Review of “Development of the Droplet Ice Nuclei Counter Zürich (DRINCZ): Validation and application to field collected snow samples” by David et al.

General comment

In this manuscript the authors describe and characterise a large volume immersion mode drop assay (DRINCZ). The authors thoroughly characterise the horizontal temperature gradient across the 96 well plate in the system and recommend a correction which is of use to other instrumental setups. The authors report a ± 0.9 ˚C uncertainty for DRINCZ and go on to validate the instrument by comparing to literature data of NX-illite. A field study investigating snow melt samples is also undertaken which shows agreement (mostly) with previous snow melt measurements. The authors then relate the INP concentrations measured to airmass trajectories and propose scavenging of INP by precipitation led to the lowest INP concentrations measured.

This manuscript is well written and presents results which are of interest to the ice nucleation community. The manuscript is in the scope of AMT and I support its publication after the following comments have been properly addressed.

Major comment

Although I like the manuscript and find the results of use to the ice nucleation community, I am unclear on the novelty of this instrument compared to others that have already been presented. The authors have acknowledged that the technique is based on the design of previous instruments. Is the method used to characterise and correct the horizontal gradients in the plate the only novelty? If so, I suggest this is made clearer in the final manuscript or that the unique traits of this instrument are further clarified.

The horizontal gradients of the plate have been characterised but the vertical gradients within the wells have not been explored. These should be discussed in the text. A reference to Beall et al. (2017) would be appropriate as they characterise the gradient within 50 µL droplets within wells with a similar profile (PCR plate).

Figure 9 displays data for NX-illite dilutions. I find the text a little misleading in presenting the data as though there are only a “few” outliers for the 0.01wt% dilution. The vast majority of data for all three triplicates for this dilution give higher freezing temperatures than higher weight percent suspensions (at the same value of ns). This is in contradiction to what we expect of ns. I believe the source of this error is different to the uncertainties characterised in previous sections as the data is consistently offset to higher freezing temperatures. This issue is not seen (in most cases) in the dilutions for the snow melt study and suggests this discrepancy may be material dependent and related to the distribution of particles. Although the authors do mention this issue, the extent of the discrepancy between dilutions is glossed over in the text. I must stress that I do not believe this inconsistency in the dilutions is a result of an error in the instrument but rather an error as a result of the material or sampling method. With this said, the results should be presented in the text to acknowledge the true extent of this discrepancy.

Minor comments

Line 67-77: Dilution is not the only means of changing the measurable range of INP. Concentrating the particles per droplet can also extend the range. I suggest this is added to the discussion.

Line 116: At what temperature does the ethanol bath start at and what temperature does it end, i.e. 0 ˚C to -30 ˚C.

In addition to this, if the sample is added to an ethanol bath at 0 ˚C (as suggested by line 165) is the sample allowed time to equilibrate? If not, this could lead to thermal gradients not just horizontally across the plate but vertically in the wells (see major comment). I suggest adding information on the cooling profile of the bath to this section.

Section 2.1.2: This describes the detection of freezing events in wells. Is this similar to other methods, e.g. (Stopelli et al., 2014)? Clarify what is different.

Section 2.2: What is the error of the sensor? What will be the fluctuation in the ethanol level? Do the authors consider it negligible?

Line 182-183: K type thermocouples can have large uncertainties, commonly ± 2.2 ˚C, compared to other thermocouple types (e.g. T type). Were these K type thermocouples calibrated other than by the manufacturer? What is the error of these? I suggest showing these errors in the figures (or an example of the errors).

Line 183-184: Were the wells completely filled with ethanol or 50 µL? If completely filled does this represent the gradients that would be present in the wells and plate in a typical 50 µL experiment?

Section 3.2: The characterisation of the horizontal gradient across the plate is very useful for the community. However, has the vertical gradient within the wells been considered (see major comment)? This system uses a similar well profile to that used by Beall et al. (2017) who found that a vertical stratification of 0.5 ˚C can be found in wells in which the headspace (air above the wells) is ≥6 ˚C warmer. Please discuss this in the text and reference Beall et al. (2017) where appropriate.

Also, you use the median freezing temperature of SA water to determine the offset in temperatures across the plate. Is there not a random probability of the SA water freezing at different temperatures without a temperature bias to start with? Would there also not be uncertainties due to accidental contamination in the individual wells during the setup of the experiment? Does this not create uncertainty in this experiment? What was the rationale for using SA water? If you were to use freezing temperatures (rather than direct measurements of the wells) then would something that froze more consistently at the same temperatures be a better standard to use, i.e. pollen has a narrow window for freezing.

Section 3.3: It is unclear to me why you assess the uncertainty of the instrument and combine this with the variability in the freezing temperature of SA water at this point. The water baseline in other studies, e.g. field-based studies, could potentially be worse (or better) than what you have done in these experiments. Should the experimental error as a result of the water impurities not be considered separately in respect to the particular experiment/environment?

Section 4.2: There is no mention on how these suspensions/dilutions are made, how are the particles suspended? This could be particularly important given the results for NX-illite. I recommend adding this information in this section or the methodology.

Figure 9: it looks like the temperature intervals where no freezing events were observed are displayed in this figure, i.e. when binning the data, temperature intervals where 0 events were observed are still shown in the cumulative plots. As there are triplicates in this figure it makes it hard to discern which data points are real freezing events and which are artefacts of the binning process. This makes it difficult to interpret the data and the extent of the discrepancy between dilutions. If this is the case, I suggest removing the data points from the cumulative plots where there were no freezing events within a temperature interval for clarity.

Line 355-357: I would not definitively say that the ns is extended as expected. All three triplicates for the 0.01wt% dilutions are giving warming freezing temperatures than the higher weight percent suspensions (at the same value of ns). See major comment.

Line 357-358: In relation to the major comments, you state a few data points from the 0.01wt% suspension appear as outliers (and only at warmer temperatures), whereas all three runs for this dilution are shifted to warmer temperatures for the same value of ns. I suggest restructuring this paragraph to better represent the data and discuss the inconsistencies.

Line 358-360: In relation to the above, you reference that Harrison et al. (2018) used individually weighed suspensions rather than a single stock suspension to minimise the effect of uneven particle distributions. Why was this not done here if you believe this is the issue? This seems important as you are validating the instrument yet have inconsistent results on dilution.

Line 362-363: At temperatures colder than -15 ˚C this doesn't seem to be the case (especially if you look at the 0.1-0.05wt% suspensions). There is just as good agreement with BINARY at colder temperatures (Hiranuma et al., 2015) but no comparison is made to this instrument.

Line 374-375: Were the samples analysed at this field location or in the lab where the background freezing has been characterised? Were blank (pure SA water) experiments run at the time of these experiments to check the background signal had not changed?

Technical comments

Line 41-43: This sentence needs restructuring/ re-wording as it is a bit clunky. E.g. an ice nucleating particle (singular) cannot get immersed in multiple cloud droplets.

Line 54: Should the word 'or' be in this sentence?

Line 57-59: No available technique can detect the lowest INP concentrations that are actually present in the atmosphere. I would suggest putting in a range of the INP concentrations detected with these techniques and rewording to say "to detect lower atmospheric INP concentrations".

Line 102: What material is the 96-well plate made from? Polypropylene? I suggest adding here.

Line 170-174: Suggest removing the terms 'potential' and 'possible' as adding 0 ˚C ethanol to ethanol at -30 ˚C will create a gradient, even if only small. Cooling the ethanol to 0 ˚C simply minimises this gradient.

Line 201-201: Consider rephrasing this sentence.

Line 208-209: Consider rephrasing for ease of understanding.

Line 235-236: Harrison et al. 2018 is not a suitable reference in this instance. As I understand, they make individual temperature measurements for each well and as such, they take into account the horizontal gradient in temperature across the plate without the need for such a correction.

Figure 2c: Perhaps label the peak which signifies initial nucleation

Line 295: device not devices

Line 307-309: This representation of the background you present is for DRINCZ in this particular lab environment. The baseline may change in field studies. I suggest rephrasing this section.

Line 356: Suggest changing to "samples overlap to an extent"

Line 445: missing bracket

Figure 9: the triangular symbols are hard to distinguish from one another. Suggest using different symbol shapes.

References

Beall, C. M., Stokes, M. D., Hill, T. C., DeMott, P. J., DeWald, J. T., and Prather, K. A.: Automation and heat transfer characterization of immersion mode spectroscopy for analysis of ice nucleating particles, Atmospheric Measurement Techniques, 10, 2613-2626, 10.5194/amt-10-2613-2017, 2017.

Harrison, A. D., Whale, T. F., Rutledge, R., Lamb, S., Tarn, M. D., Porter, G. C. E., Adams, M. P., McQuaid, J. B., Morris, G. J., and Murray, B. J.: An instrument for quantifying heterogeneous ice nucleation in multiwell plates using infrared emissions to detect freezing, Atmospheric Measurement Techniques, 11, 5629-5641, 10.5194/amt-11-5629-2018, 2018.

Hiranuma, N., Augustin-Bauditz, S., Bingemer, H., Budke, C., Curtius, J., Danielczok, A., Diehl, K., Dreischmeier, K., Ebert, M., Frank, F., Hoffmann, N., Kandler, K., Kiselev, A., Koop, T., Leisner, T., Möhler, O., Nillius, B., Peckhaus, A., Rose, D., Weinbruch, S., Wex, H., Boose, Y., DeMott, P. J., Hader, J. D., Hill, T. C. J., Kanji, Z. A., Kulkarni, G., Levin, E. J. T., McCluskey, C. S., Murakami, M., Murray, B. J., Niedermeier, D., Petters, M. D., O'Sullivan, D., Saito, A., Schill, G. P., Tajiri, T., Tolbert, M. A., Welti, A., Whale, T. F., Wright, T. P., and Yamashita, K.: A comprehensive laboratory study on the immersion freezing behavior of illite NX particles: a comparison of 17 ice nucleation measurement techniques, Atmos. Chem. Phys., 15, 2489-2518, 10.5194/acp-15-2489-2015, 2015.

Stopelli, E., Conen, F., Zimmermann, L., Alewell, C., and Morris, C. E.: Freezing nucleation apparatus puts new slant on study of biological ice nucleators in precipitation, Atmospheric Measurement Techniques, 7, 129-134, 10.5194/amt-7-129-2014, 2014.