Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-225

Response to Anonymous Referee #2 comments

The authors use measurements of aerosol number and composition along with updraft variability to identify the role each plays in determining simulated cloud droplet number concentrations. I have many concerns with this manuscript. The authors reference anthropogenic and biogenic aerosol precursors as a possible driver of climate over the southeast united states, however there is little to no discussion of this feedback. Also, simulations of cloud droplet concentrations are not compared to any actual measurements of cloud droplet concentrations.

***Response**: We thank the referee for the thoughtful and careful review. Some of the issues were also raised by Reviewer #1 and are addressed hereafter. The reference to anthropogenic and biogenic precursors was to provide the context for the SENEX campaign. The emissions-aerosol-cloud link, although very important, is not the focus of this study. Here we focus primarily on the aerosol-cloud link, and the underappreciated role of vertical velocity covariance. Unfortunately, there were no cloud data available, as the aircraft was operating in visual flight mode most of the time. However, we have shown in other studies (e.g. Kacarab et al., 2020) that our approach for calculating Nd values agree with observed droplet number in non-precipitating boundary layer clouds, therefore the conclusions are robust.*

One of the major key points of the manuscript relies on comparisons of night flights and day flights however there are only 3 night flights and a total of 10 day flights. It is hard to keep track of which cases are night time and which are day, though the diuranl variability of sigmaw is a key point of the paper. This makes it hard to follow this point. You refer to several different flights, and honestly the flight number is somewhat meaningless to the reader. Referring to the flights by certain properties (I.e. night flight 1, night flight 2) would be more useful.

***Response**: This issue was also raised by Reviewer #1. We now more clearly distinguish between daytime and nighttime flights (sun for day and moon for night) so the diurnal variability of σw is easier to see. Finally, the added characterization of the two pairs of flights as "Urban" and "Rural" now facilitates keeping track of which flights are mentioned.*

The Figure quality is inconsistent and a few figures are repetitive, showing the same result in slightly different ways. Some Figures and tables do not contain data from all cases, leaving the reader to wonder why the other cases were omitted.

***Response**: This was pointed out by Reviewer #1 as well, and now Supplementary material includes organics mass fractions and estimated cloud droplet number for all studied flights. Furthermore, new figures are now plotted in the revised version which point out important findings for the vertical velocity vs. aerosol limiting regimes and figures from the previous version showing similar results are now moved to the supplementary material.*

It is not clear that any result came from section 3.1. Section 3.2 is confusing as it mainly involves a comparisons of individual cases and many sentences and paragraphs do not relate to one another. I was so lost that I stopped reading in this section. It is unclear what data (from tables) was used to calculate many of the numbers listed in this section.
***Response**: These sections have been rewritten for clarity (with additional analysis) and numbers-figures are cited in the supporting discussion.*

The main result appears to be that updraft velocity and variability are higher during the day, leading to more "simulated" cloud droplets, which is not surprising or new. Comparisons to the contribution of organic mass and particle concentration is also not new. Overall, the manuscript lacks new and measurement supported results, lacks organization, contains figures of low quality, and hard to follow discussion. I am not suggesting rejects of the manuscript only because the measurements published are of high value. I suggest the manuscript be reconsidered after substantial revisions are made to the overall message and clarity of the text, and quality of the figures.
***Response:** Most aspects of warm cloud physics and especially droplet formation are known for decades. However, droplet formation remains at the heart of the aerosol indirect effect, so ensuring that models capture droplet number for the "right reasons" is critical for constraining aerosol-cloud-climate interactions. The latter aspect is where a huge knowledge and data gap exists – and where our study provides important constraints (vertical velocity, aerosol number, potential droplet number) and insights (covariance between σw and Na and their role on the Na-Nd relationship in the SE US). The additional insights on the limiting droplet number, and its explicit dependence on σ_w is also new and important, and offers a new possibility for remote sensing. Given that model assessments of aerosol–cloud-climate interactions do not evaluate for vertical velocity (or covariance with other parameters), our work here shows that this can lead to an unresolved source of hydrometeor variability and bias. We have made these points very clear now.*

Specific comments:
Line 25: Different how? Explain how it is different before you talk about why.
***Response:** The abstract has been rewritten and no longer includes a reference to differing climate trends.*

Line 94: Can you provide a source for the WLOPC?
***Response:** We have added Brock et al. (2011) as a source.*

Line 194: I believe you meant to cite Table 1. What do you mean by "overall values"?
***Response:** We thank the reviewer for pointing out this inconsistency. The overall value is the 0.25±0.05 stated just before, a row has been added in Table 1 giving the average values and a clarification is added to the revised text.*

Line 196: For what? It would be helpful to lead the reader more currently it is hard to see where this text is going. Are these the distributions in Figure 2? If so cite them in this sentence.

Response: *Aerosol number size distributions are crucial for the calculation of the total aerosol number during each flight, as they enter as input in the droplet number parameterization. This is added as a clarification in the revised text.*

198: I don't think you need this statement twice within 10 lines of each other.
Response: *The repetition in now omitted from the revised version.*

Line 203: You only chose 4 distributions for each plot in Figure 2. How did you choose which flights to include/exclude? I suggest making your y axis the same for each flight. It would be more obvious that the concentrations are different. A log scale for the y axis may be helpful if the authors choose. At the very least please use the same notation for the y axis tick labels.
Response: *These are good points. Each plot represents a grouping based on e.g., passes in free tropospheric conditions (a) or nighttime flights (d). The selection of the different pass types reflects the need to represent daytime-nighttime contrasts (which is important for droplet number calculations as shown below) as well as the shape of the size distribution. As each flight had around 5 passes in a constant altitude (e.g., Table 2) presenting all average particle size distributions in one graph is cumbersome. Instead size distributions are grouped with common characteristics, and the data from 9/13 flights are represented in the figure, while the rest mostly fall in one of the represented categories. The vertical axes for the three plots are now similar (apart from the nighttime flights, which is maintained different because during nighttime the aerosol number as well as the variability was lower).*

Line 205: " the modal diameters did not vary much" Why is that significant?
Response: *This is important as it dictates particles of which mode will activate, depending on* S_{max}.

Line 209: You previously mentioned that the organic mass fraction was high during a night flight, but here you are saying 'contrasts between day and nighttime aerosol characteristics/variability may not be as large' Are you saying contrast in composition should be small between night and day? Are you saying the difference in accumulation mode concentration between night and day plays a bigger role in determining cloud droplet number concentration than aerosol a characteristics/variability? It is not clear and if you are saying the latter then you should reference you partial derivatives that you mentioned in line 164 to confirm. If you are going to "discuss the variability of the total aerosol number on droplet number in section 3.2" then it should probably not be mentioned here.
Response: *Large part of the discussion has been revised to promote a more coherent and comprehensive flow in the text.*

Line 212: It is not clear that "Cont kappa" and "Cont Na" is the partial derivative in Table 3/4. Be consistent with your abbreviations. "contribution" is listed as 'Cont' and 'Contrib' which is confusing.
Response: *We sought to determine the relative contribution of aerosol composition (expressed by* κ*), total aerosol number and vertical velocity to variations of droplet number, using a variance-based approach. For this, we compute the partial sensitivities of droplet number to* N_a, κ, σ_w *(Sullivan et al., 2016; Bougiatioti et al., 2017), multiply them with their respective variance and sum as follows to obtain the droplet number variance:*

$$\sigma^2 N_d = \left(\overline{\frac{\partial N_d}{\partial N_a}}\,\sigma N_a\right)^2 + \left(\overline{\frac{\partial N_d}{\partial \kappa}}\,\sigma\kappa\right)^2 + \left(\overline{\frac{\partial N_d}{\partial \sigma_w}}\,\sigma\sigma_w\right)^2$$

The relative contribution of Nα, κ, and σw to the droplet number variance is then estimated as follows, and their values presented in Tables 3 and 4:

$$\varepsilon_{Na} = \frac{\left(\overline{\frac{\partial N_d}{\partial N_a}}\,\sigma N_a\right)^2}{\sigma^2 N_d}, \varepsilon_\kappa = \frac{\left(\overline{\frac{\partial N_d}{\partial \kappa}}\,\sigma\kappa\right)^2}{\sigma^2 N_d}, \varepsilon_{\sigma w} = \frac{\left(\overline{\frac{\partial N_d}{\partial \sigma_w}}\,\sigma\sigma_w\right)^2}{\sigma^2 N_d}$$

This is further clarified in the revised text and all abbreviations are now consistent.

Line 217: suggest changing "chemical composition" to kappa or hygroscopicity parameter and if that is how "chemical composition" is expressed throughout the paper I suggest using one consistent term or symbol.

Response: *Changes in the hygroscopicity parameter are a direct result of chemical composition changes, and we stress this point (e.g. L230 changes in hygroscopicity (i.e. chemical composition)). Keeping the composition-hygroscopicity link is important, given that variations in hygroscopicity (chemical composition) induce variability in droplet number.*

Line 228: reference table/figure that identifies daytime sigma2 varies little and is large. Sigma w at night seems to vary less than during the day based on your next two sentence.

Response: *This is now clarified in the text.*

"The large diurnal variability in σ_w (from 0.3 m s^{-1} at night to 1.0 m s^{-1} at day) contributes considerably to the diurnal variability in N_d…."

Line 231-232: Is the data used to obtain 0.23±0.04 and 0.97±0.21 in one of these tables?

Response: *This is now clarified in the text.*

"The vertical velocity distributions observed gave σ_w =0.97±0.21 m s^{-1} for daytime flights, and σ_w =0.23±0.04 m s^{-1} for nighttime flights (Table 2 and SP3)."

Line 234: " total variability in Nd based on dNd/d_, dNd/dNa and dNd/d_w and the variances of _, Na and _w" this is repetitive.

Response: *Amended*

"…we estimate their contribution to the total variability in N_d based on the variances of κ, N_a and σ_w and the sensitivity of droplet formation to those parameters."

Line 241: you should state these "sectors" were in atlanta and alabama respectively. You haven't referred to sectors at all so far, making it confusing to suddenly mention them. This paragraph is hard to follow. There are several numbers compared for different cases at different time periods

Response: *Good point. They different areas are now stated. The paragraph has now been rewritten.*

Line 257: these exact flights and "sectors" were discussed 2 paragraphs ago. This could be better organized.

Response: *Section 3.2 has been rewritten, in response to this (and other similar) comments.*

Table 2: are times in local time? Why are some flights missing from this table? Is there a reason for the order in which flights are placed in the table? (flight 12 is listed after flight 14?)

Response: *The table header now clarifies that we refer to local time. The table contains the most relevant data from the flights that are used in the text. All flights with all segments and relevant characteristics (σ_w, $w^*$and altitude) are available in the supplementary file accompanying the manuscript. The reason flight 12 is listed after 14 is simply for aesthetic reasons.*

Figure 3: your plot sizes are inconsistent. What are the hourglass markers? You should mention these are simulated droplet numbers.
Response: *Thank you for pointing this out, all changes made. Additional information is also included in the supplement.*

Figure 4: Add units to the y axis label
Response: *Amended.*