Response to anonymous Referee #1

We thank the reviewer for the positive review and the careful reading of the manuscript. The comments are addressed below in italic.

I have reviewed the article “Ice nucleation efficiency of AgI: review and new insights” by Marcolli et al. This article covers literature dating back to the 1960s on ice nucleation on AgI. The article focuses on the molecular level processes that may cause AgI to be an efficient ice nucleus. The article is timely, in that there is increasing interest in the processes that lead to efficient ice nucleation, and it would be of great interest to the readers of Atmos. Chem. Phys. I have a number of minor suggestions for authors to consider, which are listed below:

p2 line 7: It is interesting that condensation freezing is considered separately from immersion. To what extent does condensation freezing have a different efficiency than immersion? Can it also take place below water saturation, like deposition nucleation?

Condensation freezing occurs concurrent with the activation of a particle to a cloud droplet and its efficiency might be different from the one of immersion freezing. A definition of condensation freezing is also given in Vali et al. (2015). Although it is not clear whether immersion and condensation freezing have different efficiencies, these two modes are usually discriminated. We added a section in the revised manuscript that describes the different modes of heterogeneous ice nucleation.

pg 2 line 32: Is having a defect enough even if none of these “requirements” are met?

It is hard to imagine a defect that does not influence surface charge, polarizability, hydrogen bonding or van der Waals interactions. Therefore, when there is a defect, at least some of these properties change.

pg 2 line 31: It’s odd to refer to these as requirements. Are they requirements, or simply properties that promote ice nucleation?

We agree that “requirements” might be a too strong expression in this context and replace it by “properties”.

pg 3 line 28: You should specify if the symbols in Fig 1 are filled or open.

They are filled. This is now stated in the text of the revised manuscript.

pg 4 line 6: What is meant by “adsorb iodide ions on the surface”? Does it mean that iodide in the solution is surface active or something else?

This is explained in Appendix B3: “Because of the better solubility of Ag^+ compared to I^-, a neutral AgI crystal will be negatively charged in water or aqueous solutions.” Surface activity is not meant. To make this clearer, we replace “adsorb” by “enrich”:

Fig. 1: Indicate that the orange circles and triangles overlap.

This is done in the revised manuscript.

pg 11 line 25: If AgI particles stay on the surface of the droplets during immersion freezing, how is an “inside-out” contact freezing mechanism being separated from immersion freezing?

In continuous flow diffusion chambers, particles freely float in a droplet and can take a position on the droplet surface or immersed in the droplet depending on the balance of interfacial and surface forces. Therefore, an experiment that is intended to study immersion freezing can turn into a contact freezing experiment. This circumstance has not gained much attention yet. To determine whether immersion or contact freezing inside-out is at work, the position of the particle in or on the droplet must be derived based on the wetting behavior of the particle. This is discussed in more detail in the companion paper by Nagare et al. (2016).

pg 11 line 1: A reference is needed here. Water molecules at the interface participate in fewer H-bonds and therefore may have greater mobility. In polymers for example, there is greater mobility at the air-polymer interface than in the bulk of the polymer.

The reference is Taylor and Hale (1993).

pg 12 line 13: Why should the amount of water adsorbed on a surface at a given RH matter for immersion freezing, where the nucleation presumably takes place at a location on the heterogeneous nucleus inside the water droplet?

If ice nucleation happened on the surface that is totally immersed in water, RH should not matter. However, the studies compiled in Fig. 1 show that freezing temperatures *are higher for setups where the particle can access the water surface compared with experimental arrangements where the droplet is immersed in or covered with oil. This indicates that freezing is not occurring on the surface that is totally immersed in water but either on the contact line to air or in water patches on the surface exposed to air. Water patches on the surface are indeed influenced by RH.*

pg 12 line 15: I do not understand this result for condensation freezing. Again, the surface is immersed in a solution in this mechanism, so it is hard to believe that surface adsorbed water due to RH has an effect. Activation may mean that the solution is most dilute, allowing freezing to occur.

The argumentation is based on the assumption that ice nucleation occurs on water patches on the surface exposed to air.

pg 12 line 31: Why is the observed dissolution greater for the contact freezing experiment (with the same particle diameter)? Also, is this mobility diameter?

The particle diameter is the same but the droplet diameter is different. Droplets in IMCA/ZINC are 18 – 20 µm in diameter, the ones in CLINCH are 80 µm in diameter. Therefore, dissolution is greater for the contact freezing experiment in CLINCH than for the immersion freezing experiment in IMCA/ZINC. The particles are size selected according to mobility diameter.

pg 13 line 17: As long as the surface is dissolved, the ice nucleation activity should be reduced according to the arguments in this section. Why should the amount of dissolution matter? Do you suspect that at 0.04% dissolution the surface is not fully dissolved? Are these arguments consistent with the comparison of the two particle sizes?

The amount of dissolution should not matter but it should matter whether a particle is in the process of dissolving, because the surface should be different for a dissolving particle that is not in equilibrium with the surrounding solution compared with a particle that has reached or almost reached equilibrium with the solution. Equilibrium is reached or almost reached in the case of the immersion freezing experiments because activation to a water droplet occurs already in the IMCA section at warmer temperature. In the CLINCH experiment, a particle that collided with a droplet is continuously dissolving while it passes through the chamber. This is explained in Appendix B6.

pg 14 line 8: Why does the addition of AgCl increase the ice nucleation activity? (This is stated in the conclusions, but it should be stated here as well.)

In the presence of AgCl, AgI-AgCl solid solutions are formed, which have a better lattice match with ice than pure AgI. This is discussed in Sect. 3.3. We now mention this in the revised manuscript and refer to Sect. 3.3.

Fig. A2: SAED diffractograms would be interesting to report to see the amount of different polymorphs in your sample. Were these taken?

No, we just measured XRD diffractograms of the bulk sample that revealed a mixture of the β- and the γ-phase.

Wording/Grammar: pg 3 line 9: “Such an analysis should also allow us to identify factors” *Done*

pg 6 line 27: “They noted that a supersaturation of 20%....” *Done*

pg 8 line 29 & 30: For Miller indices, it is more typical to write 001 with a bar over the 1 rather than 00-1. *Done.*

pg 10 line 21: What is meant by “in sum”?

“*in sum” is misplaced. We rearranged the text and put it in front of “to low configurational entropy”*

pg 15 line 14: change “with different shares” to “in different proportions” *Done*

Reference:

Vali, G., DeMott, P. J., Möhler, O., and Whale, T. F.: Technical Note: A proposal for ice nucleation terminology, Atmos. Chem. Phys., 15, 10263–10270, doi:10.5194/acp-15-10263-2015, 2015.