Review of Connolly et al manuscript on ice crystal aggregation

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1 Overall impression

This paper describes laboratory experiments where ice crystals are nucleated and grown from vapour at the top of a tall (10m) cloud chamber. These crystals are then observed as they aggregate and fall through the chamber, and the evolution of the particle size spectrum is quantified.

This is clearly a highly significant study, essentially the first attempt to simulate the aggregation process in the laboratory under semi-realistic conditions. Previous laboratory work (Hosler et al, over 50 years ago) has involved drawing miniscule (10μ) crystals past a much larger fixed target, rather than actually observing the differential sedimentation process itself. This new paper is therefore a substantial step forward, and it is important that the details of these experiments should be published.

The authors use their data, along with a numerical model, to derive estimates of the aggregation efficiency This is valuable information for numerical simulation of the aggregation process in models. They also compute a 'confidence interval' for their estimate of E_{agg}: disappointingly, this interval is very wide, and somewhat detracts from the value of the main results. This requires further comment from the authors and I address this issue below.

The observation that aggregation is much more rapid in the region of phase space where branched planar crystals grow is a significant conclusion, and adds weight to inferences made previously by Hobbs and others.

Overall the manuscript is sound, original and interesting, with subject well suited to ACP. I recommend it be published once the following minor issues are addressed:

2 Detailed comments

1. In equation 1 you outline the differential sedimentation kernel. It is worth noting here that the version which you are proposing is an approximation, since you are equating $(A_i^{0.5}+A_j^{0.5})^2$ with the collision area. An alternative approximation would be to use the more conventional 'close approach' area

$\frac{\pi}{4}(D_i+D_j)^2$ instead, ie the area corresponding to when two particles come within touching distance of one another (based on their maximum spans D). Finally, one could actually sample the true collision area directly, as done by Westbrook et al (2004). It is worth pointing out the differences between these options because which of them you choose will give you a different physical meaning and numerical value for E_{agg}.

2. Page 25662, line 24-29, shattering. Say 'concerns have been raised'. Is there a citable paper for this? (perhaps Schwarzenboeck et al 2009?). Say that impact velocities in lab are 'orders of magnitude lower' - this is perhaps a little bit overstated: typical velocity on aircraft is 100m/s vs your 4m/s in the lab, so that's a factor of 25. I agree that shattering is probably minimal in your experiments however, particularly since the crystals involved are quite small.

3. Page 25663, line 4: pounds per sq inch. Most journals demand SI units only these days.

4. line 14: 'homogeneous nucleation of the liquid drops that were present'. From this description it sounds like the ice crystals are being produced from freezing the droplets which are already present in the chamber. I think the current understanding of how rapid expansion produces large numbers of ice crystals is slightly different to this. That extremely rapid adiabatic cooling by 50°C or more means that the humidity is, for a split second, many times the saturation value. In that split second, homogeneous condensation of droplets is possible, followed by immediate homogeneous freezing of those new droplets. Diffusion quickly returns the air to saturation of course; however homogenous condensation is an exceptionally rapid process (Mason 1971) and dominates the formation of ice germs in such an expansion. See Westbrook and Davies (2010), Foster and Hallett (1993) and Vonnegut, Mason (1981) for more details.

5. Page 25664, line 5. I don't really understand how this Peclet number is calculated or what it refers to physically. Is Re the Reynolds number of the falling ice crystal? If so, for the crystals in your experiment I would estimate Re$\sim$1 and Sc for air is about 0.6 so then the Peclet number is also of order unity, rather than 10^{11}. I guess what you need to assess is whether differential sedimentation or diffusion is dominating the aggregation process. To do this you could do a back-of-the-envelope calculation with the relevant collision kernels:

$$K^{diffusion} = 2\pi(D_1 + D_2)(\kappa_1 + \kappa_2)$$

where κ are the diffusion coefficients of the particles $= \frac{kT}{3\pi\eta D}$, [see eg. Jacobson (2005)]. This gives you:

$$K^{diffusion} = \frac{2}{3} \times \frac{kT}{\eta} \times (D_1 + D_2)\left(\frac{1}{D_1} + \frac{1}{D_2}\right)$$

now lets say $D_1 = 100\mu$ and $D_2 = 50\mu$. Take T=250K: the dynamic viscosity of air at this temperature is $\eta = 1.6{\times}10^{-5}$kg/m/s. The Boltzmann constant k is $1.38{\times}10^{-23}$m^{2}kgs^{-1}K^{-1}. So then $K^{diffusion} \approx 2{\times}10^{-17}$m^{3}s^{-1}. Now for the differential sedimentation kernel you have:

$$K^{sedimentation} = \frac{\pi}{4}(D_1 + D_2)^2 \Delta V$$

- lets say ΔV=0.1m/s. Then $K^{sedimentation} \approx 2{\times}10^{-9}$m^{3}s^{-1}. So $K^{sedimentation}/K^{diffusion} \sim 10^8$, ie sedimentation is completely dominant relative to Brownian collisions.

6. Page 25668, lines 4-10. This seems like unecessary detail and could be deleted - why not just say rime is not observed, so was switched off in the simulation.

7. Page 25671, lines 4-7. It took me a while to figure out exactly what your cost function was here - so it's just $(N^{middle}_{model} - N^{middle}_{observed})^2 + (N^{bottom}_{model} - N^{bottom}_{observed})^2$ where N are the peak concentration values? Is the model value the peak in the simulation, or the simulated concentration at the point in time where the observed peak was found? [one penalises poor timing of the peak, the other doesn't]

8. Page 25671, lines 20-30. The fact that there is a trailing distribution of ice crystals behind the main pulse in the observations is attributed to depletion of water vapour behind the main pulse (ie glaciation leading to lower growth rates, and slower fall speeds). Is an alternative explanation that there is some turbulence or mixing occurring within the chamber, smudging out the sharp concentration gradient? Do you have any air velocity measurements in the chamber to check this?

9. Page 25672, CPI images. It would be good to blow this image up to span a full page (maybe landscape format) on the final manuscript as the individual elements are rather small. Zooming in on my computer, I agree with your assessment of the habits between -5 and -20C. Interestingly the plates seem quite thin at -10 and -20 compared to previous lab work (eg Takahashi et al 1991) which has suggested an almost 1:1 aspect ratio at these temperatures. At -25C it looks like there is a mixture of habits, with some plates, columns and polycrystals. It's not really obvious to me what type the polycrystals are (ie rosette-type or planar). At -30C the images look predominantly column-like, near 1:1 aspect ratio, again with a few polycrystals. The images look similar to the crystal habits grown in free-fall at water saturation by aufm Kampe et al (1951) - it would be good to reference this paper in addition to the more recent static diffusion chamber work of Bailey and Hallett.

10. Page 25674 - how was the aspect ratio determined from the CPI images?

11. Page 25676: construction of the confidence intervals. I don't really understand the methodology here. From the description, my understanding is you:
    - pick lots of E_{agg} values at random between 0 and 1
    - calculate the residuals for each sample
    - calculate the PDF or CDF of those residuals
    - then find 25th and 75th percentiles of that residuals distribution, and the corresponding E_{agg} values are your confidence interval.

    So is the idea that you treat the residuals as inverse probabilities for that particular E_{agg} value being true? And then you construct the interval such that there is $>$50% likelihood of E_{agg} lying within those bounds? (assuming everything else about the experiment is perfect)
    You should spell out what this will quantify. It would be instructive to plot out example time series for the E_{agg} values corresponding to the 25th and 75th percentiles to see (visually) how those simulations compare with reality, and whether those values do give you a result which looks realistic.

12. Page 2567, line 20-27. Sintering more favourable for crystals similar in size. This is subtle - it must also depend not purely on the terminal velocity of the crystals in isolation from one another, but on the detailed flow around the particles as they come into contact, which may act to stabilise one crystal on top of another for certain shapes/configurations but not others. This is pretty hard to determine.

13. Page 2568, line 9-14. Is it worth referencing Hobbs et al (1974) here? They inferred a maximum aggregation efficiency at -15C based on snowfall measurements at the surface, and investigate the environmental factors / physical mechanisms that might be involved.

14. Page 2569, line 25. The conclusion that E_{agg} does not increase as the temperature increases is surprising. It's well observed at the surface that snowflakes are much larger when the temperature is close to 0C; similarly radar reflectivity often increases rapidly in the region between -5C and the bright band. It would be interesting to investigate warmer temperatures (-5 to zero) in future work.

15. Conclusions: it would be useful to suggest any ways that the methodology/instrumentation used here could be improved in order to reduce the uncertainties in E_{agg}. In particular, can you shed any light on why the differences between model and experimental data is apparently so insensitive to E_{agg} (as evidenced by the confidence intervals). Does this imply that it doesn't really matter too much what value of this parameter you choose for your aggregation model? Or does it just mean that in future analysis you need a more discerning cost function to test the simulations against the experimental data (maybe using different moments of the size spectrum)?

16. Figure 11 - would the minima be clearer on a log scale?

References

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[2] Schwarzenboeck et al (2009) ‘Indications for stellar-crystal fragmentation in Arctic clouds’ *Atmos. Res.* 92 220-228

[3] Mason (1971) *The physics of clouds*, Oxford U.P.

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[9] aufm Kampe et al (1951) ‘The influence of temperature on the shape of ice crystals growing at water saturation’ *J. Meteorol.* 8 168-174

[10] Hobbs et al (1974) ‘The dimension and aggregation of ice crystals in natural clouds’ *J. Geophys. Res.* 79 2199–2206