Response to Referee #1

We would like to thank the referee for their insightful comments and have responded below. The referee comments are highlighted in red with our responses in black.

Review of "An instrument for quantifying heterogeneous ice nucleation in multiwell plates using infrared emissions to detect freezing" by Harrison et al. General comments: I support publication of this manuscript in AMT. The research aligns well with the scope of AMT. The reviewer finds the application of release of latent heat for detecting a freezing event in immersion mode ice spectrometer unique. The authors successfully present the applicability of their technique (IR-NIPI) to characterize immersion freezing efficiencies of three different forms of the sample (incl. chips, powder and ambient particles collected on the filter and scrubbed with water) at T > -22 ◦C. In particular, its applicability to the atmospheric sample seems promising the reviewer finds Figs. 8 and 9 nice and elegant. With further improvements in the temperature uncertainty (±0.9 ◦C is reported in the manuscript) and applicability in different droplet volumes (so, wider T coverage), IR-NIPI may become very versatile in the INP research (specifically for biological high-T INPs). The review has only minor (but important) comments.

Comments

P1 L11-13: The main focus of the presented work is on novel application of latent heat in immersion freezing spectrometry, and the reviewer finds the discussion of online vs. offline unnecessary (especially in the abstract). L12-13 is erroneous – some cloud simulation chambers can assess multiple Ts. The reviewer strongly suggests removing "While instruments . . . Hence,".

Accepted, we agree and have removed this piece of text from the abstract.

P2 L36-38: Reference suggestion - Hande, L. B., and Hoose, C.: Partitioning the primary ice formation modes in large eddy simulations of mixed-phase clouds, Atmos. Chem. Phys., 17, 14105–14118, 2017.

Thank you. We have now added this reference.

P2 L40: ~1 L-1 at what temperature? Please clarify to the readers.

The study was of three case studies of cloud fields. The temperature at which this concentration was reached depended on the local INP spectrum. It is quite challenging to insert this information without distracting from the main point of the statement hence we would rather leave it as is.

P2 L42-43: Plus developing realistic but computationally inexpensive parameterization is also a key to what is addressed here by the authors.

We have changed the text to read "The ability to quantify INP spectra (INP concentrations as a function of temperature) and test the efficiency of proxy materials for ice-nucleating efficiency is invaluable for improving our understanding of cloud glaciation and developing computationally inexpensive parameterisations for atmospheric models."

P2 L45: Quantitatively define "warmer temperatures" perhaps with specific reference(s). L51-53 implies -11 ◦C as warmer temperatures?

We have altered the text to read "However it is not a trivial task, in part because INP concentrations are low (<0.1L^{-1}) (DeMott et al., 2010) and the sites on the surfaces which cause nucleation at warm temperatures (Whale et al., 2017; Vali, 2014) are rare. "

P3 L61-63: What about the Arctic? Some discussions may benefit the paper.

We agree that the Arctic is similarly low in INP concentrations, hence expect a similar situation to the southern ocean. We have now discussed this. "This can be improved with aerosol concentrators (Prenni et al., 2013; Tobo et al., 2013), but is still above the INP concentrations models suggest influence the properties of certain cloud types, such as low/high latitude cold-sector clouds (Vergara-Temprado et al., 2018)."

P3 L71: Reference suggestion - Stopelli, E. et al.: Freezing nucleation apparatus puts new slant on study of biological ice nucleators in precipitation, Atmos. Meas. Tech., 7, 129–134, 2014.

This has been added. In addition we have added Conen et al. 2012.

P3 L71: The reviewer thinks the discussion of previous studies applying latent heat release as an asset for ice nucleation research will benefit the paper. Please consider include and discuss; e.g., Marcolli, C. et al.: Efficiency of immersion mode ice nucleation on surrogates of mineral dust, Atmos. Chem. Phys., 7, 5081-5091, 2007.

We agree a discussion of this paper should be made and have added the following text in a new section where we also discuss other instruments one of the other referees highlighted: "While many instruments use optical cameras to detect freezing events (Whale et al., 2015; Budke and Koop, 2015; Häusler et al., 2018; Beall et al., 2017), , some researchers have used techniques which detect the release of latent heat associated with freezing.. For example, differential scanning calorimetry (Marcolli et al. 2007; Pinti et al. 2012) and infrared emissions (Zaragotas et al., 2016; Kunert *et al*. 2018) have been used.

P4 L99: ns(T)

Done

P5 L140-141: Very important statement – recap this point (sharp rise in T = +2 ◦C) in the abstract and/or conclusion section.

We have recapped this in the conclusion "A freezing event is detected as a sharp rise in freezing temperature to the equilibrium melting point and a novel calibration method has been proposed which relies on the return of water droplets to the equilibrium melting temperature of water, 0°C, after initial freezing." The sharp rise in temperature is already referred to in the abstract.

P6 L142-145: The authors may want to rephrase this part and explain the points more intuitively

We have reworded this section so that the reader may better understand the points made. "The 2°C threshold occasionally needs to be optimised to capture freezing events while eliminating the detection of false freezing events. For example, samples that freeze above -3°C are more difficult to detect because there is less heat released on initial freezing and crystallisation happens over a longer period of time (see section 2.4). Manual inspection is required in this temperature regime and the 2°C threshold adjusted accordingly. "

P7 L179-182: The authors may want to extend this part and explain the points more intuitively

We have rewritten this section to explain the process more clearly "Using the analysis code, an event is identified and recorded. The code then reads the temperature of the frame directly after this freezing event and calculates the difference of this value compared to 0°C to give an offset correction value, i.e. if the frame after freezing read 2°C then the correction factor for this well would be -2°C. This offset value is then subtracted from all of the temperature recordings for that specific well. The average correction value calculated for the IR camera via this method is -1.9°C with a standard deviation ±0.5°C."

P7 L189-192: The reviewer is curious if using different droplet volume can improve this uncertainty. The reviewer does not intend to ask any additional measurements (especially since ±0.9 ◦C uncertainty is well justified in L193-207), but doe the authors have any estimates of the maximum/minimum droplet volume that IR-NIPI can deal with?

We have not completed any thorough experiments with different droplet volumes but the 96 multiwell plates can hold 200μL droplets. The IR-NIPI should have no problem monitoring these volumes although the gradients within the wells will become larger. Volumes below 50μL's maybe possible but would be starting to come close to the limits of the IR cameras resolution. If the IR cameras resolution were to be improved then smaller droplet sizes should be possible.

P8 L214: Delete "see".

Done

P11 L298-300: 16.7 L/min * 100 min = 1670 L. . . The authors might want to check their nINP (L-1) since they might have employed a wrong Vs (Eqn. 2).

Thank you for noticing this. This was a typo and has now been corrected.

P11 L304: Was a dilation used to prepare suspensions for the ambient sample analysis? If not, no worries. But, if yes, the dilution factor is missed in Eqn. 2.

No dilution was used.

Fig. 7A: There seems some outliers within this T-ns(T) scale (i.e., 0.01 wt% run 2). What is responsible for them? Perhaps, it is due to what is addressed in L280-293? Please clarify.

We believe this may be related to the issues discussed in L280-293. We have added a sentence to emphasise this in the results discussion of Fig. 7A. "The $n_s(T)$ values derived from IR-NIPI with 0.01, 0.1 and 1 wt% NX-illite are shown in 7a. They are in good agreement with one another with lower wt% suspensions yielding data at lower temperatures and higher $n_s(T)$ values, as expected. The few data points from the 0.01wt% NX-illite run 2 which appear as outliers may indicate that the particles were not evenly distributed throughout the droplets."