Response to Referee #1

In this manuscript the authors test different substrates, water sources, droplet matrixes, and droplet sizes with the goal of improving the droplet freezing technique. Since the droplet freezing technique is being used by several groups to quantify and understand ice nucleating particles in the atmosphere, this manuscript is useful and appropriate for AMT. This manuscript will be especially useful for new researchers to the field of atmospheric ice nucleating particles. I suggest publication in AMT after the authors have had a chance to address the following comments.

We thank the referee for their positive response to our manuscript. We have responded to each comment made in turn along with the revision made to the manuscript where appropriate.

Title. I would delete "cleaning up our water" from the title, since the manuscript includes more than just experiments to remove impurities in water.

While we understand the referee's suggestion, we prefer to retain the original title. The freezing artifacts caused by substrate interactions can appear to have a similar and difficult to distinguish effect on the background freezing temperature in "pure" water controls, and thus we feel our manuscript does largely focus on the effects of real and apparent water impurities. Gabor Vali made similar a similar assessment in his comment on the discussion paper. Furthermore, if substrate interference can be eliminated (such as in our microfluidic approach), then any remaining background freezing above the expected homogeneous freezing temperature are due to any trace and difficult to eliminate water impurities. We think the title will make the focus and purpose of our manuscript clearer and thus reach a wider audience.

Page 9, Lines 275 – 285. Can mineral oil have different freezing temperatures depending on the average molecular weight? Maybe the WISDOM technique used a different type of mineral oil? I suggest the authors add additional information on the conditions used in WISDOM and the conditions used in the current experiments.

Molecular weight certainly plays a role in the freezing temperature of compounds. The difficulty with mineral oil is it is a mixture of dozens or hundreds of compounds derived from petroleum refining. There is no defined structure for mineral oils. Reicher et al. (2018) used mineral oil from Sigma Aldrich. Sigma has 11 different products called "mineral oil", all of which have the same CAS number despite their different physical and chemical properties. Reicher et al. almost certainly used a different mineral oil than we had, since we used mineral oil from VWR. We have included in this manuscript as much detail as was provided by Reicher et al. (2018) and believe we have adequately described the freezing of mineral oil found in this study. The relevant text has been modified as follows:

"However, the WISDOM microfluidic DFT device uses mineral oil for droplet creation and storage (Reicher et al., 2018). The device has successfully measured homogeneous ice nucleation down to -36 °C. Perhaps the surfactant (Span80, 2 wt%) used to stabilize the immersed droplets prevents freezing of the mineral oil. There are also a wide range of different mineral oils available from common chemicals suppliers and the specific type of oil used in WISDOM is not known. Alternatively, the optical fogging may not be visible when such a small volume of oil is above the droplets, as is the case for microfluidic devices. Despite the promising results from the WISDOM method, we are wary to suggest that any other groups attempt the use of mineral oil for droplet freezing measurements before further investigating how the oil's freezing may impact water droplet freezing. For all oil-immersion experiments mentioned in the following sections, squalene oil was used as the oil matrix, following the method of Wright and Petters (2013). Previously, we have shed light on squalene oil reducing the observed ice nucleation activity of Snomax bacterial particles and concluded this was due to hydrophobic partitioning of large protein aggregates (Polen et al., 2016). This was only observed in droplet refreeze experiments of Snomax, and we do not observe this effect on any other particle sample type we have tested. Squalene oil remains our recommended immersion oil for most droplet freezing experiments."

Page 9, Line 299. This sentence suggests that the water was filtered for many weeks. I assume that this is not correct.

Yes, this was unclearly worded. One filter was reused multiple times to filter several samples of HPLC water over the course of weeks. The phrasing has been changed as follows:

"The biggest deviation came from runs of HPLC water that was filtered multiple times over many weeks using the same Anotop filter."

Page 10, Lines 317-319 and Page 19, Lines 581-582. The issues with the MilliQ produced water were blamed on the particle membrane filter. How do the authors know that the particle membrane filter was the source of the problem? I would have guessed any issue associated with the particle membrane filter would be rectified with the 0.02 micrometer post-filter.

That is correct, with the addition of the Anotop 0.02 micrometer post filtering we should see similar results. However, we only saw an improvement once the MilliQ system particle filter was replaced. At this time we decided to switch to bottled HPLC water to not have to deal with any uncertainty and variability associated with the MilliQ system. We do not know why Anotop filtration did not prevent this contamination. We have clarified this in the text as follows:

"The MilliQ-produced water can result in very inconsistent results for pure water droplet controls if the particle membrane filter is not changed on a regular basis. These

contaminants were apparently not removed by filtering the poor-quality MilliQ water with a 20 nm pore Anotop filter, for reasons unknown to us."

On Page 14, it was not clear how the Vaseline surfaces were made. Line 431 suggests Vaseline was spread on the sample dish. What was the sample dish made of? On page 19, Line 563, it sounds like the Vaseline was spread on the hydrophobic glass slides? Please clarify.

The Vaseline was spread on the aluminum sample dish directly. The later statement on page 19 was meant to compare the Vaseline to hydrophobic coverslips as separate tests, not to imply that coverslips were used underneath the Vaseline. The phrasing of Line 561 has been changed to: "compared to hydrophobic cover slips".

The format of the references needs to be improved in several cases.

Thank you for pointing this out. The citations for Lohmann and Fleichter (2005), Wheeler et al. (2014), Wilson et al. (2015), and Tobo (2016) references have been corrected.