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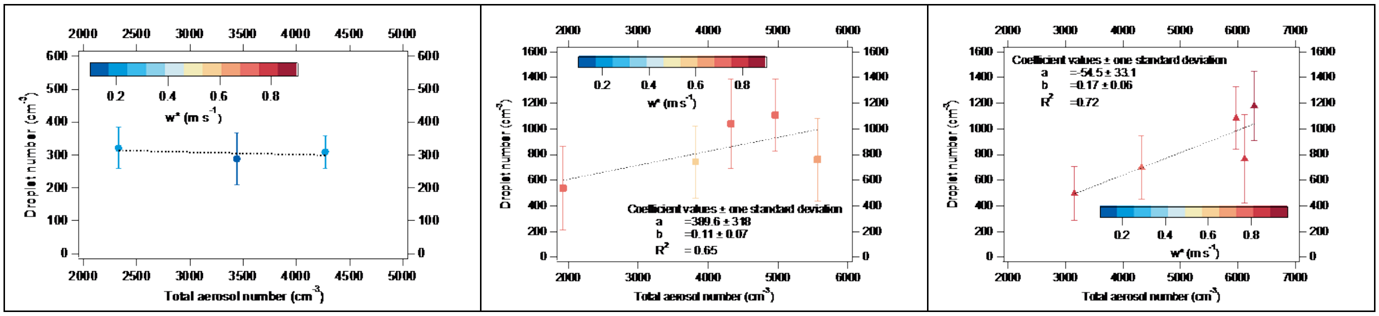
Response to Anonymous Referee #1 comments

This manuscript focuses observations of aerosol and vertical velocities over the southeast US and how it impacts predicted cloud droplet number concentration. The work uses data from the 2013 field campaign SENEX with 13 flights over the Southeast US. They find that aerosol amount and vertical velocity are responsible for up to 90% of cloud droplet number variability. They stress, early in the manuscript, that most studies do not include the impact of vertical velocity. There are some edits required, though other than that it is a fell written manuscript that will be of interest for the aerosol community.
My recommendation to accept this work with revisions and modifications to figures.
Response: *We thank the anonymous referee for the thoughtful review. Suggestions and comments for the modification of the tables and figures addressed in the revised manuscript.*

Main comments: 1) Do you have access to actual cloud data? How do the calculated N_d values compare to the calculated N_d values presented in this paper? I find it hard to believe that there were no cloud data available. Even a simple discussion about how realistic the calculated N_d values are in comparison to what was seen in in situ observations is necessary.
Response: *Unfortunately, cloud data is not available as cloud sampling was avoided (the aircraft navigating in visual flight mode most of the time). It has been shown elsewhere (e.g. Kacarab et al., 2020 and others) that our droplet number calculation methodology gives good closure with observed droplet number.*

2) For figures 5 and 6 there could be additional discussion in the manuscript. When looking at Figure 5: the first thought I had was it would be nice to see a comparison of cases when the sw* was the same and you could see how droplet number and Na were related. That seems more important than looking at a range of Na and simultaneously looking at a range of w*. Maybe a three panel figure with "Low w*," "medium w*" and "high w*" like mentioned in the text but then plot Nd and Na? Secondly, for Figure 6, Could the difference in Na with w* be due to the vertical transport? Since the w* values are higher more aerosol can be brought up from the surface.
Response: *These are great points. We have followed up on the reviewer's suggestion and splitting Figure 5 in three different graphs for low, medium and high w* values, the covariance of the total aerosol number with the vertical velocity becomes even more apparent; for low w* (during nighttime) changes in total aerosol number do not have a direct impact on calculated droplet number. On the other hand, for higher w* there is a direct correlation between total aerosol number and droplet number, which for the highest observed w* is even more accentuated, denoting*

the fact that the covariance of Na with w results in a higher variance in droplet number. Indeed, differences in Na with w* can be partially due to the entrainment of more aerosol from the surface due to higher w*, and this has been added in the revised manuscript. The respective discussion has been updated in the revised version.*



In addition, we have included a discussion on the "limiting" droplet number that develops under high aerosol number, and its dependence on σ_w (shown in Figure 6). The implications of these findings are also discussed and quite interesting.

Table Comments: 1) All the tables are ok, though it might be helpful to note daytime vs. nighttime in some way, either by shading or some type of annotation (sun and moon perhaps?)

Response: *Good suggestion! A sun and moon symbol is now added next to the number of each flight to denote whether it was a daytime or nighttime one.*

2) For Table 4: Perhaps add a mean row at the bottom for the contributions for K, Nd and sigmaw? A quick average gives k = 4.2, Nd = 75.2 and sigmaw = 13. Nd + sigmaw = 88.2. Is this where the 90% comes from that is mentioned in lines 313-314?

Response: *We added a mean row with the respective averages for dNd/Nd and the contribution of each one of kappa, chemical composition and vertical velocity to the droplet number.*

Figure Comments: 1) Figure 1: The 3D flight paths are hard to see in such a small format. Perhaps just 2D would be better, or make each panel larger. How many flights look like (a) and how many flights look like (b)? Could you include statistics about this? Otherwise it looks like your cherry picking examples.

Response: *We replaced the 3D flight paths with 2D ones, showing the values of the organics mass fraction at the different altitudes throughout the flight. All figures will be added as supplementary material and a discussion about the similarity (or not) between flights is now added in the revised manuscript. We also fixed one of the color scales that was accidentally in reverse.*

2) Figure 2: Make all the panels larger, the legends are hard to read. Why are the words and numbers together in the legends (e.g. Flight15 pass2): : : spread them out Flight 15 pass 2. Also, in the caption Line 523: you say "flights" did you mean passes? In panel (c) you have Flight 14 pass 6 and in (b) you also have Flight 14 put pass 1. Also, on panel c) consider different colors for the lines. If someone was colorblind they would not be able to tell the difference between the pink/red lines and the greenish ones.

Response: *All panels and legends are now larger and words spread within each legend. Indeed in Figure 2 we have different passes of the same flight shown in different panels; in panel (c) we have Flight 14 pass 6; in (b) Flight 14 pass 1; in panel (b) Flight 10 pass 7; in panel (c) Flight 10 pass 3; in panel (a) Flight 11 pass 6;in panel (c) Flight 11 pass 1. The transects were often made at different altitudes, thus exhibiting different characteristics each time, which were subsequently compared to other similar passes. Different colors are now used for the lines in order to make them stand out more.*

3) Figure 3: Suggestion: 4) Figure 4: same comment as in Figure 3: Add annotations to the figures to label the columns "Day" and "Night" and the rows "Alabama" and "Atlanta"

Response: *We would like to thank the reviewer for the suggestion, the specific annotation for the columns and the rows are now added to both figures (Figure 3 and 4).*

5) Figure 5: In the caption (line 547) "shading" is mentioned but is not visible in the figure. Also, the yellow marker for Flight 15 (I think) is difficult to see.

Response: *The tinted background denoting nighttime flights is now darker, thus the marker for Flight 15 is now more easily visible.*

6) Figure 6: In the caption (line 552) "shading" is mentioned but is not visible in the figure. What is the "constant altitude" that is referred to in this figure? Include the altitude somehow.

Response: *The tinted background denoting nighttime flights is now darker. As far as the constant altitude is concerned, is it clear from Table 2 that it is not the same even within each flight, let alone between flights. We do not see how it would be easy to include this information in the graph.*

Line by line comments: Line 37: Try not to use symbols in the abstract, just describe in words (it's clearer).

Response: *We have left few symbols in the abstract, because we believe it helps with conveying our message more concisely.*

Line 45: remove "the" before "incoming"

Response: *Amended*

Line 182: Specify Figure 1b here. Figure 1a does NOT show the significant decrease in organic mass fraction.

Response: *We have changed how Figure 1 is presented along with the accompanying discussion in the text.*

Line 236: how do you define what an "important contributor" is? What percentage do you consider important?

Response: *Values for σ_w during daytime flights are in the range of 0.7-1.22 with standard deviations between 0.07 and 0.31, while during nighttime flights the range of σ_w is of 0.2-0.33 and standard deviations <0.04. Therefore during a whole day the variation in σ_w values is more than a factor of 3.*

Line 242: Specify that the "first pair of flights" is for the Alabama flights.

Response: *Amended as follows:*

"The first pair of flights were conducted over a rural area under moderate aerosol number conditions…"

Line 244: Specify that the "second pair of flights" is for the Atlanta flights.

Response: *Amended*

"…while the second pair exhibited somewhat higher aerosol numbers owing to its proximity to the Atlanta metropolitan area."

Lines 253-256: The sentence that starts with "Figure 3" would be better up after "(see Fig 3.)" on line 240. It doesn't make sense where it is now.

Response: *We would like to thank the reviewer for pointing out this inconsistency. The description of Fig. 3 is now moved to L259 where Fig. 3 was introduced.*

Line 264: "characteristic", should be "characteristic,"

Response: *Section 3.2 has been rewritten, and the sentence is question no longer appears in the revised text.*

Line 313-314: How do you get the 90% number?

Response: *When adding the contribution of N_a and σ_w to the variability of the total droplet number, for each flight this added contribution is more than 90%.*

Line 319: "S.Atlantic" should be "Southeast Atlantic"

Response: *Amended*

Line 523: you say "flights" did you mean passes? In panel (c) you have Flight 14 pass

6 and in (b) you also have Flight 14 put pass 1.
Response: *Indeed so, amended*

Line 527: add “calculated” between “showing” and “cloud”
Response: *Amended*

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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:*

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

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:

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

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.*

Drivers of cloud droplet number variability in the summertime Southeast United States

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Abstract

~~The Southeast United States has experienced a different climate warming trend compared to other places worldwide. Several hypotheses have been proposed to explain this trend, one being the interaction of anthropogenic and biogenic aerosol precursors that synergistically promote aerosol formation, elevate cloud droplet concentration and induce regional cooling. We examine these aerosol-cloud droplet links by analyzing~~Here we analyze regional scale data collected onboard the NOAA WP-3D aircraft during the 2013 Southeast Nexus (SENEX) campaign to study the aerosol-cloud droplet link and quantify the sensitivity of droplet number to aerosol number, chemical composition and vertical velocity ~~on a regional scale. The~~. For this, the observed aerosol size distributions, chemical composition and vertical velocity distribution ~~(Gaussian with standard deviation σ_w)~~ are introduced into a state-of-the-art cloud droplet parameterization to show that cloud maximum supersaturations in the region are low, ranging from 0.02 to 0.52% with an average of 0.14±0.05%. Based on these low values of supersaturation, the majority of activated droplets correspond to particles of dry diameter 90 nm and above. ~~Droplet number shows little sensitivity to total aerosol owing to their strong competition for water vapor. Given, however,~~An important finding is that the ~~width of spectral dispersion~~standard deviation of the vertical velocity (σ_w) exhibits considerable diurnal variability (ranging from 0.16 m s^{-1} during nighttime to over 1.2 m s^{-1} during day~~), its covariance~~) and it

tends to covary with total aerosol number (N_a~~) during the same period~~). This σ_w-N_a covariance amplifies the predicted response in cloud droplet number (N_d) to N_a increases by 3 to 5 times~~. Therefore, correct~~ - which is important, given that droplet formation is often velocity-limited, and therefore should normally be insensitive to aerosol changes. We also find that N_d cannot exceed a characteristic concentration that depends solely on σ_w. Correct consideration of ~~vertical velocity~~σ_w and its covariance with time and ~~aerosol amount~~N_a is important for fully understanding aerosol-cloud interactions and the magnitude of the aerosol indirect effect. ~~Datasets and analysis such as the one presented here can provide the required constraints for addressing this important problem.~~Given that model assessments of aerosol–cloud-climate interactions do not routinely evaluate for overall turbulence or its covariance with other parameters, datasets and analyses such as the one presented here are of the highest priority to address unresolved sources of hydrometeor variability, bias, and the response of droplet number to aerosol perturbations.

1. Introduction

Atmospheric particles (aerosols) interact with incoming solar radiation ~~through scattering and absorption processes which~~and tend to cool the Earth, especially over dark surfaces such as oceans and forests (~~Brock~~Charlson et al., ~~2016a).~~1992; Seinfeld and Pandis, 1998). Aerosols also act as cloud condensation nuclei (CCN~~) and subsequently~~), form ~~cloud~~ droplets in clouds and indirectly affect climate ~~through modification of~~by modulating precipitation patterns and cloud radiative properties~~ – an effect which constitutes one of~~. Aerosol-cloud interactions constitute the most uncertain aspects of anthropogenic climate change (Seinfeld et al., 2016). Studies often highlight the importance of constraining the aerosol size distribution, particle composition and mixing state for predicting CCN concentrations (Cubison et al., 2008; Quinn et al., 2008~~).~~; Riemer et al., 2019). Model assumptions often cannot consider the full complexity required to comprehensively compute CCN – which together with other emissions and process uncertainties lead to CCN prediction errors that can be significant (e.g., Fanourgakis et al., 2019). Owing to the sublinear response of cloud droplet number concentration (N_d) to aerosol perturbations, prediction errors in CCN generally result in errors in N_d which are less than those for CCN (Fanourgakis et al., 2019). The sublinear response arises because elevated CCN concentration generally increases the competition of the potential droplets for water vapor; this in turn depletes supersaturation and the N_d that can eventually form (Reutter et al., 2009; Bougiatioti et al., 2016; Fanourgakis et al., 2019; Kalkavouras et al., 2019). A critically important parameter is the vertical velocity~~; so important in fact~~, as it is responsible for generation of supersaturation that drives droplet formation and growth. Droplet number variability may be driven primarily by vertical velocity variations (Kacarab et al., 2020; Sullivan et al., 2019). Compared to aerosols, vertical velocity is much less observed, constrained and evaluated in aerosol-cloud interaction studies, hence may be a source of persistent biases in models (Sullivan et al., 2019).

The Southeast United States (SEUS) presents a particularly interesting location for studying regional climate change, as it has not considerably warmed over the past 100 years – except ~~for~~during the last decade (Carlton et al., 2018; Yu et al., 2014; Leibensperger et al., ~~2012b~~2012a,b). These trends are in contrast with the trends observed in most locations globally (IPCC 2013), and several hypotheses have been proposed to explain this regional phenomenon, including the effect of involving short-lived climate forcers such as secondary aerosols combined with the enhanced humidity in the region and their impact on clouds (Carlton et al., 2018; Yu et al., 2014). Here, we analyze data collected during the Southeast Nexus of Air Quality and Climate (SENEX) campaign in June-July 2013, which was the airborne component led by the National Oceanic and Atmospheric Administration (NOAA), of a greater measurement campaign throughout the SEUS, the Southeast Atmosphere Study (SAS; Carlton et al., 2018). Here we analyze data collected onboard the NOAA WP-3D and apply a state-of-the-art droplet parameterization to determine the maximum supersaturation and N_d achieved in cloudy updrafts, for all science flights with available number size distribution and chemical composition data. We also determine the sensitivity of droplet formation to vertical velocity and aerosol, with the purpose of understanding the drivers of droplet variability in the boundary layer of the SEUS by obtaining regional-scale, representative values of the relationship between the driving parameters and cloud droplet number.

2. Methods

2.1 Aircraft instrumentation

The analysis utilizes airborne, in situ data collected during the June-July 2013 SENEX mission, aboard the National Oceanic and Atmospheric Administration (NOAA) WP-3D aircraft (typical airspeed ~100 m s^{-1}) based in Smyrna, Tennessee (36^{o}00’32’’N, 86^{o}31’12’’W). In total, twenty research flights were conducted. Based on the availability of the relevant data described below, thirteen flights are analyzed in this work. ~~Description~~Table 1 provides a synopsis of the analyzed research flights where times are ~~provided in Table 1.~~local (UTC-5). Detailed information on the instrumentation and measurement strategy during the SENEX campaign ~~can be found in~~is provided by Warneke et al. (2016).

Dry particle number distributions from 4 - 7000 nm were measured using multiple condensation and optical particle counters. 4-700 nm particles were measured by a nucleation mode aerosol size spectrometer (NMASS; Warneke et al., 2016) and an ultra-high sensitivity aerosol spectrometer (~~UHSA~~UHSAS; Brock et al., 2011), while for larger particles with dry diameters between 0.7 and 7.0 µm, a custom-built white-light optical particle counter (WLOPC) was used~~.~~ (Brock et al., 2011).

Measurements of the composition of submicron (< 0.7 µm vacuum aerodynamic diameter) non-refractory ~~aerosol (less than 0.7 µm diameter)~~particles were made with a ~~Compact Time~~compact time-of-~~Flight Aerosol Mass Spectrometer~~flight aerosol mass spectrometer (C-ToF-AMS; Aerodyne, Billerica, Massachesetts, US) (Canagaratna et al., 2007; Kupc et al., 2018) customized for aircraft use, with a 10 s time resolution (Warneke et al., 2016). Particles entering the instrument are focused and impacted on a 600 ^{o}C inverted-cone vaporizer. The volatilized vapors are analyzed by electron ionization mass spectrometry, providing mass loadings of sulfate, nitrate, organics, ammonium and chloride. For the C-ToF-AMS, the transmission efficiency of particles between 100 and 700 nm is assumed to be 100% through the specific aerodynamic focusing lens used while mass concentrations are calculated using a chemical composition-dependent collection efficiency (Middlebrook et al., 2012; Wagner et al., 2015). The C-ToF-AMS ~~only~~ measures only non-refractory aerosol chemical composition, therefore this analysis provides mass loadings of sulfate, nitrate, ammonium and organic constituents with a 10 s time resolution and neglects the contribution of black carbon (BC). The calculation of the average volume fractions from the mass loading follows that of Moore et al. (2012). An average organic density of 1.4 g cm^{-3} is used, characteristic of aged aerosol (Moore et al., 2011; Lathem et al., 2013) while for the inorganic species the respective densities are used, assuming the aerosol to be internally mixed.

The aircraft was equipped by the NOAA Aircraft Operations Center (AOC) flight facility~~, incorporating~~ with a suite of instruments to provide information on exact aircraft position as well as numerous meteorological parameters (Warneke et al., 2016). The analysis in this work makes use of vertical wind velocity, aircraft ~~radar~~pressure altitude, and ambient temperature, pressure and relative humidity (RH) provided by NOAA AOC. ~~Location~~The location of the instrumentation on the aircraft ~~can be found~~is described elsewhere (Warneke et al., 2016). For measurements inside the fuselage, a low turbulence inlet (Wilson et al., 2004) and sampling system (Brock et al., 2011; 2016a) was used to decelerate the sample flow to the instruments. The C-ToF-AMS was connected downstream of an impactor with 50% efficiency at a 1.0 µm aerodynamic diameter (PM1) cut-point (Warneke et al., 2016).

2.2 Aerosol hygroscopicity parameter

The aerosol hygroscopicity parameter (Petters and Kreidenweis, 2007), κ, is calculated assuming a mixture of an organic and inorganic component with volume ~~fraction~~fractions ε_{org}, ε_{inorg} and characteristic ~~hygroscopicity~~hygroscopicities κ_{org}, κ_{inorg}, respectively ($\kappa=\varepsilon_{inorg}\kappa_{inorg}+\varepsilon_{org}\kappa_{org}$). The organic and inorganic volume fraction are derived from the C-ToF-AMS data. Since throughout the summertime SEUS, aerosol inorganic nitrate mass and volume fraction are very low (Weber et al., 2016; Fry et al., 2018), κ_{inorg} =0.6, representative for ammonium sulfate, is used. For the organic fraction, a hygroscopicity value of κ_{org}=0.14 is used, based on concurrent measurements conducted at the ground site of the SAS at the rural site of Centreville, Alabama (Cerully et al., 2015). This value is also in accordance with the cumulative result of studies conducted in the Southeast US using measurements of droplet activation diameters in subsaturated regimes, providing κ_{org} of > 0.1 (Brock et al., 2016a).

2.3 Cloud droplet number and maximum supersaturation

Using the observed aerosol number size distribution (1 s time resolution~~) and~~), the hygroscopicity derived from the chemical composition measurements (10 s time resolution~~),~~) and vertical velocity, we calculate the (potential) cloud droplet number (N_d) and maximum supersaturation (S_{max}) that would form in clouds in the airmasses sampled. Droplet number and maximum supersaturation calculations are carried out ~~at a regional scale~~ using an approach similar to that of Bougiatioti et al. (2016) and Kalkavouras et al. (2019) with the sectional parameterization of Nenes and Seinfeld (2003), later improved by Barahona et al. (2010) and Morales Betancourt and Nenes (2014a). A sectional representation of the size distribution is used ~~and provided~~ for each 1-s data point ~~of each flight (per second, (~~(e.g. for Flight 5, n=23213 data points). Given that chemical composition is provided with a 10 s time resolution, the same hygroscopicity values are used for 10 successive size distributions ~~during each~~throughout the flight. Temperature and pressure required for droplet number calculations are obtained from the NOAA AOC flight facility dataset.

~~Droplets form from activation of aerosol in cloudy updrafts, so here we use the available measurements of vertical velocity together with the aerosol measurements to derive a potential cloud droplet concentration.~~ Given that vertical velocity varies considerably inside the boundary layer, we ~~represent~~obtain a droplet number ~~with~~that is representative of the vertical velocity distribution – the average concentration that results from integrating over the distribution (probability density function, PDF) of observed updraft velocities. To accomplish this, each flight is divided ~~in~~into segments where the aircraft flew at a constant height. For each segment, the ~~positive~~non-negative vertical velocities are fit to the positive half of a Gaussian distribution with mean of zero and ~~width of spectral dispersion~~standard deviation σ_w. ~~Positive~~Only positive vertical velocities ("updrafts") were used in this fit, as they are the part of the vertical velocity spectrum that is responsible for cloud droplet formation. The σ_w values derived from the level leg segments are then averaged into one single σ_w value (and standard deviation) to represent ~~the~~each flight. ~~Application of~~The PDF-averaged droplet number concentration is then obtained using the "characteristic velocity" approach (of Morales and Nenes~~,~~ (2010~~) then gives the PDF-averaged droplet number concentration by calling~~), where applying the droplet parameterization at a single "characteristic" velocity, $w^*=0.79\sigma_w$ (Morales and Nenes, 2010~~). This calculation approach~~) gives directly the PDF-averaged value. The flight-averaged σ_w

and subsequently the respective w^* is applied to each size distribution measured. Apart from its theoretical basis, this methodology has shown to provide good closure with observed droplet numbers in ambient clouds (e.g. Kacarab et al., 2020).

In determining σ_w, we consider horizontal segments ~~that are expected~~most likely to be in the boundary layer~~:~~. 91~~-~~% of the segments are below 1000 m above sea level (mean altitude ~700 m; Table 2~~) typically corresponding to the height of~~ and SP3 for all flights), within the boundary layer in the summertime US (Seidel et al., 2013). 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~~). Because of this strong diurnal variation in σ_w, potential droplet formation is evaluated at four vertical velocities that cover the observed ranged, namely 0.1, 0.3, 0.6 and 1 m s^{-1}.~~ and SP3).

Potential droplet formation is evaluated at four characteristic velocities w^* that cover the observed range in σ_w, namely 0.1, 0.3, 0.6, and 1 m s^{-1}. The σ_w = 0.3 m s^{-1} case is most representative of nighttime conditions, while σ_w = 1 m s^{-1} should is most representative of the daytime boundary layer.

We also compute the ~~sensitivity~~variance of the derived N_d, estimated from the sensitivity to changes in aerosol number concentration ~~(N_a)~~, κ and σ_w, expressed by the partial derivatives $\partial N_d/\partial N_a$, $\partial N_d/\partial \kappa$ and $\partial N_d/\partial \sigma_w$ computed from the parameterization using a finite difference approximation (Bougiatioti et al., ~~2016~~2017; Kalkavouras et al., 2019~~).~~) using:

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

These sensitivities, together with the observed variance in N_a, κ, and σ_w are also used to attribute droplet number variability to variations in the respective aerosol and vertical velocity parameters following the approach of Bougiatioti et al. (~~2016~~2017) and Kalkavouras et al. (2019~~).~~):

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

3. Results and Discussion

3.1. Particle composition and size distribution

For the determination of the different aerosol species present, neutral and acidic sulfate ~~species~~salts are distinguished by the molar ratio of ammonium to sulfate ions in the aerosol. A molar ratio higher than 2 indicates the presence of only ammonium sulfate, while values between 1 and 2 indicate ~~the presence~~a mixture of ~~both~~ ammonium sulfate and bisulfate (Seinfeld and Pandis, 1998). For most ~~of the~~ flights, the molar ratio of ammonium versus sulfate was ~~well~~ above 2~~, having a~~ (mean value ~~of~~ 2.41±0.72 ~~(~~and median of 2.06). For the nighttime flights, the values were somewhat lower (mean value 1.91±0.42 and median of 1.85~~, respectively~~). Nevertheless, ammonium sulfate is always the predominant sulfate salt. Organic mass fractions for the SENEX research flights are provided in Table 1. Overall, organic aerosol ~~was found to dominate~~dominated the composition during all flights, contributing 66%-75% of the total aerosol volume. Most of the remaining aerosol volume ~~consists~~consisted of ammonium sulfate, ranging from 12%-39% (with a mean of 23%±6%). The organic mass fraction during the flights ~~was found to vary~~varied with height (see ~~Fig.~~Figure 1). This vertical variability of the chemical composition can have a strong impact on droplet number within the boundary layer, as air masses from aloft may descend and interact with that underneath. Figure 1 represents the organic mass fractions during Flights 6, 12 and 16, with all flights provided in the supplementary material (~~Fig.~~Figure S1). The lowest organic mass fractions overall were observed during Flight 12 (36%±10% with values almost two-fold higher for altitudes >3000 m, Fig. 1b) while the highest ~~ones~~organic mass fractions were observed ~~at~~during flights over predominantly rural areas (Flights 5, 10, and 16 (Fig. 1c)). During Flight 5 the organic mass fraction was high (68%±5%), with the highest values found in the free troposphere at altitudes >~~-~~3000 m, as was the case for 4 other ~~4~~ flights (5/13 in total, Fig. S1). High organic mass fractions were also found during ~~a~~ nighttime ~~flight~~Flight 9 that included portions of the Atlanta metropolitan area, with values up to 78%. The impact of the ~~chemical~~aerosol composition variability on droplet number is discussed in section 3.2.

The predominance of the organic fraction is also reflected in the hygroscopicity parameter values, with an overall κ = 0.25±0.05, ~~which is~~ close to the proposed global average of 0.3 (~~Seinfeld and Pandis, 1998).~~Pringle et al., 2010). The highest values ~~are~~of κ, as expected~~, for~~ are observed during flights exhibiting the lowest organic mass fraction, namely Flight 12 with a κ = 0.39 (Table ~~2~~1). The rest of the κ-values are close to the overall ~~values, as the~~value of 0.25, corresponding to an organic mass ~~fractions are~~fraction of around 0.~~65~~60.

Median aerosol size distributions and the respective total aerosol number are obtained from the median and interquartile range in each size bin from the aerosol size distribution measurements during segments where the aircraft flew at a constant height. ~~The impact of~~Aerosol size distributions and changes in them during each flight are crucial as they are used as input for the ~~variability of the total aerosol number on~~ droplet number ~~is discussed in detail further in section 3.2.~~parameterization. Overall, ~~number concentrations~~N_a ranged from around 500 to over 100000 cm^{-3} with number size distributions varying markedly over the course of a flight~~. In general, free~~ (Figure 2). Free tropospheric distributions exhibited characteristics of a bimodal distribution with a prominent broad accumulation mode peak (80-200 nm) and an Aitken mode peak (30-60 nm) (Fig. 2a) while boundary layer size distributions exhibited a more prominent accumulation mode (Fig. 2b). There was considerable variability in the contributions of the nucleation, Aitken, and accumulation modes to ~~the~~ total ~~aerosol number~~N_a, depending on altitude and proximity to aerosol sources (Fig. 2c). Nevertheless, the modal diameters did not vary ~~much.~~considerably, dictating that mostly particles in the same mode will activate, depending on the developed supersaturation. Distributions during nighttime flights exhibited similar ~~total aerosol number~~N_a and variability between them; nevertheless, size distributions were more complex - exhibiting ~~even~~up to three ~~different~~distinct modes (20-40, 70-100 and 130-200 nm; Fig. 2d). ~~Considering that mostly particles in the accumulation mode activate into cloud droplets (particles with diameters >90 nm), contrasts between day and nighttime aerosol characteristics/variability may not be as large, and driven primarily by the total aerosol number in the accumulation mode.~~

3.2 Potential cloud droplet number and maximum supersaturation

~~The~~ We first focus on calculation of the potential N_d and S_{max}~~, was carried out~~ for data from all thirteen research flights~~.~~ and for the four prescribed values σ_w that represent the observed range. These calculations help understand the sensitivity of droplet formation to N_a and κ for all the airmasses sampled – without considering the added variability induced by changes in turbulence expressed by σ_w (considered later). Results from this analysis are ~~given in Tab. 3 for the four different values (0.1, 0.3, 0.6 and 1 m s^{-1}) of σ_w. Overall it can be seen that for all flight~~provided in Table 3. The highest N_d were found for Flights 6 and 10, which correspond to ambient conditions ~~and for low σ_w, N_d shows a low variance (mean of 132±20 for 0.1 m s^{-1} and 350±100 for 0.3 m s^{-1}).~~with the highest N_a, consistent with the sampling of the Atlanta urban environment. For a given σ_w, the variance of N_d is predominantly ~~attributed to relative~~caused by changes in N_a rather than changes in ~~the~~ hygroscopicity (i.e., chemical composition~~ (expressed by changes in the hygroscopicity parameter, κ). The highest relative contribution of the chemical composition~~). The highest influence of κ to N_d variability is found for Flight 18 (12% and 35% for 0.1 and 0.3 m s^{-1}, respectively~~) to the variation of N_d is found for Flight 18,~~), during which ~~the total aerosol number was the lowest. Indicative of a "cleaner" environment;~~ N_a was the low, and the organic mass fraction was ~~relatively lower and the hygroscopicity parameter was higher. Even though the lowest organics mass fraction and highest κ were observed during Flight 12, droplet formation~~ ~50%. The contribution of κ to the N_d variability is ~~much~~ as high as 37% (for 0.6 m s^{-1}); despite this large contribution, droplet formation is usually considerably more sensitive to changes in aerosol concentration than to variations in composition. Overall, the relative contribution of the hygroscopicity to the variation of N_d increases from 5±3% for σ_w =0.1 m s^{-1}, to 12.3±8% for σ_w =0.3 m s^{-1}, to 14.5±10% for σ_w =0.3 m s^{-1} and 16.5±9% for σ_w =1 m s^{-1}.

As ~~the vertical velocity~~ σ_w increases, so does supersaturation and consequently ~~the droplet number (~~N_d. On average, N_d increases by 62% as σ_w increased from 0.1 to 0.3 m s^{-1}, 70% as σ_w increased from 0.3 to 0.6 m s^{-1} and another 39% ~~from 0.6 to 1 m s^{-1}). The relative contribution of the chemical composition to the~~

~~variation of cloud droplet number increases from 5±3% for 0.1 m s^{-1}, to 12.3±8% for 0.3 m s^{-1}, to 14.5±10% for 0.3 m s^{-1} and 16.5±9% for 1 m s^{-1}. The highest droplet numbers are estimated for Flights 6 and 10, which included urban environments during daytime (Atlanta). Overall during daytime, when σ_w varies little and is large, and N_a is high, the relative contribution of N_a to the variation of N_d is the highest (more than 90%) while the relative contribution of κ is limited (less than 10%) (see Table 3, Flights 10, 11, 12, 17 and 19). Turbulence is limited during nighttime when σ_w is the lowest (0.23±0.04); therefore, the σ_w = 0.3 m s^{-1} case is most representative of nighttime conditions. During daytime, when σ_w is high (0.97±0.21), σ_w = 1 m s^{-1} should be considered as most representative.~~as σ_w increased from 0.6 to 1 m s^{-1}. Tripling σ_w from 0.1 to 0.3 m s^{-1} results in 31% increase in S_{max}, while doubling σ_w from 0.3 to 0.6 m s^{-1} results in 26.2% increase in S_{max} and a further σ_w increase to 1 m s^{-1} leads to an additional 20.7% increase in S_{max}.

~~As σ_w varies considerably throughout the day (more than a factor of 3), we estimate its contribution together with variations in N_a and κ, to the total variability in N_d based on $\partial N_d/\partial\kappa$, $\partial N_d/\partial N_a$ and $\partial N_d/\partial\sigma_w$ and the variances of κ, N_a and σ_w (Table 4).~~Considering the changes in vertical velocity between flights (Table 4), we observe that average σ_w during daytime varied little between flights and was large, ranging between 0.85 and 1.2 m s^{-1} with a mean of 0.97±0.21 m s^{-1}. Under such conditions, water availability during droplet formation is aerosol-limited, so that N_d is sensitive to N_a. The degree of water availability is expressed by the S_{max}, which for all the evaluated SENEX data, is 0.14±0.05%. This level of maximum supersaturation activates particles of roughly >90 nm diameter (e.g., accumulation mode particles) into cloud droplets. The highest S_{max} ranged from 0.2 to 0.3% and was found during flights which exhibited large and highly variable σ_w (Flights 4, 5, 12 and 19) while the lowest S_{max} was below 0.10% and was found during nighttime flights (Flights 9, 15 and 16). Contrasts in droplet formation between day and nighttime conditions may be driven primarily by the total aerosol number in the accumulation mode, and not be affected by ultrafine particles.

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. To understand the relative importance of all parameters affecting droplet formation (σ_w, N_a, κ) 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. The results of the analysis is summarized in Table 4. The σ_w variation during nighttime, although small (always less than 10%), consistently remains an important contributor to N_d variability, because droplet formation tends to be in the updraft velocity-limited regime. At higher values of σ_w (Table 4), the contribution of ~~σ_w~~N_a variability becomes a relatively dominant contributor to N_d variability~~ is reduced and dominated by N_a variability.~~ .

~~To explore~~Another way to express the importance of vertical velocity and aerosol ~~compared~~number for the levels of droplet number is to ~~updraft velocity~~compare flights where aerosol number or σ_w vary in a similar way. For this, we focus on two day/night flight pairs ~~of flights~~ (Flights 5 and 15, and Flights 6 and 9), shown in Fig. 3. The first pair of flights were conducted ~~in two sectors (Alabama and~~ over a rural area under moderate aerosol number conditions, while the second pair exhibited somewhat higher aerosol numbers owing to its proximity to the Atlanta ~~regions), from each sector one during day- and one during night-time (see Fig.3), where the N_d were calculated using the observed σ_w.~~metropolitan area. The size of the markers in Fig.3 represents the potential number of droplets in clouds forming in each airmass sampled, while ~~the~~their color ~~scale~~reflects the respective total aerosol number. In both pairs of flights ~~(Flight 5 and 15, and Flight 6 and 9), ,~~ σ_w varies about the same between night and day (Table 4). For the ~~first~~Flights 5, 15 pair ~~of flights (Alabama),~~, the ~~daytime variability~~difference in N_d between day and night (which is 69~~%)~~% higher during daytime) is ~~to within 75%~~ driven primarily by aerosol characteristics (69% by N_a and 7% from κ) and only 24% by σ_w. For nighttime~~,~~ (Flight 15), the variability in N_d is driven 58% by aerosol (51% by N_a and 7% from κ) and 42% ~~of the variability is driven by~~ by σ_w. For the second pair of night/day flights (~~Atlanta~~Flights 6, 9), N_a is on average similar, σ_w varies by a factor of 4.0 between day and night and κ varies by 13%. ~~Attribution calculations suggest that the diurnal variability~~ The difference in N_d ~~(~~between day and night (where daytime values are 72.1% higher than nighttime) is ~~3, 54 and 43% for κ, N_a and σ_w, respectively during day and 7, 76, and 17%~~almost equally driven by ~~κ,~~ N_a and σ_w~~,~~ changes during the day (54% and 43% respectively~~ during night (Table 4). In the second sector, 57% of the variability in N_d is~~), while predominantly driven by ~~aerosol~~N_a during the night (76%; Table 4). Overall, in the proximity of an urban environment with higher aerosol concentrations, 57% of the N_d variability is driven by aerosol (N_a and κ) during the day and 83% during the night. Figure 3 presents the calculated N_d for the four aforementioned flights, namely Flights 5 (Fig. 3a), 15 (Fig. 3b), 6 (Fig. 3c) and 9 (Fig. 3d) using the observed σ_w. The size of the markers represents the estimated number of droplets, while the color scale the respective total aerosol number. Droplet number is lower during nighttime owing to the limited turbulence, i.e., lower σ_w.

~~As expected, droplet number (N_d) and maximum supersaturation (S_{max}) increases as σ_w becomes larger. The highest S_{max} are around 0.2-0.3% and found for flights which exhibited large and highly variable σ_w (Flights 4, 5, 12 and 19) while the lowest S_{max} are around 0.10% and found for the nighttime flights (Flights 9, 15 and 16). All other flights yield similar S_{max}, which are around 0.13%. Based on the calculated S_{max} for every flight, the majority of the activated droplets correspond to particles of 90 nm diameter and above.~~

~~Figure 4 shows N_d relative to N_a for flights conducted in two sectors, during day and night (Flights 5 & 15, and Flights 6 & 9, respectively). It can be seen that throughout these flights, N_d reaches a plateau, where any additional aerosol does not translate to any significant increase in N_d. This plateau is caused by strong water vapor limitations and is different for day and night. S_{max} is lower during night because vertical wind velocity, ambient T and RH are lower. The same factors cause that for Flight 6 & 9 (Fig. 4c & d) where N_a~~

~~was almost the same, N_d is almost 3.5 times lower during night (Flight 9). For the whole dataset (13 flights), results are summarized in Figure 5, where droplet numbers are calculated based on the observed σ_w and the respective "characteristic", mean velocities, w^*. Under low w^* conditions, N_a variability does not result in an important change in N_d. On the contrary, when w^* tends to increase and N_a increases, as is characteristic of polluted regions, during daytime, then the impact on droplet number is more notable. This point is evident in Figure 6, comprising the different segments of the flights when the aircraft sampled at practically the same altitude within the boundary layer. It can be seen that N_a is enhanced as w^* increases. The lowest w^* values (shaded area) correspond to the segments of the flights during nighttime.~~

~~Overall, S_{max} of clouds from all the evaluated SENEX data, is 0.14±0.05%. Tripling σ_w from 0.1 to 0.3 m s^{-1} results in 31% increase in S_{max}, while doubling from 0.3 to 0.6 m s^{-1} results in 26.2% increase in S_{max} and a further σ_w increase to 1 m s^{-1} leads to an additional 20.7% increase in S_{max}. Overall effect of updraft velocity on calculated N_d: tripling σ_w from 0.1 to 0.3 m s^{-1} results in a 61.9% increase in N_d, doubling from 0.3 to 0.6 m s^{-1} results in a 40.5% N_d increase; increasing σ_w to 1 m s^{-1} leads to an additional 26.9% increase in N_d. Furthermore, for a given σ_w, despite of the presence or not of a large number of aerosol (e.g. Flight 10 where N_a is 2.7 times higher than N_a in Flight 15) the difference in calculated N_d for 0.6 m s^{-1} is only 1.3 times higher for Flight 10 than Flight 15. This highlights the relative insensitivity of N_d to variations in N_a for constant σ_w.~~

Figure 4 shows N_d relative to N_a for flights conducted in the two aforementioned areas, during day (Flights 5 and 15) and night (Flights 6 and 9). For high enough N_a, N_d becomes insensitive to additional amounts of aerosol and reaches a "limiting" N_d, which Kacarab et al. (2020) denotes as N_d^{lim}. This limit in N_d is reached when the competition for water vapor to form droplets is strong enough to inhibit the formation of droplets with further increase in N_a. The intense competition for water vapor is reflected in the low value of S_{max}, which drops below 0.1% when N_d is in the vicinity of N_d^{lim} (Figure 4). The availability of water vapor during droplet formation is driven by σ_w, hence droplet formation is limited by σ_w and thus by velocity, when N_d approaches N_d^{lim}. Figure 5 illustrates these effects, by presenting the relationship between N_a and N_d for "low" w^* (<0.25 ms^{-1}; upper panel), "medium" w^* (0.5-0.7 ms^{-1}; middle panel), and "high" w^* (0.75-1 ms^{-1}; lower panel) for all flights. Under low w^* conditions, changes in N_a does not result in an important change in N_d, so its value corresponds to N_{lim}. When w^* increases to "medium" values (Figure 5b), then N_d becomes sensitive to N_a, which is further amplified at "high" values of w^* (Figure 5c). The covariance of aerosol number and vertical velocity (Figure S3) means that the latter significantly enhances the inherent response of N_d to N_a, which points to the importance of constraining vertical velocity and its variance to correctly capture the aerosol-cloud droplet relationship in any model. The covariance, also observed in other environments (e.g., Kacarab et al., 2020), may result from a more effective convective transfer of aerosol-rich air to cloud forming regions, but requires further investigation.

Analysis of Figure 4 also shows that N_d^{lim} varies between 1200 cm^{-3} during day and around 350 cm^{-3} during night, which points to its strong dependence on σ_w. Indeed, when the N_d^{lim} for all flights (except Flights 4, 12, for which insufficient aerosol is present to reach N_d^{lim}) is expressed as a function of σ_w, a remarkable correlation emerges between the two parameters (Figure 6). Even more interesting is that this relationship is quantitatively similar to the corresponding N_d^{lim} - σ_w relationship Kacarab et al. (2020) found for biomass burning - influenced boundary layer clouds in the Southeast Atlantic. The implication of the N_d^{lim} - σ_w relationship, and its potential universality, is that when N_d approaches N_d^{lim}, its variability is a reflection of vertical velocity variability alone, not variability in N_a. This opens up the possibility to infer the vertical velocity distribution from the droplet number concentration retrievals in regions where considerable amounts of aerosol are present.

4. Summary and Conclusions

Measurements of vertical wind velocity, ambient ~~conditions (*T, RH*),~~temperature, humidity, aerosol number size distribution and composition in the SEUS obtained during the SENEX 2013 project are used to analyze the drivers of droplet formation. Overall, 13 research flights are studied, covering environments over sectors with different aerosol sources, ~~impacting total~~ aerosol number, size distribution~~ and~~, chemical composition~~.~~ and updraft velocity. Aerosol volume is largely dominated by an organic fraction resulting in ~~a calculated~~an estimated hygroscopicity of 0.25±0.05.

Based on the calculation of cloud droplet number concentration (N_d) and maximum supersaturation (S_{max}), we find that ~~on a~~at the regional scale, ~~most of the~~N_d variability ~~of N_d~~ is ~~due to the~~ largely driven by fluctuations in N_a (Table 4), in accordance with other recent studies (e.g., Fanourgakis et al., 2019~~).~~; Kalkavouras et al., 2019; Kacarab et al., 2020). Nonetheless, N_d levels are also sensitive to ~~fluctuations in~~vertical velocity variations, σ_w~~, as a variation by~~; a factor of 4.0 change in σ_w on its own may lead to an ~~N_d variation of~~ almost ~~a~~ proportional change in N_d (factor of 3.6~~ and at~~). These responses however occur over the ~~same time~~diurnal timescale, during which N_a also changes; the ~~N_d~~ covariance between σ_w with N_a enhances the apparent response of N_d to ~~different~~changes in N_a levels ~~may be enhanced~~ by a factor of 5 (Figure 4). ~~S_{max} changes in response to aerosol concentration, in a way that tends to partially mitigate N_d responses to aerosol. Overall, maximum supersaturation levels remain quite low (0.14±0.05%) with predicted levels being much lower in lower altitudes (0.05±0.1%). Because of the strong competition for water vapor (expressed by the low S_{max}), cloud droplet number exhibits enhanced sensitivity to aerosol number variations throughout the flights, regardless of aerosol composition. On the other hand, droplet~~

~~concentration especially within the boundary layer approaches a "plateau" that is strongly driven by vertical velocity (turbulence) and the resulting supersaturation, but also aerosol concentration.~~ In "cleaner" environments where total aerosol number is ~~lower~~not impacted by local sources, the relative ~~contribution~~response of ~~vertical velocity~~N_d to ~~cloud droplet number~~σ_w is almost ~~half during nighttime (24% vs. 42%~~ twice as great at night than during the day (24% for daytime~~) while~~ Flight 5 vs. 42% for nighttime Flight 15). On the other hand, the relative ~~contribution of N_a to the variance in~~ response of N_d to N_a is ~~somewhat higher (69% vs. 51%~~slightly lower during ~~daytime) even though N_a is 2-fold lower~~the night than during ~~night. On the contrary, in~~the day (51% at night vs. 69% during the day). In environments with elevated concentrations of accumulation-mode particles, the majority of ~~cloud droplet number~~N_d variations ~~(54% during nighttime vs. 76% during daytime)~~ can be attributed to changes in ~~total aerosol number~~N_a and to a lesser extent to ~~vertical velocity (43% during nighttime vs. 17% during daytime). The relative contribution of the total aerosol number to the cloud droplet number dominates over variations~~changes in σ_w. Variations in chemical composition (expressed by κ~~). There are cases however where chemical composition~~) do not contribute substantially to droplet number variability ~~contributes a non-negligible (~9%) contribution to droplet number variability.~~in most cases. As expected, S_{max} partially mitigates the response of N_d to N_a. Overall, maximum supersaturation levels remain quite low (0.14±0.05%) with the lowest levels (0.05±0.1%) estimated closest to surface. As a result, particles with diameters >90 nm were the most substantial contributors to CCN.

~~Overall, our results show that atmospheric dynamics is~~Our analysis also reveals the importance of the variance in vertical velocity as a key driver of cloud droplet formation and its variability in the region. ~~Especially in cases when~~When the boundary layer turbulence is low (e.g. during nighttime~~),~~) and water vapor supersaturations are low~~ vertical velocity, generating only small supersaturation~~, σ_w and its variability, can be as important a contributor to ~~droplet number variability as aerosol number. For cases with high vertical velocities and high aerosol number concentration, it is the aerosol concentration that dominates the variability in cloud droplet number.~~N_d as is N_a. On average, the two variables (N_a and σ_w) contribute almost equally to the variability in ~~cloud droplet number concentration (N_d) and together account~~N_d, accounting for more than 90% of the variability. This finding is consistent with recent modeling studies noting the importance of vertical velocity variability as a driver of the temporal variability of global hydrometeor concentration (Morales Betancourt and Nenes, 2014b; Sullivan et al., 2016). Furthermore, the N_d ~~enhancement~~response from changes in N_a is magnified up to 5 times ~~from concurrent~~by correlated changes in σ_w. A similar situation ~~has also been observed~~ was seen in ~~smoke-influenced~~ marine boundary layers influenced by biomass burning in the Southeast Atlantic (Kacarab et al., 2020). ~~Altogether, these findings carry~~Finally, we identify an upper limit to the number of droplets that can form in clouds which depends only on σ_w. This upper limit value tends to be achieved near the surface, where N_a tends to be higher. Whenever N_d approaches this upper limit, observed droplet variability is driven by σ_w and as a consequence by vertical velocity changes only.

Many aspects of warm cloud physics and especially droplet formation are known for decades. Ensuring that global models simulate N_d for the "right reasons" (i.e., aerosol variability and/or vertical velocity variability) is critical for constraining aerosol-cloud-climate interactions. Our study provides important ~~implications for~~ constraints on the relationships between σ_w, N_a, potential N_d, and S_{max}, and shows the importance of covariance between σ_w and N_a in controlling the N_d that can result from a given value of σ_w. Given that global model assessments of aerosol ~~indirect climate forcing (e.g., Leibensperger et al., 2012a) and aerosol-cloud interaction studies using remote sensing, as patterns of cooling (although consistent~~ cloud-climate interactions do not evaluate for vertical velocity or its covariance with ~~aerosol and cloud fields) may omit the covariance of vertical velocity with aerosol number, therefore neglecting this important driver~~other parameters, our work shows that this omission can lead to an underappreciated source of hydrometeor variability~~.~~ and bias, and to a biased response and attribution of droplet number to aerosol levels.

Data Availability: The data used in this study can be downloaded from the NOAA public data repository at https://www.esrl.noaa.gov/csd/projects/senex/. The Gaussian fits used for determining σ_w and the droplet parameterization used for the calculations in the study are available from athanasios.nenes@epfl.ch upon request.

Author Contributions: ~~conceptualization, A.B.~~AB and ~~A.N.;~~AN designed and initiated the study. Analysis methodology~~, A.B.~~ and ~~A.N.;~~ software~~, A.N.; formal~~ were developed and provided by AN. The analysis~~, A.B. and A.N.; investigation, A.B., A.N. and J.J.L.; data~~ was carried out by AB and AN, with comments from JJL, CB, AMM. Data curation~~, A.B., J.J.L., C.B., J.A.G., J.L., A.M.M., A.W.; writing—original draft preparation, A.B. and A.N.; writing—review~~ was provided by AB, JJL, CB, JAG, JL, AMM, AW. The manuscript was written by AB and ~~editing, A.B., A.N., J.J.L., C.B., A.W.,~~AN with additional comments ~~by A.M.M.; visualization, A.B. A.N. and J.J.L.; supervision, A.N.; project administration, A.N.; funding acquisition, A.N~~provided by JJL, CB, AW, AMM. All authors reviewed the manuscript.

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Table 1: Research flights from the SENEX 2013 campaign used in this study. The symbol "☼" next to each flight number refers to daytime flight, and "☾" refers to a nighttime flight.

Flight	Date	Local Time (~~CDT,~~ UTC-5 ~~hrs~~)	~~κ~~Hygroscopicity Parameter κ	Organic mass fraction
4☼	10/6	09:55-16:30	0.23±0.02	0.62±0.11
5☼	11/6	11:30-17:57	0.20±0.00	0.68±0.05
6☼	12/6	09:48-15:31	0.21±0.01	0.68±0.07
9☾	19/6	17:30-23:29	0.24±0.01	0.66±0.06
10☼	22/6	10:01-17:09	0.21±0.02	0.68±0.08
11☼	23/6	10:08-17:22	0.25±0.03	0.58±0.07
12☼	25/6	10:18-17:25	0.39±0.02	0.35±0.18
14☼	29/6	10:26-17:39	0.22±0.03	0.62±0.07
15☾	2/7	20:08-02:51	0.28±0.05	0.55±0.09
16☾	3/7	19:56-02:55	0.22±0.05	0.67±0.09
17☼	5/7	09:52-16:24	0.23±0.05	0.59±0.14
18☼	6/7	09:19-16:18	0.31±0.02	0.52±0.08
19☼	8/7	10:11-16:44	0.23±0.04	0.62±0.08
Average			0.25±0.05	0.60±0.09

Table 2: Flight number, time interval, ~~spectral dispersion~~standard deviation of vertical wind velocity (σ_w) and characteristic vertical velocity $w^*=0.79\sigma_w$ during flight segments where the aircraft flew at a constant altitude.

Flight (pass)	Time ~~Range~~ Interval (Local Time)	σ_w (m s^{-1})	w^* (m s^{-1})	Altitude a.s.l. (m)	Flight (pass)	Time ~~Range~~ Interval (Local Time)	σ_w (m s^{-1})	w^* (m s^{-1})	Altitude a.s.l. (m)
5 (1)	12:31-12:58	1.02	0.81	549± 58	9 (1)	18:44-18:58	0.~~255~~25	0.~~202~~20	797±2.01
5 (2)	13:16-13:29	0.82	0.65	982±11	9 (2)	19:20-19:29	0.~~249~~25	0.~~197~~2	740±1.23
5 (3)	13:34-13:50	1.01	0.80	502±13	9 (3)	19:33-19:48	0.~~217~~22	0.~~171~~17	740±1.23
5 (4)	13:53-14:08	1.03	0.81	614±27	9 (4)	19:51-20:25	0.~~218~~22	0.~~173~~17	776±1.22
5 (5)	14:20-15:00	0.91	0.72	603±40	9 (5)	20:34-20:39	0.~~232~~23	0.~~183~~18	597±1.19
5 (6)	15:35-15:41	0.87	0.69	533±18	9 (7)	20:56-21:10	0.~~201~~20	0.~~158~~16	773±1.11
5 (7)	16:17-16:30	0.77	0.61	638±23	9 (8)	21:31-21:45	0.~~191~~19	0.~~151~~15	725±1.18
5 (8)	16:31-16:39	0.55	0.44	559±18	9 (9)	22:24-22:31	0.~~257~~26	0.~~203~~20	745± 1.36
5 (9)	17:10-17:22	0.53	0.42	686±40	9 (10)	22:48-22:54	0.~~221~~22	0.~~175~~17	804± 1.37
14 (1)	12:34-12:49	0.94	0.75	558±2	15 (1)	21:09-21:52	0.~~236~~24	0.~~186~~19	505±6.64
14 (2)	13:57-14:17	0.97	0.77	658±3	15 (2)	22:19-22:31	0.~~301~~30	0.~~238~~24	633±1.21
14 (3)	14:22-14:46	0.95	0.75	737±3	15 (3)	22:42-22:54	0.~~255~~25	0.~~202~~20	600±1.17
14 (4)	14:58-15:33	0.55	0.43	746±23	15 (4)	23:26-23:37	0.~~329~~33	0.~~260~~26	908±1.56
14 (5)	15:55-16:08	0.57	0.45	714±3	15 (5)	00:02-00:19	0.~~297~~30	0.~~235~~23	1208±1.23
14 (6)	16:11-16:21	0.77	0.61	801±3	15 (6)	00:43-1:08	0.~~253~~25	0.~~199~~20	592±1.37
14 (7)	16:33-16:41	0.45	0.35	793± 2	15 (7)	1:10-1:24	0.~~276~~28	0.~~218~~22	676±1.02
					15 (8)	1:37-2:02	0.~~207~~21	0.~~164~~16	713±19.5
12 (1)	11:50-12:34	0.96	0.75	484±3	19 (1)	11:20-11:41	0.~~622~~62	0.~~492~~49	1014±2.27
12 (2)	12:48-13:18	1.09	0.86	503±3	19 (2)	12:09-12:23	1.~~203~~20	0.95	652±3.34
12 (3)	13:34-13:50	1.12	0.88	894±3	19 (3)	12:51-13:10	0.~~873~~87	0.~~689~~69	537±2.51

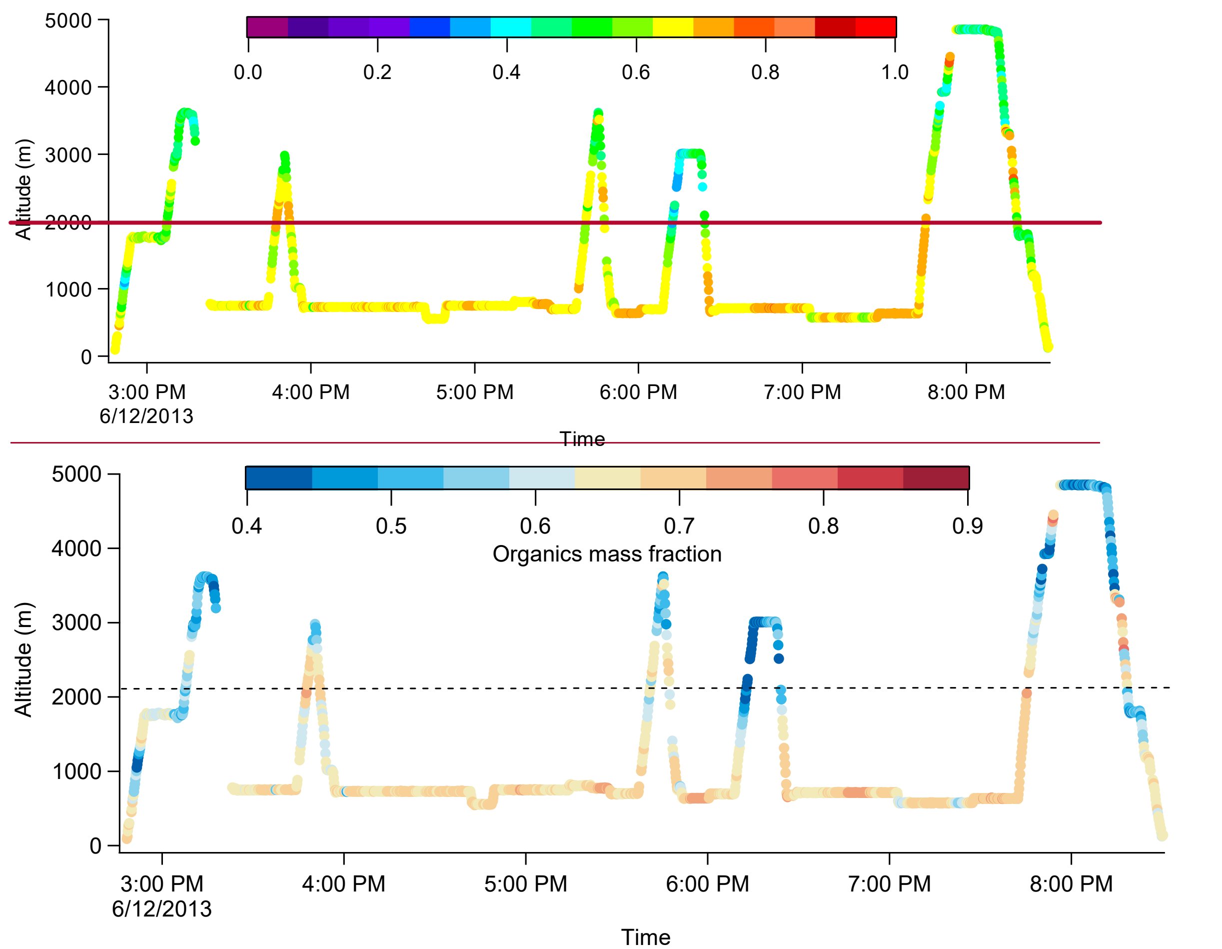
12 (4)	14:06-14:40	1.04	0.82	479±4	19 (4)	13:22-13:49	1.~~294~~29	1.~~022~~02	518±22.6
12 (5)	15:21-15:32	1.10	0.87	521±3	19 (5)	14:44-14:57	1.~~361~~36	1.~~075~~07	528±3.26
12 (6)	15:43-16:02	0.99	0.78	475±3	19 (6)	15:04-16:06	0.~~896~~90	0.~~708~~71	524±2.8

Table 3: Derived cloud parameters (maximum supersaturation, droplet number) and relative contribution of chemical composition and total aerosol number for different vertical velocities. Numbers in parentheses indicate standard deviation values. The symbol "☼" next to each flight number refers to daytime flight, and "☾" refers to a nighttime flight.

Flight	N_a	Std Dev N_a ~~variab~~	σ_w=0.1 m s^{-1}				σ_w=0.3 m s^{-1}				σ_w=0.6 m s^{-1}				σ_w=1.0 m s^{-1}			
			S_{max}	N_d	Contrib κ	Contrib N_a	S_{max}	N_d	Contrib κ	Contrib N_a	S_{max}	N_d	Contrib κ	Contrib N_a	S_{max}	N_d	Contrib κ	Contrib N_a
4☼	6118	4520	0.11 (0.06)	122 (41)	0.08	0.92	0.16 (0.09)	315 (114)	0.20	0.80	0.21 (0.12)	520 (212)	0.23	0.77	0.26 (0.17)	737 (321)	0.2	0.8
5☼	4324	2598	0.08 (0.04)	139 (31)	0.09	0.91	0.1 (0.06)	388 (104)	0.15	0.85	0.14 (0.08)	712 (216)	0.17	~~0.83~~083	0.17 (0.1)	1063 (360)	0.21	0.79
6☼	4958	3054	0.07 (0.07)	151 (24)	0.03	0.97	0.08 (0.04)	422 (70)	0.11	0.89	0.1 (0.06)	773 (171)	0.08	0.92	0.13 (0.07)	1162 (302)	0.07	0.93
9☾	4271	3095	0.07 (0.02)	152 (18)	0.05	0.95	0.12 (0.04)	367 (68)	0.17	0.83	0.16 (0.05)	533 (115)	0.17	0.83	0.19 (0.06)	680 (126)	0.12	0.88
10☼	6286	7201	0.07 (0.03)	158 (24)	0.02	0.98	0.1 (0.05)	422 (86)	0.02	0.98	0.14 (0.07)	748 (180)	0.04	0.96	0.18 (0.08)	1063 (295)	0.09	0.91
11☼	5969	7271	0.04 (0.01)	137 (19)	0.01	0.99	0.06 (0.01)	381 (61)	0.04	0.96	0.08 (0.02)	695 (134)	0.03	0.97	0.10 (0.02)	1025 (226)	0.03	0.97
12☼	3154	5150	0.06 (0.03)	110 (45)	0.03	0.97	0.1 (0.04)	274 (117)	0.05	0.95	0.14 (0.04)	404 (179)	0.08	0.92	0.17 (0.05)	486 (207)	0.07	0.93
14☼	5564	5891	0.07 (0.02)	118 (41)	0.05	0.95	0.10 (0.03)	328 (125)	0.17	0.83	0.13 (0.04)	590 (240)	0.25	0.75	0.16 (0.05)	842 (361)	0.27	0.73
15☾	2328	1428	0.05 (0.01)	135 (22)	0.03	0.97	0.09 (0.02)	339 (67)	0.12	0.88	0.12 (0.02)	557 (137)	0.21	0.79	0.16 (0.03)	717 (203)	0.3	0.7
16☾	3440	4507	0.08 (0.06)	158 (37)	0.03	0.97	0.12 (0.1)	403 (120)	0.06	0.94	0.17 (0.13)	670 (235)	0.07	0.93	0.23 (0.16)	917 (374)	0.1	0.9
17☼	3813	4645	0.05 (0.02)	129 (41)	0.06	0.94	0.07 (0.03)	342 (130)	0.1	0.9	0.1 (0.04)	593 (248)	0.06	0.94	0.13 (0.05)	841 (371)	0.06	0.94
18☼	1925	983	0.08 (0.04)	90 (58)	0.12	0.88	0.12 (0.05)	233 (157)	0.35	0.65	0.15 (0.06)	379 (262)	0.37	0.63	0.19 (0.07)	499 (346)	0.27	0.73
19☼	4323	7261	0.06 (0.02)	121 (33)	0.02	0.98	0.08 (0.02)	314 (96)	0.06	0.94	0.12 (0.03)	526 (177)	0.11	0.89	0.15 (0.03)	670 (249)	0.13	0.87

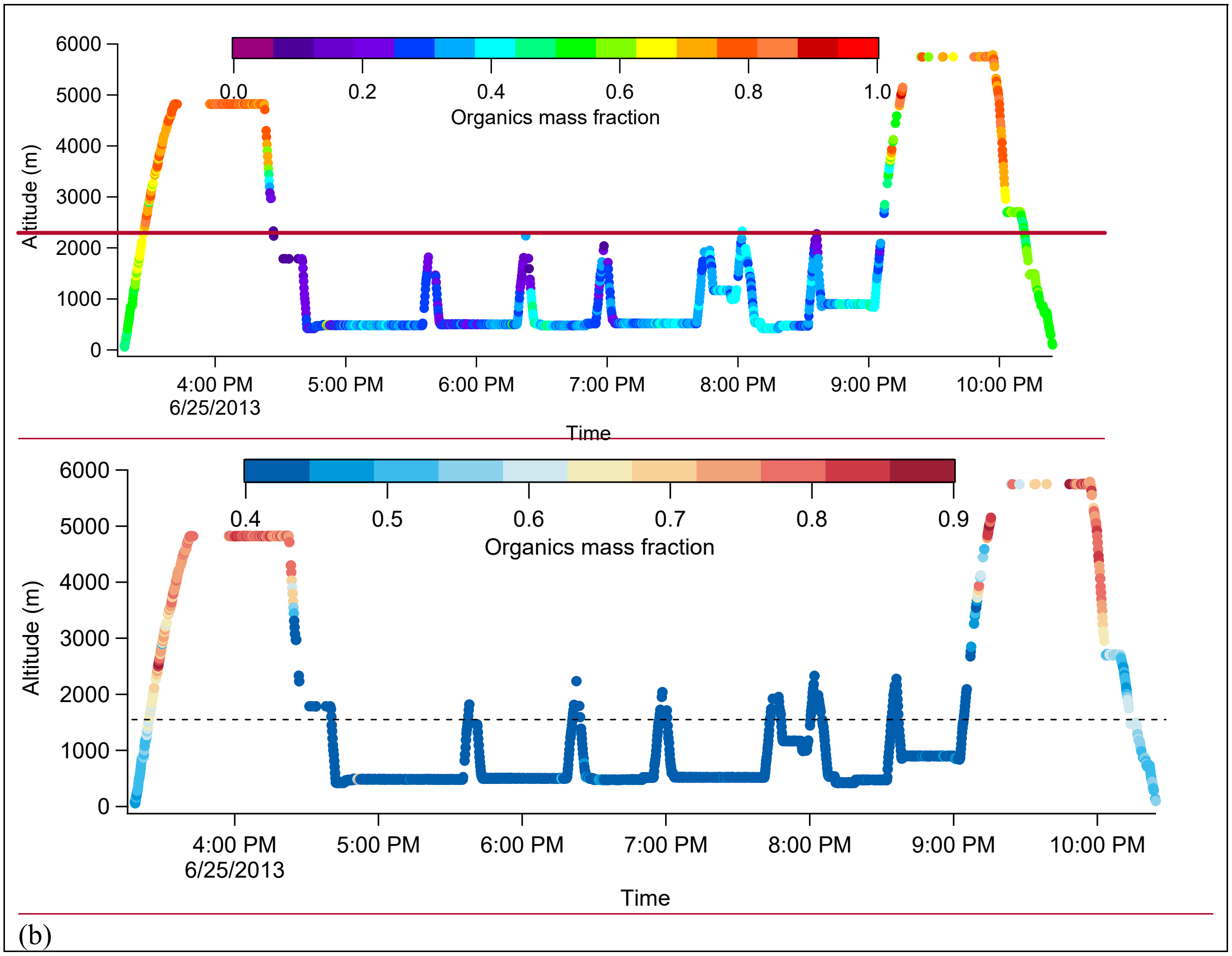
Table 4: Derived S_{max}, N_d, σ_w for all research flights along with the estimated contribution of each parameter to the variability of the droplet number. The symbol "☼" next to each flight number refers to daytime flight, and "☾" refers to a nighttime flight.

Flight	σ_w (m s^{-1})	$\frac{\Delta\sigma_w}{\sigma_w}$	S_{max} (%)	N_d (cm^{-3})	$\frac{\Delta N_d}{N_d}$	**Contrib.** κ	**Contrib.** N_a	**Contrib.** σ_w
4☼	1.03±0.25	0.243	0.29±0.19	707±343	0.485	4%	79%	17%
5☼	0.97±0.1	0.103	0.17±0.10	1040±350	0.337	7%	69%	24%
6☼	0.94±0.18	0.191	0.13±0.07	1108±283	0.255	3%	54%	43%
9☾	0.23±0.02	0.043	0.10±0.03	309±51	0.165	7%	76%	17%
10☼	1.22±0.11	0.090	0.12±0.03	1177±271	0.230	1%	90%	9%
11☼	1.08±0.04	0.037	0.11±0.03	1082±242	0.224	1%	83%	16%
12☼	1.05±0.07	0.067	0.18±0.05	495±210	0.424	2%	96%	2%
14☼	0.85±0.2	0.024	0.15±0.04	761±321	0.422	9%	72%	19%
15☾	0.28±0.04	0.143	0.08±0.02	321±63	0.196	7%	51%	42%
16☾	0.20±0.04	0.200	0.10±0.08	289±79	0.273	2%	65%	33%
17☼	0.71±0.26	0.366	0.15±0.11	742±280	0.377	1%	71%	28%
18☼	0.90±0.06	0.067	0.31±0.18	538±325	0.604	7%	83%	10%
19☼	0.99±0.31	0.313	0.15±0.03	699±248	0.355	4%	88%	8%
~~Avg~~Average					0.334	4%	75.2%	20.6%

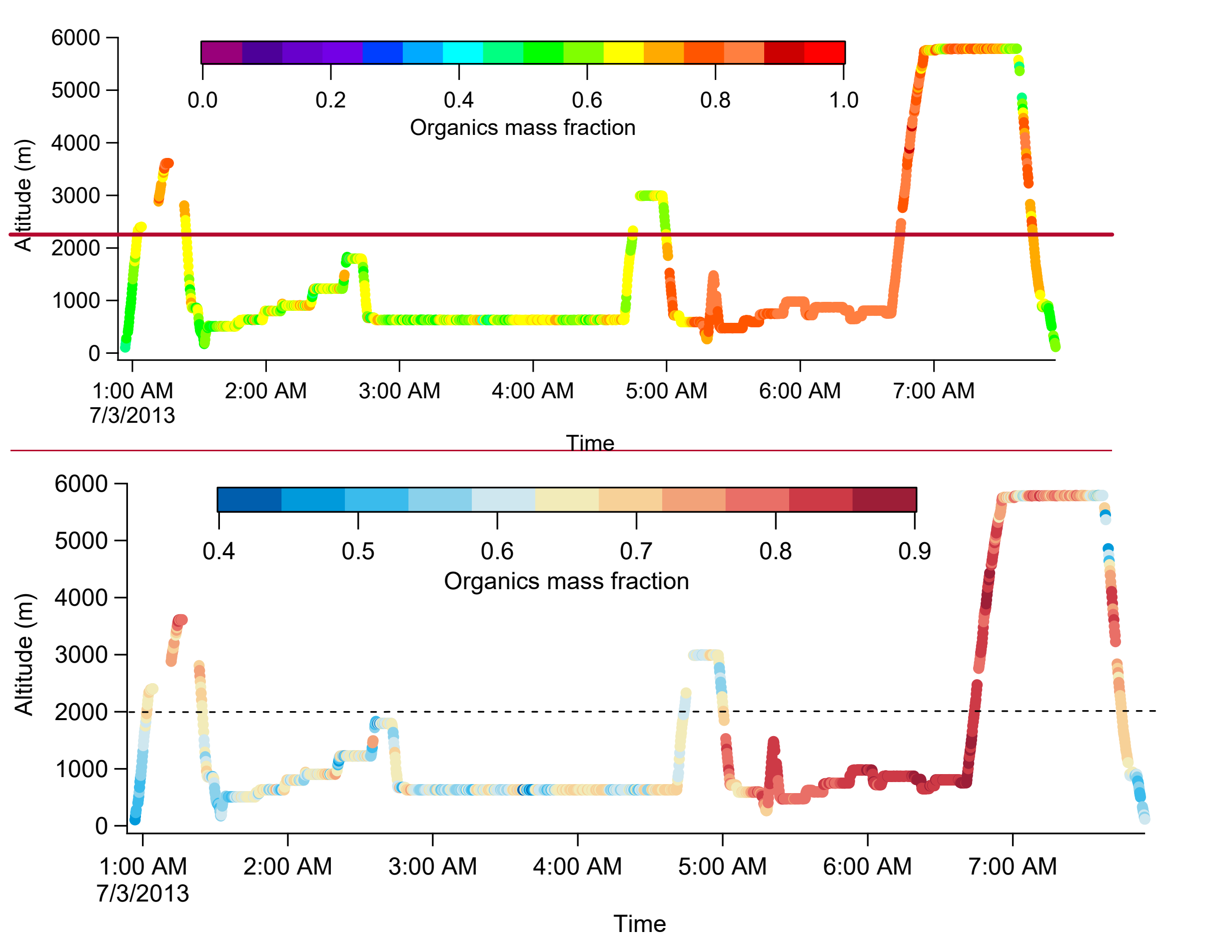


Altitude (m)
Time
3:00 PM
6/12/2013
4:00 PM
5:00 PM
6:00 PM
7:00 PM
8:00 PM
Organics mass fraction

(a)


6000
5000
4000
3000
2000
1000
0
Altitude (m)
0.0
0.2
0.4
0.6
0.8
1.0
Organics mass fraction
4:00 PM
6/25/2013
5:00 PM
6:00 PM
7:00 PM
8:00 PM
9:00 PM
10:00 PM
Time
6000
5000
4000
3000
2000
1000
0
Altitude (m)
0.4
0.5
0.6
0.7
0.8
0.9
Organics mass fraction
4:00 PM
6/25/2013
5:00 PM
6:00 PM
7:00 PM
8:00 PM
9:00 PM
10:00 PM
Time

(b)



(c)

~~**Figure 1:**~~**Figure 1:** Altitude as a function of time (UTC) colored by organic mass fraction. Spatial and vertical distribution of the organics mass fraction (a) for Flight 6, (b) for Flight 12 and (c) for Flight 16, denoting the difference in chemical composition, which in turn, may influence cloud droplet number concentration. ~~The color scale denotes the percentage of the organics mass fraction.~~The dashed line represents the boundary layer height.

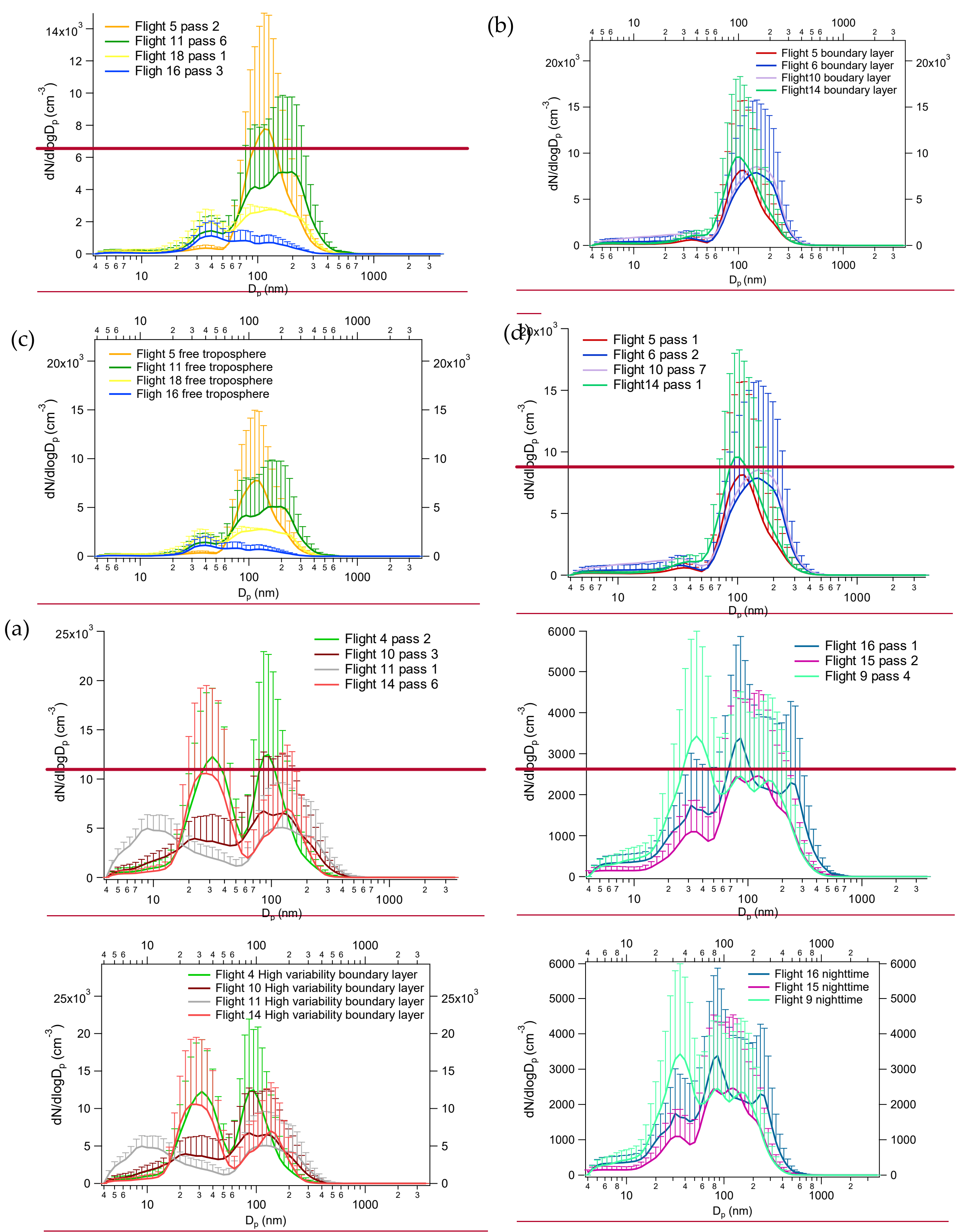
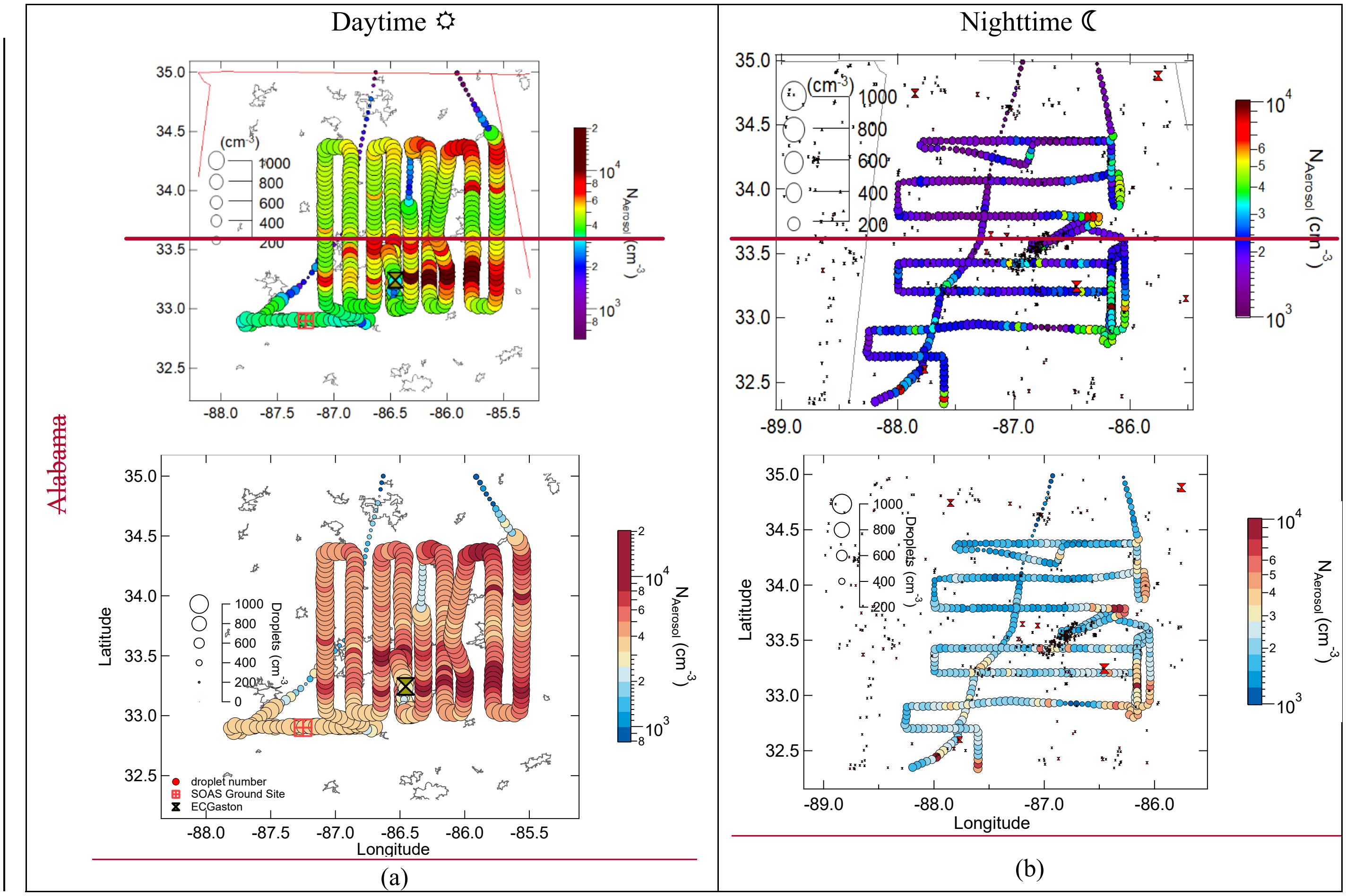

(a)
(b)
(c)
(d)
Flight 5 pass 2
Flight 11 pass 6
Flight 18 pass 1
Fligh 16 pass 3
Flight 5 boundary layer
Flight 6 boundary layer
Flight10 boudary layer
Flight14 boundary layer
Flight 5 free troposphere
Flight 11 free troposphere
Flight 18 free troposphere
Fligh 16 free troposphere
Flight 5 pass 1
Flight 6 pass 2
Flight 10 pass 7
Flight14 pass 1
Flight 4 pass 2
Flight 10 pass 3
Flight 11 pass 1
Flight 14 pass 6
Flight 16 pass 1
Flight 15 pass 2
Flight 9 pass 4
Flight 4 High variability boundary layer
Flight 10 High variability boundary layer
Flight 11 High variability boundary layer
Flight 14 High variability boundary layer
Flight 16 nighttime
Flight 15 nighttime
Flight 9 nighttime
dN/dlogD$_p$ (cm^{-3})
D$_p$ (nm)

Figure 2: Average particle number size distributions for: (a) free tropospheric conditions, (b) within the boundary layer, (c) during ~~passes~~segments with high variability in total aerosol number, and (d) during nighttime passes. Error bars represent the 75th percentile of the distributions within each ~~pass~~segment.



Daytime
Nighttime
Alabama
(cm-3)
1000
800
600
400
200
NAerosol (cm-3)
Latitude
Longitude
Droplets (cm-3)
droplet number
SOAS Ground Site
ECGaston
(a)
(b)

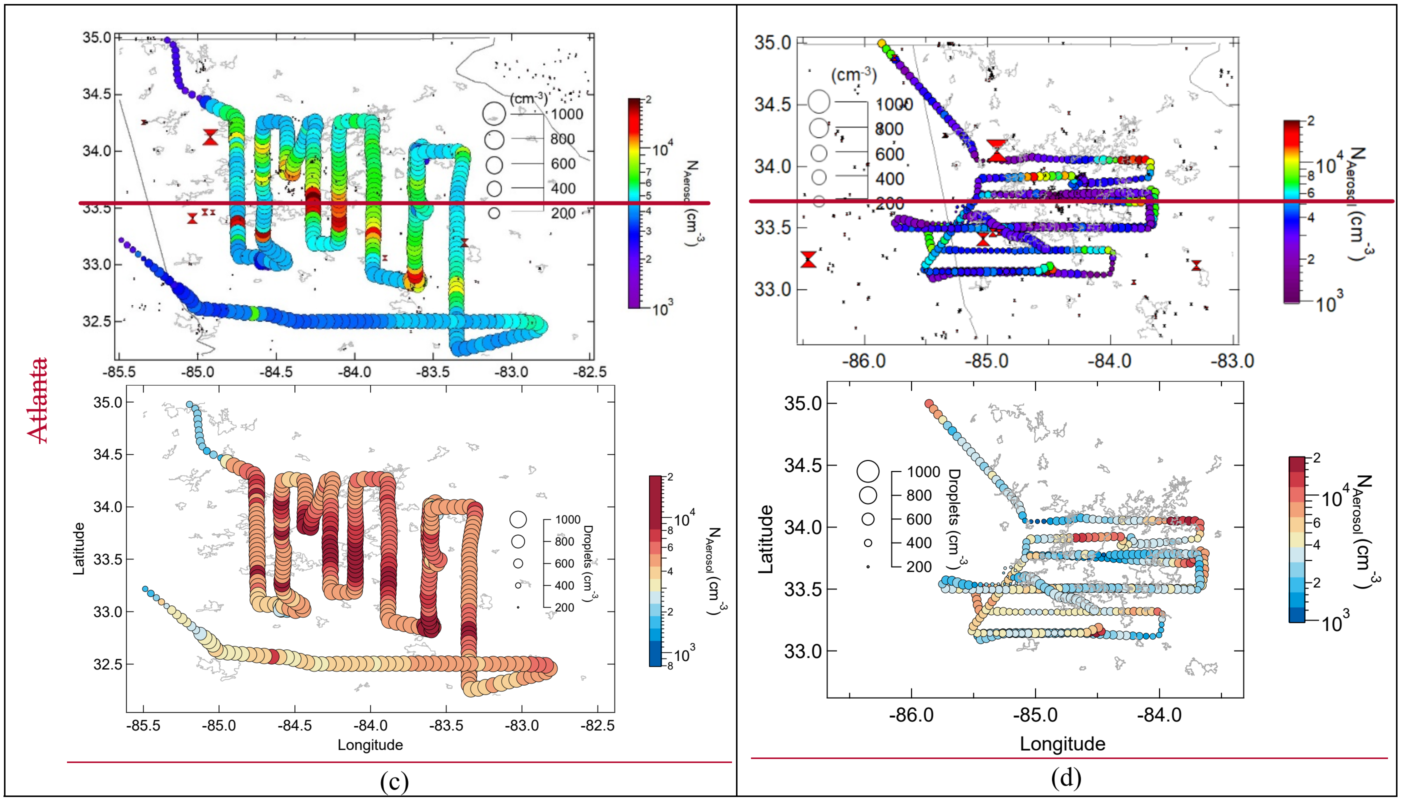
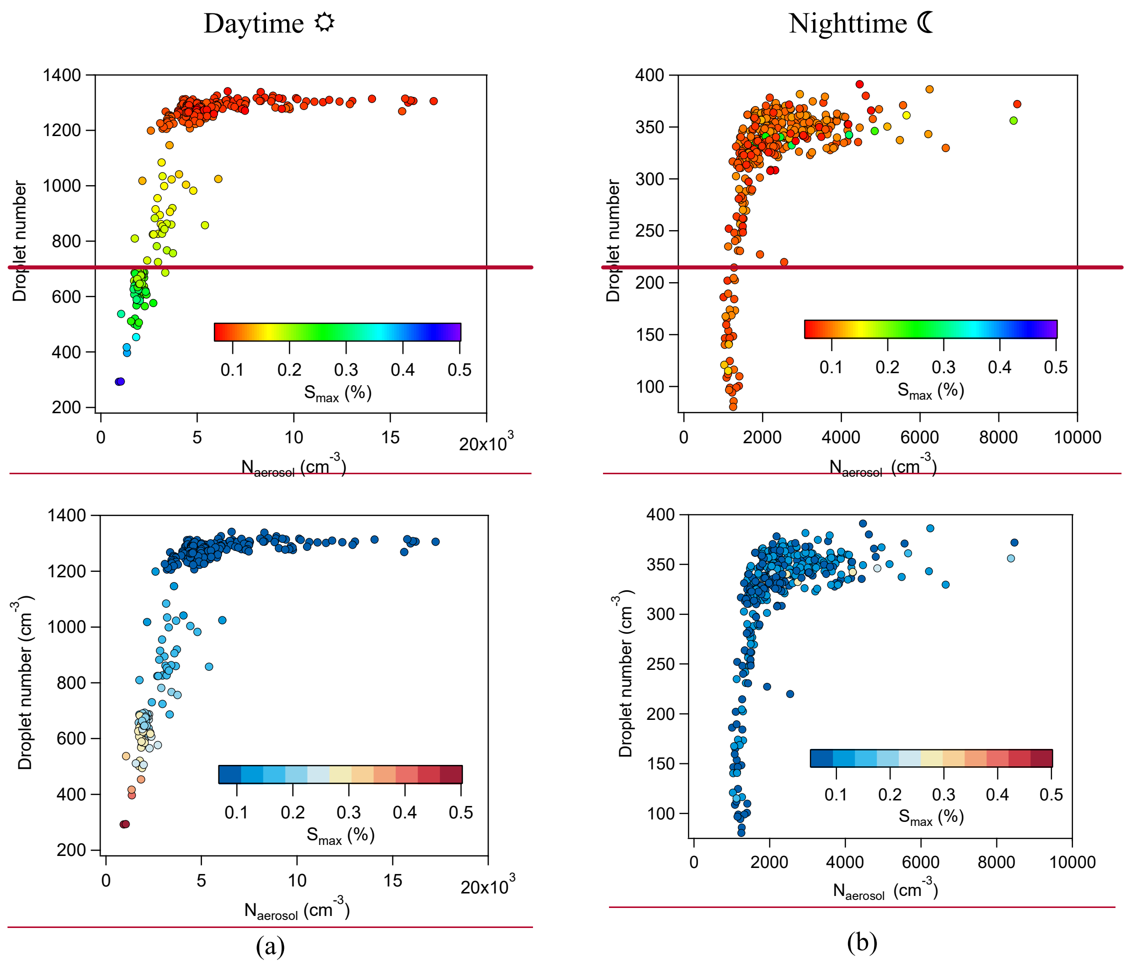


Figure 3: ~~Flight trajectories~~Map of aircraft flight track showing ~~calculated~~calculated cloud droplet number (indicated by marker size (cm^{-3})) and total aerosol number (indicated by marker color) for the observed characteristic vertical velocity (w*). (a) for the ~~Alabama~~rural sector during daytime (Flight 5) and (b) nighttime (Flight 15). (c) for urban Atlanta during daytime (Flight 6) and (d) nighttime (Flight 9). Note that the data are plotted at less than 1 Hz in order to better show the size and color of the markers.

~~Alabama~~Rural



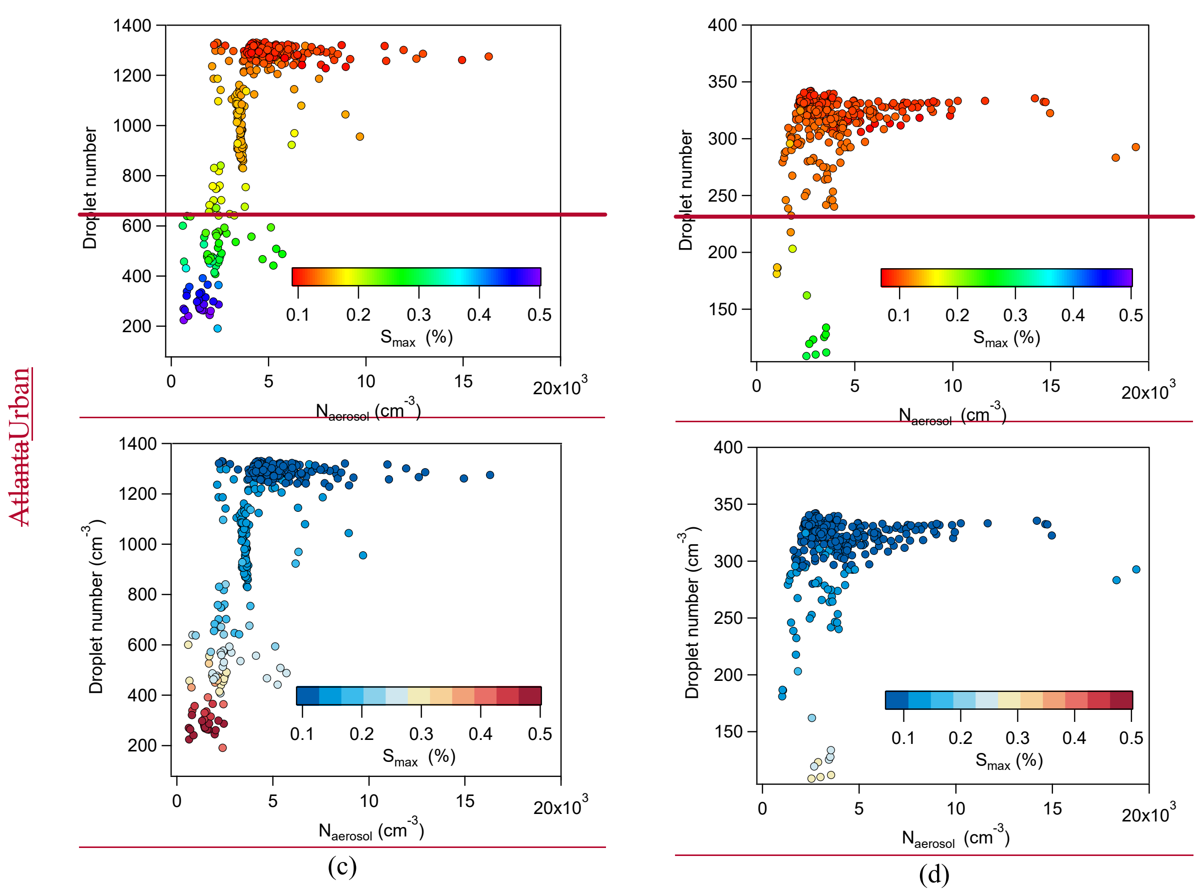
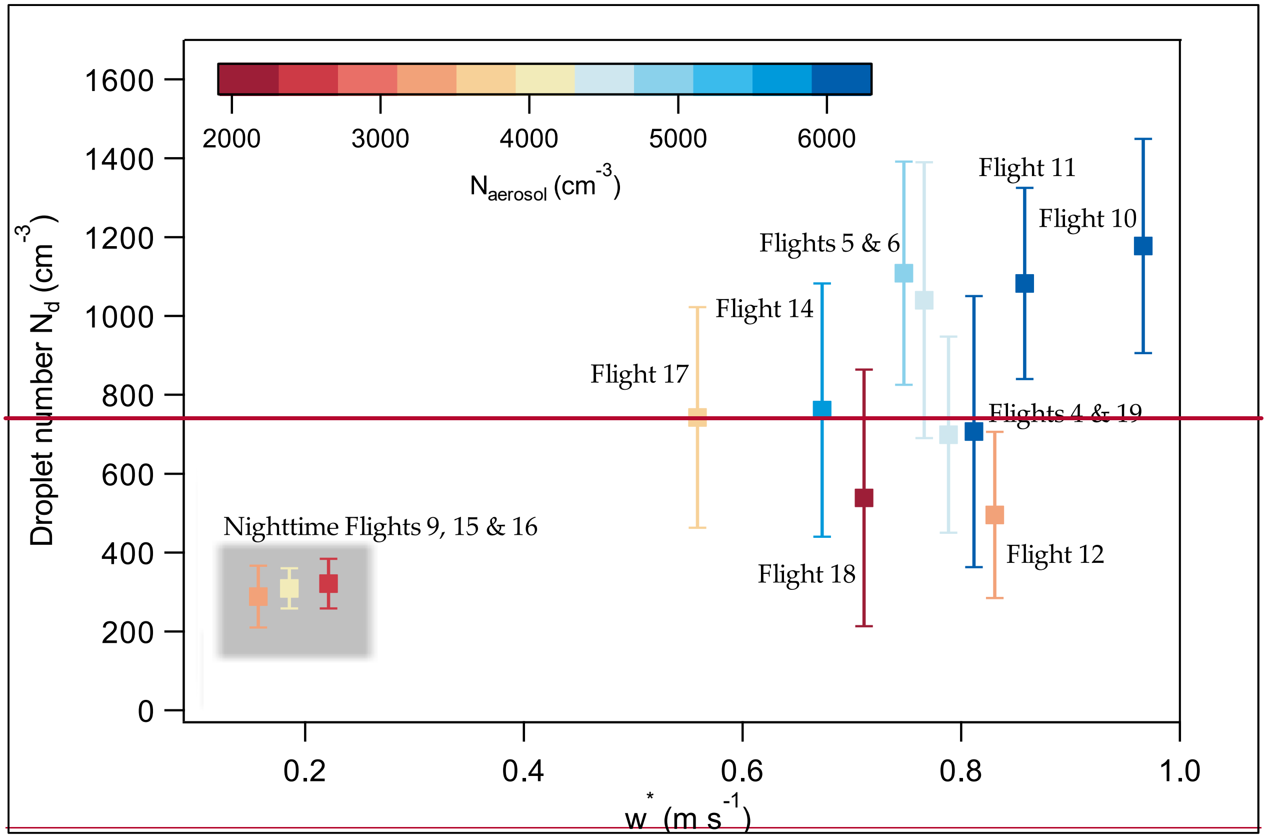
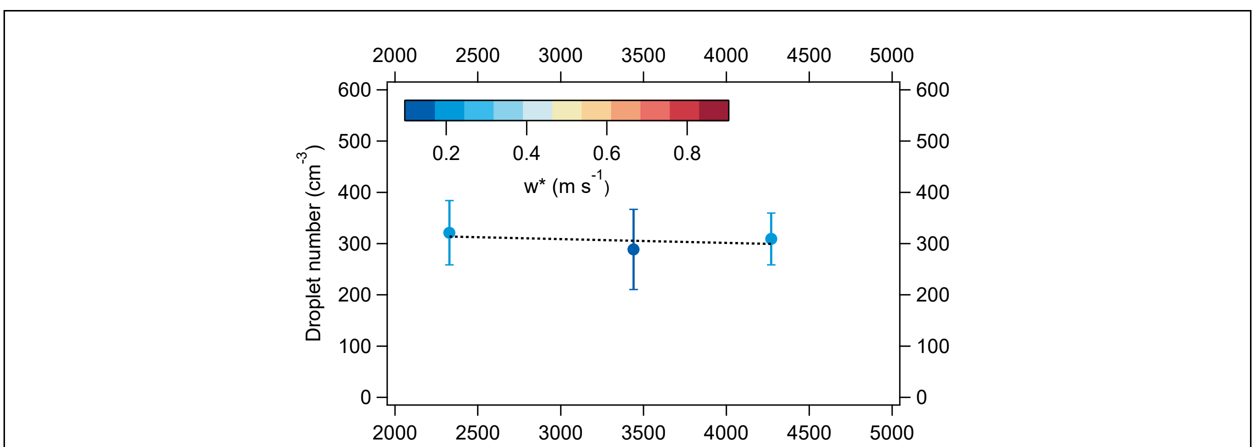


Figure 4: Cloud droplet number vs. total aerosol number for the derived characteristic vertical velocity (w~~*).~~*) of each flight (Table 4). (a) for the ~~Alabama~~rural sector during daytime (Flight 5) and (b) nighttime (Flight 15). (c) for ~~Atlanta~~urban Altanta during daytime (Flight 6) and (d) nighttime (Flight 9). Data are colored by maximum supersaturation.


Droplet number N_d (cm^{-3})
1600
1400
1200
1000
800
600
400
200
0
2000
3000
4000
5000
6000
$N_{aerosol}$ (cm^{-3})
Flight 11
Flight 10
Flights 5 & 6
Flight 14
Flight 17
Flights 4 & 19
Nighttime Flights 9, 15 & 16
Flight 18
Flight 12
0.2
0.4
0.6
0.8
1.0
w^* (m s^{-1})


2000
2500
3000
3500
4000
4500
5000
600
500
400
300
200
100
0
0.2
0.4
0.6
0.8
w* (m s^{-1})
Droplet number (cm^{-3})
Total aerosol number (cm^{-3})

(a)

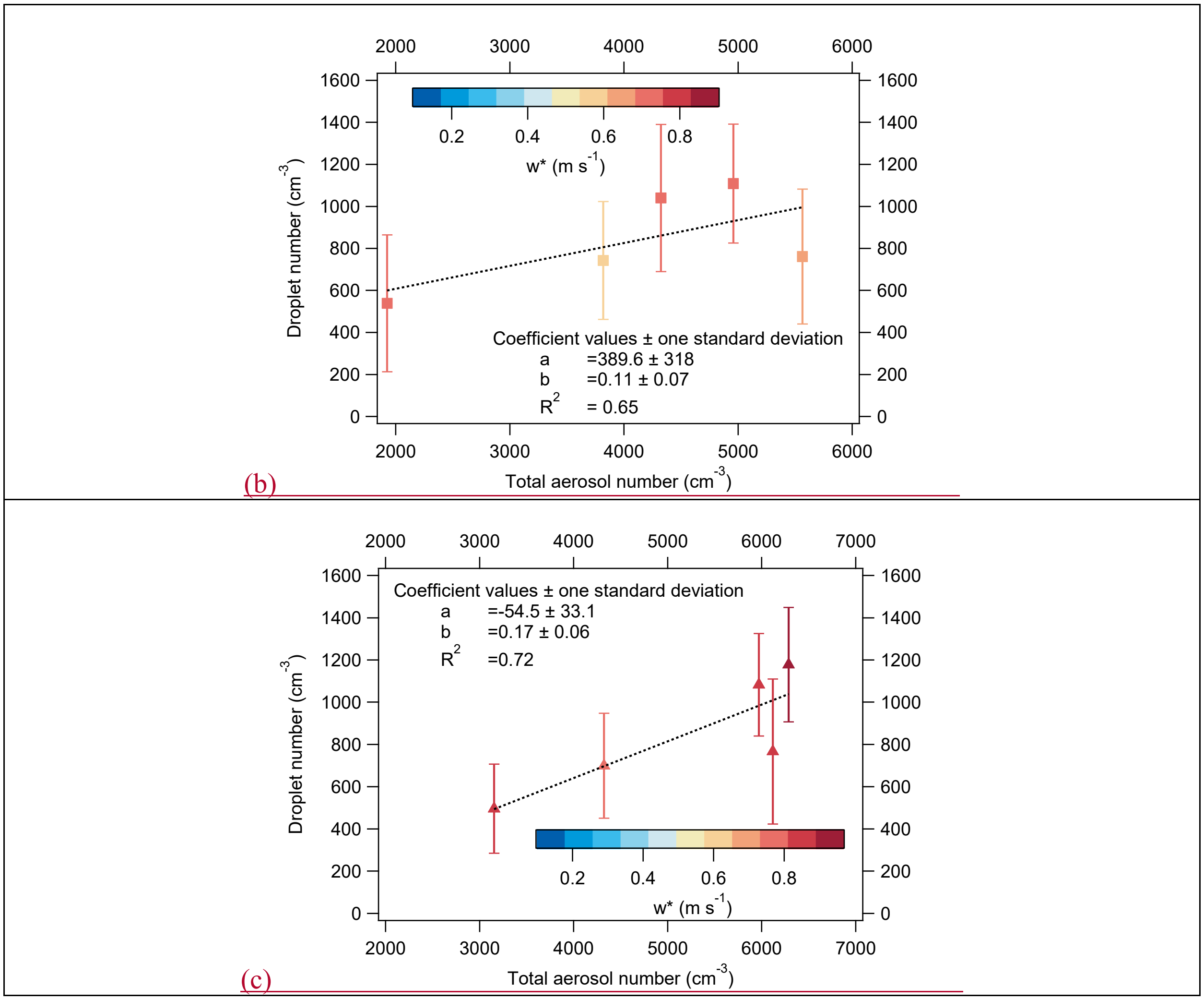
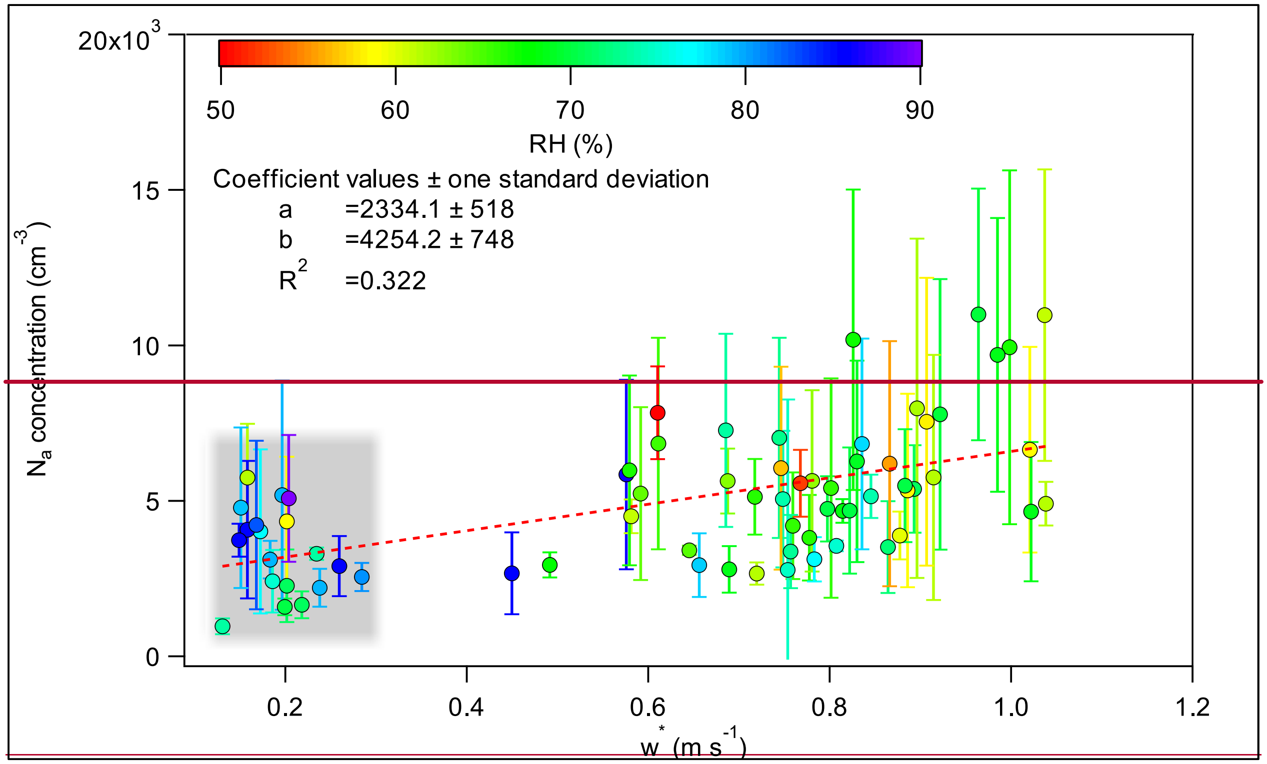


Figure 5: Average cloud droplet number vs. total aerosol number, colored by characteristic velocity ~~during the 13 research flights, colored by total aerosol number.~~ w* for each flight. Error bars represent the standard deviation of cloud droplet number during each flight. ~~The shaded area represents the flights conducted during nighttime.~~


20x10^3
15
10
5
0
50
60
70
80
90
RH (%)
Coefficient values ± one standard deviation
a =2334.1 ± 518
b =4254.2 ± 748
R^2 =0.322
N_a concentration (cm^-3)
0.2
0.4
0.6
0.8
1.0
1.2
w* (m s^-1)

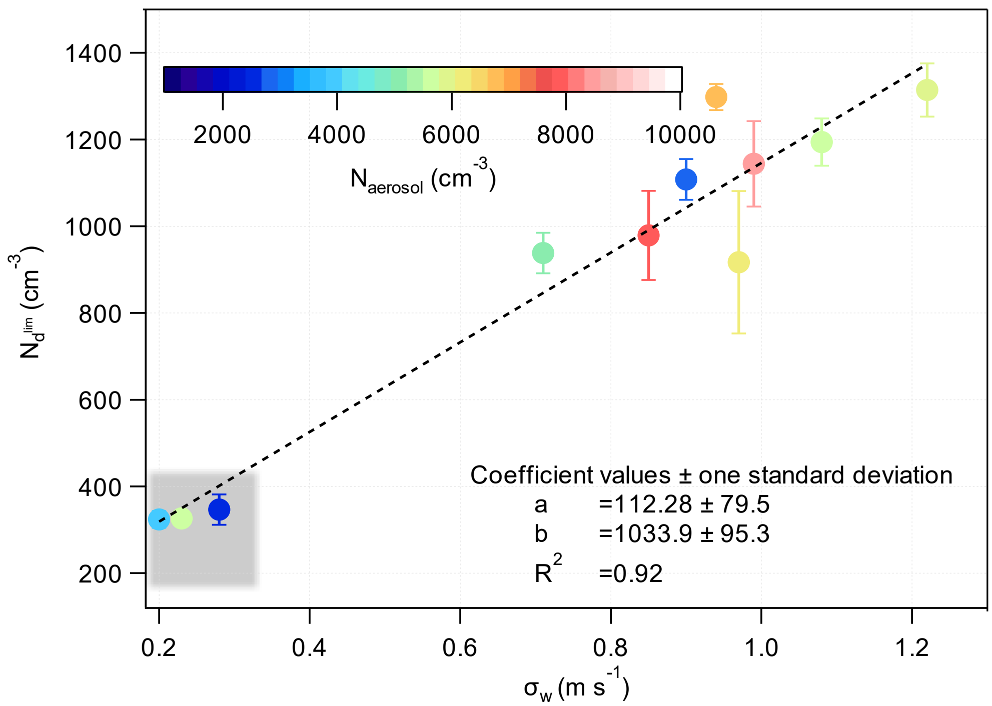


Figure 6: ~~Total aerosol~~Limiting droplet number vs. ~~characteristic~~standard deviation of vertical velocity during ~~the segments of the flights when the aircraft remained at a constant altitude within the boundary layer, colored by relative humidity.~~flights where a velocity-limited regime is reached (all except Flights 4, 12). The shaded area represents the segments of the flights conducted during nighttime while color scale denotes total aerosol number levels.