Authors' response to reviewers' comments for "The Microfluidic Ice Nuclei Counter Zürich (MINCZ): A platform for homogeneous and heterogeneous ice nucleation" by Florin N. Isenrich, Nadia Shardt, Michael Rösch, Julia Nette, Stavros Stavrakis, Claudia Marcolli, Zamin A. Kanji, Andrew J. deMello, and Ulrike Lohmann

We are grateful for the anonymous reviewer's comments and constructive suggestions that improved our manuscript. Below we outline our point-by-point replies and revisions to the manuscript. Page and line numbers refer to the uploaded document with tracked changes.

Reviewer #2

Comment

This paper describes the development of an improved microfluidic device that has lower temperature gradients and less water and gas permeability. The paper is suitable for AMT and should be publishable after some revisions.

This paper does need a thorough editing for content and especially length. The paper should be considerably shortened before final publication. There are also grammatical errors and statements that are not quantified. Some of these are called out specifically below but I encourage the authors to edit and shorten the paper before resubmission.

Authors' response

We have implemented the specific suggestions of the reviewer for shortening the paper, and we have checked the whole manuscript for opportunities to condense where possible.

Comment

Abstract:

1. The Abstract is too long and reads like an introduction to a paper. Please remove extraneous details and cut the text could by 50% which can be done without loss of important content. Please concentrate on the instrument being presented.

Authors' response

We have now significantly shortened the abstract; please see the tracked changes in the updated manuscript.

Comment

2. "requirements: (i) high accuracy and precision in measuring droplet temperatures within 0.2 K (ii) ability to reach the homogeneous freezing point of pure water, with a median freezing temperature of 237.3±0.1 K…" These appear to be capabilities, not requirements? Also, here and throughout the paper : the uncertainties here seem contradictory. Is the latter the uncertainty or spread in the freezing temperature? The numerous uncertainties, specifically in temperature, used throughout the paper need to be explicitly stated for clarity.

Authors' response

We are now more exact in our language around uncertainty, and we explicitly refer to accuracy and precision instead. Immediately below, we only list changes to the abstract, and later, in response to a follow-up comment by the reviewer we list the remaining changes.

Changes to manuscript

Page 1, lines 31–32: changed "high accuracy and precision in measuring droplet temperatures within 0.2 K" to "high accuracy of 0.2 K in measured droplet temperature"

Page 1, lines 33–34: changed "median freezing temperature of 237.3 ± 0.1 K" to "median temperature of 237.3 K with a standard deviation of 0.1 K".

Comment

3. The Title and Abstract state MINCZ is going to provide homogeneous freezing data but the above quote and paper then says this is restricted to only pure water – something that doesn't exist in the atmosphere. This means a qualification – "…a platform for homogenous water and …" needs to be added.

Authors' response

There is consensus that the homogeneous nucleation rate of pure water is important in the atmosphere (e.g., Murray et al. Nature, 434(7030), 202, 2005). We demonstrate that MINCZ can be used to observe both homogeneous and heterogeneous freezing by investigating pure water and microcline suspensions. The platform is not limited to these specific cases and could also be used in the future to investigate the freezing behaviour of other solutions and suspensions.

Comment

4. "to detect mediocre and poor ice-nucleating particles" please define what mediocre and poor means (in temperature and saturation)? Please note that throughout the text words that are qualitative are often used when quantitation is necessary.

Authors' response

In shortening the abstract, we have deleted this phrase. However, we also used this wording in the main text of the manuscript, and we have now better defined what we intend to convey.

Changes to manuscript

Page 3, lines 78–79: changed "with mediocre or poor activity" to "that are active at temperatures between that of homogeneous freezing and the melting point of water"

Page 13, lines 376–377: changed "due to mediocre ice-nucleating particles" to "due to the presence of ice-nucleating particles"

Page 16, line 475: changed "due to mediocre or poor" to "catalysed by"

Comment

Materials and Methods

1. Many parts of Figure 1 seem extraneous and the figure overall confusing. Inclusions of things such as syringes, computer, cooling bath complicate the figure without adding any detail not in the text. Parts c and d are the most important and could be combined with a very simple composite of a and b to improve the figure. Please consider what really needs to be in the figure and can't be in the text and eliminate for clarity.

Authors' response and change to manuscript

We have considered the reviewer's suggestion, and we have revised Figure 1 to only display the essential components of the instrument. We have removed panel (a), and

retained panels (b) through (d), as suggested, and we have updated the caption and main text accordingly (see tracked changes in document).

Comment

2. Can the authors explain in the text why a ~75 μm droplet size was chosen? Is this a fabrication limit? While it is understood microfluidic devices can't attain the small size of most atmospheric droplets it is important to detail why this size was chose and what implication this volume has in relation to the atmosphere.

Authors' response

The lowest possible droplet diameter can be seen as a balance between the available diameter of the PFA tubing and the practicalities of generating such small droplets. A diameter of 75 μm was chosen, because it was one of the available dimensions of commercially available PFA tubing into which droplets are loaded after droplet generation. We also tested PFA tubing with an inner diameter of 50 μm, but due to the high pressure drop arising from such a small inner diameter, stable droplet generation became more challenging. The high pressure also increased the frequency of PDMS delamination from the glass slide. Additionally, it is difficult to detect the freezing of smaller droplets unless a higher magnification objective is used (and then fewer droplets can be investigated simultaneously due to the smaller field of view).

From the perspective of homogeneous ice nucleation, the droplets themselves should be small enough to avoid heterogeneous freezing caused by impurities in the pure water (such as the gradual increase in frozen fraction at higher temperatures, as seen in Peckhaus et al. (Atmos. Chem. Phys., 16(18), 11477, 2016) and Brubaker et al. (Aerosol Sci. Technol., 54(1), 79, 2019)). At the same time, to investigate heterogeneous ice nucleation, it is better to investigate larger droplet volumes so that the surface area of ice-nucleating particles is distributed more uniformly amongst the generated droplets.

Comment

3. What information is detailed in Figure 2 that is not given in the text? It appears this figure simply repeats what the text says in a flow chart format.

Authors' response and change to manuscript

We have considered the reviewer's suggestion, and we have deleted Figure 2 from the manuscript.

Comment

4. The caption of Figure 3 is a method description, which belongs in the text, not a figure caption.

Authors' response

We have shortened the caption of Fig. 3 (now Fig. 2) by condensing two of the sentences into one, removing unnecessary explanation, and we have moved one sentence to the main text.

Changes to manuscript

Fig. 3 (now Fig. 2) caption: replaced the second and third sentences in the caption with "In the first step, locations where droplets potentially froze are automatically screened (highlighted in blue pixels for the two consecutive images and in green pixels for comparison to the image two time steps prior to I_t)."

Page 12, lines 340–341: deleted “To reduce the number of potential droplets that must be classified by the user”

Page 12, lines 349–351: added “Together, the above criteria aid in removing false positives from consideration and limit the number of potential freezing events that need to be presented to the user for visual classification.”

Comment

Results

1. See also abstract. Can the authors explain the temperature uncertainties in the paper, specifically this section? For example, 2 significant figures in “237.41 ± 0.04” is in excess of the earlier statements on the equipment capabilities of .2. Figure 4 then appears to show a yet larger range in data which seems to be the most important uncertainty. A comprehensive explanation of the uncertainties and data ranges would greatly improve this paper.

Authors’ response

We clarify that the accuracy of our reported temperature is 0.2 K. To better convey the meaning of the number following the ±, we instead explain it in words.

Changes to manuscript

Page 1, lines 31–32: changed “high accuracy and precision in measuring droplet temperatures within 0.2 K” to “high accuracy of 0.2 K in measured droplet temperature”

Page 1, line 33–34: changed “median freezing temperature of 237.3 ± 0.1 K” to “median temperature of 237.3 K with a standard deviation of 0.1 K”.

Page 10, line 276: changed “uncertainty in our temperature measurement” to “accuracy of our temperature measurement”

Page 13, lines 360–361: changed “reproducible within a narrow temperature range of 237.3 ± 0.1 K” to “237.3 K with a precision of 0.1 K (standard deviation of the three experiments)”

Page 13, lines 364–365: changed “an even narrower median temperature range of 237.41 ± 0.04 K” to “a better precision of ± 0.04 K (standard deviation) in median temperature”

Page 14, lines 395–396: changed “244.6 K ± 0.7 K” to “244.6 K, with a spread of ± 0.7 K (standard deviation)”

Figs. 4 and 5 (now numbered 3 and 4): last sentence in each caption, replaced “uncertainty” with “accuracy”

Comment

2. The captions in Figures 4 and 5 are too long and include detail that is not a description of the figure. This text needs to be move to the main text.

Authors’ response

In the caption of Fig. 5, we were able to shorten some of the text and move the details of the Peckhaus et al. (2016) data to the main text.

Change to manuscript
Fig. 5 caption: deleted "where 0.2 nL aqueous droplets with 0.05 wt% microcline suspension were printed onto a solid substrate and cooled at 1 K min^{-1}."

Page 16, lines 449–450: added "in printed 0.2 nL droplets"

Comment
Conclusions

1. Consider not using "homogeneous" to describe the droplet size here as it is then used for the freezing mechanism; this is confusing.

Authors' response
We agree with the reviewer's suggestion.

Change to manuscript
Page 16, line 466: changed "homogeneously-sized" to "monodisperse"