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Atmospheric Measurement Techniques Discussions

Interactive comment on "The portable ice nucleation experiment PINE: a new online instrument for laboratory studies and automated long-term field observations of ice-nucleating particles" *by* Ottmar Möhler et al.

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General Comments As a scientist in this research area who both continues strong collaborations with some of the group represented on this paper and has promoted use of continuous flow diffusion chambers (CFDCs) for ice nucleation measurements over many years, I figure it is important to self-identify in this review. In this manuscript, the PINE instruments are introduced, appearing to represent a great new tool for the community, and with promise for meeting certain INP monitoring and experimental study needs. The new results for unhindered immersion freezing are very encourag-

ing, admirable in being achieved over a relatively short period of development. From the standpoint of a publication documenting a new method, there were a few things missing for me as a very interested reader. Hence, I list a number of specific comments/questions below, driven by my desire to understand the instrument clearly. In short summary, 1) there was not a full description of principles and device(s) in order to understand what challenges may be met in applying the method for the range of ice nucleation studies inferred to be possible (deposition and immersion freezing to $-60°$C (i.e., only immersion freezing is thus far discussed to the point of homogeneous freezing conditions); 2) uncertainties were given relatively limited discussion (especially at the limit of detection); 3) there was no discussion of consistency of results with physical expectations that might be revealed from, for example, microphysical modeling considerations; 4) relatedly there seemed more cursory consideration given to defining the relevant temperature associated with a measurement (I did follow the arguments, although the confirmation was mostly by comparing to AIDA), the role of growth time and sedimentation if any, clear separation of water and ice given that the latter occurs usually a few orders of magnitude lower than the water drop concentrations; and 5) finally, the introduction of field data and field instrument was rather abbreviated considering the nature/nuances of that application and considerations that will impact operation across the stated T and supersaturation range of the device in the presence of varying atmospheric conditions and full aerosol distributions. The field data only serve the purpose of demonstrating a range of data collected during automated operation for a period, as there is no other discussion of the data provided. I expect that some of the lack of clarity that I sensed will be resolved in review here. I understand, of course, that full information on any new device is often revealed over some time, often in a number of separate publications. This is clearly underway as indicated by a paper in preparation (and other intercomparison studies I am aware of), but it suggests then that some of the statements herein may require a few caveats because supporting data are not fully shown. Hence, I might even suggest consideration of a title change to include something like "An introduction..." or "A first evaluation..." or "First description and results from…" or something to that effect. That is not an adamant request, simply a suggestion. The paper is otherwise well-written and an anticipated and welcome addition to the literature.

Specific Comments

Introduction:

1) The introduction was comprehensive, perhaps overly so for a paper describing a new instrument. It was long, and not so much related to the development itself other than attempting to meet motivations.

2) Lines 87-89: It seems clear that both low and high time resolution are desirable things for different scenarios. High time resolution is arguably not useful if one is attempting to document the most special INPs, the ones that even the PINE instrument may struggle to measure in all but INP-rich environments such as shown later in this paper. I see higher time resolution measurements as highly useful, but not sufficient, unless their resolution can match higher volume collections. Some of the studies referenced to preface this statement were made with instruments capable of even higher time resolution than the PINE, but the issue I am speaking of is resolving low INP concentrations in some environments. Those other methods have been developed for automation as well, a point I will raise next.

3) Lines 97-100: I would say to be fair that these statements need modification or qualification for other studies in the recent literature. I think that the continuous flow chamber developments reflected in Bi et al. (2019) and Brunner et al. (2020) meet the criteria of operating more than "periodically" and of saving "intensive man-power and time for operation or offline analysis." Such developments are advancing rapidly across the discipline. Those instruments also appear to be capable of higher time resolution than is demonstrated for the PINE instrument in this paper. It is also implied here that time resolution of minutes will somehow solve the INP size and chemistry resolution issue, although due to statistics (sample volume and particle numbers) it

is hard to imagine this as yet being achievable for single INPs except in high loading situations. Rather, this would occur by correlation to independent high resolution composition measurements for all aerosols, which sometimes does not work for comparing directly to specialized INPs. Hence, I see high resolution INP capabilities as one piece needed in the course of a full development.

4) Line 103: The stated temperature range is what the instrument is designed for, but no exploration of capabilities to make useful measurements to as low as $-60°$C are given in this manuscript. It appears as a potential capability, only in that the temperature can be achieved in PINE-c. One can imagine that challenges in operating and interpreting data to that lower limit could be significant (e.g., control on final T and RH of expansion, low water vapor pressure and slow ice crystal growth rates), and not simply depend on the capacity of the cooling system (line 121). I suggest to stick to what is demonstrated in this paper, as far as confirmed operational capabilities and to clearly identify capabilities that remain to be defined.

Basic principles and milestones of the PINE development:

1) Line 121 repeats the assertion that likely requires "potential" as a caveat. No low temperature data are shown excepting the homogeneous freezing onset for grown droplets.

PINE instrument setup:

1) Line 169 or thereafter: Have particle losses been characterized through the nafion dryer system? For that matter, I realize in reviewing these comments that particle transfer versus size into the PINE systems has not been discussed.

2) Lines 172-173: Perhaps this is irrelevant since an aircraft system is not yet described, but I wondered about the use of the nafion system on aircraft where the pressure drop will be limited at higher altitudes. Will the system work over the needed ranges in this scenario?

3) Lines 177-180: I am curious about the later tests shown for background, simply because I did not understand the implications of no background particles found after five runs. Why does it take five runs to decrease, and does it mean that any background is then absent from thence forward in time? Have you explored this systematically, and/or after hours of operation? My personal understanding from an overlapping study in time with the one at the SGP site, is that the dewpoint was $-10°$C in that case, and that background counts at some level were always detected, if minimal. Hence, the basic question is if it is understood what dewpoint is sufficient for frost-free operation at any given T?

4) Lines 183-184: It was not clear to me what actually constitutes the cooling system? Is it a plenum around the chamber and this is fed by the large chiller reservoir?

5) Line 191: I expected that the minimum air temperature achieved would be colder than the minimum cooling temperature? Why are they the same?

6) Line 195: Can you explain the Stirling cooler method of cooling the wall of PINE-c for those of us unfamiliar with the exact cooling mechanism? E.g., fluid versus expansion cooling or whatever it is. The details on cooling systems in general does not match the later attention to detail of the OPCs.

7) Line 198-200: Again, the cooling is understood, but the utility for performing low temperature ice nucleation experiments, especially where this will presumably involve more special control over the expansion conditions to meet some final peak relative humidity, is not yet discussed or demonstrated herein.

8) Line 222: Does this more limited volume used to define ODV explain the higher value of lowest detection limit concentration listed in Table? Perhaps worth noting here, since It only comes up again at the end of section 4.

PINE operating principle:

1) Line 250: To this point, the definition of ice crystals versus drops has not been made.

Perhaps add a short note about this, "…as discussed later in this section"? Otherwise, this raised a number of questions immediately.

2) Lines 269-270: Figure 3 is an important figure, and it raises a number of questions that were mostly answered in time over this section. However, I will list a number of them here. Immediately I wondered why the lowest temperature measured was used. As an aside, this point (lowest T used) should also be stated in the figure caption, for clarity. What differences are seen in these temperatures, and what uncertainty does this create? Are concentrations referenced to the entire integrated time interval and volume of expansion (and will this be the case also for the PINE-c), and do they represent the lowest temperature achieved (e.g., there is a $4°$C cooling shown in the figure over the time of the expansion)? Hence, is it one measurement or many, and how are the sub-intervals defined? A range of apparent ice crystal sizes are shown in Fig. 3, up to 100 microns. Are these ice sizes consistent with expectations of grown sizes for the conditions and growth times? The PINE chamber is quite small compared to the AIDA chamber where volumetric concentrations are assessed in situ. Is there sedimentation that could impact inferred concentrations and their reference temperature for the smaller geometry of the PINE? Have any such calculations been made at this time, or are they planned?

3) Lines 273-275: Regarding the starting vapor saturation ratio for expansion, you assumed this or you set that partial pressure based on a room temperature RH measurement? Why would it be ice saturated if there is no ice on the walls? Or is it close enough as determined on some other basis? This would seem important for future use toward other measurements than immersion freezing.

4) Line 288: Here an important distinction may arise, but perhaps the authors can correct any misconception I have. While described as purely immersion freezing, the temperature is already cold at the point of expansion, and so does the measurement also not integrate some proportion of INPs from any/all INP mechanisms, other than contact freezing, that ensue as the air rapidly cools and ultimately exits the chamber

through the OPC? That is, somewhat similar to CFDCs when they are operated for bringing air to a final RH that is well above water saturation?

5) Line 317-318: Concerning addressing the size threshold setting for ice crystals, I struggled a bit to reconcile Figures A5 and 5. In A5, the scale is frequency, and it spans about three orders of magnitude out to 10 microns. Is there an issue in the fact that if cloud droplet concentrations range up to 1000 per cubic centimeter, and activated INP concentrations could range down to 1 per liter, then assessment of cloud drop frequency would have to be made over a greatly extended period of time to capture the tail of the distribution? Or is it simply the case that repeated experiments like the one in Fig. A5 never indicated a drop even in the size range greater than 10 microns? It might help to add the time and/or volume of assessment represented in Fig. A5. Clearly, Fig. 5 shows particle numbers appearing in these larger size ranges at 4-5 orders of magnitude below cloud droplet concentrations at least. This is an issue that perhaps deserved more attention in the paper, but if I understand, sensitivities of the ice cut size threshold will be more extensively covered in Adams et al. It would be good to add a reference for that paper, if it is now in submission.

6) Lines 348-349 and lines 359-360: Note that the first statement repeats from earlier in the manuscript. One example is provided in Fig. A3. Perhaps repeating myself also, is this the very start of operations, or a period during the midst of operations? Why does it take 5 cycles at all, and does the background then stay that low in all cases? What does this depend on? The question arises again in the later sentence where long time operating detection limits are listed. Do not these very low detection limits listed for long operation imply the need for validating backgrounds being below such levels over such long times?

Laboratory tests of the prototype version PINE-1A:

1) Lines 374-375: In Fig. 6, there looks to be up to 1C temperature uncertainty in defining the lowest temperature attributed to ice nucleation. Since T is not spatially

uniform in the chamber, do you anticipate a bias in sampling only part of the flow as in PINE-1A versus all of the flow in PINE-c? Also, Figure 6 and its caption could use a little attention to description. At present the data are described as “all single ice crystals measured.” Should it say something like “Data points indicate all single ice crystal event temperatures...”?

2) Line 391: Just a note that there seems an inconsistency between the statement of a minimum pressure reduction every 5th cycle versus what is shown in Fig. 8 (and stated in that caption). It looks like 4 cycles. It is 5 cycles in Fig. 10.

3) Lines 401 to end of section: The basic agreement shown between AIDA and PINE in Figures 7, 9 and 11 (over a more limited range) is excellent. I again wonder here about the percentage uncertainties being constant over the entire dynamic range of ice concentrations. For example, at the LOD, the true uncertainties must be larger, no? That statistical uncertainty does not appear to be captured in defining uncertainties based on the OPC ODV alone. I guess I expected based on statistical count considerations that the uncertainties should be larger for lower INP concentrations. Additionally, given that ice concentrations are integrated over the range of temperatures present throughout the volume, and if some of the crystals grow in that time to 50-100 microns (would be good to state the typical mode size), does sedimentation assuredly not impact/skew the results attributed to one temperature? There could be differences as to how this is measured temporally in situ in AIDA versus drawing the entire tank flow from the PINE, and there is some room for not discerning that in the comparisons shown. Nevertheless, a minor point overall.

Field measurements with PINE-c:

1) Lines 416-417: With an expansion mode time of 60-90s, a question arises as to the applicability of the discussion of temperature attribution and method for calculating INP concentrations with the PINE-c versus PINE-1A. Were they exactly the same (lowest T used, etc) for these presented analyses?

2) Fig. 12: This is a nice compilation of results, if leaving a lot of room for discussion of their meaning still (i.e., variability of 2 orders of magnitude temporally at any T,) and raising all of the questions listed in the last sentence of this section. It is a minor concern for showing them in this manner, simply as a demonstration that the data were collected more or less autonomously over this period (maintenance or other attention needed were not discussed). Let me ask one thing though. The flattening of the INP concentrations toward the higher temperature limit of detection is interesting, but raises a question regarding the confidence in these results. The uncertainties are based on relative standard errors. The percentage errors are quite small and I wonder how these can be the same at the LOD as they are at any other conditions. This is the same question raised for PINE-1A.

3) Lines 423-424: I am not sure what is meant by "warranted" here. Possible? Also, can the point regarding the dewpoint temperature be clarified? Dewpoint is not controlled somehow? It would be much higher in summer and much lower in winter. How might this affect the operational range, background etc, or does this remain to be investigated?

4) Lines 425-427: What exactly is meant by deconvolution of nucleation modes? Meaning different operation of the PINE than discussed in this paper, which is immersion freezing? Or meaning resolving what I mentioned earlier in this review, the temporal evaluation of data during single expansions? This is a point that should be clarified, as it is important to state which potential aspects of PINE measurement capabilities are demonstrated in this first publication and which remain.

Other editorial comments:

Line 96: typo – based

Line 266: Suggest "one of" after "An example of. . ."

Line 282: "so" not needed before "calculated"

Line 287: Suggest "than" for "as"

Figure 3 caption: Suggest to add "Calculated" at start of sentence starting "Liquid water. . ."

References:

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