Response to Referee #2

The authors would like to thank Prof. Snider for carefully reading the revised manuscript and once more providing very thorough and constructive remarks. Reviewer comments are provided in blue font with our responses in black fonts.

1. L546 - L548. "The Gaussian fit to the updraft velocities gave a distribution with $\sigma_w = 0.24$ and 0.16 ms⁻¹ for the first two clouds present on the 7th of March, and, $\sigma_w = 0.37$ ms⁻¹ for the cloud system observed on the 8th of March."

Two comments:

1) The σ_w data (these are 1-hour averages) plotted in Fig. 5e indicate a factor of four spread over the Cloud-1 interval. Consequently, the σ_w evaluated for Cloud-1 (0.24 m/s; see L546 - L548) does not seem reasonable for either the first part of Cloud-1 (time < 17:00) or for the second part of Cloud-1 (time > 17:00).

2) It's not clear how to reconcile the σ_w averages in Fig. 5e with the σ_w evaluated for Cloud-2 (0.16 m/s; see L546 - L548). By eye (Fig. 5e), the value for Cloud-2 is $\sigma_w \sim 0.12$ m/s. It must be that there is there more updraft variability during the times you have cloud data (Fig. 5c, Cloud-2).

Since the N_d closure (Fig. 6) is central to your paper, I think it is appropriate to explore further this aspect of your analysis. Here is my recommendation. Please present averages of σ_w (in and updated Figs. 5e and 5f, or in a response) for intervals shorter than 1 hour. For example, present a 10-minute average corresponding to 120 updraft samples (temporal resolution 5 s max). In my opinion, this would make clear the basis for the σ_w you report in L546 - L548. It could also make it simpler for you. For example, should you care to rationalize splitting Cloud-1 into an earlier (time < 17:00) and later interval (time > 17:00) interval. Or, it could make it easier for you to argue that the σ_w average for time < 17:00 (7 March) is biased low by updraft measurements collected prior to start of Fig. 5e at ~ 16:30.

Thank you for raising this point. The temporal resolution of the wind lidar products is variable, with the maximum resolution being 5 s (as mentioned in the manuscript). During the two cloud events on 7 March the maximum temporal resolution of the wind lidar was ~ 30 s, allowing us to use ~ 10-20 vertical velocity samples (updrafts + downdrafts) for the suggested 10-min calculations. Given that we fit only the updraft velocities to the half-Gaussian PDFs, which is just a subset of these 10-20 samples, the σ_w calculated from the 10-min interval PDF is just too uncertain to be useful. Using the hourly σ_w resolves this problem.

During Cloud-1, the in-cloud updraft variability is indeed high, with higher σ_w values recorded after 17:00. A discussion is now added in the revised manuscript mentioning that the σ_w derived for Cloud-1 might be biased low by the lower σ_w values measured before 17:00. Nevertheless, the updraft averaging used in the droplet closure study corresponds to the N_d averaging timeperiod and, therefore, we do not expect the degree of closure to be affected.

 L556 – L563 "The good agreement between measurements and predictions - even under mixed-phase conditions, reveals that processes like condensation freezing and.." It's not clear what you are getting at with "condensation freezing." It's established, by your group and others (Korolev et al. 2017), that pathways generating ice beyond a few tens per liter, within moderate updraft (≤ 1 m/s), significantly reduce the Smax. I think this is what Sud et al. (2013) and Barahona et al. (2014) were getting at. Can you rewrite L556 – L563 for clarity?

The sentence has been rephrased.

3. I recommend the following addition of "**by**":

Reutter et al. (2009) pointed out that droplet formation in clouds can be limited by the amount of CCN present (called the "aerosol-limited" regime), or **by** the vertical velocity that generates supersaturation in the cloudy updrafts (called the "velocity-limited" regime).

Thank you, corrected.

4. "..may decrease CCN activity through the formation of glassy aerosol, has not been assessed in a closure study to date." Is this speculative or is a reference missing?

We cannot provide a reference here, since to our knowledge there are no in-situ studies assessing cloud droplet closure in mixed-phase clouds. The sentence is now modified in the revised manuscript, to make this point clearer.

5. "With box model simulations, Hammer et al. (2015).." These simulations applied a closed adiabatic parcel model, I think. "Box" seems like a rigid container.

Thank you for this comment. "Box model" is a term frequently used for 1D models, like what we use here. Nevertheless, we switched to "cloud parcel model" in the revised manuscript.

6. L299 – L303 "Aiming to examine how N_d responds to different vertical velocity-aerosol situations, as a sensitivity test, potential N_d for both sites are calculated at 10 values of σ_w between 0.1 and 1.0 ms⁻¹ that cover the observed range (Section 3.2.4). Note that we use the term "potential" droplet number throughout this study, as its calculation is performed regardless of the actual existence of clouds over the measurement sites."

This application of the word "potential" is useful. Given what you are saying, Section 3.2.1 is _not_ about potential droplet number, rather it's about measured Nd and measured σ_w in (near) actual clouds. In contrast, Section 3.2.2 is about potential droplet number.

Here is what I'm advocating for: Please improve the section titles so that they apply your definition (L299 - L303) and especially so for titles of Sections 3.2, 3.2.1, and 3.2.2.

Related to this is L541 - L542:

"Note that the *potential* droplet formation is evaluated using the updraft velocity PDF calculated for each cloud period, rather than the hourly σ_w data shown in Figures 5e and 5f (Section 2.3)."

In L541 – L542, it's my opinion, you should remove the word "potential."

Thank you for this point. Suggested changes are made throughout the text, to ensure that the term "predicted N_d " is used only when comparing against the direct observations of cloud droplet numbers (i.e., in the droplet closure section), while the term "potential N_d " is adopted in the rest of the paper.

7. L606 "Lower N_d values are visible during nighttime due to the limited turbulence." Turbulence is lower near a surface at night, however, turbulence is being prescribed in Fig. 7. The diurnal cycle is explained on L320 – L321: "N_{aer} at WOP peaks in the evening, reaching up to \sim 104 cm⁻³ presumably because of BB emissions in the valley which seem to stop around midnight (Fig. 1a)."

Thank you for pointing this out. What is driving the potential N_d in Figure 7 is, indeed, the amount of aerosol particles rather than the turbulence which is prescribed. This sentence is now corrected.

8. Why did the locations of the SEA points change moderately from Figure 9 (acp-2020-1036manuscript-version3.pdf) compared to an earlier draft of Figure 9?

The SEA data points presented in Figure 9 are derived from Figure 6 in Kacarab et al. (2020). This figure illustrates N_d^{lim} for several research flights as a function of the measured characteristic vertical velocities, which are then translated into σ_w in our case ($\sigma_w = w^*/0.79$). The inset plot of this figure shows a sensitivity test of how N_d^{lim} varies for polluted, intermediate and clean conditions for w* values between 0.1 and 0.6 ms⁻¹. In the earlier version of our manuscript we superimposed the results from this sensitivity analysis, whereas in the latest version we decided to include the actual w* measurements. The latter is more consistent with the SEUS data points (derived from Fig. 6 in Bougiatioti et al., 2020), which are also obtained from aircraft observations in cloud legs.

References:

- Bougiatioti, A., Nenes, A., Lin, J. J., Brock, C. A., de Gouw, J. A., Liao, J., Middlebrook, A. M., and Welti, A.: Drivers of cloud droplet number variability in the summertime in the southeastern United States, Atmos. Chem. Phys., 20, 12163–12176, https://doi.org/10.5194/acp-20-12163-2020, 2020.
- Kacarab, M., Thornhill, K. L., Dobracki, A., Howell, S. G., O'Brien, J. R., Freitag, S., Poellot, M. R., Wood, R., Zuidema, P., Redemann, J., and Nenes, A.: Biomass burning aerosol as a modulator of the droplet number in the southeast Atlantic region, Atmos. Chem. Phys., 20, 3029–3040, <u>https://doi.org/10.5194/acp-20-3029-2020</u>, 2020.