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Atmospheric Measurement Techniques Discussions

Interactive comment on "Re-evaluating the Frankfurt isothermal static diffusion chamber for ice nucleation" *by* J. Schrod et al.

Anonymous Referee #1

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This is the review of "Re-evaluating the Frankfurt isothermal static diffusion chamber for ice nucleation" by Schrod et al. This manuscript deals with improvements of Frankfurt isostatic diffusion chamber (FRIDGE) which is an ice nucleating particle collection and online detection system for ambient measurements. The improvements are concerned with the collection and analysis techniques including a discussion of the uncertainties inherent in the measurement method and error analysis. This is meant as guidelines of all current and future measurements using FRIDGE instrumentation.

This manuscript fits well within the scope of AMT. I applaud the authors to re-evaluate the measurement performance of FRIDGE and to admit that published results between 2008 and 2012 include problematic measurements. However, before this manuscript can be recommended for publication, I have some major criticism that should be ad-

dressed concerned with the fundamental caveats of this measurement technique, collection efficiency, issues of sample storage, and representativeness of particles employed in this study. I feel these sections need further elaboration.

The FRIDGE operation is based on particles deposited on a substrate that are exposed to supersaturation for initiation of ice formation. A caveat of this technique is that once one ice crystal forms, the water vapor is likely non-uniform across the substrate area. A water vapor gradient will form instantaneously as can be inferred from any standard textbooks, leading to an inhomogeneous water vapor field. In other words, subsequent ice crystals formation likely occurred under different thermodynamic conditions. Besides mass transfer effects, is there sufficient water vapor available, once the chamber is filled, to activate the particles as cloud droplets or ice crystals? This depends on how many particles are deposited and thus is difficult to assess prior when in the field. Figure 2 shows that NaCl droplets are not growing further with time. As these droplets become larger they should grow slower, however, a constant value is either an indication of too many particles competing for the available water vapor, too little water vapor present, or diffusion limitations to supply water vapor.

A collection efficiency of 60% has been determined based on measurements of hematite and fluorescein-natrium particles. These particles are likely crystalline or solid. Are those particles representative of what one would sample in the field? Looking at the AMS community and single particle analyses by various groups, it seems there is often organic material associated with the particles. In other words, the particles are likely much more complex and if more liquid-like will likely change the collection efficiency. Similarly crucial, it is not clear if the collection results in a size discrimination of the particles? Assuming ice nucleation scales with surface (at least this is how the community parameterizes ice nucleation), this could artificially skew ice formation in terms of particle size and composition.

The sample storage discussion does not reflect issues that come up with ambient particles. First it is not clear how the samples are stored within FRIDGE – under ambient

air, argon, nitrogen, cold, dark, etc.? Second, some of the particle systems chosen give the impression that ambient aerosol is inert material (minerals) and then, obviously, storage will not have much effect. Looking at communities who analyze ambient particles for composition and morphology, it is clear that storage is a non-trivial issue. For example, coatings of organic acids can change composition and morphology of inorganic particles (see e.g. Laskin and Gilles groups). Depending how stored, particles can become liquid, semi-solid, or glassy, undergo phase separation, etc. This presents a challenge for the community in general but is even more crucial when performing ice nucleation on these particle surfaces. The applied mineral dust aerosol will not change much with time. Snowmax will also not change in time (if, it would not be a commercial product stored in tons at ski resorts). However, the soil and desert dust, likely containing organic material, show a trend to deviate from the 1:1 line in Fig. 10. The storage effect will depend crucially on the complexity of the sampled particles which are not always known prior to sampling.

Other comments:

p. 12529, l. 11: Can it be guaranteed that all particles are negatively charged? Do multiple electron charges per particle affect the electrostatic precipitation efficiency (resulting in size discrimination)?

p. 12529, l. 20 and following: In addition to my general comments, please give more details on storage, how long to sample, are particles uniformly deposited or do they coagulate and form large particles (= potentially better ice nuclei?), etc.

p. 12530, l. 18: The FRIDGE sample is first evacuated. This means volatile material is lost?

p. 12530, l. 20 and following: A value of 30 adjacent pixels indicates a surface area of 30 times 400 μm2= 12000 μm2. Depending on particle coverage, many other particles may be covered by this growing ice crystal? How can this be accounted for? Is this an issue?

p. 12531, l. 9 and following: If droplet formation was a cause of misinterpretation, why not conducting careful deliquescence experiments for different particle types at various temperatures?

p. 12531, l. 26 and following: It is not clear how many particles activated and how this mean diameter is derived. I assume all NaCl particles took up water but only a few AgI particles nucleated ice and grew large? This mean diameter is then derived for each time step? Are continuously growing ice crystals included in the derivation of this value? Does particle loading have an effect?

p. 12532, l. 25: What do you mean with “metamorphosed”? A phase transition? Also, I do not understand Fig. 5: Is this a sample that contains silver iodide and NaCl particles? At which temperature and RH was this experiment performed? It is very surprising to see ice and the presence of droplets located so closely. Shouldn’t the Bergeron-Wegener-Findeisen process lead to an instantaneous mass transfer of the droplet water to the ice crystal.

p. 12536, section 5: I suggest making a table listing the conditions (T, RH) of the wafers tested including particle loading, particle types, how often repeated, etc. As is, it is difficult to understand. E.g. Fig. 9 shows 3 wafers, so I assume 1 sample was repeated $\sim$10 times but then it says “The weighted mean relative error determined from repeated measurements of 20 wafers (3–10 repetitions per wafer) is 18.3 %.” Have 20 different particle samples (which particle types?) been employed and each has been repeated 3-10 times, meaning up to 200 experiments have been conducted? This is not clear. Also, I am not convinced that such a relationship can be derived that easily, since this depends on IN type, particle loading, T, and supersaturation. (Besides the fact that I am not convinced yet that multiple ice nucleation events can be counted as outlined above.)

p. 12536, l. 17: This sentence is not understandable. Also its conclusions (following sentence) are not clear. Why should it be representative? The statistical math and

experiments are missing for these statements. I suggest rewriting this paragraph.

p. 12537, l. 1 and following: This is also difficult to understand: What are FRIDGE sampling units? How are flow regulators involved in this. You mean uncertainty in flows? What is meant by timing?

p. 12538, section 5.1.: Do not use expression “straddled”, “myriad”, “outstrips”. See also general comment above. I would assume any deviation from the 1:1 line is a sign of uncertainty. When applying 18% uncertainty to the data and then the data do not fall on the 1:1 line, this indicates something happened to the sample?

p. 12539, section 6: For now, I doubt that the DeMott et al. parameterization can be compared to FRIDGE measurements. Please give reference for correction factors discussed on lines 23-26.

p. 12540, l. 9-23: This paragraph is highly speculative and could be omitted.

Technical comments:

p. 12526, l. 25: Please rephrase “jibe”. Do you mean “match”?

p. 12529, l. 11: “flown”

p. 12535, l. 26: Change “is both edifying and remarkable.” To “is remarkable.”

p. 12537, l. 14: Change “modulo”.

Interactive comment on Atmos. Meas. Tech. Discuss., 8, 12525, 2015.