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Atmospheric Measurement Techniques
Discussions

Interactive comment on "Development of the drop Freezing Ice Nuclei Counter (FINC), intercomparison of droplet freezing techniques, and use of soluble lignin as an atmospheric ice nucleation standard" *by* Anna J. Miller et al.

Anonymous Referee #1

Received and published: 5 December 2020

General comments:

This reviewer would support publication of this manuscript in AMT after minor (but seemingly necessary) revisions. Though the development/introduction of another DFT would not add an entirely new aspect in the research community, the reviewer advocates the authors for the fact that they carefully address all the details behind their new DFT and beyond. The amount of trivial details over the main manuscript and SI (clipped from one's thesis?) are somewhat bulky and cumbersome to read in general, but this reviewer still considers these as positive and beneficial information and contribution to

AMT.

Minor and technical comments:

*P3L71: The authors may consider adding Murray & Koop (2016) J. Chem. Phys. and/or Koop (2000) Nature as for homogeneous freezing reference(s). These papers nicely discusses on water volume dependent homogeneous freezing etc., which seems relevant to the concurrent study.

*P3L71-73: It would be nice if the authors can elaborate on how 'dominant' immersion freezing is in a bit more quantitative manner in the text. This fundamental information seems important since FINC is specifically developed to look into immersion only.

*P3L79: "to improve estimates of" –> "as well as improving overall understanding in"; there has been an ongoing discussion on the relationship between INP and ice crystal concentrations. At this stage, the discussion of 'whether chicken is first or egg is first' is not settled as it involves many aspects, such as aerosol dynamics, cloud macro-/micro-physics incl. secondary ice formation etc. In any case, the statement implying INP estimation to improve ice crystal estimation (regardless of intension) is seemingly misleading as no definite cause-effect answer is currently available. A simple modified statement separating INP from ice crystals (vice versa) may be the safest thing the authors may do. The reviewer suggest the authors to give some considerations at the least.

*P3L84-P485: How about water types, detectable T ranges, uncertainties etc.? The reviewer believes that there are many other variables to be considered in this statement.

*P4L98-99: The authors might want to briefly discuss advantages and disadvantages in detail – these information would be meaningful/useful to the reader.

*P4L102-103: The authors may include the investigable T range (and a summary of other limitations) of FINC in this statement. Carefully including caveats to the reader is as important as offering sales points in any technique papers, in the reviewer's opinion.

*P5L120: How ‘abundant’ in the atmosphere? The authors may provide brief but quantitative information here for the reader.

*P6L153: The reviewer strongly suggests replacing “unique. . .” with “updated feature of existing DFTs (Sect. S1).” At the end, FINC is one of DFTs and claiming the novelty of another DFT might not be a right approach to go with the concurrent paper. Simply reducing the tone (here and everywhere applicable for the similar context) should resolve the issue.

*P6L157: Briefly explain what the oven heating is for here. The reviewer is aware that it is discussed in the later section, but doing this may increase the overall readability for the future reader.

*P6L164: How crucial is it? Briefly explain/summarize what is discussed in David et al. It should be straightforward, and the readers would appreciate this complementary summary info appearing here rather than going through another paper themselves.

*Fig. 2: So each image # corresponds to incremental step of +0.2 dC? Then, for clarity to the future reader, the authors may consider introducing the temperature axis (on the top x-axis?). Also, where does this data/result come from? Which sample? The authors may want to briefly mention it in the caption or associated text.

*Sect. 2.4: The authors might want to briefly describe their general/specific suspension dilution methods (any systematic procedures; e.g., x10, x100 for all etc.?) as well as the data merging protocols – how they dealt with the overlapping n_INP (T) data – in this section somewhere.

*Sect. 3: Errors/uncertainties of DFTs are typically evaluated and expressed for both temperature and INP counts (or any ice nucleation efficiency metrics, such as n_m, n_s etc.). In the reviewer’s opinion, the DFT uncertainty cannot be governed by a single variable of temperature. Please discuss the uncertainty involved in FINC for its INP counts at the least in this section. For instance, the authors can compute

binomial confidence interval errors on their INP counts (e.g., at 95% - see Eqn 3.21 of https://publikationen.bibliothek.kit.edu/1000076327). In addition, related to this point, the reviewer finds it a bit strange that Figs. 4 and 6 dot not show any error bars. The reviewer urges the authors to show error ranges in any visual presentations of IN results. The reviewer is certain that the reader would appreciate it, too.

*P10L251-253: Adding Murray & Koop (2016) J. Chem. Phys. besides O & Wood (2016) plus extending the discussion of previous findings on measurable droplet homogeneous freezing would be meaningful.

*P10L259-260: if the water volume is $\sim$60 micro L, then it becomes more like bulk water, correct? Or the authors have a particular reason calling it still as ‘droplet’? Rephrasing it may be needed.

*P10L263-265: How do these measured T_50 values compared to theory (e.g., CNT)? Please elaborate it here.

*Sect. 4.3.2: I agree that there would not be inverse Kelvin effect for the given dimension, but the contact angles and all other relevant properties, that may matter for freezing, change depending on the volume used, correct? Would the authors elaborate this point a bit further within this section?

*Sect. 4.4.1: Please state if/how the authors applied background freezing corrections somewhere within this sub-section. Looking at a non-negligible contribution of pure water droplet freezing at above -25 dC (Figs. S3, S5), the authors may have corrected the other data presented in this paper for this background contribution in some ways (or not? – then, why?)? Please clarify. This may be something already mentioned in the manuscript and overlooked by the reviewer, but having an independent sub-section for the background corrections and all that may make a good, informative section.

*Sect. 4.4.4: The reviewer sees the point of all the bubble discussion. But, this section sounds a bit speculative. More quantitative proofs of supporting FINC and DFTs would have problem with bubbles? Perhaps, dealing with highly viscous solutions (e.g., high wt% of Snomax) tend to make micro bubbles (perhaps not so visible) and introduce high deviation and low reproducibility of the immersion IN spectra because of bubbles?

*Sect. 4.4.6: Keeping things in a laminar hood is a good practice in general. The reviewer wonders how the measurements prepared and carried out outside the hood would impact immersion compared to all the operation conducted in a hood. The quantitative answer incorporated in this section might strengthen the paper.

*Fig. 3: The reviewer wonders how this result compare to previously published work of the freezing depletion by solute; e.g., Whale et al. (2018) Chem. Sci. Please elaborate it in Sect. 4.6.

*Fig. 4: As mentioned earlier, it would be really nice to see x- and y-axis uncertainties in this sort of figures (even only on several representative data points). The same suggestion goes to Fig. 6 in P19.

*Sect. 6 onwards: The reviewer likes the idea of seeking a chemically inert & stable standard for DFTs. The tones of some words/phrases/sentences in this section seem too strong for given the context (e.g., P21L497-499 – including but not limited to). The reviewer suggests carefully re-phasing some parts to simply report what are observed/measured. The reviewer feels more comfortable accepting the proposed idea of suggesting lignin as one of potential DFT standards if the limitations are also offered, too (nothing could be perfect for now, correct?). No special procedure seems needed for the lignin suspension preparation other than suspending correct mass of lignin in pure water – is this right?

*P18L437: Justification of why n_m is appropriate to use as IN efficiency metric should be addressed here. Do the authors assume absolutely all lignin components are soluble without any insoluble precipitates? Would that be really the case? The reviewer is asking this question because, in P18L450, the authors states that this parameterization is limited specifically to 20 mg C Lˆ-1. Why so? If n_m is truly applicable and reasonably

representing IN efficiency of lignin, the n_m(T) of different lignin mass concentrations should overlap each other and collapse into a single spectrum with a minimum n_m(T) deviation, correct? This is a similar question/concern raised in Sect. 2.4 (i.e., how to merge/stitch different dilution spectra).

*Fig. 6: The reviewer is not so sure if the IN efficiency deviation involved in lignin is notably better than that of illite NX. The reviewer is aware that different batches were examined for Sigma Aldrich lignin in this study. Careful word choice seems necessary here.

*Cont'd: The reviewer is also aware that the authors' intension is to attempt to designate lignin and a potential standard for DFTs. Regardless, how do DFT-lignin data compare to online IN chamber results for lignin (Steinke et al., 2020, ACP)? The future reader will appreciate to see this discussion somewhere in this section.

*P18L454-455: This sounds like lignin exists in maritime sources. The reviewer suggests rephrasing this sentence.

*S1: The reviewer finds the compiled summary list of currently existing DFTs valuable. Are the authors or whoever is in charge of this data depository intending to continuously update contents of the list in the future (, which would be tremendous effort for the IN research community)? If so, please keep and log update dates etc. for each version. It may be a good idea if the authors or whoever is in charge of this data depository can contact each instrument PI to check if there are any updates periodically to keep all information valid, accurate, and up to date all the time? By the way, WT-CRAFT deals with 3 uL droplets, not 6 – just so you know. There may be other info to be revised/updated.

*S2: 0.2 dC instead of 2 dC?

*Fig. S2: What sample is this? The authors may want to briefly describe it here.

*Fig. S3 or S5: It would be nice to see the homogeneous nucleation frozen fraction curves from numerical modeling and their respective droplet volume at a cooling rate of FINC – e.g., Koop and Murray (2016) – in comparison to the FINC pure water data.

*P1L5: quantification –> estimation; this word choice seems better fitting for the given context.

*P1L21: in the research field of

*P3L65 & P4L112: ice nuclei –> ice-nucleating particles – Any reason why ice nuclei is used in these two locations rather than just suing INPs uniformly as done in the rest of the manuscript?

The reviewer enjoyed reading this paper. Hope some of suggestions/comments made here help the authors (and future readers).

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