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Comment

## ***Interactive comment on “Estimating collision efficiencies from contact freezing experiments” by B. Nagare et al.***

### **Anonymous Referee #2**

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“Estimating collision efficiencies from contact freezing experiments” describes an experiment on 80 micrometer water droplets and 200 nm AgI aerosol where collision efficiency was inferred from the formation of ice. In brief, aerosol and droplet were allowed to interact in an ice saturated chamber so that ice formation could then be detected with a custom optical counter called IODE. The paper seems to therefore indicate what is measured is an ice nucleation rate, not a collision efficiency. The authors assume that all contacts lead to freezing (which may or may not be valid, but proof of the equality is lacking in the paper).

The majority of the paper is not experimental, as the title seems to imply, instead being centered on theory of collision. As an example, section 2 is ‘Theory’ – 11 pages - while Experimental is section 3 – 3 pages. This is in no way a negative although a change in

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title seems warranted (see below). The theoretical calculations are more than an order of magnitude lower than the collision efficiency that is experimentally determined. The authors suggest that this is due to uncertainty in thermophoretic and diffusiophoretic theory – and this point is argued in the discussion section. This seems a theory that isn't supported with much evidence since it is based on only a single data point (one aerosol and one droplet size).

This topic is well suited for ACP and the work is of contemporary interest in the atmospheric sciences since ice nucleation in general, and the largely unexplored mechanism of contact ice nucleation specifically, are in need of constraint.

There are, however, some serious issues with this paper that need to be addressed before publication. The major issue is the presence of only a single data point (one aerosol and one droplet size) in the concluding Figure 6 and, realistically, there should be several points across the aerosol range to help determine where the discrepancy between experimentation and theory lies. I stress that this is not a minor change and significant experimentation is required to make this paper suitable for publication. Such a paper would be of great importance in the field. The authors appear to be in a good position to do this and I therefore recommend 'major revisions'.

Alternatives seem to be (a) simply publishing the single size aerosol and droplet data shown here but not trying to draw conclusions by comparison to theory which isn't warranted with the lack of constraint offered by a single data point or (b) publishing a theoretical review paper without data. I find these alternatives not preferable and highly recommend more experimentation and a revised paper with enough data to allow proper interpretation.

Suggestions for improvement:

1. Since the theory and experimental results differ by an order of magnitude more experimental points are warranted. This is further highlighted by the amount of text spent on theory versus experiment (roughly 4 times as much). Specifically, Figure 1 is

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data for 80 micrometer droplets and 200 nm aerosol. A great deal would be learned from variation of droplet and aerosol size and this would seem the best way to constrain the reason for the difference between theory and experiment. Just looking at Figure 6, and the plots of theory leading to this, several more points with varied aerosol size seem required to draw any reasonable conclusion. Thus I suggest that several more figures of the Figure 1 type be prepared at different aerosol and drop size. 2 or 3 more points, at variable aerosol size, would greatly help in understanding the difference here. As the adage goes, an infinite number of lines can be fit to a single data point. At this time, all the authors show is that the single data point is high by more than an order of magnitude but this does not make a satisfactory conclusion for the paper. Furthermore, experiments could be devised to test if the authors' suggestion that thermophoretic and diffusiophoretic effects might be the cause based on the variation of these forces with size. There is also a significant uncertainty in charge, stated as 39k elemental charges  $\pm 20k$ , that could contribute. Currently the authors point to a 'worst case scenario' of charge but the means of measuring charges seems unconstrained (see below).

2. Change the title. This paper is truly a comparison of theory to contact freezing experiments, not just experiments on collisions at low temperature. The authors have not demonstrated that every collision leads to an ice nucleation event and if they can not do this they can not decouple collisions per contact ice nucleation event. I agree that in this regime it is extremely likely that every collision leads to an ice nucleation event but the authors need to more explicitly state this assumption (starting in the abstract –around sentence 4) but unless the authors prove it the implied assumption they are the same is not acceptable.

3. 19k – 59 k elemental charges seems extremely large and this value caught my attention. The authors include a few statements on the atmospheric range is in the paper but these are only found in the conclusions and seem to indicate some 100s to 1000s. The text in this regard is rather vague, including statements of attraction and repulsion but little on the atmospheric relevance. The atmospheric range should be

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moved higher, into the experimental part for comparison. Please explain why such a large charge was used? Please include more data on how charge was measured and if any calibration was used (such as particles of known charge). As is stands, this seems a large area of uncertainty and one that needs to be expanded upon.

4. Regarding the assumption that all freezing is caused by contact nucleation: it is understood these are pure water droplets and conditions, per Figure 1, are outside the homogeneous regime. However, the authors need to show they don't have any freezing going on in the absence of aerosol. Please include some control experiments, e.g. in Figure 1.

5. The section "Comparison with previous experimental work" has some issues. First, as mentioned above, it needs to be clear that the authors are assuming freezing events = collisions.

6. Second, "Comparison with previous experimental work" includes a comparison to several room temperature experiments, including that of Ladino et al. (2011), which apparently comes from this same research group. This caught my attention because a comparison is made between the theory here and the old Ladino et al. (2011) experiments in the final figure, as if this was the summation figure of this manuscript. The purpose seems to be to show that the theory used here is solid and the statement is made, regarding Ladino et al., that "their experimental results are in general agreement with the theoretical predictions." First, Figure 6 should really be the concluding figure of this paper since it combines the lab and theory performed here. Figure 7 should be moved up since its purpose seems to be to show agreement of theory with previous experiments. Note that Figure 7 calls "Ladino et al. experiment" but no year is included for reference. Also, I believe with multiple data points this should be the plural, 'experiments'.

7. Finally, and most important, I think the reference here (if the text is correct) is Ladino et al. "Experimental Study of Collection Efficiencies between Submicron Aerosols and

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Cloud Droplets” (so the figure caption should be 2011b) and the copied data are Ladino et al. Figure 7. If that is the case then Ladino et al. stated in their paper “We did not have equipment to measure the droplet and particle charges.” and yet here a charge of 10k is applied for the stated agreement. Please clarify in the text and the response : is this the correct reference? How was this charged arrived upon, as a best fit or was it measured? Is this a correction to an old paper? If so that should not be done in this manuscript but as an addendum to the original journal article.

In conclusion, this is a well-written paper that should make a fine addition to ACP if more experiments to help lead to conclusions, not assumptions, are made. This is highlighted by the single data point in Figure 6 that leaves the reader unable to interpret the experiments or the theory being used to interpret them. This is no small amount of work but it is required for a publishable paper.

This review was made before reading the first Reviewer’s comments. It is worth noting that many points - on the need for more data, uncertainty, and charge quantification - were made independently and highlight the needs in these areas.

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 12167, 2015.

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