RC2 review of "The Fifth International Workshop on Ice Nucleation Phase 3 (FIN-03): Field Intercomparison of Ice Nucleation Measurements," by P. J. DeMott et al. 2024

It was a pleasure to read "The Fifth International Workshop on Ice Nucleation Phase 3 (FIN-03): Field Intercomparison of Ice Nucleation Measurements," by P. J. DeMott et al. 2024. The manuscript presents data and summarizes results from the third of a series of instrument measurement comparisons that were aimed at achieving better collective community understanding when it comes to measuring ice nucleating particles (INPs). The focus of the third effort was utilizing a subset of instruments used in the first two phases, to focus on making field measurements of ambient air. While many of the same instruments were used in FIN-021, that study's focus was on laboratory-based measurements. The field-based FIN-03 measurements add complexity due to the heterogeneous nature of ambient aerosol and the low naturally occurring concentrations of INPs. The FIN campaigns are well known and brought much of the community together, but given the summarized measurements were conducted in 2015 it has been a bit of a wait to see the outcomes of the 3rd phase published. The results summarized in the submission remain well worth reporting and valuable to the community at large. However, in its current form the manuscript lacks polish and consistency, which diminish its readability. Moreover, given the large gap between when the measurements were made and their submission in this form, the submitted work seems to, in places, lack connections to work published in the intervening years. This is not a blanket statement, as some more contemporary results (references) are used, but there is a lack of consistency that again points to a need for a systematic review of the manuscript.

I suggest that the author's take into consideration the contextualized comments I include below and resubmit an updated manuscript.

We thank the reviewer for the constructive comments regarding manuscript readability and have made what we consider strong efforts to revise accordingly and update other publications referenced. This was needed because the paper had a few evolutions, ultimately slowed by the pandemic, before it came to fruition. Many of these changes are discussed below in response to itemized comments. As here, all our replies are italicized.

Itemized Scientific and Editorial Comments:

Suggestions are given by line number taken from the downloaded pre-print PDF document:
• (Abstract, 48) strike "a subset of"
• (Abstract, 50) suggest a rephrasing to: Composition of the total aerosol was characterized using ...
• (Abstract, 55) suggest a rephrasing to: Mineral dust containing particles were ...
• (Abstract, 56) should probably be diameters

All above modified as suggested.

• (Abstract, 58) Here and throughout the text (lines 93, 186, 187 etc.) the symbol ~ is used where

≈ would be a better choice. Although not always strictly followed, generally the former, ‘similar to’, means the same order of magnitude. Most often the author’s intention it seems is the later, which is ‘approximately’.

We have modified to use “≈” throughout the manuscript, where appropriate.

• (Abstract, 67) Should this say: ....order of magnitude or more, more efficient
• (Abstract, 76) strike “at most times”

All above modified as suggested.

• (79) suggest a rephrasing to: Aerosol particles that ...

We have changed this to “Particles that…”

• (140-143) A reference (and probably in other places) to Brasseur et al.[2], seems like it should be included. Much like the campaign summarized by the Lacher et al. 3 paper, the Brasseur et al.[2] Measurement Report describes intercomparison and measurements by instruments for counting INPs, including a subset of the instruments used in FIN-03.

We were neglectful in omitting Brasseur et al. (2022). This reference has been inserted in all relevant places. A rigorous comparison between online and offline instruments was not discussed in Brasseur et al. (2022), which is why we have emphasized reference to Lacher et al. (2024) in this context.

• (171) suggest to replace “aerosol” with particle.
• (228) A Thomson et al. (2000) paper is referred to but is not found in the listed references.
• (323-324) suggest a rephrasing to: ...all ice nucleation instruments utilized in FIN-03 is provided in Table 1. Detailed operating principles, locations of samplers ....

All corrected as noted.

• (362) “Frost corrections are defined through...” The wording is strange here. Suggest replacing through with, using or utilizing, or somehow rephrasing.

We write, “"Frost corrections are defined via use of time intervals sampling..."

• (§2.2) A general comment on the presentation of instruments and instrumental setups in Åò2.2, both §2.2.1 and §2.2.2. I would suggest the author’s do a careful re-reading of the instrument sections and that they re-write the sections such that equivalent information is presented for each instrument. During reading the lack of consistency became most apparent in §2.2.2, but this forced me to return to the earlier text and notice a general lack of consistency. When presenting different instruments different details are presented, and it is not evident why or what, if any, importance these differences indicate. For example, for the NC State - CS the temperatures of sample storage are included, but not in the case of the CSU-IS. My intent is not that the author’s should include all the information or all the instruments. Rather the author’s should decide on

what are the important and relevant parameters and make sure to present them uniformly. The reader should have the same information for all apparatuses. Alternatively, they must tell the reader why some things are important for some instruments but not for others. The current presentation is both a-systematic and simultaneously uninformative as to why/how the information that is presented is chosen.

We thank the reviewer for this comment. Similar comments were made by other reviewers. We reflected strongly on these and have made many changes in how information is reported and organized in each Methods subsection. We note that on rereading the sections carefully, they do proceed with the same outline of information. Further, this information needs to account for differences in online and offline methods. The online instrument sections begin with operating principles and procedures, sampling and inlets, discuss uncertainties in calculated INP concentrations and related corrections for false counting of non-INP, and finish with any special studies that will be reported. The offline sections describe the configurations for sampling, computation of INP concentrations and confidence intervals, and any special applications reported. We have attempted to assure now that all the same factors are discussed for all instruments, adding any missing information, modestly reorganizing and removing extraneous information.

• (370) "(by 3 time)" by a factor of 3?

Changed as suggested.

• (378 and again at 407) The o.d., which I take to mean 'outer diameter' is an irrelevant dimension here, as it gives no information concerning inner diameter.

We have replaced this statement with 0.19" inner diameter and have done this throughout the manuscript.

• (411-412) The "low-pass filter" sentence seemingly comes from nowhere? Why are these counts removed? I do not follow the logic at the beginning of this paragraph.

This has been changed to read more descriptively as, "A low-pass filter was applied next to remove all 1 Hz data that exceeded a total of three counts s^{-1}, as recommended by Richardson et al. (2007) to reduce obvious frost noise that equates to INP concentrations larger than about 200 L^{-1} (>2 standard deviations above mean values discussed later) for the SPIN volume sampling rate." The reference to Richardson et al. (2007) refers to their discussion (using "IN" for "INPs")- "During IN measurements under conditions supporting only heterogeneous nucleation, 1 Hz OPC counts are dominated by counts of 0 and 1. Frost events are acute and generate 1 Hz OPC counts exceeding approximately 3 (roughly equivalent to 120 IN per liter). To prevent overestimation of IN concentrations, the 1 min data set was further corrected by setting these high 1 Hz events to 0 and re-averaging the periods in which these events occurred." This filtering of data is done on all data before lower levels of frost are corrected by comparing time periods on and off the HEPA filter. Because the CSU CFDC used an aerosol concentrator to improve statistical sampling at times and generally experienced only lower frosting issues during FIN-03, the low pass filter method was not applied in processing those data.

• (447) suggest: ...has been previously...
• (462) suggest: ...except that Teflon tape replaced stopcock grease sealing the impinger...
• (491) suggest: ...2 m distant.
• (501-502) suggest: ...suspension estimated using Eq. 1. (Vali has been cited when presenting theequation.)
• (549-550) suggest: ....specific analytical procedures differ.
• (560) suggest: ...is derived using Eqs. 1 and 2.

All above corrected as suggested.

• (563-566) This is a confusing description of the EAC's working architecture and may not even be necessary given the instrument papers available. I would suggest re-writing if it is to be included. For example, what does "12 kV against a grounded..." mean? I think that it must be that the 12 gold wires have an applied 12 kV electric potential.

We have revised and shortened this section to read, "Within the EAC aerosol particles are electrostatically precipitated onto silicon wafers, which are used as sample substrates. After sampling is completed, the analysis at select pairs of temperature and relative humidity set points follows in a separate step."

• (576) suggest: At SPL samples were taken with the EAC for ...

Changed as suggested.

• (Figure 2, plus caption) The font in panels (a) and (b) is extremely small and hard to read, make uniform with (c) which is much better. (c) is entirely missing from the caption, although I take it that it is described beginning with, "Timeline...."

We hope that the revised figure and caption are greatly improved. See below.

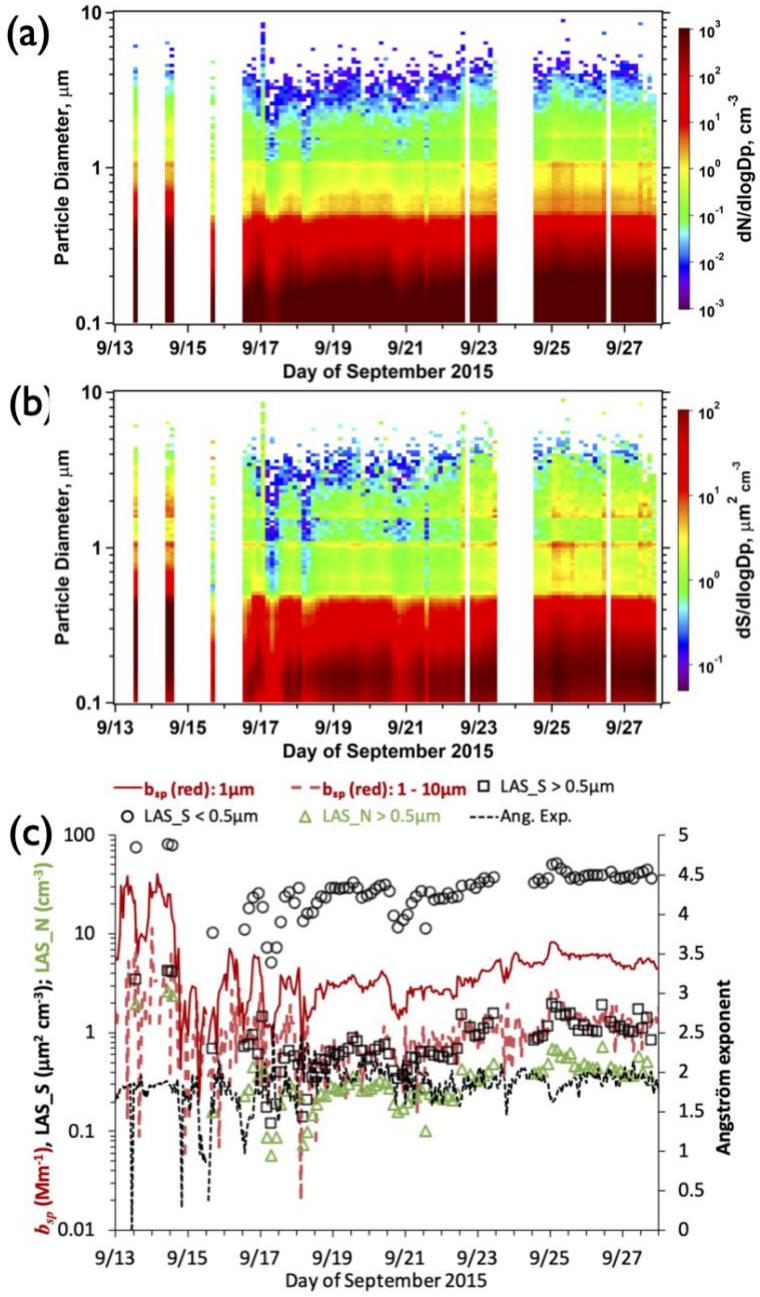


Figure 2. *Time series of dry particle number concentration distribution (ambient conditions, not STP) measured by the laser aerosol spectrometer (LAS) in a), shown as three-hour means at ambient pressure. Time series of particle surface area distribution is in b). c) Timeline of nephelometer scattering (1-hr data) in the red channel for < 1 mm and 1 - 10 mm size ranges, 3-hr LAS number concentration > 0.5 mm, 3-hr LAS surface area at sizes below and above 0.5 mm, and Angström exponent (dashed, right axis).*

• (666) extra (
• (759-760) suggest: ...during a period of warming

Above two points fixed as suggested.

• (766) I find it strange that figure S3 is referred to before figure 7 appears in the text, as S3 seems to be a distillation of Figure 7. It seems the authors should carefully consider their choices as it regards these results and figures. Moreover, both aforementioned figures strictly speaking present the 1:1 comparison incorrectly. In Figure S3 a blue FRIDGE-CS bar should show 100% agreement in the FRIDGE-CS column, and so on for all instruments compared with themselves. Currently they are missing, and the caption does not mention that the 1:1 comparison is ignored because it is 100%. The same is true of the diagonal of the figure matrix in Figure 7. In fact, S3 is a bit of a strange presentation, and perhaps some sort of matrix like presentation with a heat map (for example) would be better, but "agreement within 1 order of magnitude" is a bit awkward in general. Figure 7 is more informative but quite busy, and this might be the better figure for the supplement. Albeit I would include 1:1 or 100% agreement along the diagonal for rigor. In general, I would suggest this information could be better presented and communicated and would suggest the author's re-imagine these figures.

Fig. 7 (now Fig. 8, with reference to the response to the review of G. Vali) is referred to prior to Fig. S3 in the manuscript, but they are mentioned together since their discussion goes hand in hand.

As for the figure selection for the main manuscript and supplement, we would rather have the complete data set in the main paper (Fig. 7, now 8). We do now include the data from Fig. S3 as a table (new Table 3) in the main manuscript.

For the self-comparison of instruments, we choose to leave this out, as it appears now. This was a conscious decision to make the figure a little less busy. We do add a mention in the caption now that these comparisons are omitted.

We also find the quick "one order of magnitude" information easy to grasp, and easy to compare to other intercomparisons from previous endeavors, so we would argue to keep it. However, we may note that we have added quantitative analyses of the results in new Figure 8 as Table 2. This demonstrates average agreements from 1 to 5.5 for all matched measurement pairs of instruments over the entire study (see response to G. Vali). The question raised by this reviewer (point: page 40) and by G. Vali about whether one order of magnitude is really "good" agreement is of course open to argument. We add some discussion of this point in the Summary section in response to the review by G. Vali. We include the temperature uncertainty represented by the results as well.

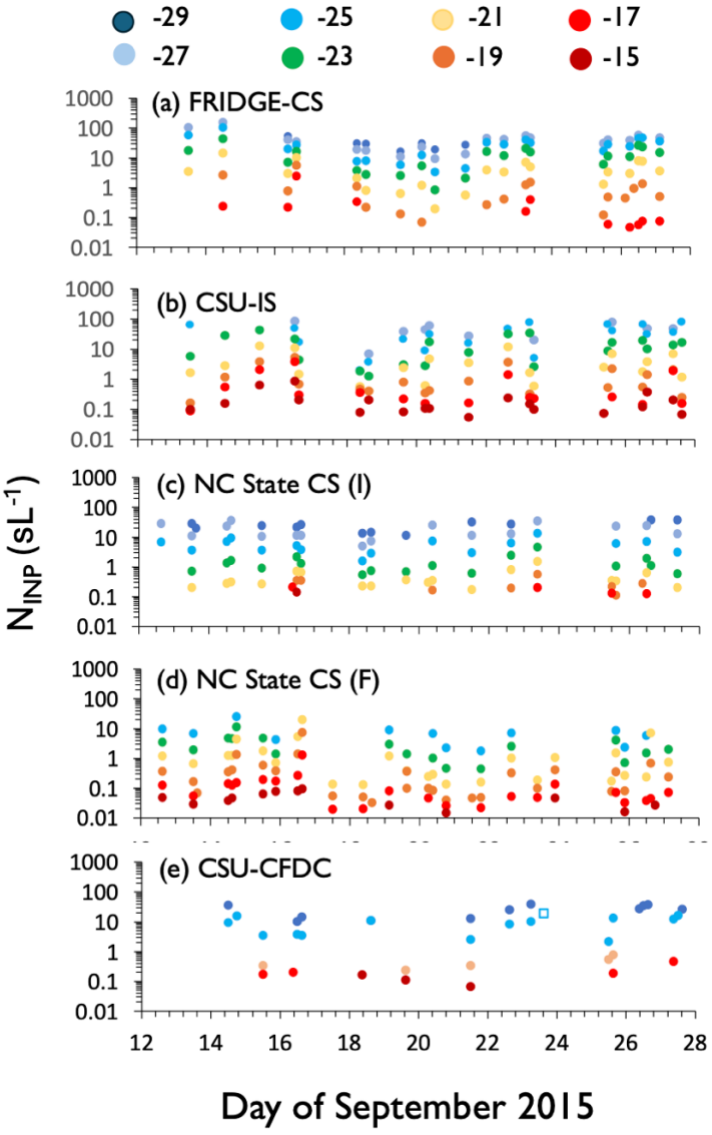
We did attempt several ways to reimagine these figures as heat maps, but these would turn out to be quite patchwork, which is the reason that a figure folding in all overlapping data (all temperatures in common) in one (Fig. S3) was used in the first place.

• (Figure 6) Are all temperatures necessary? For the main text perhaps a distilled figure focusing on a few temperatures, or results for temperature averages could be a better alternative and more effective at communicating the point. Also, some lines connect points across missing data points at the warmest temperatures, where I assume no activation was observed. I would suggest that

these points are better left unconnected, for clarity of the figure, but also because it is likely the value is actually 0 INP or below the resolution. For the CSU-CFDC panel (e) such data points are not connected, which is a much better approach than in some of the upper panels.

In the figure caption, use the acronyms as they were introduced in the text. Strike "from Goethe University Frankfurt". This kind of information appears in the text and does not need to be reproduced in a caption.

We agree that Figure 6 could be improved by limiting the amount of data shown, so have redone the figure with half as many temperature points and no lines connecting data points (these were only intended to show trends for the eye but fail for reasons mentioned and others). Because CFDC data was primarily collected at select temperatures and with only few data points made at intermediate temperatures, the primary temperatures shown for these data are -30, -25, -20, and -15 ºC) in this panel, with slight but proximal colors used to compare to the data shown only at odd temperatures for other instruments. We prefer not to take temperature averages. The new figure appears as below:



• (page 40) A general question when using the comparison that 1 order of magnitude is a good agreement. It would be worthwhile to present a simple calculation to quantify how many particles (i.e., activated droplets or volumes in immersion freezing) result in an order of magnitude change. At some temperatures and concentrations, the counting statistics may be rather poor, it would be interesting to address this given the already dilute nature of the ambient sampling.

This is not a straightforward exercise, as might be imagined, because different instruments have different number of droplets, droplet volumes, volumes of washing water, and air sample volumes, and therefore the number of freezing events resulting in a one order of magnitude difference would be different for each instrument and be different for different temperatures (number of unfrozen droplets). It is the case though that counting statistics only become an issue at the highest temperatures when limits of detection are approached. We discussed this in reply to the RC1 review in relation to understanding apparent differences between the onset temperature of freezing detectable by the NCSU impinger and filter methods. Otherwise, confidence intervals are described for each instrumental method, and these are never as large as one order of magnitude.

• (Table 2) I see the CF constant discussed in the text, but no discussion of the other constants in the Table. Are these taken from previously published literature? Or used as fitting parameters here in some minimization scheme? Please illuminate.

It was implicit that these constants listed were described/derived in the publications listed in the table.

• (989) "trends better" what is meant here? correlates?

This has been revised to, "The structure of the timeline of predicted N_{INP} resembles that of the observed N_{INP} only below -20 ºC..."

• (1200) again the Brasseur et al.[2], seems like it should be included.

Added as suggested.

• (1240-1241) Rephrase, "factor of a few increases" Something here is awkward and unclear. Deposition freezing increases with increasing RH?

We rephrase to: "The deposition INP concentration obtained by FRIDGE-DC increases from 95% RH to 99% RH on average by a factor of 3.3."

• (1246) Again awkward phrasing, "...achieved statistical significance from the CSU-CFDC data."

Revised to, "For the online instruments, only limited periods of deposition INP measurements with the CSU-CFDC achieved statistical significance."

• (References) In addition to the previously mentioned Thomson text, the Burrows, 2022 paper that is referred to is not found in the Reference list.

We thank the reviewer for these notes. All have been corrected.