Referee comments on “A new method for operating a continuous flow diffusion chamber to investigate immersion freezing: assessment and performance study” by G. Kulkarni et al. 2020

In the submitted manuscript Kulkarni et al. describe a new method for operating a Continuous Flow Diffusion Chamber (CFDC) and show both system modeling results and results from testing using various experimental test aerosol and some ambient air sampling. I find the manuscript generally well written and presented. The idea for the new CFDC operation principle is original and enticing. This idea potentially expands the operational range of CFDC instruments and could be a significant contribution to the community. However in its current form the submitted work lacks clarity in some key areas. Some additional work also needs to be done with respect to the figures, where either interpretation is difficult and/or mistakes appear to have been made with labeling in the main text etc.

Nucleation Temperature and Crystal Growth: I think the primary question that the authors must clarify is related to quantifying the ice nucleation temperature and ice crystal growth within the evaporation (now nucleation) section of the CFDC. The authors have done a nice job of trying to model the droplet growth in the ‘conditioning’ section, but have not shown analogous results for modeling the crystal changes in the evaporation section (I recognize they posit that given the saturation condition is RH$_{ice}$ = 100% there are no changes – but consider comment below). My interpretation of Figure 4 and many of the Supplemental figures is that if the system behaves as modeled then liquid droplets quickly evaporate within the nucleation section – on approximately the same time scale as the temperature and RH fields equilibrate. This suggests that the nucleation occurs in this transition region and that the fixed nucleation section temperature in fact controls the gradient between the two sections but does not necessarily represent the actual nucleation condition. What size water droplet must nucleate into ice in order to grow to reach the quoted 3 μm OPC cutoff for ice? If there is a lower bound on this value then one might interpret before what point along the droplet evaporation curves ice must form. Likewise it would be interesting to understand the range of potential ice crystal sizes depending on at what point entering the chamber a droplet nucleates. Clearly at the warmest temperatures the gradients between the two chambers are weaker and thus the constraints on thermodynamic forcing will be better, but at the colder temperatures I remain to convinced that the nucleation occurs at the equilibrated chamber conditions.

Further evidence already included: on page 6 the authors state that, “ice particle size measured by the OPC can be representative of the size of the droplet while freezing.” However, all simulations of droplet growth suggest maximum droplet sizes between 2 and 2.5 μm. Figure 5 shows peak OPC concentrations from about 3.57 to 5.02 (diameter) which more-or-less corresponds to the peak predicted particle sizes, and those droplet diameters only occur immediately in this transition region and not within the equilibrated portion of the chamber. However, also to consider is that, although the equilibrated chamber represents RH$_{ice}$ = 100%, as long as droplets do exist ice particles can grow due to scavenging...to what extent? Perhaps this is minimal? Will the droplet evaporation go back to the walls?

Below I present an itemized list of additional thoughts and comments as I came to them in the text, which I hope helps to further contextualize my thoughts.

Itemized Scientific and Editorial Suggestions:

Specific Suggestions by Page and Line Number (page, line):

- (1,26) enough to say sampled from ‘am ambient aerosol inlet’. The location etc. is described later.
- (2,42) replace toward with for
- (2,50) percent
- (2,50) CFDCs also
- (2, 61) particles are activated not ‘all aerosol’. Remember the strict definition of aerosol is the gas, particle mixture thus activation of all aerosol seems strange.
- (3, 68) the Compact Ice...

- (3, 70) thermally isolated or insulated? How much thermal contact do the 2 sections actually have?

- (3,78) Here begins the use of many symbols $\sim$, $=\sim$, etc. which continues throughout the manuscript in an ill-defined manner. I presume most often these are being used to indicate approximately, for which I suggest $\approx$. Although definitions are a bit muddled the use of similar to $\sim$ to many, including me, denotes an order of magnitude (-ish) approximation. I am sure the authors intention is to convey a more approximate value than that in many of their uses here and throughout.

Here also the RHw is indicated as 106%. Later in the numerical modeling section 2.2 a RHw of 113% is chosen and this value also seems to be chosen in the experimental descriptions that follow. I am left confused, why these differences?

- (3, 80) An OPC

- (3,93) Please also include here the saturation condition that results from the choice of temperatures – it would be nice to also have the value in terms of ice saturation.

- (4, 103) 'The choice of steady-state cooling....' I think the manuscript would benefit from a longer discussion related to the cooling rate. The empirical choice of cooling rate as being satisfactory is supported by the filled symbols on Figure 6, which I understand were measurements made with the chamber at static conditions. However, did the authors try any other cooling rates? Do they have any evidence of what a maximum cooling rate might be? I think any additional information that might have been gathered with regard to the operational limits would add value to what the authors have done.

- (4,110-113) More clarity is needed with respect to the pulse experiments. The pulse duration is quoted as 10.5 s. In Fig. 1 of the supplement the dashed line is used to indicate the limit after which particles are considered to be outside of the lamina. However, if the pulse duration is 10.5 s and the residence time is ≈ 10s shouldn't pulsed particles continue to arrive until 20.5 s? This would presumably significantly alter the 16% number in the text. How is my understanding deficient?

- (4,113-116) Final sentence of this section seems to be better suited to introduce the following section.

- (5,133-134) such a geometry; I am confused by the end to this sentence. "...it was coupled with energy and viscous heating to enable the species..." I think this needs to be reworded. What was coupled exactly? Is energy conservation meant? Please clarify this sentence.

- (5,141) I found the relevant information is S.1 not S1, but this appears very far into the supplement. It would be useful to order the supplement in an order that corresponds to how it is referenced in the text.

More notes with regard to S.1: What is meant with e_∞ and e_r? The use of 'environment' is confusing. I think e_∞ represents the far field vapor pressure, while e_r represents the equilibrium vapor pressure at the surface. Similarly the temperature terms should be precisely defined. Furthermore, the D_v term introduces another temperature T and pressure p that seem to have the same definitions as T_∞ and e_∞. Please use uniform notation and be clear.

Finally r_0 is the initial radius of the droplet, but by my reading, for the purposes of this manuscript r_0 has been set to equal the dry aerosol particle diameter. However, we know that at deliquescence (DRH) any soluble aerosol particle will have a sharp transition terms of growth factor (GF). For example at DRH the GF for NaCl jumps suddenly from 1 to ≈ 1.6[1]. How is this discontinuity accounted for? Even for mineral surfaces one would expect the r_0 to be potentially, importantly different when it is completely coated in bulk water versus when it is dry or just has adsorbed water present.

- (5,143) Figure 2 is referred to but I believe the intent is Figure 4a perhaps?

- (5, 154) Figure S2-5: I found myself spending a lot of time digesting these figures and wonder if the authors should revisit what in fact is best to include in the main text. Perhaps they might hybridize some current figures to add some detail to the main text that only appears now in the supplement. I would also suggest that in Figures S2-S4 the authors choose different color maps for time and RH. With 2 color maps and an offset perhaps panels b and c could potentially be combined. Even if not flipping between figures would be easier if the color maps differed. Figure S5 is missing a legend. Also in this figure the red droplet radius points seem problematic. Firstly, they seem to show a discontinuity at the chamber transition that none of the other curves indicate. Second, one would intuitively expect their values to perhaps lie between the black and pink, but also the red temperature seems to be lower than the black as it gets close to the transition. Why does the particle further from the cold wall have a colder temperature than that which is closer? I find that a clear explanation of this figure, and especially the reason the red points stand out is lacking.

- (6, 167) Table S1: replace very small with $\leq X$.

- (6, 169) evaporating droplet

- (6, 175) 200 nm? Why not use 300 nm to match the simulations? Perhaps a comment on this choice would be useful.

- (6,177) space betwen RHw and conditions

- (6,180s) See my comment above with regard to the OPC spectra and interpretation based on water droplet size predictions. Also, as a reader it became confusing that the authors switched from discussing droplet radius to droplet diameter when they begin discussing OPC data. I suggest that one dimension is chosen and all discussions and figures converted to this for consistency.

- (6, 183) How are ice particles of 2.0 μm observed, when previously it was stated a cutoff of 3 μm is used to select ice? Is this a result of my radius versus diameter confusion?

- (7, 200) “It can be seen that RHw values close to 113% are required before all the AS particles are activated to droplets.” I believe the observation is of ice, not of droplet activation. The DRH of ammonium sulfate is $\approx 82\%$ and weakly dependent on temperature. Thus all ammonium sulfate particles should activate at much lower RH. The value of RH here is what is needed for them to grow to a size, subsequently freeze, and remain big enough to be measured as ice.

- (7, 207) Here again another size 400 nm mobility diameter particle is used, perhaps a word as to why this choice was made, relative to the 200nm or 300 nm used in other contexts in the text?

- (7, 211) 7% and 3%

- (7, 222) was once .....is now

- (7, 225) I suggest the authors stick with SI units – mph to m/s.

- (8, 232) particles were also collected.... Were the same particles collected on the SEM films after the CFDC or was this sampling run in parallel?

- (8, 252) froze at the highest

- (9, 263) See previous comment related to temperature ramping.

- (9, 265) allows a comparison with other....

- (9, 271) But citations in order from earliest to latest.

- (9, 278) Here error in n_s is mentioned but does not lead to any uncertainty plotted in Figure 7. In addition to the error bars plotted from other studies it would be nice to have error bars plotted for this study.

- (11, 339) Perhaps the authors could spend some more time attempting to explain why their results seem to be systematically high relative to the other studies (Figure 7). Are there good physical explanations for this?

- (conclusion) From the conclusions I am missing a discussion of whether other existing CFDCs could employ this technique. What for example are the physical constraints in terms of evaporation section length? Given the published geometries of instruments like ZINC[2], SPIN[3] etc. could these instruments hope to run using the operational mode introduced here? Alternatively, if new chambers were being designed what features should be introduced or geometry utilized to enable operation in both traditional and this new mode? Recommendations to the community would strengthen the paper.

- (Figure 1) Can basic chamber dimensions be included, space appears plentiful.

- (Figure 2) Why include temperature from when initial cooling began? Why not just the shaded region, or shaded plus rewarming?

- (Figure 5a) See previous comment with regard to radius versus diameter. Also, why the arbitrary scale? Is this a result of OPC binning? Can scale be changed to be linear? This plot is very hard to interpret in its current form.

- (Figure 6 caption) Suggest a change in text: Other solid square markers represent data collected when the chamber was operated in a steady-state temperature mode (instead of steady cooling).

Summary:

I have enjoyed reading the submitted manuscript and find that this is an intriguing new idea. In order to recommend the manuscript for publication I think the authors need to state more convincingly that they constrain the conditions for the observed nucleation. Furthermore, I think the conclusion would be significantly enhanced by describing whether or not other existing CFDC systems could run or test run such a mode of operation. I also encourage the authors to conduct a round of editing to ferret out small mistakes that I found numerous enough that not all could be included here.

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[1] Castarède, D. and Thomson, E. S. (2018). A thermodynamic description for the hygroscopic growth of atmospheric aerosol particles. *Atmospheric Chemistry and Physics*, 18(20):14939–14948.

[2] Stetzer, O., Baschek, B., Lueoeond, F., and Lohmann, U. (2008). The zurich ice nucleation chamber (zinc) - a new instrument to investigate atmospheric ice formation. *Aerosol Science and Technology*, 42(1):64–74.

[3] Garimella, S., Kristensen, T. B., Ignatius, K., Welti, A., Voigtländer, J., Kulkarni, G. R., Sagan, F., Kok, G. L., Dorsey, J., Nichman, L., Rothenberg, D. A., Rösch, M., Kirchgäßner, A. C. R., Ladkin, R., Wex, H., Wilson, T. W., Ladino, L. A., Abbatt, J. P. D., Stetzer, O., Lohmann, U., Stratmann, F., and Cziczo, D. J. (2016). The spectrometer for ice nuclei (SPIN): an instrument to investigate ice nucleation. *Atmospheric Measurement Techniques*, 9(7):2781–2795.