Anonymous Referee #2

General comments

The paper demonstrates that the single-scattering properties of small ice crystals, with a maximum dimension of 50 μm, are highly dependent on the assumed idealized shape. This dependence on shape becomes exacerbated when the authors introduce their own small idealized ice crystal model called the "budding Bucky ball", which is based on laboratory grown ice crystal analogues described in Ulanowski et al. (2004). The Ulanowski paper demonstrated, in part, that the then current generation of in situ microphysical probes may not be resolving the detailed structure of small ice crystals due to their limiting resolving paper. This lack of resolution causes the automatic classification algorithm, of say the CPI, to report back "quasi-spherical" ice crystals. This appearance of "quasi-spherical" ice crystals has led a number of authors to assume spheres, spheroids, and Chebyshevs to represent the light scattering properties of small ice crystals. More recently more complex models such as Gaussian random spheres and Droxtals have been assumed. None of the currently assumed idealized small ice crystal models contain protruding hexagons from a common center similar to that described in Ulanowski et al. (2004). The authors demonstrate substantial single scattering (phase function and asymmetry parameter) differences between their Bucky ball model and previously developed small ice crystal models. For instance, for the most non-spherical particles the uncertainty in g can be as large as 14% which would imply an uncertainty in the instantaneous short-wave forcing of about ±42 Wm^{-2}, such an implied uncertainty is very large and g for small ice crystals must be further constrained.

The question is how is it best to do this? The problem with such uncertainties is that they generate a cottage industry of papers which essentially just tweak one of the parameters, and this can go on ad infinitum, without adding anything of real significance, just a series of tweaks with endless sensitivity studies without constraint. What is the point of this? Is it not better to obtain further atmospheric state parameters coupled with ice crystal growth models so that small ice crystals can be predicted from first principles rather than end up with n small ice crystal models? The authors do elude to this in their conclusion, but why not just do it? The point of the Ulanowski paper was to demonstrate *the potential deficiency of current imaging probes by showing for the same resolution, as the imaging probe, a more complex particle may appear "quasispherical". The model adopted by the authors was not meant to characterise actual small ice crystals that might exist in cirrus, it was merely used to demonstrate the potential inadequacy of current imaging probes. Moreover, the Ulanowski paper does not mean that small ice crystals are not "quasi-spherical". At the moment we simply do not know how non-spherical small ice crystals may or may not be. The present paper is found to be unconvincing and further analysis is required before the paper can be accepted for publication, the major concerns are listed below together with a number of more minor issues.*

We thank the review #2 for the careful reading of the manuscript and the many helpful suggestions that have improved the quality of the manuscript.

We absolutely agree with the reviewer's concerns on small ice crystals. We have tried to search ice crystal growth models to predict shapes of small ice crystals. However, current growth models cannot predict the shapes of small ice crystals. At this point, our knowledge of ice crystal growth is very poor. Because of this, current idealized models that represent the shapes of small ice crystals depend heavily on in-situ measurements rather than laboratory experiments. For example, a droxtal is the most widely used model for small ice crystals. This model has been developed based on images of ice crystals from ice fogs. Right now, almost all studies that require a database for the single-scattering properties of small ice crystals use that of Yang et al. (2005) for droxtals. Many conclusions about the importance of small ice crystals for both climate and remote sensing studies have been reached assuming that small ice crystals are droxtals. Thus, it is sensible to examine fundamental assumptions used in such studies, namely small crystal shape can be adequately described by droxtals. If we are forced to wait to publish uncertainties about these fundamental assumptions until our knowledge of nucleation and initial ice crystal growth is advanced enough to predict the shape of small ice crystals or until the next generation of cloud probes is able to better resolve the shapes of small crystals, some potentially misleading conclusions might be ingrained into the scientific literature. Not only is the budding Bucky ball a potentially better representation of a small crystal, but also this paper examines the sensitivity of the single-scattering properties to the shape of small crystals: both are original contributions. We certainly agree that further study is needed to test the suitability of the budding Bucky ball in predicting satellite radiances or in-situ measurements of distributions of scattered radiation. We want this paper published so that others can be aware of these issues and also include 3B in their tests. This is better than waiting for several years and publishing the complete solution to how small crystals impact radiation.

It should also be noted that we don't argue that a budding Bucky ball represents all real small ice crystals in ice clouds. At this point, we don't know. Because small ice crystals from Bailey and Hallet (2009) and ice analogues from Ulanowski et al. (2004) showed budding arms on small ice crystals, it is possible that this model better describes the shape of small ice crystals than those currently used. We attempted to quantify differences on single-scattering properties due to arm structures on small ice crystals. Further, there has been no study to compare single-scattering properties between current models (i.e., Chebyshev particles, droxtals, Gaussian random spheres). Thus, this study is meaningful because it compares current models and it emphasize possible errors due to uncertainties in measuring small ice crystals using state-of-the-art cloud probe, which will stimulate to improve current cloud probes.

Major concerns

1. The authors do not present any quantitative evidence that the Bucky ball is representative of real small ice crystals that might exist in cirrus. All that is cited is a paper by Bailey and Hallet (2009) based on laboratory grown ice crystals, some of which appeared to have budding arms. Can the authors be more quantitative here, what fraction of ice crystals had budding arms and how many arms were observed? What were the assumed growth conditions for these ice crystals in terms of temperature and humidity? What were the activation nuclei or what was the doping property that was put onto the substrate? Are the resulting ice crystals a function of doping property? Laboratory experiments such as these may not be representative of actual cirrus conditions due to the nature of particle growth in such experiments.

From Bailey and Hallet (2009), crystals were grown from 5 μm drops generated with a sonic nebulizer that nucleated, either homogeneously or by contact with background aerosol, while falling 5 m in the refrigerated DRI fall tower into a large cold box at temperatures of around -42°C. Although the paper of Bailey and Hallet (2009) is not quantitative, they note that "most of the particles that look quasi-spherical at lower magnification are faceted or show emerging facets at a size of 5-10 μm, while somewhat larger compact shapes are clearly faceted or are budding rosettes". This information has been added to the manuscript.

2. The Bucky ball phase function is invariant with respect to area ratio as is expected since the aspect ratios of the columns are invariant. With the number of arms adopted this significantly increases side-scattering and hence g becomes quite low. Why was the number 20 assumed? Is this number of arms representative of laboratory experiments of Bailey and Hallet (2209) and/or Ulanowski et al. (2004)? A further model that has been used to study ice crystal scattering is the Polycrystal due to Macke et al. (1996)[J.Atmos.Sci.53,2813-2825]. This model at 0.55 um gave an asymmetry parameter of about 0.74, invariant with respect to size since the aspect ratio of the Polycrystal was near unity. Therefore, how does the side-scattering of the Polycrystal compare with the Bucky ball model? This must be shown in Figure 11 for the same size, since if the side-scattering of the Bucky ball exceeds the Polycrystal or is very similar then we can dismiss this new model as being representative of cirrus scattering since we know from POLDER measurements that the Polycrystal side-scattering is too high [Labonnote et al. 2001. JGR 106,12139-12153].

As it is shown in Fig. 3 of this paper and in Ulanowski et al. (2004), the number of arms of small ice crystal is larger than that of conventional bullet rosette (~ 6 to 8 branches). Thus, we need another model to represent the arm structure shown in the ice analogue that has more than 10 arms. In a budding Bucky ball, we can change the number of arms. Note that the development of the 3B and polycrystal were guided by very different goals.

The polycrystal was never designed to represent the shapes of individual crystals, but rather was designed to represent the scattering properties of distributions of crystals. The 3B is designed to represent the shapes, and hence scattering properties of small quasi-shapes ice crystals: calculating the scattering properties of a distribution would require integration over all shapes and sizes of ice crystals.

Figure 1 compares P_{11} and g of 3B and Macke's polycrystal. The side scattering of the 3B is comparable with that of the 2^{nd} and 3^{rd} order of polycrystal. Since the 3B represents small ice crystals rather than a distribution of crystals, we have calculated the bulk scattering properties of tropical cirrus using observed size and habit distributions and libraries of single-scattering properties assuming that small crystals can be represented by a 3B. The calculated mean P_{11} is featureless and its side and backward scattering is smaller than polycrystal and 3B. Further, the calculated mean g is similar with the direct measurements of g the using Polar Nephelometer (Auriol et al., 2001; Gayet et al., 2004).

In addition, the normalized intensity calculated from TWP-ICE data and that measured by a polar nephelometer (PN) during several field campaigns are compared in Fig. 2. The calculated relative intensity using TWP-ICE data agree well with that measured by a PN during South Pole Ice Crystal Experiment (SPICE) although there is spread with other field campaigns, which depends on meteorological conditions, contributions of different crystal habits, and other factors. Thus, it seems that there is no reason to dismiss 3B to calculate the bulk scattering properties of cirrus. The bulk scattering properties of cirrus determined using the 3B and other crystal models will be presented in a subsequent paper under preparation.

Auriol et al.: In situ observation of cirrus scattering phase functions with 22 and 46 halos: Cloud field study on 19 February 1998, J. Atmos. Sci., 58, 3376-3390.

Gayet et al.: Cirrus cloud microphysical and optional properties at southern and northern midlatitudes during the INCA experiment, J. Geophys. Res., 109, D20206, doi:10.1029/2004JD004803, 2004.

Shcherbakov et al.: Light scattering by single natural ice crystals, J. Atmos. Sci., 63, 1513-1525.

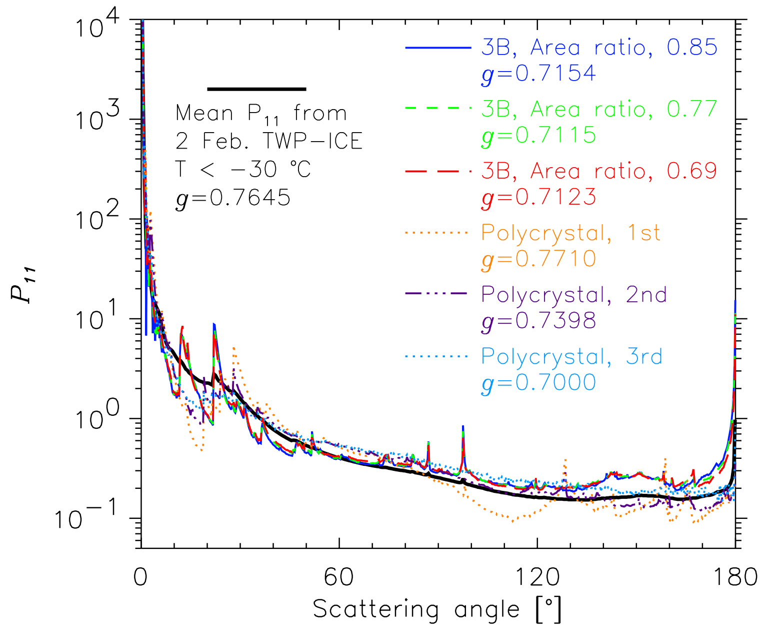

Fig. 1. A comparison of scattering phase function and asymmetry parameter for 3B, Macke's polycrystal, and those calculated from TWP-ICE

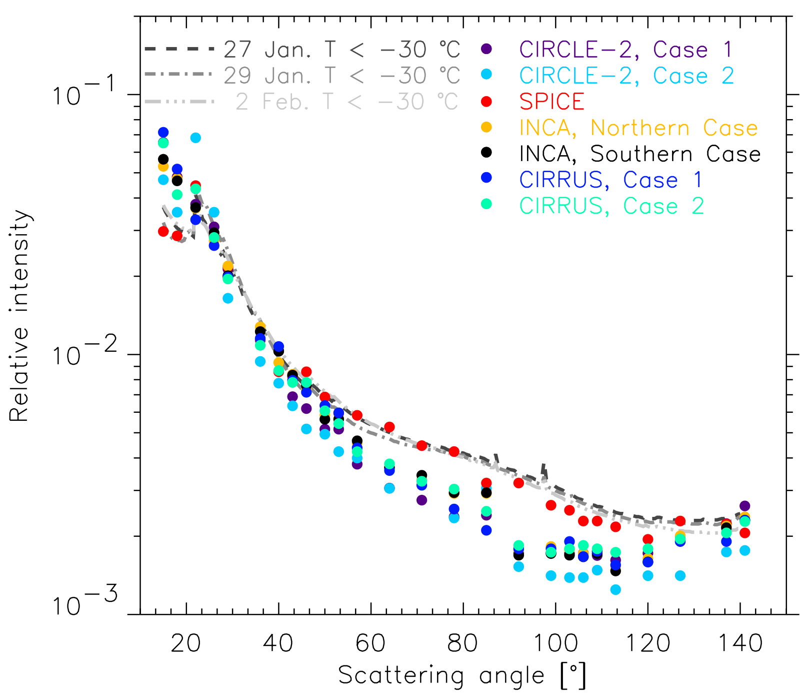

Fig. 2. Relative intensity calculated using TWP-ICE data (lines) and that measured by a PN during several field campaigns (circles).

3. The authors do not test their model against actual satellite-based measurements or in situ estimates of the bulk extinction, which could be easily derived from the CPI instrument that they used. Is the Bucky ball predicted area compatible with measurements? If the test suggested in point 2 above is inconclusive then a further test using PARASOL data on a pixel-by-pixel basis for appropriate thin cirrus cases should be performed. It is believed at the moment that the paper is too premature since the authors present no tests of their model scattering predictions against measurements.

The CPI does not measure the bulk extinction. It only provides size and shape information of ice crystals. It has a small and poorly defined sample volume that means size distributions cannot be derived. If the size distributions could be derived then the extinction could be estimated from the projected area of the budding Bucky ball model. But, what then could we compare it against? Measures of bulk extinction from probes such as the Korolev extinctiometer are currently in their infancy. The Korolev probe made measurements of bulk extinction during the 2008 Indirect and Semi-Direct Aerosol Campaign (ISDAC), but those data have yet to be released because Korolev is still working on determine the calibration of the probe needed to determine the bulk extinction. There was no such probe used in the TWP-ICE experiment analyzed here. In any event, the projected area of the budding Bucky ball can be changed according to the projected area of the quasi-spheres in the in-situ measurements. In fact, we choose the area ratio of the 3B to match the projected area of the in-situ measured ice crystals.

As mentioned previously, the budding Bucky ball is used to calculate the single-scattering properties of individual ice crystals, rather than distributions of ice crystals. To compare against PARASOL data, the bulk scattering properties of cirrus must be computed first by weighting the single-scattering properties of different sizes and shapes of ice crystals according to their observed concentrations (e.g., following equations in McFarquhar et al. 2002 and others). Comparing satellite data with the single-scattering properties of one crystal habit would be misleading and cause large errors. To calculate bulk scattering properties is beyond scope of this study. Further, to compare against satellite data (e.g., PARASOL data), in-situ data in thin cirrus at time of a PARASOL overpass would be needed: we are not aware of any such measurements. At the minimum, we would need a PARASOL overpass in conditions similar to those in which the observations discusses in the paper were obtained (i.e., aged anvil cirrus in the Tropics).

4. The behaviour of the model at absorbing wavelengths should also be investigated at for instance at 1.6 μm, where obviously the single-scattering albedo will be less than 1. How does this single-scattering albedo compare with other models including the polycrystal? Or how does the volume-to-area ratio compare to other models used for small ice crystals, including the Polycrystal? It is also known that due to the compact nature of the polycrystal the single-scattering albedo was too low when compared against aircraft measurements described in Francis et al. (1999) [JGR 104,31685-31695]. The same could also be true of the Bucky ball model? The authors need to prove their model against observation.

This study is aimed to determine the dependence of the single-scattering properties of small ice crystals on the assumed model. The polycrystal has been used to represent bulk scattering properties of ice clouds rather than single-scattering properties of one specific crystal habit. To compare scattering properties of the polycrystal with other small ice crystal models is inappropriate. Calculations of single-scattering properties of small ice crystals at infrared wavelengths require the use of a code other than the geometric ray-tracing code used here. These calculations will be done in subsequent papers.

5. Further analysis on the polarization properties of the Bucky ball would also be helpful, how do the other scattering matrix elements compare against other small ice crystal models? Especially the P12 element.

We agree. The P_{12} element has been added. See the text in the paper discussing the differences.

6. The present paper is also incomplete as it only considers non-absorbing wavelengths. The A-train samples cirrus across the electromagnetic spectrum simultaneously. It is important to develop ice crystal models that are consistent across the electromagnetic spectrum. How does their model compare against others at wavelengths in the 8-12 um region? It is suggested that the authors use the publicly available DDA method to complete this study. With the latest DDA method size parameters of 40 should be attainable.

As it is stated in question 4, calculations of single-scattering properties of small ice crystals at infrared wavelengths will be done in following studies. We are planning such studies.

It is believed that the present paper is incomplete for the reasons listed above; in essence the authors must provide the evidence that shows their model is at least representative of the scattering properties of optically thin cirrus. If they are able to prove their model against currently available observations and measurements then the contribution becomes significant.

We are in total agreement with the reviewer that our study has not answered all questions regarding how small ice crystals affect scattering. However, this paper has made significant progress in developing an alternate model of a small ice crystal that is most likely more consistent with observed crystal shapes, and by demonstrating the sensitivity of single-scattering properties to assumptions about small crystal shapes. Given the wide number of papers assuming that droxtal (about which there is also no observational evidence) can be used to determine the single-scattering properties of small ice crystals, it is important that these differences be used so that the limitations of past remote sensing and climate studies can be understood. By making this model available to the community now, we are hopeful that others will pursue the use of the model, and hopefully obtain collocated in-situ and remote sensing observations that would allow it to be more thoroughly tested.

Minor points.

1.Introduction: Line 14 page 28111. The parameterizations and retrieval methods also depend on the assumed randomization of the model ice crystals.

The randomization is included in the assumed shape distribution. See response below.

2.Introduction line 24 page 28111. The aggregate of columns proposed by Baran and Labonnote (2007) are highly randomized such that their scattering properties become independent of the initial monomers making up the ice aggregate. Therefore, from the scattering pattern it would not be known that the particles were aggregates of columns. Other models tend to retain the hexagonal symmetry such that the scattering properties of individual monomers making up the aggregate are retained.

Thank you. A note on the importance of randomization has been added to the manuscript.

3.The statement on page 28113 line 8 the single-scattering scattering properties of ice crystals also depend on size and assumed randomization.

A statement has been added that the scattering properties also depend on assumed randomization.

4.In the discussion of shattering on page 28114 section 2 the paper by Field et al. (2003) [J. Atmos.Ocean.Tech. 20,249-261] should also be cited as the authors also find that for *narrow PSDs shattering is not likely to be a major problem.*

A reference to Field et al. (2003) has been added.

5.Section 4 page 28120 line 21 please give the reference for the value of the refractive index of ice used in the calculations.

Thank you for pointing out. It has been added.
Warren, S. G., and Brandt, R. E.: Optical constants of ice from the ultraviolet to the microwave: A revised compilation, J. Geophys. Res., 113, 2008.

6.Section 5.1 page 28122 line 22. The authors should also note that in the review paper of Baran (2009) [JQSRT 110,1239-1260] it is shown that the g value of aggregates tend to asymptote as a function of aggregation for columns and plates due to the spatial nature of the resulting ice aggregates.

It has been added.

7.Page 28125 line 7. The term multiple scattering is used. Since the paper is based on the method of ray-tracing the term multiple reflections should be more appropriate.

Multiple scattering has been replaced with multiple reflections. Thank you.

Figures.

Fig 1. The maximum dimensions in each image are very unclear can these be made clearer?

The 200 μm scale bar in the figure is now labeled.

Fig 7. What do the authors mean by irregular hexagonal columns? In what way are the columns irregular? It is difficult to see the irregularity in the figure.

It is an imperfect hexagonal column. It is also shown in Fig. 8 and corresponding reference has been added.

Fig 10. The phase functions for the more irregular particle show some degree of noise at scattering angles between about 50 -180 degree. This suggests that there are insufficient random orientations assumed in the Monte-Carlo simulations.

We increased random orientations several times, but got similar phase functions and derived asymmetry parameters. We have added a note in the manuscript that our results are not dependent on the number of orientations used.

Fig 11. Same as fig 10.

Please see the answer for Fig. 10.

Fig 13. What does the g value for the 3B model with aspect ratio of 1 increase with decreasing area ratio?

As an area ratio of 3B with aspect ratio 1 decreases, the *g* of 3B increases.