

## ***Interactive comment on “Automation and Heat Transfer Characterization of Immersion Mode Spectroscopy for Analysis of Ice Nucleating Particles” by Charlotte M. Beall et al.***

**Anonymous Referee #2**

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### General Comments

The paper presents a modified immersion mode ice nucleation measurement technique. One improvement of the apparatus is automated control and analysis that enables for higher amounts of measurements leading to better statistics. But my opinion is that the more important part of the paper is the discussion of new heat transfer simulations supposed to get higher temperature accuracies in the development of such instruments. Especially this point is of great interest for the community in order to characterize and assess different instruments. However, the authors do not show the relationship between temperature measurement and sample temperature in detail. At the moment the description does not totally convince me that the new instrument is im-

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proved by the simulations. I have some specific comments and suggestions regarding the simulations and the literature comparison, and several technical comments.

### Specific Comments

P4, L14: Additional reasons for high freezing temperatures might be impurities in the water that become important at  $\mu\text{L}$  volumes.

P5, L33: ... is smaller than ... (If the intensity decreases with freezing,  $\eta$  should be a negative number (see also Fig. 4) or the opposite difference should be used:  $I(t_{i-1}) - I(t_i) > \eta$ )

P6, L1:  $I(t_i) - I(t_{i-1}) < \eta$  (The correct equation is used in Fig. 3)

P6, L5: Please mark the position or height of the thermistor in Fig. 6-8.

P8, L6: Where is something yellow in the inset of Fig. 5 (see also P9, L21)? I guess the black arrow shows the boundary conditions ("Data used in simulation"). This would mean a data range from about 800 s ( $T_{\text{well}} = 0^\circ\text{C}$ ) to 3000 s ( $T_{\text{well}} = -27^\circ\text{C}$ ). Why does the simulation cover only 1320 s of cooling?

P9, L20: Was the experimental bath temperature change with time used in the first simulation, too?

P9, L23: What does "a doubling of the warming mean"? The experimental temperature difference between the coolant bath and the headspace gas is about 10-11  $^\circ\text{C}$  after 800 s (below  $0^\circ\text{C}$ ).

P10, L11: ... +12  $^\circ\text{C}$  offset between the coolant bath and the headspace gas, ...

The paper would benefit from a graph which compares the simulation results. The interesting points for the reader would be the offset between the temperature of the water sample and the measured value at the well base, and the degree of stratification (the temperature gradient in the water sample).

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P10, L15: The experimental temperature difference between the well and the headspace gas changes, and values from 3 to 5 °C are found in the well temperature region between 0 and -27 °C. The first simulation shows that stratification is present at 5.6 °C with an offset and a temperature gradient of about 3 °C. What are limit values for stratification and a temperature bias?

P10, L32: If the temperature offset should be quantified, why is that not a part of the current paper? Please comment on the difficulties of such a measurement here or add regarding results. I understand that the temperature measurement in the sample volumes does introduce a contamination, but a calibration using pure water or solutions with similar thermal properties should make a part of the interesting temperature range available. The authors do not use the simulation to quantify the offset. Why is the simulation not a reliable tool?

P11, L14: The dilution is a useless information without the starting concentration.

P11, L17: Why are no measurements done with lower concentrations? Reliable values should be possible down to freezing temperatures of -25 °C.

P11, L27: Comment on the reason why an offset to the data is not applied. Do probably show a plausible temperature range in Fig. 9 using error bars.

P12, L1: A quite high starting concentration of 10 mg/mL ( $\sim 1$  wt%) and a dilution of 1:25,000 would correspond to a mass concentration of  $4 \cdot 10^{-4}$  mg/mL and a weight fraction of about  $4 \cdot 10^{-5}$  wt%, assuming a density of 1 g/cm<sup>3</sup> and a negligible amount of solute. Please check values and units.

P12, L12: As the CSU-IS and the new SIO-AIS are almost identical focus on the comparison of these two instruments. The CSU-IS used 60  $\mu$ L samples and weight fractions of 0.5 to  $3 \cdot 10^{-6}$  wt% in the literature study. How can it be explained that the AIS data is in the high concentration range (low surface site densities and high freezing temperatures) whereas the concentrations are in the lower end?

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P12, L12: To compare the AIS to all the other instruments in Fig. 9 additional data at lower concentrations and freezing temperatures between -20 and -25 °C are needed to ensure overlap. What is the reason for the difference to the other techniques? Can it be argued that the new one has a higher accuracy or are there other effects that have to be taken into account?

P13, L1: The experimental temperature differences are larger than 3 and 6 °C below a well temperature of -5 °C. Does this mean that stratification is present there?

P14, L1: See the comment on P12, L12.

P14, L6: The amount of samples per hour depends on the number of wells per sample. In principle the AIS can process 192 samples per cooling cycle. But to adequately characterize an IN it is necessary to investigate several concentrations and an amount of individual volumes larger than 24.

P14, L7: If the offsets are just identified but not quantified it does not lead to higher accuracies.

P16, Table 1: The Handbook of Chemistry and Physics gives thermal conductivities for air of 0.018 and 0.026 W m<sup>-1</sup> K<sup>-1</sup> at 200 and 300 K, respectively. Comment on the used values and add references.

To minimize a temperature bias between the aluminium block and the water sample the use of PCR wells made of thermally conductive plastics ( $k = 5\text{--}10$  W m<sup>-1</sup> K<sup>-1</sup>) might be interesting.

P21, Fig. 5: Why is the well temperature higher than the headspace gas temperature at the beginning? Is the system in equilibrium at the start ( $t = 0$  s)?

P25, Fig. 9: There is less data for the CSU-IS (bulk) than presented by Hiranuma et al. (2015). Add the additional points if available.

There seems to be an amount of points that does not belong to one of the given in-

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struments. Are these the missing CSU-IS values? Some points are similar but not identical. I suggest the use of different symbol types additionally to the colors and a legend for the attribution.

#### Technical Corrections

Page 1, Line 28: The AIS is compared to 6 other instruments in Fig. 9, not 5. Please correct that number whenever the comparison is discussed.

P2, L14: ... clouds. Accurately ...

P3, L18: ... the University of Colorado Raman Microscope Cold Stage (CU-RMCS) ...

P3, L24: ... frozen droplets ... grow by taking up water ... (the liquid droplets shrink)

P3, L31: ... change. Additionally, the heat transfer properties of the new SIO-AIS (Scripps Institution of Oceanography - Automated Ice Spectrometer) are characterized in the current paper through ...

P5, L9: ... temperature air away from the well ...

P5, L12: Change the figure numbers to the order of their appearance in the text: Fig. 5 → 3, and following.

P5, L17: ... National Instruments' LabVIEW software ...

P7, L10: ...  $\pm 0.3$  °C ...

P8, L6: ... is warmest in the simulation range, a maximum of 5.3 °C warmer ...

P8, L28: where  $Q_{convection}$  is the rate ...

P9, L1: ... as two vertical plates, calculating the Nusselt number  $N$ , and using  $h = Nk/H$ , where  $H$  is ...

P9, L6:  $Ra$  and  $Pr$  are the Rayleigh and ...

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P9, L8:  $Ra = \frac{g\beta(T-T_{\infty})D^3}{\nu^2} \cdot Pr$  (the viscosity has to be squared)

P9, L8: define the acceleration of gravity  $g$

P9, L13: ... diffusivity. Since  $\beta, \nu, \alpha$ , and  $k$  are ...

P11, L26: ... calculated using Eq. (1) (Vali, 1971). Although ...

P12, L1: ... The AIS measurement results in terms of cumulative INPs per volume were converted to the surface site density  $n_{s,BET}$  using the mass concentration and the specific surface area as follows: ...

P12, L7: Add the BINARY here to get the 6 instruments of the comparison.

P12, L11: ... favourably to those of ...

P12, L26: ... from having to constantly monitor ...

P15, L27: Change the doi text color to black.

P15, L27: Insert a line break between the two articles.

P19, Fig. 3: Add the Tolerance  $e$  to the Ramp option.

The text is still quite difficult to read in a printed version. Please use the font size of "Left plate" for the whole figure.

P21, Fig. 5: Clean the figure for ? symbols and use the same type of axis labels in all figures, e.g. "Temperature / °C".

Grids in both graphs similar to Fig. 9 would be beneficial here, too.

P21, L3: ...  $\pm 0.3$  °C ...

P21, L8: ... from 0 °C to -27 °C, see ...

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