

Dear Dr. Tim Garrett,

On behalf of myself and my co-authors, I would like to thank the editor and reviewers for the valuable feedback on our revised manuscript.

We agree that the abstract should be shortened and are glad for the opportunity to make the suggested improvements for the final version. The revised abstract is now close to the 250 word limit and makes a stronger statement on the potential implications for ice nucleation in the atmosphere. We would like to confirm with the editor that our statements are reasonable for this publication (not too provocative)?

The additional comments from the reviewers have each been addressed and our responses are below in blue text. We greatly appreciate the attention to detail from the reviewers, which has ensured that the final manuscript is of a good quality.

Kind Regards,  
Elise Rosky

### Reviewer #1

The authors have addressed the comments of the reviewers carefully and in detail. There are only few points left that need to be corrected or clarified.

Line 60: “The molar volume difference between water and ice is negative...” The way it is formulated, it remains ambiguous whether the difference “water – ice” or “ice – water” is negative. The formulation should be improved to remove this ambiguity.

Thank you for this suggestion. The improvement has been made near Line 60 where we now express the explicit definition  $\Delta v_{is} = v_l - v_s$ .

Line 247: “...dependent, the...” instead of “...dependent; The...”

Done.

Line 274–275: “...from Qiu et al. (2018) which identified mW water at the mW-Carbon interface as having thermodynamics similar to that of the bulk liquid.” This sentence is grammatically incorrect. Should “having” be removed?

Done.

Line 303: “We find that the data follows a linear trend as anticipated.” Consider a more cautious formulation: “We find that the data can be described by a linear trend as anticipated”.

Done.

Figure 4: Fonts are in general rather small and the one chosen for the x-axis in panel b is much too small and must be increased. The label below “1 atm reference value” on the x-axis of panel b is not readable.

Fonts have been increased.

Line 401: the abbreviation for “ice nucleating particle” is usually “INP” and not just “IN”.

This is true. We also use the term “ice nuclei” in this paragraph, which can be abbreviated as “IN”. In the equations discussed in this paragraph we use the abbreviation “IN” in the subscripts. In this case it’s a little more convenient to stick with “IN” in order to not muddle the subscripts which also use “P” to denote pressure. We hope this abbreviation is still clear enough to the readers.

Lines 432–434: “Insofar as our results depend on the water activity (Nemec, 2013), they are consistent with previous efforts to cast nucleation in that framework (Koop et al., 2000; Knopf and Alpert, 2013).”: Note that water activity and pressure both act on the chemical potential difference between ice and the water phase, but they are not the same or interchangeable.

The authors agree with this comment. We have adjusted the text accordingly to clarify that pressure and water activity are related through their contribution to the chemical potential difference between liquid and ice.

Lines 443–445: “Experimental measurements of freezing inside porous material observed no freezing enhancement in pores with confinement on similar size scales as studied here (~2 nm diameter) (e.g., Marcolli, 2014),...”: Note that the experiments compiled in Marcolli (2014) are slurry experiments. The pores have no air-water interface. Therefore, no Laplace pressure is expected. Moreover, ice nucleation does not occur within the pores but ice grows into the pores when the temperature is low enough. A valid reference should be given for this statement.

The presence of Laplace pressure is not necessary in this argument because this paragraph is considering how different confinement geometries will result in mixed influences on the ice nucleation rate. One may anticipate an increase due to Laplace pressure, but this could be suppressed or further enhanced if confinement effects are present.

Figure 3 of Marcolli 2014 shows freezing point depression in completely filled pores with increasingly small diameters. This is a good summary of experimental results showing that small scale confinement does not always result in enhancement in nucleation rate.

In the final manuscript we keep the same reference but adjust the text so that there is no longer a discrepancy between our statement and the provided reference. The revised section can be found in the second-to-last paragraph of Discussion section.

*Marcolli, C.: Deposition nucleation viewed as homogeneous or immersion freezing in pores and cavities, Atmospheric Chemistry and Physics, 14, 2071–2104, 2014.*

Lines 448–449: “The key mechanism for rate enhancement in the slit pore configuration is the density oscillations induced by a flat interface (Cox et al., 2015; Bi et al., 2016; Lupi et al., 2014), which is destroyed by curved geometry.” Please explain what you mean with this sentence. Up to here, the enhancement in pores has been explained by the Laplace pressure arising in pores with air-water interface. Density oscillations are a completely new explanation.

The reviewer makes a good point. We now introduce this concept at an earlier point in the manuscript, when we first describe the confinement effect in the 18A tall simulation cell (Section 3.3 Paragraph 1).

## Reviewer #2

I commend the authors on the greatly improved manuscript that has resulted from their thorough revisions. The authors have addressed all my questions, comments and concerns. I have only one comment/suggestion and a suggestion/request of correction that I hope the authors will address in the final version of the manuscript:

Line 272) "This could indicate the thermodynamic properties of mW water are less influenced near the substrate compared to the ML-mW model. This interpretation is supported by evidence from Qiu et al. (2018) which identified mW water at the mW-Carbon interface as having thermodynamics similar to that of the bulk liquid."

My comment/suggestion: Note that the reason why the thermodynamics of mW at the carbon surface is almost same as for bulk water is because the contact angle for the liquid on that surface is almost 90 degrees. The authors could use the equations of that paper to derive how a contact angle of 50 degrees for the surface with ML-mW impacts the thermodynamics, and whether that explains the differences they see for these two water-surface models.

We will incorporate this important point into the text. We thank the reviewer for the suggestion and look forward to exploring this idea in more detail in future work.

Line 356) "This lack of heterogeneous ice nucleation in the immediate vicinity of the air–water interface could be related to premelting at the ice–vapor interface, described for the mW model by Qiu and Molinero (2018)."

My comment/request: Qiu and Molinero 2018 demonstrates that heterogeneous ice nucleation cannot occur on a surface that exhibits premelting presented using solely thermodynamics and nucleation theory. It is a general derivation that does not depend on results for any particular model. Please replace "described for the mW model " by "as deduced using thermodynamics and nucleation theory"

Done.