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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Size-resolved measurements of ice nucleating particles at six locations in North America and one in Europe" *by* R. H. Mason et al.

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The problem addressed in this paper - the sizes of atmospheric ice nucleating particles - is important and is far from having been adequatly resolved in previous studies.

The paper does leave the impression of a somewhat brief, preliminary study for exploring the potential of the relatively recent MOUDI-DFT technique. That is a legitimate goal and it has been successful. But there are some shortcomings that deserve to be mentioned. No ancillary data are presented, such as aerosol size distributions.

The aerosol were not brought to a standardized humidity so the sizing would be more consistently comparable. Possible aging of the samples was not cotrolled for. Other questions and suggestions are listed in the following sections. Some of the points are perhps marginal, as this paper focuses on the sizes of the INPs not on their concentration; nonetheless, the authors should discuss these problems and reflect on possible implications for the results. Some caveats would be in order and some changes should be included when the paper is revised

1 Technical issues

Since the MOUDI-DFT method employed in this paper has been described in at least two previous publications, and is used here with no changes that would require special attention, the method is taken as established and few details are given. Nonetheless, there are some questions that can be esked regarding some aspects of the method.

1.1 Background

Unless I missed it, it seems that the background level, or detection limit, has not been established. There is mention of sudden freezing at -25C (pg. 20531 line 14) and this may be an indication of the detection limit. But the background levels at other temperatures are still open to question. Tests with the intake air passed through an efficient fiter upstream of the sampler would have yielded blanks for establishing the background. This may have made the condensation of drops difficult but that could have been circumvented by some steps like higher supersaturation, producing drops meachanically, etc. None of this is simple, of course.

1.2 Sample volume

The volume of the air sample that is evaluated for INP content by Eq. 1 is taken as the ratio of the DFT view area to the area of the glass slip while the total volume is for the entire MOUDI sample. Isn't the ratio of glass slip to MOUDI disk missing here?

1.3 INP content of drops

The assumption of equal drop sizes from Vali (1971) has been maintained in Eq. 1 in spite of the large range of droplet sizes (a factor 20 or more in volume) that form via condensation in these experiments. This can't be simply ignored.

The issue raised in the previous paragraph is linked to the question what is the relevant measure of INP content per drop in these experiments. The Vali (1971) proceedure is formulated for the case when all drops are derived from the same bulk sample of water with a random partitioning of INPs in the drops. In that case, all drops can be assumed to have the same probability of containing any type of INP. With the condensation method employed in these experiments the validity of this assumption needs to be examined carefully, considering what determines where the drops form on the plates and what particles they contain. If condensation is initiated on the INPs themselves, the question reduces to whether there is any dependence of the supersaturation required for condensation and the INP activity of that particle. This in turn depends on what kind of internal mixtures consitute the INPs. If the INPs become included into the drops as those drops grow from a condensation nucleus that is not an INP, than the assumption of random probability of INP content per drop seems to be fulfilled. The main point is that the authors should justify the use of the Vali (1971) formula for these experiments. It is not obvious how the original assumptions apply here.

Additional possible problems relate to the adhesion of collected particles on the supporting surface. Perhaps the authors have looked into whether the drops move par-

ticles along the surface as they expand during condensation, or cover them up. The same question needs to be asked for the period of evaporation: are particles left behind as the drops shrink? There is a hint that perhaps this is the case in Fig 2(c) of Mason et al. (2015a) with particles(?) seen in circular patterns.

1.4 Cooling rate

The reported cooling rate in the freezing experiments is -10C min^{-1}. Thermal lags throughout the system, specially within the drops but also within the stage, could be a problem with such rapid cooling. Has that been considered? How uniform was the rate of cooling throughout an experiment?

1.5 Correction factors

Two corrections factors are included in Eq. (1). The first one, f_{nu} is explained in some detail in the referenced paper (Mason et al. 2015a). The second one, f_{ne} is less clear as the referenced paper (Koop et al. 1997) doesn't show this quantity directly and here it is applied to a formula (Vali 1971) that is differently derived. This requires some explanation. Numerical values for the correction factors should be given.

2 Results

2.1. Were the overall average and median concentrations reported calculated with weighting by the number of samples for each location? In light of the sample numbers varying from 1 to 34, giving equal weight to each results in some distortion of the data.

2.2. On page 20534 long-range transport is stated to preferentially reduce the numbers of large particles. That is not an obvious result for INPs, specially since the mixture of

components that make up the INPs is unknown.

2.3. There are many facets of the results obtained in this work and the authors have selected meaningful diagrams to present those results. Yet, it would have been useful to see the results also in the form of temperature versus concentration. That would also bee a good way to compare present results with other published works.

2.4. Results from the Colby site lie at the upper ends of the data both in concentration and in size. It is stated that sampling was during and following combine operations. The range of variability in concentrations and in sizes for these 3 samples is no different from other sites. Is this relative uniformity related to having combine operations in the area, if not directly near the site, throughout the sampling periods? On page 20258 it is indicated that sampling was 3-10 m from the fields. Isn't that an error and kilometers were meant?

2.5. There is interesting aspect of the data that the authors might want to explore. The question arises: are the INPs of different sizes of the same composition and have the same surface properties? If so, the probability of a nucleating site being found on any size particle would be the same. A rough estimate of the relative proportions indicated in Fig. 3 of the paper for two sizes indicates a negative answer to the question. Using a ratio of about 1.3 for INPs greater than 1μm to 2.5μm and using those as nominal sizes whose surface areas would differ by a factor of 6.25, the indication is that the larger particles have a much lower probability per unit surface area to contain an ice nucleus. That would be something to ponder. This calculation could be made more precise and extended to the full range of sizes covered by the sampling. There are broad implications of this type of analyses so presentation of the results in this form would make a valuable addition to the paper.

2.6. It is pointed out in the paper that the size range of INPs examined is restricted to $> 0.1\mu$m aerodynamic size and that further studies are needed to examine the contributions of smaller size particles to INP populations. An additional aspect of INP sizing

would be worth mentioning. The size distributions of atmospheric particles undergo many changes due to capturing, scavenging and other processes. Few particles are of single composition. The measurements here reported refer to sizes of internally mixed particles and also of aggregates. Thus, small INPs may be detected at larger sizes if they adhere to other particles. This may take place already at the origin (soil especially) but also during atmospheric transport. A difficult issue, no doubt, but one that need to be recognized.

3 Supplement

It is good to have the authors' analyses of the works cited for comparison of size determinations. It helps to illustrate the sparsity of data on this important point.

All but one of the works cited employed aerosol samplers. The exception is the Vali (1966) report which is based on size discrimination by filtering of liquid samples. It would be helpful to readers to point out this difference and its possible implications, namely that soluble components of the INP get removed and particle might break up when immersed in water. Both of these factors introduce a bias in the comparison of sizes, with corresponding aerosol sizes likely to have been considerably larger than the size found in water. Coagulation of particles in the liquid would add a bias in the opposite directions but is less likely to have been a factor.