Review of "On the drivers of droplet variability in …" by Georgakaki et al., submitted to ACP, November 2020

By Jeff Snider, University of Wyoming

The authors have done a good job of improving their ACPD manuscript. Below, I provide a detailed review of their ACP submission. In my opinion, the following things should be addressed before the manuscript is published in Atmospheric Chemistry and Physics.

Most Important:

In my opinion, it is subjective to bin σ_w as 0.1 0.3, 0.6 and 0.9 m/s (Figure 8). What happens if you re-bin with a different set of σ_w values? Please note, I do not feel that a complete answer is on L610 to L614. Given that I consider your setting of σ_w to be subjective (Figure 8), and what you say on L610 to L614, it is my opinion that you need to provide evidence (and discussion) of the robustness of the following procedure: Your setting of σ_w values, your drawing of horizontal lines (e.g., in Figure 8), and your picking of $N_d^{\lim} - \sigma_w$ pairs for Figure 9.

Related to what I say above, did the papers that resulted from SEA and SEUS examine the robustness of their procedure for determining $N_d^{\lim} - \sigma_w$ pairs? Was their procedure the same as yours?

Here is a related question: Why not draw a horizontal line at the flat-tops (plateaus) of Figures 8d or 8g and conclude that those $N_d^{\lim} - \sigma_w$ pairs should also be included in Figure 9?

Also in my opinion, to refer to the derived $N_d^{\lim}$ (i.e., where the horizontal lines are drawn in Figure 8) as the velocity-limiting concentration requires a caveat. The caveat is that only those aerosol size distributions corresponding to points close to the horizontal line are actually velocity-limited. Consequently, I cannot understand this statement, mostly because it is not clear what you are implying by "regime": "Within the velocity-limited regime of droplet formation, we can notice that the corresponding Smax values are low (<0.1 %),

reflecting the severe water vapor limitation that allows only a few particles to activate into cloud droplets." I'm looking at Figures 8a, 8d, and 8e and in all cases, below where you have drawn the horizontal line, I see data points with Smax > 0.1%. This is particularly evident in Figure 8e. These cases have relatively large N_d-to-N_{aer} ratios. Those relatively large ratios concord with those points having relatively a large Smax (i.e., larger than 0.1 %). It follows that those cases *could have* achieved larger N_d, for example, had they had the same N_{aer} but fewer large particles (recalling that N_{aer} is controlled by smaller particles), or if they had an Aitken mode at critical supersaturations larger than 0.1 %. In either case, those cases are not velocity-limited, rather, they are aerosol limited. If I have it right, this is your main point. This aerosol limitation is in the sense envisioned by Twomey (1993) where he states that "In a general way, increasing particle numbers must reduce the maximum supersaturation achieved, which means that some previously activated particles may now not be activated. This factor itself tends to reduce droplet numbers somewhat. Hence it is not necessarily true that increasing particle numbers mean a proportional increase in droplet numbers. It is not too difficult to invent distributions of particles such that an increase in their total concentration leads to *reduced* numbers of final droplets". I feel that the authors need to better incorporate what Twomey (1993) was/is getting at. He was explicit! Overall, this paragraph (L577 to L601) needs to be rewritten with improved definition of what you mean by "regime" and with recognition of the shoulders you rest on.

Related to what I said in the previous paragraph, I feel you need to spell out what you mean by velocity-limited and aerosol-limited. Perhaps it's best to do this with a table. As I understand it, velocity-limited implies Nd ~ $N_d^{\lim}$ and Smax < 0.1%. Conversely, aerosol-limited implies Nd < $N_d^{\lim}$ and Smax > 0.1%. Still, I'm puzzled by two things. 1) You use N_{aer} to classify aerosol-limited behavior (L689 to L682) and you also use Smax to classify aerosol-limited behavior (L693 – L696). Which of the second conditions (the Smax second condition or the N_{aer} second condition) is best, in your estimation, to guide future investigators and especially those conducting experiments at locations other than your field site? 2) The table

you can also help those who wonder how to think about a situation with Nd ~ $N_d^{\lim}$, and with Smax > 0.1 %, and what to call that situation.

The issue of who to cite, and who not to cite, is too parochial. Here is one example of this in your manuscript. If you are going to contend that the relationship shown in Figure 9 is significant, for example because it can be used to diagnose σ_w from retrievals, then it is important that you reference work that has retrieved (using airborne lidar, for example) droplet concentration. If you don't want to cite the Wyoming team, and want to focus on space borne lidar, then please do so. Perhaps you should cite Danny Rosenfeld's team, and the work they are doing in this arena.

On L721 to L724 you must be careful. The persistence/existence of mixed phase clouds can be controlled by many things other than SIP. For example, the availability of active INPs (need primary ice to get secondary ice) and the temperature regime (Hallett/Mossop occurs over a relatively narrow temperature range). As I pointed out in my pre-review, control of droplet size (and thus SIP) can also come from variation of cloud depth and entrainment, and from processes that broaden the cloud droplet size distribution (updraft variability is one of them). So, I encourage you to more carefully circumscribe what you are saying in lines L721 to L724.

Less Important, but should also be addressed:

Figure 8 - What is the purpose of the sloped dashed lines in Figures 8a, 8d, and 8e? Can they be removed? They caused me confusion. Perhaps you could put in the 1 to 10 line…maybe that is what those sloped dashed lines are indicating. If so, tell us that. Finally, better definition (longer and outward directed) of the minor ticks is needed in Figure 8, and I would remove the grid within the panels of Figure 8, its making things murky.

L474 - What about removal of droplets by riming or by completion for vapor in MP conditions? Did the HOLIMO show evidence of either of those processes in those instances where LWC was depleted relative to adiabatic?

About the CFSTGC. My understanding (Snider et al., JGR, 2010) is that two stream-wise temperature gradients are experienced by the particles moving along the flow path. Is this correct for the instrument you operated? If yes, why does your description imply that there is only one stream-wise temperature gradient?

L176 – L181 – In addition to the sensitivity of SS to pressure, the measured concentrations are also dependent on pressure via their dependence on aerosol sample flow rate. The latter is reported (by instruments) as a mass or volume flow rate. IMO statements are needed to tell the reader how CCN concentrations were calculated for comparison to the following other concentration measurements: N_{aer}, aerosol size distribution, measured cloud droplet number, and theoretical cloud droplet number. One place this is relevant is in Figure 5 where aerosol is reported per cubic centimeter at STP and droplets are expressed per cubic centimeter. There are other examples of this (Figures).

L204. Why is this statement relevant to this paper?: "HOLIMO has an open path configuration (i.e. the detection volume lies between the two instrument towers) and thus is also able to measure raindrops up to a size of ∽ 2 mm."

L209 to L211. I'm think your statement about the bulk density of liquid is getting in the way of what's important. Everyone knows that the density of liquid is a constant at 1000 kilogram per cubic meter. In my view, a hydrometeor size distribution, measured by HOLIMO, should be sufficient for calculating the (cloud) LWC and (cloud) droplet number concentration, provided the ice particles (typically much larger) can be distinguished from the smaller (cloud) droplets. Please provide discussion on how well this distinction (cloud droplets vs ice) can be made by HOLIMO and what the implication is for estimation of Nd (droplet concentration at D > 6 micrometer) and (cloud) LWC.

L210. It's not clear why "measured number concentration" is in this sentence.

Table 1. Droplets smaller than 6 micrometer can, in some instances, contribute significantly to droplet number concentration. Is this discussed?

L206 to L208. If I have it correct, the WOP is the lower of the two sites. So, you only have cloud microphysical data (HOLIMO) when the WOP was at AGL heights greater than the cloud base? Is this explained?

L231. Is the DBS acronym needed for this manuscript?

L248. What's relevant here is the value (constant ?) applied for the surface tension, not the value of the universal gas constant. BTW, you have already defined the density of liquid water (cloud microphysical section).

L286. I commented on the w-star approach in a review of the Morales and Nenes (2010). Since you are presenting σ_w in Figures 5, 7, 8, 9, and 10 please double check that the transformation from σ_w to w-star is applied in the activation calculations.

L335. The Seinfeld and Pandis (2006) book is enormous. Please specify where in the text the authors conclude this. It is better to reference journal paper (s) which concludes that in aged air the concentration is lower, the accumulation mode is pronounced, and the hygroscopicity is enhanced.

L357. IMO, you should introduce, parenthetically, Figure 2 before going into this discussion of the weather-impacted data.

L360. Precipitation rate maximizes at 1.1 mm / hr, not "up to ~ 7 mm / hr".

L372. The "…March nucleation processes.." should be described differently. You are speculating about the removal of aerosol particles that activate and then removed by precipitation, or by precipitation removing aerosol through diffusive and impaction processes. The word "nucleation" here will alert some of your readers incorrectly to "aerosol particle nucleation" (aka, NPF).

L464. I do not see a "gap" of vertical winds in Figure 5f. I do see that the red data end at ~ 16:15. The labels (e) and (f) are not correctly placed in Figure 5.

L466 to L469. In Figure 5d, I see σ_w values from 0 m/s to 0.36 m/s. In Figure 5f, I see σ_w values from 0.25 m/s to 0.47 m/s. You say that the selected values are 0.24 and 0.16 m/s for 7 March and 0.37 m/s for 8 March. Perhaps this comment is only for the beta version of the ACPD manuscript. Please check.

Did you discuss why the time resolution of σ_w is so poor in Figure 5d and Figure 5f? I read that the time resolution of the lidar is "up to 5 s" (Table 1). Do you mean that the time resolution is "no better than 5 s"?

Figures 5c and 5d are using blue to indicate three things. Error bars on averages (are these actually variabilities), a line that connects filtered/measured values, and the "three cloud events observed." This presentation needs to be improved. Thank you for correcting this. Yet, is there not a better color to connect the data points?

L523. "focused" -> "faced."

L601 "Na" -> "Naer"