

Response to Referee #2

The authors would like to thank Prof. Snider for carefully reading the manuscript and providing thorough and constructive remarks that have improved the manuscript. The replies to the comments are given below. The referee comments are highlighted in red with our responses in black fonts.

Most Important:

1. 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} - σ_w pairs for Figure 9.

In Figures 7 and 8 we have shown results of N_d for representative values of σ_w in the observed range of σ_w values derived from wind lidar measurements (shown in Figures 10c and 10d). These observations clearly show that σ_w varies between 0.1 and 1.0 m/s, with the higher values being recorded at the mountain-top site compared to the valley site.

The horizontal lines are plotted in Figure 8 only when velocity-limited conditions are met, and tend to occur when the predicted $S_{max} < 0.1$ %. The absence of a horizontal line indicates that S_{max} is systematically higher than 0.1 % and therefore an N_d^{lim} was not reached. The picking of N_d^{lim} - σ_w pairs, shown in Figure 9, shows that at Wolfgang-Pass (WOP; orange circles) N_d^{lim} is achieved only for $\sigma_w \leq 0.2$ m/s, while at Weissfluhjoch (WFJ; blue circles) N_d^{lim} is reached “even for more turbulent boundary layers” with $\sigma_w \leq 0.5$ m/s.

2. 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} - σ_w pairs? Was their procedure the same as yours?

Both studies (Kacarab et al., 2020; Bougiatioti et al., 2020) have followed the same probabilistic approach for computing cloud droplet number as the one we followed here. The only difference is that the updraft velocities were obtained from aircraft observations in cloud legs. The robustness of our approach is supported by the good degree of droplet closure achieved. This is also the case for the study of Kacarab et al. (2020), where calculated droplet numbers in non-precipitating boundary layer clouds were found to agree with the observed ones to within 20%.

3. 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} - σ_w pairs should also be included in Figure 9?

As mentioned in Comment 1 above, the horizontal line in Figure 8 is drawn only for those cases when S_{max} is found to fall below 0.1 %. This indicates which N_d^{lim} - σ_w pairs will be included later

in Figure 9. We should clarify here that N_d^{lim} is determined by calculating the averaged N_d achieved whenever $S_{max} < 0.1\%$ for each examined σ_w value.

4. 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 S_{max} 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 $S_{max} > 0.1\%$. 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 S_{max} (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.

Indeed, Twomey (1993) and others before and after (e.g., Jensen and Charlson, 1984; Ghan et al., 1998, Nenes et al., 2001 and Reutter et al., 2009) recognized the role of water vapor competition on droplet formation. Twomey (1993) discusses this conceptually and states that competition may be fierce enough to reduce droplet number with increasing aerosol (which was later demonstrated by Ghan et al. 1998 to occur for mixtures of sulfate aerosol and sea spray). Reutter et al. (2009) did not focus on such extreme conditions of water vapor competition, but rather situations that are consistent with dominance of anthropogenic pollution in clouds. Indeed, for high enough aerosol amount, droplets in clouds become insensitive to aerosol perturbations, giving rise to the so-called “velocity limited cloud formation”. Formally this condition can be expressed in terms of the partial derivative of droplet number to aerosol– the smaller the derivative, the more clouds are velocity-limited (e.g., Morales-Betancourt and Nenes, 2014). Kacarab et al. (2020) used such derivatives to express when clouds respond weakly to aerosol perturbations, i.e., became velocity-limited, for a wide range of ambient size distributions. The derivative became small (i.e., the clouds became velocity-limited) when supersaturation dropped below 0.1% because of the increasingly fierce competition for water vapor during droplet formation. Bougiatioti et al. (2020) also found similar results. We build upon these findings and apply the above to the Alpine aerosols and clouds sampled during the study. We will make these discussions very clear.

5. 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 $N_d \sim N_d^{lim}$ and $S_{max} < 0.1\%$. Conversely, aerosol-limited implies $N_d < N_d^{lim}$ and $S_{max} > 0.1\%$. Still, I'm puzzled by two things. 1) You use N_{aer} to classify aerosol-limited behavior (L689 to L682) and you also use S_{max} to classify aerosol-limited behavior (L693 – L696). Which of the second conditions (the S_{max} 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 $N_d \sim N_d^{lim}$, and with $S_{max} > 0.1\%$, and what to call that situation.

This is a good point. N_{aer} alone is a weak constraint and it can be used only as a rough indicator for aerosol- or velocity-limited conditions. The primary condition for velocity-limited clouds is $S_{max} < 0.1\%$ and $N_d \sim N_d^{lim}$, as supported by the histogram presented in the supplementary material (Fig. S6). We will make these points very clear in the text.

6. 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.

When this conclusion was written, we cited the work that was used in assembling the specific figure. The suggestion to refer to studies that adopt airborne (e.g. Snider et al., 2017) or satellite (e.g. the review paper of Grosvenor et al., 2018) cloud droplet number retrievals is also useful. These two reference studies are now added in the revised manuscript.

7. 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.

We do not disagree with these comments. However, when we refer to aerosol effects on clouds and the size distribution (which are separate from cloud depth, entrainment and vertical velocity variations), approaching the conditions of N_d^{lim} is consistent with the maximum reduction in droplet size (even if the distribution broadens). The latter tends to be consistent with reduced riming – compared to if the cloud, everything else identical, has less aerosol present.

Less important, but should also be addressed:

1. 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.

Thank you for the suggestions. The sloped dashed lines are removed from Figures 8a, 8d and 8e, to avoid any confusion. Only the horizontal dashed lines can be found in these figures, indicating the limiting droplet number (if it is reached). The representation of the minor ticks is also improved while the grids are removed.

2. 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?

Thank you for this point. Indeed, within the seeder-feeder situation observed on March 8, HOLIMO recorded a large fraction of rimed particles and graupel, indicating that ice particles gained mass by riming and depositional growth while falling through the mixed-phase cloud layer (Ramelli et al., 2020). This comment is now added in the revised manuscript.

3. 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?

Thank you for bringing up this point. The description is based on the fundamental operation principle of the instrument. Calibration is done based on the delta T across the full column so the resulting supersaturation is calibrated against that temperature difference.

4. 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).

Thank you for this comment. N_{aer} reported in Figure 5 is now converted from STP to ambient conditions, to be consistent with the measurements shown in this figure. In Figures 1a and 2, the aerosol concentrations are reported at standard conditions since our aim was to directly compare two stations located at different altitudes – WOP and WFJ. CCN data were collected only at WFJ

and are expressed at ambient conditions in Figure 3. To determine the predicted cloud droplet number and perform the droplet closure study we used solely in-situ measurements. This is the reason why we have decided to report all concentrations per cubic centimeter throughout the manuscript, except N_{aer} in Figures 1a and 2. Statements are added in the text to clarify whether the measurements are expressed at ambient or standard conditions.

5. 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.”

Thank you. This statement is removed.

6. 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 N_d (droplet concentration at $D > 6$ micrometer) and (cloud) LWC.

The distinction between cloud droplets and ice crystals is done for particles larger than $25\ \mu\text{m}$ diameter based on the particle shape (circular vs non-circular). For particles smaller than $25\ \mu\text{m}$ it is challenging to differentiate between the ice and liquid phase owing to resolution limitations of HOLIMO (around 8 times the effective pixel size ($3\ \mu\text{m}$) is required to differentiate between liquid and ice). As noted, droplets below $6\ \mu\text{m}$ are possible, and if present would result in HOLIMO underestimating their total concentration. The influence of small cloud droplets on the reported LWC is however much smaller, as the contribution of the larger cloud droplets dominates the relevant distribution. This is now mentioned in the text.

7. L210. It’s not clear why “measured number concentration” is in this sentence.

Thank you for noting this! This is now removed.

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

Good point, a discussion is added in the revised manuscript.

9. 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?

Indeed, we have cloud microphysical data when the Holoballoon was at AGL heights greater than the cloud base.

10. L231. Is the DBS acronym needed for this manuscript?

Good point. It is now removed.

11. 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).

Thank you for this comment. Here the surface tension of pure water is assumed and this is calculated as a function of temperature.

12. 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.

The transformation from σ_w to w^* is actually carried out within the parameterization. The calculated σ_w values are among the inputs required to call the parameterization and this is the reason why we have decided to present σ_w instead of w^* in the figures throughout the manuscript.

13. 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.

The pages referring to the free tropospheric aerosols in Seinfeld and Pandis (2006) are specified in the revised manuscript (pp. 376-378). Here we have also cited some previous studies focused on the mountain-top site of Jungfraujoch (JFJ) in the Swiss Alps. For instance, when free tropospheric conditions prevail at JFJ, Baltensperger et al. (1997) provided evidence that sufficiently aged aerosol is usually found, while during PBL injections Kammermann et al. (2010) and Jurányi et al. (2011) reported an increase of aerosol loadings accompanied by a decrease in aerosol hygroscopicity.

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

Done!

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

Thank you, corrected. There was a mistake in Figure 2.

16. 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).

Thanks for this comment. Suggested changes are made.

17. 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.

Thank you for these comments. All these issues are now addressed in the revised manuscript.

18. 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”?

The temporal resolution of instantaneous velocity from the wind lidar stated in Table 1 is now changed to “5 s max”. However, σ_w is determined with much lower frequency. To calculate the σ_w values that are shown in Figures 5e and 5f, the high-resolution wind lidar data was grouped by hour and each fitted to half-Gaussian PDFs with zero mean and standard deviation σ_w (supplement Fig. S2). The σ_w values shown in Figures 5e, 5f and Figures 10c, 10d are calculated per hour.

To determine the σ_w values for the closure study we isolated the segments that correspond to the three cloud events shown in Figures 5c and 5d, and fit to half-Gaussian PDFs, being $\sigma_w = 0.24$ and 0.16 m/s for 7 March, and, 0.37 m/s for 8 March.

All the above σ_w values are then used to run the droplet number calculation – by converting each to a characteristic velocity ($w^*=0.79\sigma_w$), which is then used by the parameterization to compute droplet number – which is equal to the PDF-averaged droplet number (Morales and Nenes, 2010).

Changes have been made in the Sections 2.3 and 3.2.1 to clarify the above-mentioned points.

19. 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?

The error bars represent the standard deviation of the measured droplet numbers (hence the variabilities), not the average deviation (or the mean absolute deviation). Figure 5 is modified in the revised manuscript. The same color (cyan) is now used to represent the 2-minute averaged N_d and the corresponding error bars, while a black line is used to connect the data points.

20. L523. “focused” -> “faced.”

Amended.

21. L601 “Na” -> “Naer”

Thank you, corrected.

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