

**We would like to thank Dr. Murray for his comments and suggestions, which helped us to considerably improve the manuscript. Specific answers and manuscript modifications related to his comments are given below in bold.**

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

**We agree with you. The title of the revised manuscript was changed taking into account your suggestion ("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 mechanically 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.

**The different types of contact nucleation are explicitly distinguished in the revised manuscript.**

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.

**A new paragraph was added to the introduction where the atmospheric relevance of contact freezing is further discussed. The suggested references were also added to the revised manuscript.**

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. 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.

**We agree that there are some gaps when experimental results from immersion and contact freezing are compared in section 3.6. Therefore you are right that it cannot be ruled out that experiments of immersion freezing could find a higher freezing temperature than obtained in contact freezing mode depending of the design of the experiment. We added a word of caution.**

**However, the cold plate studies conducted by Shaw et al. (2005), Durant et al. (2005) and Fornea et al. (2009) are quantitative. Those studies clearly show that the freezing of liquid droplets occurred at warmer temperatures due to contact freezing.**

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?

**We agree that the comparison of frozen fraction curves has some limitations. The best to compare different freezing modes will be a comparison of the nucleation rates of each mode under similar conditions. That is the reason why we propose to conduct experiments where the number of IN involved in each mode (or the surface area of the IN which is in contact with water) is measured and monitored to conduct a fair comparison between these two freezing modes.**

**With contact FE as a function of temperature and immersion freezing nucleation rates one could simulate the competing mechanisms with a process model under different conditions. Therefore we first suggest a possible approach based on the ideas from Dr. Alexei Kiselev (Fig 15) for lab studies.**

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.atmosmeas-tech-discuss.net/6/3407/2013/amtd-6-3407-2013.html>).

**These papers were added in different sections of the revised manuscript**

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)

**The word amorphous was added and the suggested reference as well. The distinction between pure organics and SOA was also added.**

8) Ln 14. Avoid the use of the term 'good IN'. This term is subjective and could be deleted.  
**The word good was replaced by efficient.**

9) Ln 15-17. DeMott (1990) and Mohler (2005) do show soot to nucleate ice, contrary to what is stated.  
**Mohler et al. (2005) reference was corrected and the DeMott et al.(1990) was replaced to DeMott et al. (1999) which is the correct reference.**

10) Use of word 'believed'. To me this word implies faith rather than a fact or idea which has been arrived at through scientific reasoning.

**This word was changed along the revised manuscript.**

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

12) P7819. Ln 11. Is there also a dependence on RH, size, etc.  
**The other key parameters were added to the sentence.**

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?  
**Since the time scales for both immersion and contact freezing are very small, the impact of this mechanism on modifying the IN surface properties must be pretty small if not negligible. We included it in our manuscript in order to summarize all possible mechanisms suggested in the literature.**

**Something that it is related to this mechanism but it was not introduced by Fletcher (1970) and Guenadiev (1970) is the amount of soluble material into the IN. It is very likely that a mineral dust particle has soluble material on the particle surface. Once this particle is immersed into a liquid droplet, the soluble material gets dissolved and "new" active sites may be "created". In this case immersion freezing would be more efficient than contact freezing because these "new" active sites would not be available for contact freezing.**

14) Also, replace 'it is believed' with 'They suggest'.  
**The suggestion was added**

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.  
**We agree with you. This paragraph was re-phrased.**

16) P7824, Ln 10. Mention the material used by Gurganus et al. Maybe this result is specific only to this material.  
**The material is mentioned in the revised manuscript and the new paper from the same group was also added.**

17) P7826, Ln 23-26. The two consecutive sentences are contradictory.  
**The paragraph was corrected**

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.  
**The word was replaced**

19) P7827, Ln 25. Why is this interesting?  
**We found it interesting because the bacteria profile looks pretty similar to a typical homogenous freezing activation curve (i.e., a full activation in a very small temperature range and/or time). This was not the case for the other tested materials, which showed a profile typical for a heterogeneous freezing activation. We added this argumentation.**

20) P7828, Ln 15. Add references.  
**Relevant references were added.**

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.

**Although we observed a small difference in the “onset” freezing values, we agree that this is not enough to claim a significant difference. This discussion was modified accordingly.**

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?

**The freezing efficiencies reported in Ladino et al. (2011b) are overestimated due to an overestimation in the droplet size when calculating the collision efficiencies. The droplet size estimation along CLINCH was not possible due to technical limitations. Additionally, this overestimation was partially caused by the used laser in the IODE detector (this is now added to the revised manuscript).**

**We do not think that this needs to be discussed in much detail here, since the take home message of Figure 12 is the poor agreement between the reported freezing efficiencies. The limitations of each technique were included in sections 3.1-3.4.**

23) P7835, In 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 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.

**In order to propose a better way to compare contact freezing and immersion freezing in section 3.6, the available laboratory data was first introduced. As mentioned in the answer of your comment 4, we agree about the limitations that these data sets have. This is clearly stated in the revised manuscript. However, we kept this comparison, which is useful to provide a context and to guide the readers and the scientific community on what is needed in future experiments.**

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

**The limitations of this comparison are clearly presented in the original manuscript. We do not state that this is a quantitative comparison and that we should draw definitive conclusions based on this data. We kept the CLINCH/IMCA data since it nicely introduced the limitations of the available data and presents the needs for futures experiments similar to the wind tunnel data. In the revised manuscript it is clearly mention that this is a qualitative comparison and the presented data are not used to draw any conclusion.**

**Note that the closer we look into the details the obvious it is that both mechanisms cannot easily be compared, even though this has been done in the past. This is now added in the revised manuscript as the take-home message of these data.**

25) P7838, In 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).

**The reference was added in addition to Hoffmann et al. (2013a)**

26) P7838, In 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.atmosmeas-tech-discuss.net/6/3407/2013/amtd-6-3407-2013.html>).

**In the acknowledgements it was clearly stated that these ideas were obtained from Alexei Kiselev who is one of the scientist involved in the above articles. In the revised manuscript, in addition to the acknowledgements the Hoffmann et al. (2013a) is also cited.**

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

**Although Gorbunov et al. (2001) did not state that their experiments were representative to contact freezing, it was not possible to fully attribute their results to one single heterogeneous nucleation mode. That is the reason why Hoose and Mohler (2012) include a question mark when attributing the freezing mode of Gorbunov's data. Since the data presented in section 3.7 is based on the parameterization developed by Diehl et al. (2006) we need to include Gorbunov et al. (2001) into the discussion because their data was used to develop the above mentioned parameterization.**

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.

**We presented the available literature data on contact freezing in the lab and the limited available experimental comparisons between contact and immersion freezing. Most of the previously mentioned comparisons are qualitative but the cold plate comparisons are considered quantitative. Taking into account the quantitative comparisons it is possible to conclude that contact freezing is more efficient than immersion freezing because it takes place at warmer temperatures than immersion freezing.**

**The sentence was slightly modified to be clear that this statement is based on the cold plate results to avoid any confusion with atmospheric data or with other experimental setup.**

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!

**You are right. This sentence was removed.**

Technical comments

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

**This was corrected**

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

**This definition was taken from the definitions by the International Commission on Clouds and Precipitation (ICCP) and the International committee on Nucleation and Atmospheric Aerosols (ICNAA). This concept was summarized by Vali 1985 in his nucleation terminology paper.**

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

**This was corrected**

4) Lots of problems with bibliography.

**The bibliography was revised**

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

**In the revised manuscript it is mentioned that the NCAR counters do not have the modification conducted by Langer et al.(1978) which are needed to conduct contact and immersion freezing experiments.**