods are normally used to retrieve ice cloud properties from satellite  
24 | observations: the first is a bi-spectral method employing solar reflectance bands (the solar-  
25 | band retrieval) (Nakajima and King, 1990); and the second is based on the IR bands (the IR-  
26 | band retrieval) (Inoue, 1985; Heidinger and Pavolonis, 2009). To infer the optical thickness  
27 | and effective particle diameter of ice clouds, an ice cloud model, i.e., the optical properties  
28 | obtained from the given particle habit or habits, is fundamental for both the solar-band and  
29 | IR-band retrievals. Thus, identical cloud properties are expected to result from using the solar-  
30 | band and IR-band retrievals for the same target based on the same ice model; however, this  
31 | does not hold true for most ice cloud models. The optical thicknesses retrieved from IR-band  
32 | observations are generally smaller than those from solar-band retrievals (Baum et al., 2014).

1 Specifically, the solar-band retrieval is based on two solar reflectance bands, i.e., a weakly  
2 absorbing, visible or near-infrared window band (VIS/NIR) mainly sensitive to the cloud  
3 optical thickness  $\tau$ , and an ice absorbing shortwave infrared (SWIR) band sensitive to both  $\tau$   
4 and  $D_{eff}$ . The approach based on a VIS/NIR and a SWIR band is used by the MODIS  
5 operational cloud-property retrieval (Platnick et al., 2003). Another method to obtain  $\tau$  and  
6  $D_{eff}$  is the split-window technique (Inoue, 1985) based on multiple IR window channels (e.g.  
7 8.5, 11.0 and 12.0  $\mu\text{m}$  for the MODIS observations), and the application of the algorithm can  
8 be found in the Advanced Very High Resolution Radiometer (AVHRR) (Heidinger and  
9 Pavolonis, 2009), as well as some studies based on MODIS observations (Minnis et al., 2011;  
10 Wang et al., 2013b). Note that the IR-band retrieval is not strongly sensitive to the explicit  
11 scattering properties, because of the strong absorption within ice crystals; whereas, the  
12 scattering properties are essential for the solar-band retrievals. Thus, the optical properties of  
13 the ice cloud model at the solar bands become the key parameters to determine the spectral  
14 consistency of the models.

15 A case study, using MODIS observations, is conducted to assess the spectral consistency of  
16 optical thickness retrievals based on both solar-band and IR-band observations. The solar-  
17 band retrieval uses MODIS reflectances at 0.86 and 2.13  $\mu\text{m}$  bands and the fast radiative  
18 transfer model (FRTM) developed by Wang et al. (2013a). By using pre-computed  
19 bidirectional reflectance/transmittance distribution functions and a numerical integral over a  
20 twisted icosahedral mesh, the FRTM is approximately two orders of magnitude faster than  
21 that of the standard 128-stream discrete ordinates radiative transfer code. The IR-band  
22 retrieval is based on the three MODIS IR bands at 8.5, 11, and 12  $\mu\text{m}$ , and a fast high-spectral  
23 resolution radiative transfer model (HRTM) developed by Wang et al. (2013b), is used to  
24 simulate radiances and resulting brightness temperatures at the three bands. The HRTM  
25 accounts for the gas absorption using a pre-computed transmittance database, and the optical  
26 properties of the ice cloud model are used to calculate the look-up-tables for cloud reflectance,  
27 transmittance, effective emissivity, and effective temperature functions.

28 Datasets from the Aqua/MODIS and the Modern Era Retrospective-Analysis for Research and  
29 Applications (MERRA) are used for the retrievals. The MODIS level-1B calibrated radiances  
30 (MYD021KM) product provides top of the atmosphere radiance/reflectance and brightness  
31 temperatures for the solar and IR bands. The 1km-resolution geolocation and solar-satellite  
32 geometry are obtained from the MOD03 datasets. The MODIS level-2 cloud product  
33 (MYD06) is used to give cloud phase, cloud optical thickness and cloud top height. The over-

ocean pixels identified as ice cloud by MYD06 are retrieved, and because the IR-band retrieval is inherently less sensitive to optically thick clouds, the cases with MODIS optical thicknesses larger than 5 are ignored. The atmospheric profile used for radiative transfer simulations and gas absorption is collocated from the MERRA data. The MERRA 3-hourly instantaneous atmospheric profile data (Int3\_3d\_ams\_Cp) provides temperatures, water vapor densities, and ozone densities at 42 pressure levels with a spatial resolution of  $1.25^\circ \times 1.25^\circ$ .

Figure 12 shows the retrieval results based on the single-column model and the THM, and the Aqua/MODIS granule used. The case study is carried out for a granule at 03:50 UTC on 24 February 2014 and shown in panel (a). Panel (b) shows the retrieved optical thicknesses of thin ice cloud pixels, and the results are obtained based on the present solar-band retrieval with the THM. Approximately half of the granule pixels show relatively small optical thicknesses (less than 5 from MYD06 Collection 6 data), and are used for both the solar- and IR-band retrievals. Panels (c) and (d) are comparisons of optical thicknesses inferred from the solar-band and IR-band retrievals. The color contours indicate the occurrence of optical thickness values from the two retrievals, and the warm colors indicate higher values of the occurrence frequency. To facilitate interpretation of the results, a 1:1 line is included in the figure. Based on the single-column model, the solar-band retrieved optical thicknesses are clearly shown to be higher than the IR-band, and the differences increase as the optical thickness increases. However, the THM shows much better spectral consistency with the high occurrence frequency closely following the 1:1 line. This is mainly because of the relatively small asymmetry factors of the THM at visible wavelengths (as shown in Fig. 9), which yield larger optical thickness from the solar-band retrieval compared with those based on the SCM.

## 5.2 Comparison between the simulated and observed polarized reflectivities

The polarization property of ice clouds obtained from the PARASOL satellite is important and useful perspective for evaluating the performance of numerical models, because the measured polarized reflectivity is very sensitive to the  $P_{12}$  element of the phase matrix. We use PARASOL observations over ocean at  $0.865 \mu\text{m}$  from 1 August 2007, and the dataset details can be found in Cole et al. (2013). Data from only one day of observations is used, because a previous study (Baum et al., 2014) indicates that the occurrence frequency of the PARASOL polarized reflectivities exhibits a very similar pattern over time. A vector adding-doubling radiative transfer model is used (Huang et al., 2015), and the simulation assumes a single-layer ice cloud with an optical thickness of 5 at a height of 9 km over an ocean surface. Cole et al. (2013) demonstrated that an ice cloud with an optical thickness of 5 is sufficient for the

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1 polarized reflectivity to be saturated. The simulated polarized reflectivities based on the THM  
2 are not strongly sensitive to the particle effective diameter, and the optical properties of ice  
3 clouds with effective diameters of 50  $\mu\text{m}$  are used.

4 [Figure 13](#) illustrates ice cloud polarized reflectivities from the PARASOL measurements over  
5 ocean (color contours) and the simulations (black dots) based on the bulk scattering properties  
6 developed using the single-column (left panel) and two-habit model (right panel). The same  
7  $D_{\text{eff}}$  of 50  $\mu\text{m}$  is used for both models. The color contours in Fig. [13](#) are the occurrence  
8 frequency of the PARASOL polarized reflectivities of ice clouds over ocean, and the red color  
9 indicates the region of high occurrence for the measurements. The black dots in the figure  
10 correspond to the model calculations of a given set of solar-satellite geometries (i.e., solar  
11 zenith, viewing zenith and relative azimuth angles), and 3000 different geometries from the  
12 PARASOL data are used for the simulations. Due to the scattering peaks in both the phase  
13 function and the other phase matrix elements for the smooth hexagonal column, the single-  
14 column-based results show significant oscillations as well, and exhibit very different  
15 variations than the PARASOL observations. However, the numerical results based on the  
16 THM accurately match the satellite observations over the entire range of scattering angles.  
17 Considering similar patterns in the occurrence frequency of the PARASOL polarized  
18 reflectivities over time and the similar scattering phase matrices of the THM at different  $D_{\text{eff}}$ ,  
19 the THM is expected to perform consistently in matching the observed polarized reflectivities  
20 of ice clouds.

21 Again, the THM not only infers similar optical thickness from the solar-band and IR-band  
22 retrievals, but also provides polarization properties similar to satellite observations. The  
23 excellent performance of the THM indicates a great potential for the remote sensing  
24 applications. More research is needed to [further confirm](#) whether the THM is a robust model  
25 for referring ice cloud properties based on observations from different wavelengths and  
26 sensors.

## 28 6 Conclusion

29 This study constructs an ice cloud model with two particle habits, and the performance and  
30 consistency of the THM in representing the microphysical and optical properties of ice clouds  
31 are investigated in detail. The THM includes a hexagonal column with an aspect ratio of unity  
32 and an aggregate containing 20 hexagonal columns, and both hollow structure and surface

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roughness are considered. The habit fractions of the two particle habits are determined to match the in situ measurements of ice cloud microphysical properties and the general trends, from analyses of particle imagery data sets, in the percentages of simple and complex crystals (i.e., more complex particles as particle maximum dimension increases). The simulated  $IWC$  and  $D_{mm}$  values based on the THM agree closely with the in situ data sets. Furthermore, an expression for ice crystal volume as a function of particle maximum dimension is also presented, which leads to the aforementioned agreements in the cases of  $IWC$  and  $D_{mm}$ .

The optical properties of the THM are calculated with a combination of the II-TM, PSTD and IGOM models for particle sizes from 2 to 10,000  $\mu\text{m}$  at wavelengths of interest, and the data library contains the extinction coefficient, single-scattering albedo, asymmetry parameter, and six independent nonzero phase matrix elements. The simulated phase functions based on the THM show excellent agreement with both the laboratory and in situ measurements at 0.67 and 0.80  $\mu\text{m}$  as well as satellite retrievals at 1.38  $\mu\text{m}$ .

In addition to the excellent performances in representing the microphysical and optical properties of natural ice clouds, an initial retrieval analysis demonstrates that the THM significantly improves the spectral consistency in the remote sensing of ice cloud properties from different satellite sensors or wavelengths. The optical thicknesses retrieved based on the two MODIS solar bands show close agreement with those inferred from the MODIS IR window measurements. Furthermore, a comparison between the simulated polarized reflectivities based on the THM and those measured from the PARASOL satellite indicates that the THM can closely represent the polarization properties of ice clouds.

We focused on the development and performance of the THM in representing ice cloud properties, but their effect on radiative forcing is not tested. Developing the THM optical property database over the whole spectral domain, obtaining the parameterized optical properties, performing retrievals over all ranges of viewing and illumination conditions, and investigating the radiative effects in the RTMs and GCMs are straightforward and will be discussed in further studies.

Furthermore, we would like to emphasize that the SCM used for comparison is based on pristine ice crystals with smooth surfaces and particular aspect ratio values, and the findings based on the assessment of the performance of SCM in remote sensing applications may not necessarily be applicable to a different single column/plate model, particularly, when particle surface roughness is considered.

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## 1 Appendix A: Geometry of the hexagonal aggregate

2 The THM uses an aggregate of hexagonal columns as the complex particle, and 20 hexagonal  
3 columns with different sizes and aspect ratios are used to build the aggregate. Four steps are  
4 necessary to build the final aggregate as shown in Fig. 2(b).

5 First, we randomly generate each of the column elements by giving its length and aspect ratio:

$$6 \quad L = [(1 - A_1) + 2A_1\xi_1]L_o, \quad (A1)$$

7 and

$$8 \quad \frac{2a}{L} = A_2 + (1 - A_2)\xi_2, \quad (A2)$$

9 where  $A_1$  and  $A_2$  are constants related to the geometries of the hexagonal columns, and  $\xi_1$  and  
10  $\xi_2$  are independent random numbers distributed uniformly in  $[0, 1]$ .  $A_1$  determines the [range](#)  
11 of the column sizes, and  $A_2$  limits the minimum aspect ratio. We use values of 0.2 and 0.8 for  
12  $A_1$  and  $A_2$ , respectively, to generate the 20 hexagonal columns used to build the aggregate.  
13 Here,  $L_o$  is the column average [length](#). Once the aggregate is generated, the dimensions can be  
14 scaled to fit the ice crystal size in the single-scattering computations.

15 Secondly, the 20 hexagonal columns are attached to form an aggregate without overlapping.  
16 An improved particle-cluster aggregation algorithm, normally used for a fractal aggregate  
17 (Filippov et al., 2000; Liu et al., 2012c) with spherical monomers, is adapted, and the only  
18 difference is that each hexagonal column is randomly rotated to attach to another. For  
19 simplification, the rotation is managed to make only a vertex and a surface point-attached  
20 (without overlapping or surface-surface attachment). [The criteria used by Xie et al. \(2011\) to](#)  
21 [detect overlapping between two hexagonal particles are used to avoid intersecting particle](#)  
22 [faces](#).

23 The third step is to introduce a hollow structure into the hexagonal columns by replacing one  
24 hexagonal surface of each column using the hollow structure as shown in Fig. 1. The depth of  
25 the hollow is fixed at  $d/L=0.25$ . To ensure the attachment of the aggregate, the hollow  
26 structure is only added to a surface without any attached particles, and a hexagonal column  
27 with both hexagonal surfaces connected with other monomers is kept solid. The aggregate has  
28 only one solid column.

29 As a final step, surface roughness is added to the particle by replacing each of the smooth  
30 surfaces with roughened ones. In the II-TM and PSTD simulations, explicit particle  
31 geometries are achieved by following the rough surfaces defined by Liu et al. (2013). The

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1 titled-facet approximation (Yang et al., 2008) is applied for the IGOM simulations, because of  
2 the efficiency without significant loss of accuracy (Liu et al., 2013).

3 The completed roughened aggregate is shown in panel (b) of Fig. 2. Numerically, the  
4 aggregate is defined by the explicit vertices and surfaces of the columns. Thus, the volume  
5 and averaged projected area of the aggregate can be rigorously and numerically calculated,  
6 and will be used for the microphysical and optical properties of the THM. Note that the  
7 surface roughness has little effect on the microphysical properties of the THM, but its  
8 influence is shown in the optical properties.

9 The geometrical parameters used to determine the aggregate geometry are listed in Table A1,  
10 including the lengths, aspect ratios, coordinates of three points and hollow depths of 20  
11 monomers. It should be noticed that all length parameters are normalized to  $L_0$ . The  
12 coordinates of three points for each monomer, i.e. the center of a column (point O in Fig. 1),  
13 the center of a particle face (point A in Fig. 1), and a vertex (point B in Fig. 1) are listed. The  
14 last column in Table A1 indicates whether a monomer has a hollow structure as shown in Fig.  
15 1. The maximum dimension of the aggregate is numerically calculated, which is  $7.137L_0$ . In  
16 addition, the volume and projected area of the aggregate are numerically found to be  
17  $0.0255D^3$  and  $0.260D^2$ , respectively.

18 Although Baran (2009) demonstrated that adding hexagonal monomers with the element  
19 number beyond 3 does not significantly alter the asymmetry factor, in this study we select 20  
20 monomers for three reasons: 1) as an appropriate particle geometry is sought to mimic the  
21 complicated morphologies of realistic aggregates within ice clouds and the use of only a few  
22 monomers seems to be an oversimplification; 2) an aggregate geometry corresponding to a  
23 potentially lowest value of the asymmetry factor is desired, and it is found that the asymmetry  
24 factor slightly decreases as the number of monomers increases; 3) with the trial and error  
25 method, the use of 20 monomers is optimal in terms of the balance between the computational  
26 effort in light scattering simulation and the performance of the particle habit model in fitting  
27 the measured microphysical properties (specifically,  $IWC$  and  $D_{mm}$ ).

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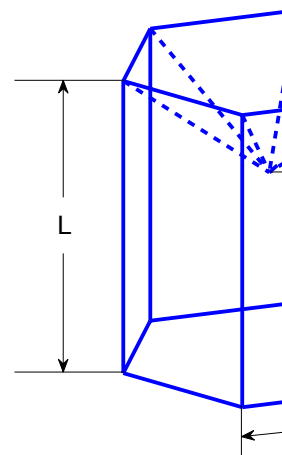
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Table 1. Relative errors of the THM in representing the microphysical properties obtained during 11 field campaigns. The mean and standard deviation (STD) of the relative errors of the theoretical median mass diameter ( $D_{mm}$ ) and ice water content ( $IWC$ ) are listed. The details of the 11 field campaigns can be found in Heymsfield et al. (2013) and Baum et al. (2014).

Field Campaign	Number of PSDs	Relative Errors of $D_{mm}$ (%)		Relative Errors of $IWC$ (%)	
		Mean	STD	Mean	STD
ARM-IOP	1420	-0.56	4.4	-2.4	7.3
TRMM	201	4.0	4.6	0.28	3.2
CRYSTAL-FACE	221	4.4	4.8	148	50
Pre-AVE	99	2.3	4.1	89	24
MidCiX	2968	-1.3	2.9	4.5	16
ACTIVE Hector	2583	2.1	5.4	17	17
ACTIVE Monsoon	4268	0.75	4.0	14	9.3
ACTIVE Squall Line	740	-0.56	4.0	9.7	11
SCOUT	358	-17	6.6	25	14
TC-4	877	-2.6	2.1	18	11
MPACE	671	-1.7	3.8	3.2	7.2
All	14406	-0.27	5.2	13	24

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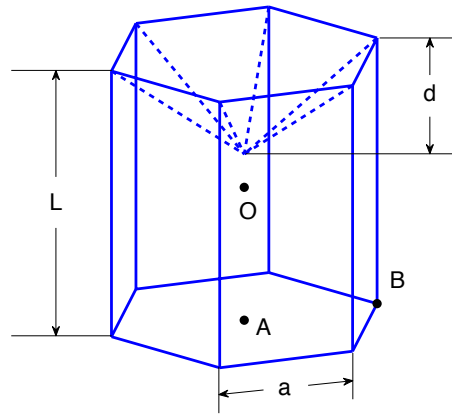
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Table A1. Geometric parameters of the hexagonal aggregate with twenty monomers. All length parameters are normalized by  $L_o$ , the average monomer length. The last column indicates whether the monomer has a hollow structure as shown by Fig. 1 (“Y” indicates “yes”, and “N” indicates “no”). Note, only the third monomer does not have a hollow structure.

NO.	L/ $L_o$	2a/L	O			A			B			H
			x/ $L_o$	y/ $L_o$	z/ $L_o$	x/ $L_o$	y/ $L_o$	z/ $L_o$	x/ $L_o$	y/ $L_o$	z/ $L_o$	
1	0.823	0.854	0.063	0.339	0.093	0.063	0.339	-0.319	0.239	0.035	-0.319	Y
2	0.974	0.859	-0.660	-0.082	-0.150	-0.548	0.045	0.307	-0.406	-0.342	0.379	Y
3	1.161	0.817	-0.008	-0.695	0.142	-0.098	-0.659	-0.431	-0.162	-0.191	-0.391	N
4	1.155	0.903	-0.642	-0.087	0.958	-1.028	-0.448	1.189	-0.876	-0.306	1.667	Y
5	1.168	0.870	0.317	-0.229	1.400	0.310	-0.756	1.148	0.307	-0.975	1.607	Y
6	1.176	0.944	1.134	-0.165	0.467	1.442	-0.046	0.953	1.484	0.485	0.797	Y
7	0.824	0.821	2.188	0.162	0.969	2.531	0.360	0.856	2.450	0.314	0.530	Y
8	0.955	0.985	-0.294	-0.248	2.322	-0.750	-0.338	2.209	-0.724	-0.748	2.438	Y
9	0.932	0.836	1.9678	0.957	0.626	1.891	1.068	0.180	2.274	1.106	0.124	Y
10	1.174	0.915	0.143	0.929	2.281	0.082	0.692	2.815	-0.041	0.220	2.591	Y
11	0.883	0.829	-1.144	0.326	-1.051	-0.752	0.125	-1.071	-0.888	-0.160	-0.888	Y
12	1.069	0.884	0.337	-0.547	-1.153	-0.095	-0.454	-0.854	-0.051	-0.882	-0.658	Y
13	0.922	0.816	-0.458	-0.558	-1.790	-0.758	-0.770	-2.068	-1.038	-0.569	-1.918	Y
14	1.173	0.905	-0.323	1.140	-1.166	-0.092	1.572	-0.842	-0.230	1.314	-0.399	Y
15	1.079	0.933	0.594	0.800	-1.809	0.588	0.803	-1.270	0.997	1.095	-1.267	Y
16	0.830	0.873	-0.262	0.824	3.369	-0.574	0.931	3.620	-0.368	1.192	3.764	Y
17	0.970	0.992	0.256	-0.278	-2.286	0.620	-0.053	-2.059	0.812	0.066	-2.484	Y
18	1.156	0.973	-0.320	-1.560	-1.552	-0.332	-1.467	-2.122	-0.781	-1.803	-2.168	Y
19	1.099	0.943	-0.445	0.479	-2.554	-0.310	0.515	-2.023	-0.666	0.885	-1.957	Y
20	1.130	0.885	-1.378	-0.442	2.205	-1.636	-0.572	2.690	-1.638	-0.088	2.818	Y



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3 Figure 1. Geometry of a hexagonal column with a hollow structure.  $L$  is equal to the  
4 maximum dimension  $D$  for the hexagonal column.

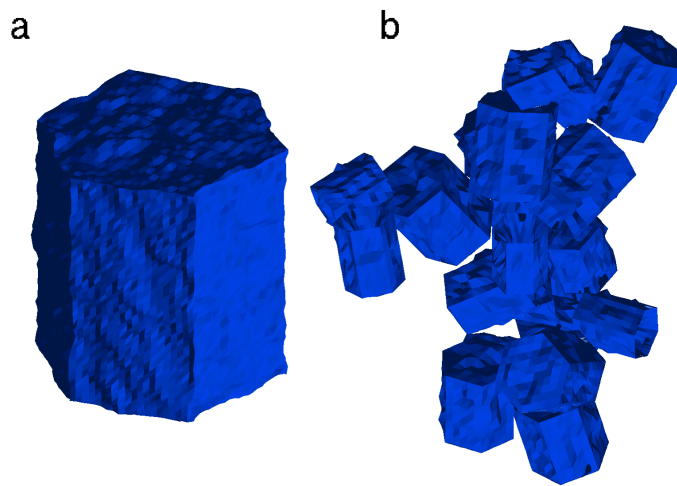


Figure 2. Particle geometries for the two-habit model (THM): (a) single hexagonal column with an aspect ratio of unity, and (b) hexagonal aggregate with 20 solid or hollow columns.

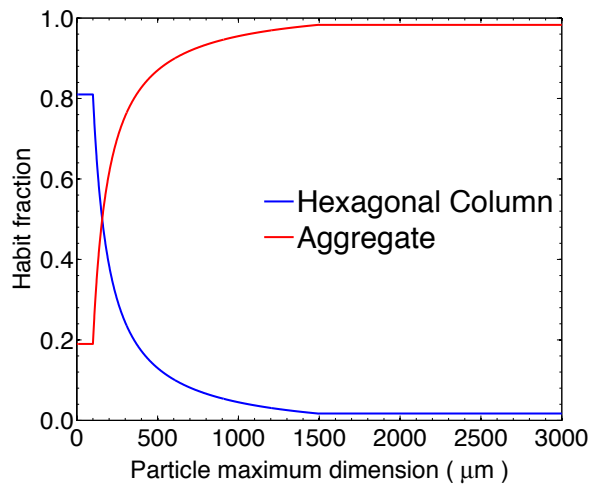
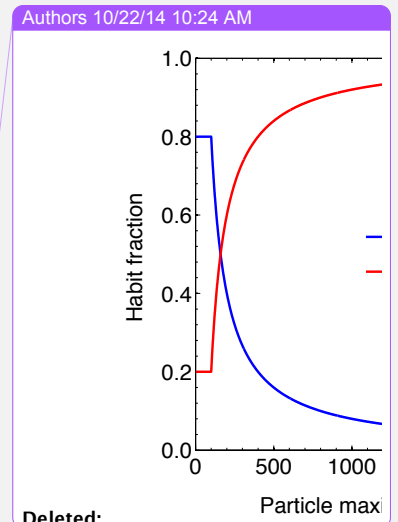


Figure 3. Ice crystal habit fraction as a function of particle maximum dimension for the two-habit model.



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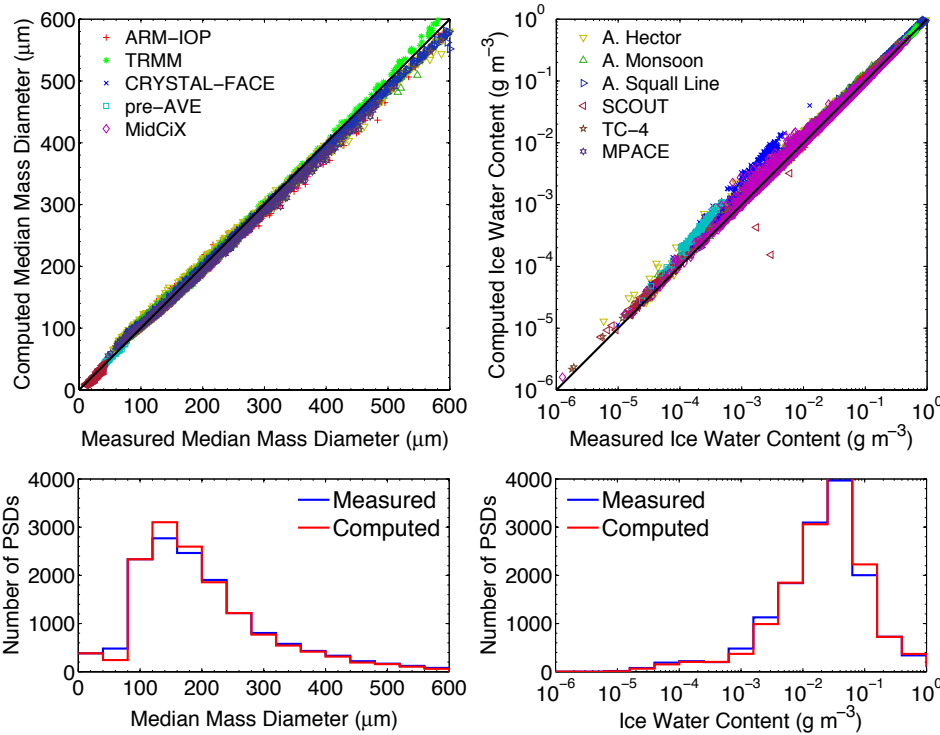
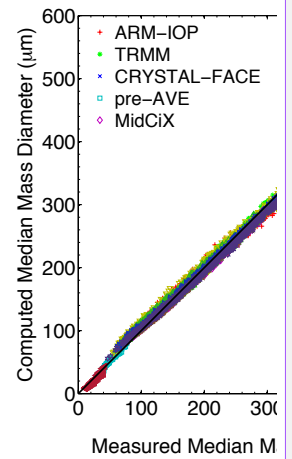


Figure 4. Upper panels: Comparison between the measured and calculated microphysical properties ( $D_{mm}$  and  $IWC$ ) for each of the PSDs from 11 field campaigns. Lower panels: Histograms of the distributions of the measured and calculated  $D_{mm}$  and  $IWC$ .

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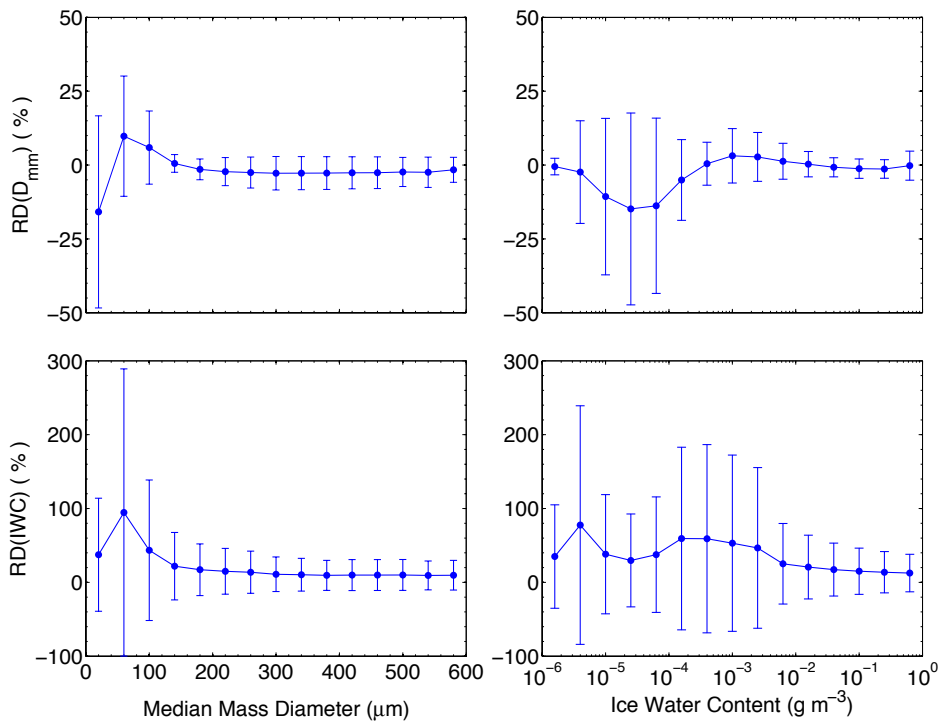
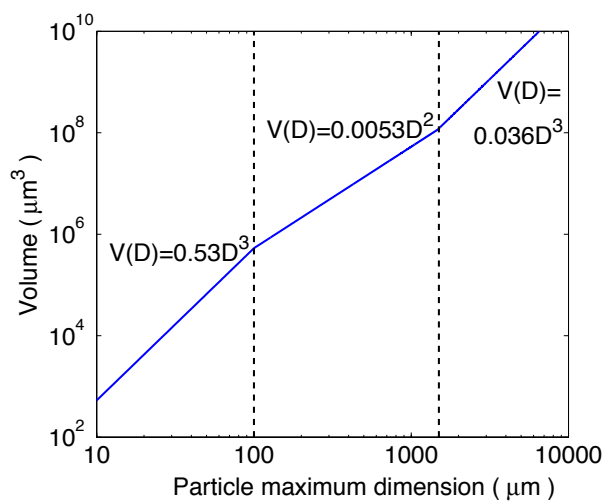
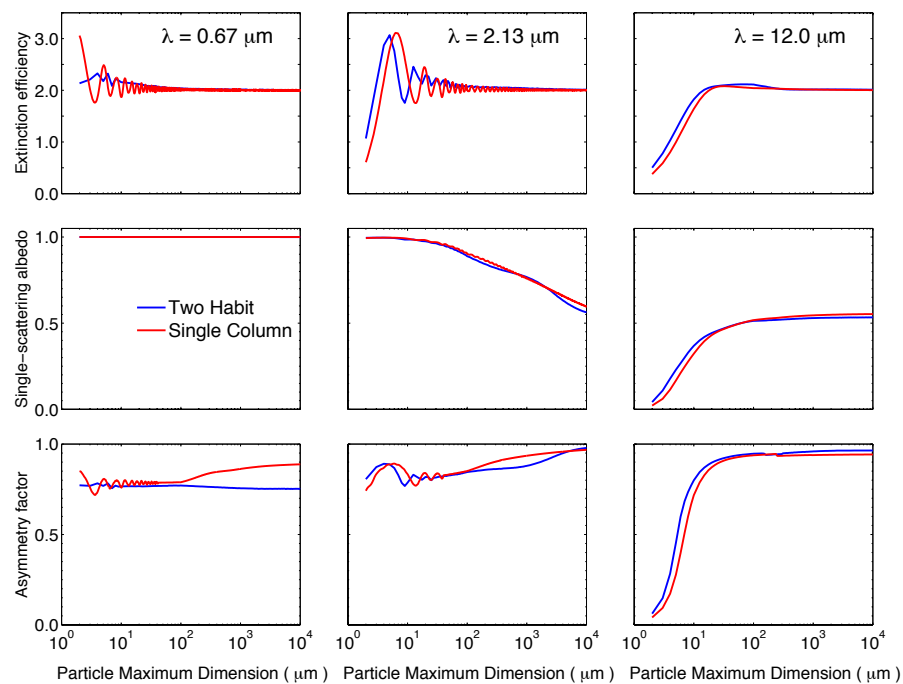


Figure 5. Relative differences (RD) of the calculated microphysical properties at different bins of median mass diameter (left panels) and ice water content (right panels). Error bars indicate the corresponding standard deviations.



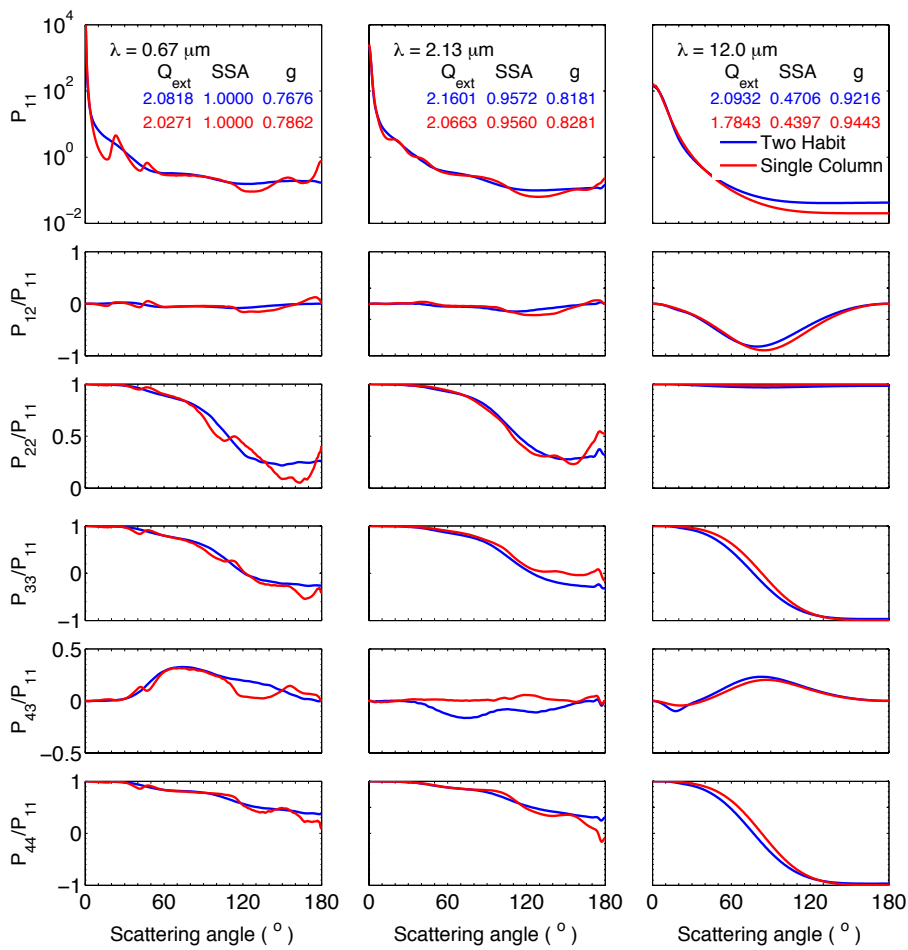
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Figure 6. Relationship between ice crystal volume and maximum dimension.



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Figure 7. Extinction efficiency (upper), single-scattering albedo (middle), and asymmetry factor (lower) of the single-column and two-habit model as functions of particle maximum dimension at wavelengths of 0.67, 2.13 and 12.0  $\mu\text{m}$ .



**Figure 8.** Comparison of bulk non-zero phase matrix elements of the two-habit model and the single-column model with an effective diameter of 30 μm at wavelengths 0.67, 2.13 and 12.0 μm.

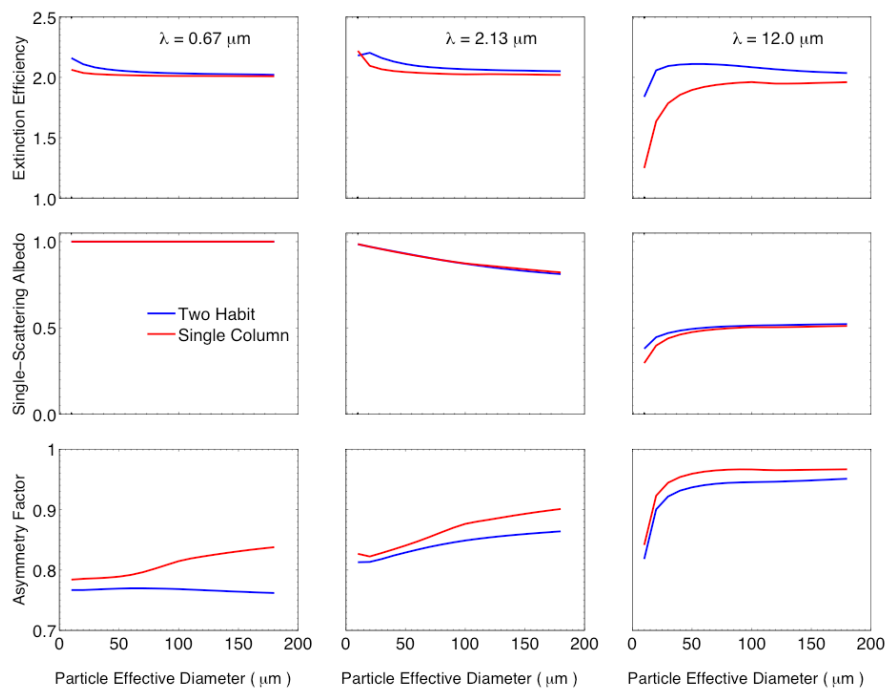
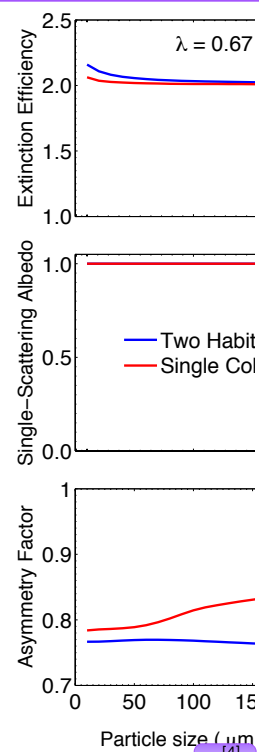


Figure 9. Bulk extinction efficiency (upper), single-scattering albedo (middle), and asymmetry factor (lower) of the two-habit model and the single-column model at wavelengths 0.67, 2.13 and 12.0  $\mu\text{m}$ .

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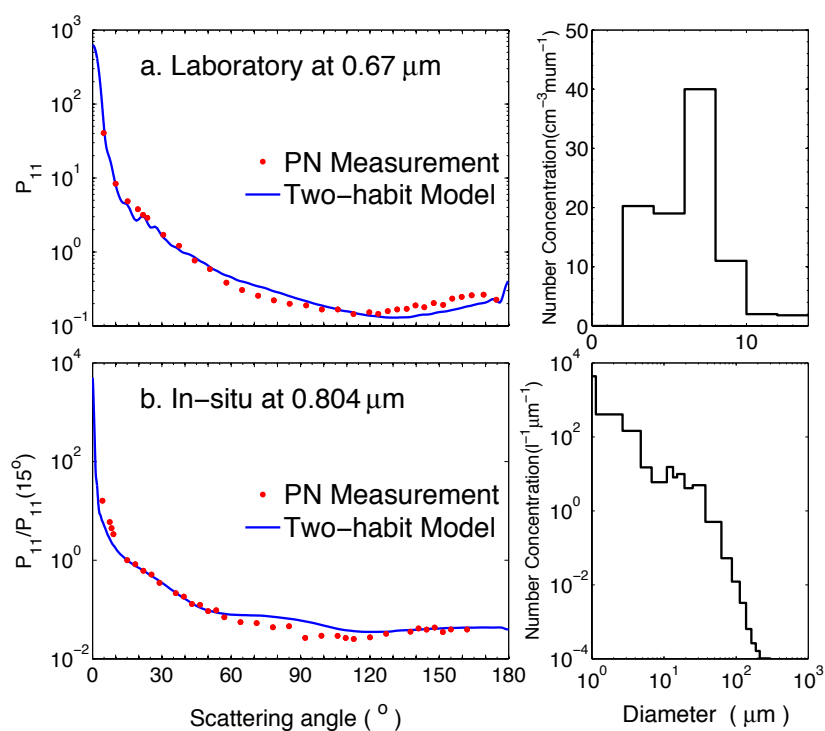


Figure 10. Comparison between the phase functions (left panels) from the two-habit model and the polar nephelometer (PN) measurements from: (a) laboratory at a wavelength of 0.67 μm and (b) in situ at a wavelength of 0.804 μm. The right panels are observed particle number concentration of the corresponding measurements.

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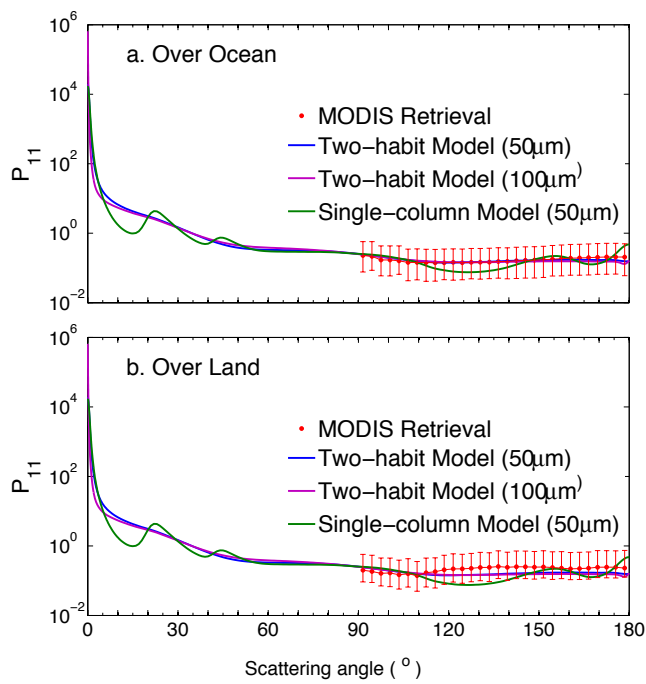
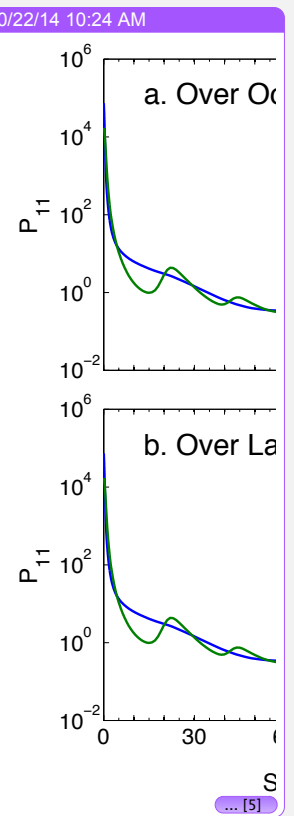


Figure 11. Comparison between the phase functions from the numerical models (both single-column and two-habit models) and MODIS retrieval at a wavelength of 1.38  $\mu\text{m}$  (Wang et al., 2014). The effective diameter used for the THM is 50  $\mu\text{m}$ .



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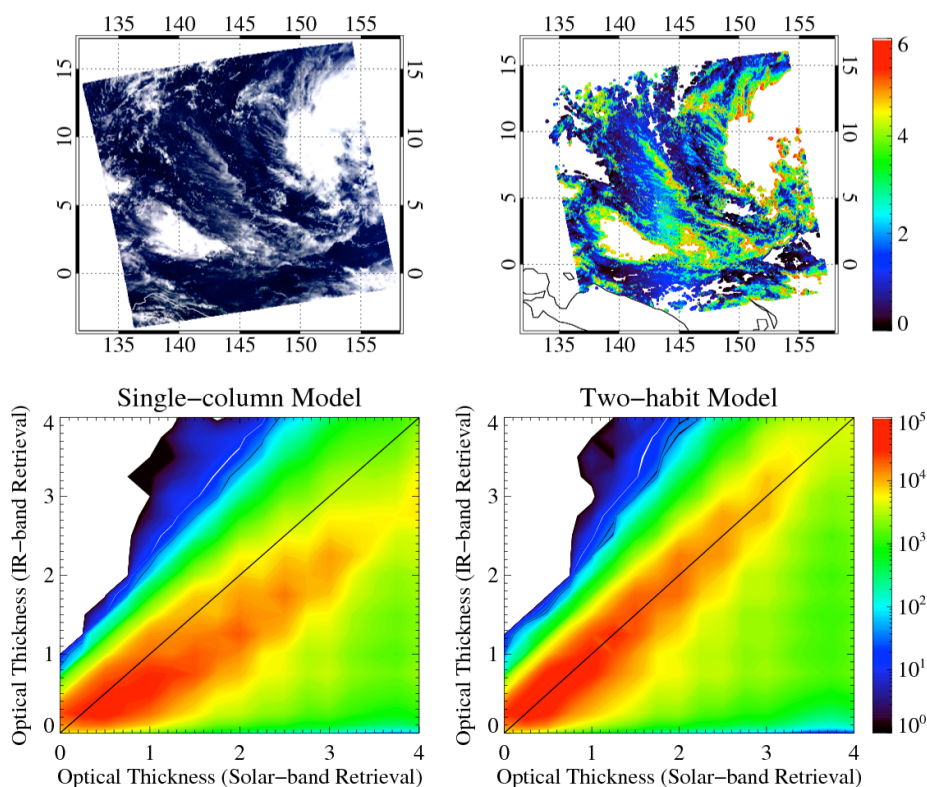


Figure 12. (a) RGB image of an Aqua/MODIS granule from 24 February 2014 at 03:50 UTC. (b) Retrieved optical thickness of thin ice clouds. (c) and (d) are comparisons of ice cloud optical thicknesses retrieved from the MODIS solar bands and IR bands, and the results are based on the single-column model and two-habit model, respectively. The histograms illustrate occurrences of thin ice cloud pixels, and the red color corresponds to the high frequency of occurrence.

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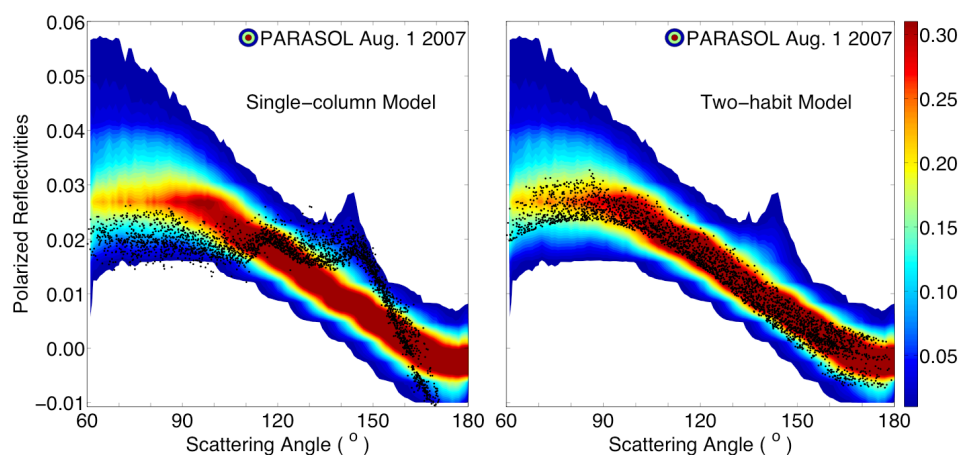
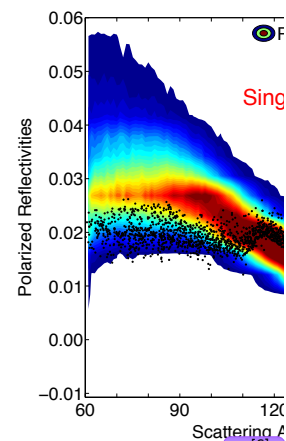


Figure 13. Comparisons between the normalized polarized reflectivities obtained from one day of PARASOL data over ocean (color contours) and calculations (black dots) based on the single-column model (left) and the two-habit model (THM, right).

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