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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Contact freezing: a review" *by* L. A. Ladino et al.

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Contact freezing is a mechanism of ice particle production which our community urgently needs to address. Hence, I am very pleased to see a review focused on this subject. Other recent reviews of ice nucleation (Hoose and Mohler, and Murray et al.) which came out in 2012 are thorough in covering the other modes of nucleation, but do not address the subject of contact nucleation in any detail. The authors have done a good job of introducing the complexities and summarising the available laboratory data and experimental methodologies and I strongly recommend that a version of this paper is suitable for publication in ACP. However, there are several issues which I think need to be addressed before acceptance.

Major issues: 1) Title: This review is focused on laboratory work with some thought given to theory. It does not cover how contact nucleation should be treated in cloud

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models and does not address field work. Hence, I recommend making the title more focused. For example: 'Contact freezing: a review of experimental studies'.

2) The different types of contact nucleation need to be explicitly distinguished. The inside out contact freezing or particles mechanical pushed into droplets observed by Shaw and co-workers may be fundamentally different to contact nucleation by a particle colliding with a supercooled droplet. In many places in the paper these mechanisms are discussed as being the same.

3) In the introduction it would be very helpful to include a section discussing how important contact nucleation is likely to be in the atmosphere. Several authors have attempted to estimate this and several have suggested contact is not important – some strong arguments in favour of contact nucleation and why the community needs to spend a lot of time and effort working on it is essential. Two studies which come to mind which need to be addressed are: i) Cui et al. (Z. Q. Cui, K. S. Carslaw, Y. Yin and S. Davies, J. Geophys. Res., 2006, 111, D05201.) who quantitatively shows that contact nucleation is not important in convective clouds. ii) Phillips et al. (V. T. J. Phillips, L. J. Donner and S. T. Garner, J. Atmos. Sci., 2007, 64, 738–761.) suggests that contact is only important in evaporating droplets through phoretic arguments and therefore that contact nucleation is of secondary importance. This seems to be the prevailing view in the literature (e.g. see discussion in Murray et al. (Chem Soc Revs, 2012) and it needs to be counteracted in this review article. My own opinion is that we know so little about contact nucleation that we cannot say if it is important or not, hence this review is very useful.

4) In multiple prominent places throughout the paper (including abstract and conclusions) it is stated that contact freezing can initiate freezing at the 'highest temperatures'. I see no convincing evidence that supercooled droplets will be more likely to freeze due to collisions than due to immersed IN. As the authors go to lengths to explain the experiments are not done in a way in which a direct comparison can or should be made. For example, when comparing wind tunnel data for immersion and contact.

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The amount immersed is some arbitrary amount and the number of collisions was also arbitrary. We know that increasing the surface area per droplet will increase the freezing temperature, so it is conceivable that someone could repeat these experiments and find the opposing result: that immersion causes freezing at warmer temperatures simply because they decided to make droplets with more solid particles inside them.

5) The authors have chosen to base the comparison of efficiency between immersion and contact nucleation on one particle per droplet vs one collision. I commend the authors for trying to come up with a way of making a comparison, but this needs some discussion. This definition of a basis of comparing fraction frozen curves may be pragmatic, but it is not definitive. For example, if we try to translate this information to a cloud, what does it mean? Will contact or immersion be most important under cloud conditions?

6) There are a few very recent papers (published since the article was submitted) which should be discussed in the final version:

(http://pubs.rsc.org/en/content/articlepdf/2013/fd/c3fd00033h) (http://www.atmos-meas-tech-discuss.net/6/3407/2013/amtd-6-3407-2013.html).

Other comments 7) P7813, ln 8-10. Important to emphasise 'amorphous' here, perhaps before 'Organic'. Wang looked at SOA, whereas the others looked at proxy materials. Also there is a new article which should be cited: Wilson, T. W., et al., Glassy aerosols with a range of compositions nucleate ice heterogeneously at cirrus temperatures , Atm. Chem. Phys., 12, 8611-8632, (2012)

8) Ln 14. Avoid the use of the term 'good IN'. This term is subjective and could be deleted.

9) Ln 15-17. DeMott (1990) and Mohler (2005) do show soot to nucleate ice, contrary to what is stated.

10) Use of word 'believed'. To me this word implies faith rather than a fact or idea which

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has been arrived at through scientific reasoning.

11) P7815. Ln 25-27: Insert word 'may'. No one has proven that this process is important!

12) P7819. Ln 11. Is there also a dependence on RH, size, etc.

13) P7820. S 2.4.1: This needs a critical evaluation. Do you think that this mechanism is sensible given what is now in the literature?

14) Also, replace 'it is believed' with ' They suggest'.

15) P7822, ln 8-15. I do not think it is possible to claim that a difference has been observed between contact and immersion in these experiments. See comment 4 above.

16) P7824, ln 10. Mention the material used by Gurganus et al. Maybe this result is specific only to this material.

17) P7826, ln 23-26. The two consecutive sentences are contradictory.

18) P7827, ln 8. Replace 'avoid' with 'reduce'. I think the air in the wind tunnel could be maintained up to ice saturation, but not above.

19) P7827, ln 25. Why is this interesting?

20) P7828, ln 15. Add references.

21) P7832, ln 27. I don't see a strong difference between the two experiments. To me the two sets of data are scattered over one another. The subsequent discussion needs to be removed or modified.

22) Section 3.5. Add a comment on the Ladino results being above unity. The limit should be unity, so why are they well above this?

23) P7835, ln 24-4. This paragraph discusses the different freezing temperatures between contact and immersion. Given my comment 4 and the author's subsequent discussions it is not appropriate to make these comparisons. This section needs to

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be reworked. A better approach might be to discuss the problems with comparing immersion and contact and then go onto say what the experiments tell us.

24) Similar to the above comment, it is not clear to me that the CLINCH/IMCA comparison is valid.

25) P7838, ln 4. This needs a reference. The only study to experimentally show scaling with surface area and time in the way described is Murray et al. (Heterogeneous freezing of water droplets containing kaolinite particles , Atm. Chem. Phys., 11, 4191-4207, 2011).

26) P7838, ln 10-20. These ideas are similar to what Leisner and co-workers have very recently published and these new articles should be mentioned: (http://pubs.rsc.org/en/content/articlepdf/2013/fd/c3fd00033h) (http://www.atmos-meas-tech-discuss.net/6/3407/2013/amtd-6-3407-2013.html).

27) P7839, ln 8-10. This comment is very odd. Gorbunov (2001) says nothing about contact nucleation. Amend the sentence accordingly.

28) P7839. 'why contact freezing is the most efficient ice nucleation mode' and also the last sentence of the next paragraph. These statements cannot be made! There is nothing in this paper that quantitatively shows contact nucleation, as in collision of an aerosol particle with a droplet, is more efficient. The way this is written will be used by people less familiar with the literature to say that contact is always the most important mode of nucleation, including in clouds as well as in experiments.

29) P7841, ln 10-13. I don't understand this, the crystals used in the cold plate experiments were 100's micrometres – much larger than those in the wind tunnel!

Technical comments 1) Ln 21: 'act' not 'acts'.

2) P7814, ln 2-5. Sentence doesn't make sense.

3) P7817. Ln 18 'get in' should be 'come into'

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4) Lots of problems with bibliography.

5) P7830. Ln 14-16: Not sure what the 'previously mentioned method is'. Revise.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 7811, 2013.