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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A laboratory investigation into the aggregation efficiency of small ice crystals" *by* P. J. Connolly et al.

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Review of Connelly et al. ACPD manuscript titled: A laboratory investigation into the aggregation efficiency of small ice crystals

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PLEASE SEE SUPPLEMENT FOR ACCURATE TEXT AND EQUATIONS

1. General Comments

The paper describes an interesting ice crystal aggregation experiment using a 10 m cloud chamber where ice crystals aggregate in free-fall and a detailed bin model for

calculating aggregation efficiencies from the cloud chamber observations. The bin model predicting the evolution of the ice particle size distribution is robust in certain respects that in themselves are noteworthy of publication. This investigation into the aggregation efficiency, Ea, of ice-bearing clouds between -5° and -30°C is a large improvement over previous laboratory studies on aggregation, and definitely this paper should be published in ACP after major revisions are made. Very few laboratory studies of Ea have been made over the last half century, which is surprising since aggregation affects cloud optical properties and is relevant to climate modeling.

In spite of the great deal of thought and effort that went into this study, there are still shortcomings in my opinion that should be addressed before this manuscript is suitable for publication in ACP. These are detailed below.

2. Specific Comments

1. First, I'd like to follow-up on Chris' comment about the aggregation collection kernel. The results in Fig. 12 show that Ea is extremely sensitive to the ice crystal aspect ratio due to its influence on Ai and Aj, underscoring the importance of accurately estimating ice particle area projected to the flow. I too had the same concern as Chris, but upon further investigation, I discovered that the two expressions for collision area are identical provided one assumes spherical particles:

(Ai0.5 + Aj0.5)2 = (π/4) (Di + Dj)2 .

This can be derived mathematically or shown by representing A as (π/4)D2. But the collision area may still be ambiguous when confronted with real ice particle shapes since A0.5 would seem to be the radius of an equivalent area sphere (see Fig. 14-1 in Pruppacher and Klett). If we represent A as A = c Dd, where D is ice particle maximum dimension, perhaps some improvement is achieved with the collection kernel becoming:

K(i,j) = [c (Did/2 + Djd/2)]2 Ea | vi – vj| .

But there are other concerns about the description of Eq. 1 where Ea is described as the ratio of sticking events to collisions between ice particles. This would be true if the collision efficiency between particles was unity (1.0), but clearly this is not the general case as shown in Fig. 14-1 of P&K. Rather, Ea = Ec $\times$ Es where Ec = collision efficiency (determined by the hydrodynamic flow lines around the particle) and Es = sticking efficiency. For the small ice crystals used in this experiment, Ec < 1.0.

2. The authors appear unaware of relevant work on aggregation that is not mentioned in the introduction. A case study on aggregation in cirrus clouds found that the cloud-averaged Ea was $\sim$ 0.5 in Mitchell et al. (1996) and in more recent work (Mitchell et al. 2006), a new snow growth model (SGM) was developed that found the evolution of the ice particle size distribution (PSD) in “typical” frontal clouds was best modeled when Ea = 0.07. In a case study characterized by temperatures producing dendrites near cloud top, Ea = 0.55 gave the best agreement with the observations. The PSD evolution for this case suggested that ice nucleation occurred almost exclusively near cloud top.

This more recent work describes a SGM formulated from moment conservation equations of the 0th and 2nd moments with respect to mass (0th and 6th moments with respect to spherical particle size). Thus the PSD predicted are weighted by number concentration N and radar reflectivity. This differs from the SGM in Mitchell (1988) and elsewhere where the PSD where weighted by ice water content (IWC) and radar reflectivity. This new formulation produces Ea values generally lower than those predicted by the earlier SGM formulation, with these more recent Ea estimates appearing more consistent with those obtained in this aggregation study. However, the shape factor assumed in Mitchell et al. (2006) was 0.75 Dmax, which is too high based on Field et al. (2006b) who found the shape factor was $\sim$ 0.25 Dmax. Therefore Ea in Mitchell et al. (2006) may have been underestimated.

3. Section 4.1: Planar ice crystal growth at -25°C was also observed by Bailey and Hallett (2009, JAS).

4. Section 4.1: There is no information on ice particle sizes in this paper, although it can be inferred that since the ice fall speeds were $\sim$ 4 cm/s, ice particles were less than 100 μm (Mitchell 1996, JAS, Sec. 4e). This information is critical to the interpretation of results. Regarding Eq. 6, please include a figure showing representative PSD observed in this experiment, also showing the best fit line in log-linear space.

5. Comparison of Fig. 5 with Fig. 8: Figure 5 depicts the conceptual approach of this experiment and is well done. It shows the ice particle pulse near the base of the chamber occurring after the pulse was detected in the middle of the chamber. Figure 8 shows the opposite; the concentration peak in the middle of the chamber comes after the peak near chamber base. This makes no sense and I hope this is a labeling error. 6. Figure 9 and associated discussion: The text says “for all experiments the distributions get broader towards the bottom of the chamber”, but the caption in Fig. 9 states the opposite. I hope this is a labeling error, and the following discussion assumes this is the case.

The change in PSD slope parameter λ in Fig. 9, from higher values in the middle of chamber to lower values near chamber base, has been interpreted as solely due to the process of aggregation. That is, the decrease in number concentration peak from middle to bottom of chamber is due solely to aggregation. However, some panels in Fig. 9 also show a subtle increase in λ with time regarding the black line (corresponding to the bottom of chamber). This pattern can be explained by the phenomena known as “size-sorting”, where ice particles are not all the same size, with larger ice particles falling faster than smaller crystals, leading to an enrichment of larger particles having lower concentrations lower in the chamber. While all ice crystals may eventually reach the chamber bottom, as ice crystals grow the PSD broadens and fall speed differences increase, spreading the ice population over a greater volume. Thus it appears that size-sorting could also contribute to the ice concentration behavior shown in Fig. 5, used to determine Ea. While the CPI images show that aggregation is clearly occurring, can the authors exclude the possibility that size-sorting is also playing a role in lowering concentrations near chamber base? This issue appears critical since Ea is based on the first pulse of ice crystals which size sorting is most likely to impact near chamber base. Size-sorting may contribute to a smaller λ (larger particles) near chamber bottom, with the lower chamber λ slightly increasing with time in some cases as smaller ice particles replace the larger ones that sediment out.

If this allegation cannot be dismissed, is it possible to reanalyze the results to account for size-sorting?

7. Figure 12: The dependence of Ea on aspect ratio is understandable (i.e. aspect ratio affects collision kernel area) but also troublesome. This underscores the sensitivity of Ea to ice particle projected area to the flow or "A". The treatment of A on p. 12 is very approximate, relying on results from other studies to characterize the A-D expression used. Also, assuming the area of a circle for hex-plates overestimates A, noting that the basal area of a plate is only $\sim$ 2/3 the area of a circle having same maximum dimension. Perhaps the treatment of A could be improved if CPI data from the chamber could be used to generate A-D power-law relationships that could be used in the bin-model. Each temperature used would require a separate A-D expression.

8. Figure 15: Where is the black dashed line?

9. Lines 560-561: For T > -40°C, cirrus clouds have a broad spectrum of ice particle sizes; cirrus PSDs are not well approximated as mono-disperse as suggested here.

10. Lines 603-4: "The fact that the planar crystals observed at -10°C did not have dendrites suggests that interlocking is the likely reason for the maximum in Ea at -15°C." This conclusion was also arrived at by Mitchell (1988) where Ea was made a function of temperature for steady-state snowfall such that ice crystals produced in a given habit regime were assigned an Ea appropriate for that regime. Tracking the numbers of ice crystals produced in each habit regime, the overall Ea at a given level was based on the relative weighting of ice crystal concentration from each habit regime and corresponding Ea value. While the assumption that ice crystals near cloud top

descend through cloud base may be questionable, the scheme nonetheless assigned Ea in terms of crystal complexity or branching and tracked the contribution of crystal habit to the overall Ea. This resulted in surprising agreement between predicted and observed PSD (especially when the observed IWC profile was used; see Appendix) and strongly supports the "interlocking" argument made above in this paper.

11. Paragraph beginning on line 609 and elsewhere in text: The authors have stated that they have used the Mitchell (1988) snow growth model (SGM) to estimate Ea from the change in PSD slope or λ (between mid- and lower-chamber) but that this overestimated Ea (with Ea sometimes exceeding 1.0). They state that when Ea was set to zero (diffusion growth only), the SGM predicts that λ increases (i.e. the mean ice particle size becomes smaller) from mid-to-lower chamber. The authors also note that the PSD evolution predicted by the SGM is sensitive to the constants used in the m-D power laws. While it is true that the SGM is sensitive to these constants, the other SGM behavior described suggests the SGM was not run correctly, as explained below.

Firstly, the SGM from Mitchell (1988) does not always predict λ increases for diffusion growth only (Ea = 0) as seen in the Appendix of that paper. For example, Fig. B5 clearly shows λ rapidly decreasing long before reaching the aggregation zone, and the simulation for diffusion growth only (shown in that figure) confirms this. The behavior of λ depends strongly on what is assumed for the mass exponent "b" in the m-D power law (Eq. 4 in this reviewed paper) since this has something to do with new ice crystal formation in the SGM.

To better understand the SGM, understand that new ice production is based on conservation of mass, and the SGM is initialized from a height-dependent IWC profile. The height-dependent equation for λ predicts the broadening of the PSD with decreasing height and increasing IWC, and this broadening may account for all or some of the increase in IWC with decreasing height. Whatever IWC in the initialization profile that is not accounted for by the decreased λ (larger particle sizes) is balanced by the production of new ice crystals. The amount of mass in the ice crystals is determined by the constants in the m-D expression. So the number of new ice crystals required to balance the IWC profile depends on the m-D expression used.

If the SGM is run for diffusion growth only and ice crystal concentration N is held constant, then λ will not be allowed to increase since only new ice production will increase λ. However, since the SGM is run off of an IWC profile, it is not easy to hold N constant, but it is possible to do this with proper care. In the aggregation experiment, since no new ice crystals below the source region were nucleated, one could argue that N should remain constant to a first approximation in the absence of aggregation, although size-sorting should expand the population volume and thus decrease N somewhat. So one thing you could try is to vary the value of "b" in the m-D expression until N is approximately held constant.

What is also not clear is how the variation in IWC within the chamber was established. An IWC profile must have been determined somehow in order to initialize and run the SGM. Was this based on the bin-model? If so, the IWC profile would be subject to uncertainties and limitations of the bin-model, which could also contribute to the results you obtained. The PSD predicted by the SGM are very sensitive to the IWC profile as shown in the appendix of Mitchell (1988).

In summary, the reported increase in λ predicted by the SGM from mid-to-lower chamber can only occur through new ice crystal production, which did not occur during the aggregation experiment. If the authors take appropriate measures to ensure that N does not increase, they will find that λ will increase from mid-to-lower chamber.

Rather than use the old Mitchell (1988) SGM that is based on the 3rd and 6th moments, why not use the Mitchell et al. (2006) SGM that is based on the 0th and 6th moments? It is more sensitive to changes in N and thus more appropriate for this experimental design. Moreover, at its current stage of development, an ice nucleation scheme has not been employed, meaning that it is initialized by establishing N at cloud top (i.e. chamber top). It does not assume an IWC profile but rather increases the IWC through

the standard diffusion growth equation for a given supersaturation with respect to ice, such as saturation for liquid water. This new SGM architecture appears well suited to the aggregation experiment. If you like, I could send you this code and I'd be happy to assist in the analysis in whatever capacity you are comfortable with.

12. Paragraph at line 640: Same comments as in (11) above.

Selected References

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Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/11/C10411/2011/acpd-11-C10411-2011-supplement.pdf

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 25655, 2011.