

Reviewer comments:

The authors have sufficiently responded to reviewer comments, improving figures, improving the clarity, making it clear what cases are night/day and rural/urban, and clearly indicating the importance of these results.

Response: We thank the anonymous referee for the thoughtful review. The proposed suggestions and comments for the modification of the manuscript helped improve the quality of the manuscript.

The figures have been improved however, I highly recommend using images for your plots instead of vector graphics. Especially in the supplement where there are so many vector graphics. Currently it takes a while to load each figure and it is cumbersome to scroll through the document.

Response: Indeed, all figures now in the supplement are in .jpeg format so that all figures are loaded rapidly and it doesn't take long to scroll through the document.

Below are additional comments/suggestions. Note: line numbers in comments are based on the text version with track changes.

Line 41 "3 to 5 times" for what scenario? If Na were consistent between night and day? Or if sigmaw were consistent? Something else?

Response: The increase refers to the increase of expected droplet number compared to what it would be based on Na changes alone. This is now clarified in the text.

Line 120 could you provide some evidence or cite a relevant publication with evidence indicating that assuming the aerosol are internally mixed is a reasonable assumption.

Response: Relevant references are now added.

Line 160 – I understand droplets are only formed for positive updraft velocities, but is sigma w calculated from the entire vertical velocity distribution (or just positive velocities)? You mention the gaussian distribution has a mean of zero (which mean there are negative and positive values), but you also say you only use the positive updraft velocities, so it's not clear.

Response: Only the positive half of the Gaussian distribution around a mean of zero were used for the sigma w calculations. This is now clarified in the text.

Line 178-180 – reference table 3 *Response: Done.*

Line 245 "These calculations help understand" - suggest rewording *Response: Amended.*

Line 256 – change 'low' to 'lowest' *Response: Amended.*

Line 307 – its driven mainly by Na (very little by kappa). I suggest changing 'characteristics' to 'concentrations' and note kappa had little influence (same for the following sentence).

Response: Amended.

Line 317-321 this was already mentioned earlier in this section. I suggest removing these last 3 sentences.

Response: Indeed, these sentences are now removed.

Line 352 – suggest changing 'strong' to 'high' *Response: Done.*

Line 358- change 'does' to 'do' and change 'important' to 'large' *Response: Done.*

Line 419 – add 'of' before 'a' *Response: Done.*

Table 2. reference equation 2 as the equations used to determine the contributions.

Response: Done.

Figure 1. What measurement or model was used to determine BL height.

Response: The boundary layer height for daytime in the area (1200 m) was taken from Wagner et al. (2015) as calculated during the same measurement campaign while for nighttime the value of 500 m was used from Seidel et al. (2012). This is now clarified in the text.

Below please find the revised manuscript with marked track changes

1 Drivers of cloud droplet number variability in the summertime

2 Southeast United States

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24 Abstract

25 Here we analyze regional scale data collected onboard the NOAA WP-3D aircraft during the 2013
26 Southeast Nexus (SENEX) campaign to study the aerosol-cloud droplet link and quantify the sensitivity of
27 droplet number to aerosol number, chemical composition and vertical velocity. For this, the observed
28 aerosol size distributions, chemical composition and vertical velocity distribution are introduced into a
29 state-of-the-art cloud droplet parameterization to show that cloud maximum supersaturations in the region
30 are low, ranging from 0.02 to 0.52% with an average of $0.14 \pm 0.05\%$. Based on these low values of
31 supersaturation, the majority of activated droplets correspond to particles of dry diameter 90 nm and above.
32 An important finding is that the standard deviation of the vertical velocity (σ_w) exhibits considerable diurnal
33 variability (ranging from 0.16 m s^{-1} during nighttime to over 1.2 m s^{-1} during day) and it tends to covary
34 with total aerosol number (N_a). This σ_w - N_a covariance amplifies the predicted response in cloud droplet
35 number (N_d) to N_a increases by 3 to 5 times, compared to expectations based on N_a changes alone. -
36 which This amplified response is important, given that droplet formation is often velocity-limited, and
37 therefore should normally be insensitive to aerosol changes. We also find that N_d cannot exceed a
38 characteristic concentration that depends solely on σ_w . Correct consideration of σ_w and its covariance with
39 time and N_a is important for fully understanding aerosol-cloud interactions and the magnitude of the aerosol

40 indirect effect. Given that model assessments of aerosol–cloud–climate interactions do not routinely
41 evaluate for overall turbulence or its covariance with other parameters, datasets and analyses such as the
42 one presented here are of the highest priority to address unresolved sources of hydrometeor variability, bias,
43 and the response of droplet number to aerosol perturbations.

44 **1. Introduction**

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

64 The Southeast United States (SEUS) presents a particularly interesting location for studying regional
65 climate change, as it has not considerably warmed over the past 100 years – except during the last decade
66 (Carlton et al., 2018; Yu et al., 2014; Leibensperger et al., 2012a,b). These trends are in contrast with the
67 trends observed in most locations globally (IPCC 2013), and several hypotheses have been proposed to
68 explain this regional phenomenon, including the effect of involving short-lived climate forcers such as
69 secondary aerosols combined with the enhanced humidity in the region and their impact on clouds (Carlton
70 et al., 2018; Yu et al., 2014). Here, we analyze data collected during the Southeast Nexus of Air Quality
71 and Climate (SENEC) campaign in June–July 2013, which was the airborne component led by the National
72 Oceanic and Atmospheric Administration (NOAA), of a greater measurement campaign throughout the

73 SEUS, the Southeast Atmosphere Study (SAS; Carlton et al., 2018). Here we analyze data collected onboard
74 the NOAA WP-3D and apply a state-of-the-art droplet parameterization to determine the maximum
75 supersaturation and N_d achieved in cloudy updrafts, for all science flights with available number size
76 distribution and chemical composition data. We also determine the sensitivity of droplet formation to
77 vertical velocity and aerosol, with the purpose of understanding the drivers of droplet variability in the
78 boundary layer of the SEUS by obtaining regional-scale, representative values of the relationship between
79 the driving parameters and cloud droplet number.

80

81 **2. Methods**

82 *2.1 Aircraft instrumentation*

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

90 Dry particle number distributions from 4 - 7000 nm were measured using multiple condensation and optical
91 particle counters. 4-700 nm particles were measured by a nucleation mode aerosol size spectrometer
92 (NMASS; Warneke et al., 2016) and an ultra-high sensitivity aerosol spectrometer (UHSAS; Brock et al.,
93 2011), while for larger particles with dry diameters between 0.7 and 7.0 μm , a custom-built white-light
94 optical particle counter (WLOPC) was used (Brock et al., 2011).

95 Measurements of the composition of submicron ($< 0.7 \mu\text{m}$ vacuum aerodynamic diameter) non-refractory
96 particles were made with a compact time-of-flight aerosol mass spectrometer (C-ToF-AMS; Aerodyne,
97 Billerica, [Massachusetts](#), US) (Canagaratna et al., 2007; Kupc et al., 2018) customized for aircraft use, with
98 a 10 s time resolution (Warneke et al., 2016). Particles entering the instrument are focused and impacted
99 on a 600 °C inverted-cone vaporizer. The volatilized vapors are analyzed by electron ionization mass
100 spectrometry, providing mass loadings of sulfate, nitrate, organics, ammonium and chloride. For the C-
101 ToF-AMS, the transmission efficiency of particles between 100 and 700 nm is assumed to be 100% through
102 the specific aerodynamic focusing lens used while mass concentrations are calculated using a chemical
103 composition-dependent collection efficiency (Middlebrook et al., 2012; Wagner et al., 2015). The C-ToF-
104 AMS measures only non-refractory aerosol chemical composition, therefore this analysis provides mass

105 loadings of sulfate, nitrate, ammonium and organic constituents with a 10 s time resolution and neglects the
106 contribution of black carbon (BC). The calculation of the average volume fractions from the mass loading
107 follows that of Moore et al. (2012). An average organic density of 1.4 g cm^{-3} is used, characteristic of aged
108 aerosol (Moore et al., 2011; Latham et al., 2013) while for the inorganic species the respective densities are
109 used, assuming the aerosol to be internally mixed ([Lance et al., 2009](#); [Bougiatioti et al., 2009](#); [Wang et al.,](#)
110 [2010](#)).

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

120 2.2 Aerosol hygroscopicity parameter

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

131 2.3 Cloud droplet number and maximum supersaturation

132 Using the observed aerosol number size distribution (1 s time resolution) and the hygroscopicity derived
133 from the chemical composition measurements (10 s time resolution) and vertical velocity, we calculate the
134 (potential) cloud droplet number (N_d) and maximum supersaturation (S_{max}) that would form in clouds in the
135 airmasses sampled. Droplet number and maximum supersaturation calculations are carried out using an

136 approach similar to that of Bougiatioti et al. (2017) and Kalkavouras et al. (2019) with the sectional
137 parameterization of Nenes and Seinfeld (2003), later improved by Barahona et al. (2010) and Morales
138 Betancourt and Nenes (2014a). A sectional representation of the size distribution is used for each 1-s data
139 point (e.g. for Flight 5, n=23213 data points). Given that chemical composition is provided with a 10 s time
140 resolution, the same hygroscopicity values are used for 10 successive size distributions throughout the
141 flight. Temperature and pressure required for droplet number calculations are obtained from the NOAA
142 AOC flight facility dataset.

143 Given that vertical velocity varies considerably inside the boundary layer, we obtain a droplet number that
144 is representative of the vertical velocity distribution – the average concentration that results from integrating
145 over the distribution (probability density function, PDF) of observed updraft velocities. To accomplish this,
146 each flight is divided into segments where the aircraft flew at a constant height. For each segment, only the
147 non-negative vertical velocities are fit to the positive half of a Gaussian distribution with-around a mean of
148 zero and standard deviation σ_w . Only positive vertical velocities (“updrafts”) were used in this fit, as they
149 are the part of the vertical velocity spectrum that is responsible for cloud droplet formation. The σ_w values
150 derived from the level leg segments are then averaged into one single σ_w value to represent each flight. The
151 PDF-averaged droplet number concentration is then obtained using the “characteristic velocity” approach
152 of Morales and Nenes (2010), where applying the droplet parameterization at a single “characteristic”
153 velocity, $w^*=0.79\sigma_w$ (Morales and Nenes, 2010) gives directly the PDF-averaged value. The flight-averaged
154 σ_w and subsequently the respective w^* is applied to each size distribution measured. Apart from its
155 theoretical basis, this methodology has shown to provide good closure with observed droplet numbers in
156 ambient clouds (e.g. Kacarab et al., 2020).

157 In determining σ_w , we consider horizontal segments most likely to be in the boundary layer. 91% of the
158 segments are below 1000 m above sea level (mean altitude ~700 m; Table 2 and SP3 for all flights), within
159 the boundary layer in the summertime US (Seidel et al., 2012). Also according to Wagner et al. (2015) the
160 mixed layer height in the area during the measurements had a median of 1.2 km while during nighttime
161 values are <0.5 km (Seidel et al., 2012). The vertical velocity distributions observed gave $\sigma_w = 0.97 \pm 0.21$ m
162 s^{-1} for daytime flights, and $\sigma_w = 0.23 \pm 0.04$ m s^{-1} for nighttime flights (Table 2 and SP3).

163 Potential droplet formation is evaluated at four characteristic velocities w^* that cover the observed range in
164 σ_w , namely 0.1, 0.3, 0.6, and 1 m s^{-1} . The $\sigma_w = 0.3$ m s^{-1} case is most representative of nighttime conditions,
165 while $\sigma_w = 1$ m s^{-1} should be most representative of the daytime boundary layer (Table 3).

166 We also compute the variance of the derived N_d , estimated from the sensitivity to changes in aerosol number
167 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

168 from the parameterization using a finite difference approximation (Bougiatioti et al., 2017; Kalkavouras et
 169 al., 2019) using:

$$170 \quad \sigma^2 N_d = \left(\frac{\partial \overline{N_d}}{\partial N_a} \sigma N_a \right)^2 + \left(\frac{\partial \overline{N_d}}{\partial \kappa} \sigma \kappa \right)^2 + \left(\frac{\partial \overline{N_d}}{\partial \sigma_w} \sigma w \right)^2 \quad (1)$$

171
 172 These sensitivities, together with the observed variance in N_a , κ , and σ_w are also used to attribute droplet
 173 number variability to variations in the respective aerosol and vertical velocity parameters following the
 174 approach of Bougiatioti et al. (2017) and Kalkavouras et al. (2019):

$$175 \quad \varepsilon_{N_a} = \frac{\left(\frac{\partial \overline{N_d}}{\partial N_a} \sigma N_a \right)^2}{\sigma^2 N_d}, \varepsilon_{\kappa} = \frac{\left(\frac{\partial \overline{N_d}}{\partial \kappa} \sigma \kappa \right)^2}{\sigma^2 N_d}, \varepsilon_{\sigma_w} = \frac{\left(\frac{\partial \overline{N_d}}{\partial \sigma_w} \sigma w \right)^2}{\sigma^2 N_d} \quad (2)$$

176
 177
 178 **3. Results and Discussion**

179 *3.1. Particle composition and size distribution*

180 For the determination of the different aerosol species present, neutral and acidic sulfate salts are
 181 distinguished by the molar ratio of ammonium to sulfate ions in the aerosol. A molar ratio higher than 2
 182 indicates the presence of only ammonium sulfate, while values between 1 and 2 indicate a mixture of
 183 ammonium sulfate and bisulfate (Seinfeld and Pandis, 1998). For most flights, the molar ratio of ammonium
 184 versus sulfate was above 2 (mean value 2.41 ± 0.72 and median of 2.06). For the nighttime flights, the values
 185 were somewhat lower (mean value 1.91 ± 0.42 and median of 1.85). Nevertheless, ammonium sulfate is
 186 always the predominant sulfate salt. Organic mass fractions for the SENEX research flights are provided in
 187 Table 1. Overall, organic aerosol dominated the composition during all flights, contributing 66%-75% of
 188 the total aerosol volume. Most of the remaining aerosol volume consisted of ammonium sulfate, ranging
 189 from 12%-39% (with a mean of $23\% \pm 6\%$). The organic mass fraction during the flights varied with height
 190 (see Figure 1). This vertical variability of the chemical composition can have a strong impact on droplet
 191 number within the boundary layer, as air masses from aloft may descend and interact with that underneath.
 192 Figure 1 represents the organic mass fractions during Flights 6, 12 and 16, with all flights provided in the
 193 supplementary material (Figure S1). The dashed line represents the boundary layer for daytime (1200 m;
 194 Wagner et al., 2015) and for nighttime (500 m; Seidel et al., 2012). The lowest organic mass fractions
 195 overall were observed during Flight 12 ($36\% \pm 10\%$ with values almost two-fold higher for altitudes >3000
 196 m, Figure 1b) while the highest organic mass fractions were observed during flights over predominantly
 197 rural areas (Flights 5, 10, and 16 (Figure 1c)). During Flight 5 the organic mass fraction was high
 198 ($68\% \pm 5\%$), with the highest values found in the free troposphere at altitudes >3000 m, as was the case for
 199 4 other flights (5/3 in total, Fig. S1). High organic mass fractions were also found during nighttime Flight

200 9 that included portions of the Atlanta metropolitan area, with values up to 78%. The impact of the aerosol
201 composition variability on droplet number is discussed in section 3.2.

202 The predominance of the organic fraction is also reflected in the hygroscopicity parameter values, with an
203 overall $\kappa = 0.25 \pm 0.05$, close to the proposed global average of 0.3 (Pringle et al., 2010). The highest values
204 of κ , as expected are observed during flights exhibiting the lowest organic mass fraction, namely Flight 12
205 with a $\kappa = 0.39$ (Table 1). The rest of the κ -values are close to the overall value of 0.25, corresponding to
206 an organic mass fraction of around 0.60.

207 Median aerosol size distributions and the respective total aerosol number are obtained from the median and
208 interquartile range in each size bin from the aerosol size distribution measurements during segments where
209 the aircraft flew at a constant height. Aerosol size distributions and changes in them during each flight are
210 crucial as they are used as input for the droplet number parameterization. Overall, N_a ranged from around
211 500 to over 100000 cm^{-3} with number size distributions varying markedly over the course of a flight (Figure
212 2). Free tropospheric distributions exhibited characteristics of a bimodal distribution with a prominent broad
213 accumulation mode peak (80-200 nm) and an Aitken mode peak (30-60 nm) (Fig. 2a) while boundary layer
214 size distributions exhibited a more prominent accumulation mode (Fig. 2b). There was considerable
215 variability in the contributions of the nucleation, Aitken, and accumulation modes to total N_a , depending on
216 altitude and proximity to aerosol sources (Fig. 2c). Nevertheless, the modal diameters did not vary
217 considerably, dictating that mostly particles in the same mode will activate, depending on the developed
218 supersaturation. Distributions during nighttime flights exhibited similar total N_a and variability between
219 them; nevertheless, size distributions were more complex - exhibiting up to three distinct modes (20-40,
220 70-100 and 130-200 nm; Fig. 2d).

221 *3.2 Potential cloud droplet number and maximum supersaturation*

222 We first focus on calculation of the potential N_d and S_{max} for data from all thirteen research flights and for
223 the four prescribed values σ_w that represent the observed range. These calculations are helpful in
224 understanding the sensitivity of droplet formation to N_a and κ for all the airmasses sampled – without
225 considering the added variability induced by changes in turbulence expressed by σ_w (considered later).
226 Results from this analysis are provided in Table 3. The highest N_d were found for Flights 6 and 10, which
227 correspond to ambient conditions with the highest N_a , consistent with the sampling of the Atlanta urban
228 environment. For a given σ_w , the variance of N_d is predominantly caused by changes in N_a rather than
229 changes in hygroscopicity (i.e., chemical composition). The highest influence of κ to N_d variability is found
230 for Flight 18 (12% and 35% for 0.1 and 0.3 m s^{-1} , respectively), during which N_a was the lowest, and the
231 organic mass fraction was $\sim 50\%$. The contribution of κ to the N_d variability is as high as 37% (for 0.6 m s^{-1});
232 despite this large contribution, droplet formation is usually considerably more sensitive to changes in

233 aerosol concentration than to variations in composition. Overall, the relative contribution of the
234 hygroscopicity to the variation of N_d increases from $5\pm 3\%$ for $\sigma_w = 0.1 \text{ m s}^{-1}$, to $12.3\pm 8\%$ for $\sigma_w = 0.3 \text{ m s}^{-1}$,
235 to $14.5\pm 10\%$ for $\sigma_w = 0.3 \text{ m s}^{-1}$ and $16.5\pm 9\%$ for $\sigma_w = 1 \text{ m s}^{-1}$. As σ_w increases, so does supersaturation and
236 consequently 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
237 increased from 0.3 to 0.6 m s^{-1} and another 39% as σ_w increased from 0.6 to 1 m s^{-1} . Tripling σ_w from 0.1 to
238 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
239 in S_{max} and a further σ_w increase to 1 m s^{-1} leads to an additional 20.7% increase in S_{max} .

240 Considering the changes in vertical velocity between flights (Table 4), we observe that average σ_w during
241 daytime varied little between flights and was large, ranging between 0.85 and 1.2 m s^{-1} with a mean of
242 $0.97\pm 0.21 \text{ m s}^{-1}$. Under such conditions, water availability during droplet formation is aerosol-limited, so
243 that N_d is sensitive to N_a . The degree of water availability is expressed by the S_{max} , which for all the evaluated
244 SENEX data, is $0.14\pm 0.05\%$. This level of maximum supersaturation activates particles of roughly $>90 \text{ nm}$
245 diameter (e.g., accumulation mode particles) into cloud droplets. The highest S_{max} ranged from 0.2 to 0.3%
246 and was found during flights which exhibited large and highly variable σ_w (Flights 4, 5, 12 and 19) while
247 the lowest S_{max} was below 0.10% and was found during nighttime flights (Flights 9, 15 and 16). Contrasts
248 in droplet formation between day and nighttime conditions may be driven primarily by the total aerosol
249 number in the accumulation mode, and not be affected by ultrafine particles.

250 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
251 the diurnal variability in N_d . To understand the relative importance of all parameters affecting droplet
252 formation (σ_w , N_a , κ) we estimate their contribution to the total variability in N_d based on the variances of
253 κ , N_a and σ_w and the sensitivity of droplet formation to those parameters. The results of the analysis are
254 summarized in Table 4. The σ_w variation during nighttime, although small (always less than 10%),
255 consistently remains an important contributor to N_d variability, because droplet formation tends to be in the
256 updraft velocity-limited regime. At higher values of σ_w (Table 4), the contribution of N_a variability becomes
257 a relatively dominant contributor to N_d variability.

258 Another way to express the importance of vertical velocity and aerosol number for the levels of droplet
259 number is to compare flights where aerosol number or σ_w vary in a similar way. For this, we focus on two
260 day/night flight pairs (Flights 5 and 15, and Flights 6 and 9), shown in Fig. 3. The first pair of flights were
261 conducted over a rural area under moderate aerosol number conditions, while the second pair exhibited
262 somewhat higher aerosol numbers owing to its proximity to the Atlanta metropolitan area. The size of the
263 markers in Fig.3 represents the potential number of droplets in clouds forming in each air mass sampled,
264 while their color reflects the respective total aerosol number. In both pairs of flights, σ_w varies about the
265 same between night and day (Table 4). For the Flights 5, 15 pair, the difference in N_d between day and night

266 (which is 69% higher during daytime) is driven primarily by aerosol ~~characteristics-concentrations with κ~~
267 ~~having limited influence~~ (69% by N_a and 7% from κ) and only 24% by σ_w . For nighttime (Flight 15), the
268 ~~majority of~~ variability in N_d is driven ~~58% once more again~~ by aerosol ~~concentrations and to a limited extent~~
269 ~~only κ~~ (51% by N_a and 7% from κ) and 42% by σ_w . For the second pair of night/day flights (Flights 6, 9),
270 N_a is on average similar, σ_w varies by a factor of 4.0 between day and night and κ varies by 13%.

271 The difference in N_d between day and night (where daytime values are 72.1% higher than nighttime) is
272 almost equally driven by N_a and σ_w changes during the day (54% and 43% respectively), while
273 predominantly driven by N_a during the night (76%; Table 4). Overall, in the proximity of an urban
274 environment with higher aerosol concentrations, 57% of the N_d variability is driven by aerosol (N_a and κ)
275 during the day and 83% during the night. ~~Figure 3 presents the calculated N_d for the four aforementioned~~
276 ~~flights, namely Flights 5 (Fig. 3a), 15 (Fig. 3b), 6 (Fig. 3c) and 9 (Fig. 3d) using the observed σ_w . The size~~
277 ~~of the markers represents the estimated number of droplets, while the color scale the respective total aerosol~~
278 ~~number. Droplet number is lower during nighttime owing to the limited turbulence, which results in a lower~~
279 ~~σ_w .~~

280 Figure 4 shows N_d relative to N_a for flights conducted in the two aforementioned areas, during day (Flights
281 5 and 15) and night (Flights 6 and 9). For high enough N_a , N_d becomes insensitive to additional amounts of
282 aerosol and reaches a “limiting” N_d , which Kacarab et al. (2020) denotes as N_d^{lim} . This limit in N_d is reached
283 when the competition for water vapor to form droplets is ~~strong-high~~ enough to inhibit the formation of
284 droplets with further increase in N_a . The intense competition for water vapor is reflected in the low value
285 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
286 vapor during droplet formation is driven by σ_w , hence droplet formation is limited by σ_w and thus by
287 velocity, when N_d approaches N_d^{lim} . Figure 5 illustrates these effects, by presenting the relationship between
288 N_a and N_d for “low” w^* ($<0.25 \text{ ms}^{-1}$; upper panel), “medium” w^* ($0.5\text{-}0.7 \text{ ms}^{-1}$; middle panel), and “high”
289 w^* ($0.75\text{-}1 \text{ ms}^{-1}$; lower panel) for all flights. Under low w^* conditions, changes in N_a ~~does~~ not result in an
290 ~~important-large~~ change in N_d , so its value corresponds to N_{lim} . When w^* increases to “medium” values
291 (Figure 5b), then N_d becomes sensitive to N_a , which is further amplified at “high” values of w^* (Figure 5c).
292 The covariance of aerosol number and vertical velocity (Figure S3) means that the latter significantly
293 enhances the inherent response of N_d to N_a , which points to the importance of constraining vertical velocity
294 and its variance to correctly capture the aerosol-cloud droplet relationship in any model. The covariance,
295 also observed in other environments (e.g., Kacarab et al., 2020) may be a result of more effective convective
296 transfer of aerosol-rich air to cloud forming regions, but requires further investigation.

297 Analysis of Figure 4 also shows that N_d^{lim} varies between 1200 cm^{-3} during day and around 350 cm^{-3} during
298 night, which points to its strong dependence on σ_w . Indeed, when the N_d^{lim} for all flights (except Flights 4,

309 12, for which insufficient aerosol is present to reach N_d^{lim}) is expressed as a function of σ_w , a remarkable
300 correlation emerges between the two parameters (Figure 6). Even more interesting is that this relationship
301 is quantitatively similar to the corresponding $N_d^{\text{lim}} - \sigma_w$ relationship Kacarab et al. (2020) found for biomass
302 burning - influenced boundary layer clouds in the Southeast Atlantic. The implication of the $N_d^{\text{lim}} - \sigma_w$
303 relationship, and its potential universality, is that when N_d approaches N_d^{lim} , its variability is a reflection of
304 vertical velocity variability alone, not variability in N_a . This opens up the possibility to infer the vertical
305 velocity distribution from the droplet number concentration in regions where considerable amounts of
306 aerosol are present.

307

308 **4. Summary and Conclusions**

309 Measurements of vertical wind velocity, ambient temperature, humidity, aerosol number size distribution
310 and composition in the SEUS obtained during the SENEX 2013 project are used to analyze the drivers of
311 droplet formation. Overall, 13 research flights are studied, covering environments over sectors with
312 different aerosol sources, aerosol number, size distribution, chemical composition and updraft velocity.
313 Aerosol volume is largely dominated by an organic fraction resulting in an estimated hygroscopicity of
314 0.25 ± 0.05 .

315 Based on the calculation of cloud droplet number concentration (N_d) and maximum supersaturation (S_{max}),
316 we find that at the regional scale, N_d variability is largely driven by fluctuations in N_a (Table 4), in
317 accordance with other recent studies (e.g., Fanourgakis et al., 2019; Kalkavouras et al., 2019; Kacarab et
318 al., 2020). Nonetheless, N_d levels are also sensitive to vertical velocity variations, σ_w ; a factor of 4.0 change
319 in σ_w on its own may lead to an almost proportional change in N_d (factor of 3.6). These responses however
320 occur over the diurnal timescale, during which N_a also changes; the covariance between σ_w with N_a enhances
321 the apparent response of N_d to changes in N_a levels by a factor of 5 (Figure 4). In “cleaner” environments
322 where total aerosol number is not impacted by local sources, the relative response of N_d to σ_w is almost
323 twice as great at night than during the day (24% for daytime Flight 5 vs. 42% for nighttime Flight 15). On
324 the other hand, the relative response of N_d to N_a is slightly lower during the night than during the day (51%
325 at night vs. 69% during the day). In environments with elevated concentrations of accumulation-mode
326 particles, the majority of N_d variations can be attributed to changes in N_a and to a lesser extent to changes
327 in σ_w . Variations in chemical composition (expressed by κ) do not contribute substantially to droplet
328 number variability in most cases. As expected, S_{max} partially mitigates the response of N_d to N_a . Overall,
329 maximum supersaturation levels remain quite low ($0.14 \pm 0.05\%$) with the lowest levels ($0.05 \pm 0.1\%$)
330 estimated closest to surface. As a result, particles with diameters >90 nm were the most substantial
331 contributors to CCN.

332 Our analysis also reveals the importance of the variance in vertical velocity as a key driver of cloud droplet
333 formation and its variability in the region. When the boundary layer turbulence is low (e.g. during
334 nighttime) and water vapor supersaturations are low, σ_w and as a consequence vertical velocity variability,
335 can be as important of a contributor to N_d as is N_a . Even for situations with both high σ_w and N_a , the large
336 σ_w enables enough water vapor availability to activate significant numbers of particles to cloud droplets.
337 On average, the two variables (N_a and σ_w) contribute almost equally to the variability in N_d and together
338 account for more than 90% of variability. This finding is consistent with recent modeling studies noting the
339 importance of vertical velocity variability as a driver of the temporal variability of global hydrometeor
340 concentration (Morales Betancourt and Nenes, 2014b; Sullivan et al., 2016). Furthermore, the N_d response
341 from changes in N_a is magnified up to 5 times by correlated changes in σ_w . A similar situation was seen in
342 marine boundary layers influenced by biomass burning in the Southeast Atlantic (Kacarab et al., 2020).
343 Finally, we identify an upper limit to the number of droplets that can form in clouds which depends only
344 on σ_w . This upper limit value tends to be achieved near the surface, where N_a tends to be higher. Whenever
345 N_d approach this upper limit, observed droplet variability is driven by σ_w and as a consequence by vertical
346 velocity changes only.

347 Many aspects of warm cloud physics and especially droplet formation have known for decades. Ensuring
348 that global models simulate N_d for the “right reasons” (i.e., aerosol variability and/or vertical velocity
349 variability) is critical for constraining aerosol-cloud-climate interactions. Our study provides important
350 constraints on the relationships between σ_w , N_a , potential N_d , and S_{\max} , and shows the importance of
351 covariance between σ_w and N_a in controlling the maximum N_d that can result from a given value of σ_w .
352 Given that global model assessments of aerosol–cloud-climate interactions do not evaluate for vertical
353 velocity or its covariance with other parameters, our work shows that this omission can lead to an
354 underappreciated source of hydrometeor variability and bias, and to a biased response of droplet number to
355 aerosol perturbations.

356

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

361

362 **Author Contributions:** AB and AN designed and initiated the study. Analysis methodology and software
363 were provided by AN. The analysis was carried out by AB and AN, with comments from JJL, CB, AMM.

364 Data curation was provided by AB, JLL, CB, JAG, JL, AMM, AW. The manuscript was written by AB and
365 AN with additional comments provided by JLL, CB, AW, AMM. All authors reviewed the manuscript.

366

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374

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376

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

Flight	Date	Local Time (UTC-5)	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

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Table 2: Flight number, time interval, 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 Interval (Local Time)	σ_w (m s ⁻¹)	w^* (m s ⁻¹)	Altitude a.s.l. (m)	Flight (pass)	Time 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	0.25	0.20	797±2.01
5 (2)	13:16-13:29	0.82	0.65	982±11	9 (2)	19:20-19:29	0.25	0.2	740±1.23
5 (3)	13:34-13:50	1.01	0.80	502±13	9 (3)	19:33-19:48	0.22	0.17	740±1.23
5 (4)	13:53-14:08	1.03	0.81	614±27	9 (4)	19:51-20:25	0.22	0.17	776±1.22
5 (5)	14:20-15:00	0.91	0.72	603±40	9 (5)	20:34-20:39	0.23	0.18	597±1.19
5 (6)	15:35-15:41	0.87	0.69	533±18	<u>9 (6)</u>	<u>20:44-20:52</u>	<u>0.16</u>	<u>0.12</u>	<u>484±1.14</u>
5 (7)	16:17-16:30	0.77	0.61	638±23	9 (7)	20:56-21:10	0.20	0.16	773±1.11
5 (8)	16:31-16:39	0.55	0.44	559±18	9 (8)	21:31-21:45	0.19	0.15	725±1.18
5 (9)	17:10-17:22	0.53	0.42	686±40	9 (9)	22:24-22:31	0.26	0.20	745± 1.36
					9 (10)	22:48-22:54	0.22	0.17	804± 1.37
14 (1)	12:34-12:49	0.94	0.75	558±2	15 (1)	21:09-21:52	0.24	0.19	505±6.64
14 (2)	13:57-14:17	0.97	0.77	658±3	15 (2)	22:19-22:31	0.30	0.24	633±1.21
14 (3)	14:22-14:46	0.95	0.75	737±3	15 (3)	22:42-22:54	0.25	0.20	600±1.17
14 (4)	14:58-15:33	0.55	0.43	746±23	15 (4)	23:26-23:37	0.33	0.26	908±1.56
14 (5)	15:55-16:08	0.57	0.45	714±3	15 (5)	00:02-00:19	0.30	0.23	1208±1.23
14 (6)	16:11-16:21	0.77	0.61	801±3	15 (6)	00:43-1:08	0.25	0.20	592±1.37
14 (7)	16:33-16:41	0.45	0.35	793± 2	15 (7)	1:10-1:24	0.28	0.22	676±1.02
					15 (8)	1:37-2:02	0.21	0.16	713±19.5
12 (1)	11:50-12:34	0.96	0.75	484±3	19 (1)	11:20-11:41	0.62	0.49	1014±2.27
12 (2)	12:48-13:18	1.09	0.86	503±3	19 (2)	12:09-12:23	1.20	0.95	652±3.34
12 (3)	13:34-13:50	1.12	0.88	894±3	19 (3)	12:51-13:10	0.87	0.69	537±2.51
12 (4)	14:06-14:40	1.04	0.82	479±4	19 (4)	13:22-13:49	1.29	1.02	518±22.6
12 (5)	15:21-15:32	1.10	0.87	521±3	19 (5)	14:44-14:57	1.36	1.07	528±3.26
12 (6)	15:43-16:02	0.99	0.78	475±3	19 (6)	15:04-16:06	0.90	0.71	524±2.8

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570 **Table 3:** Derived cloud parameters (maximum supersaturation, droplet number) and relative contribution (Equation 2) of chemical composition
571 and total aerosol number for different vertical velocities. Numbers in parentheses indicate standard deviation values. The symbol “☼” next to each
572 flight number refers to daytime flight, and “☾” refers to a nighttime flight.

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Flight	N_a	Std <i>Dev</i> N_a	$\sigma_w=0.1 \text{ m s}^{-1}$				$\sigma_w=0.3 \text{ m s}^{-1}$				$\sigma_w=0.6 \text{ m s}^{-1}$				$\sigma_w=1.0 \text{ m s}^{-1}$			
			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	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

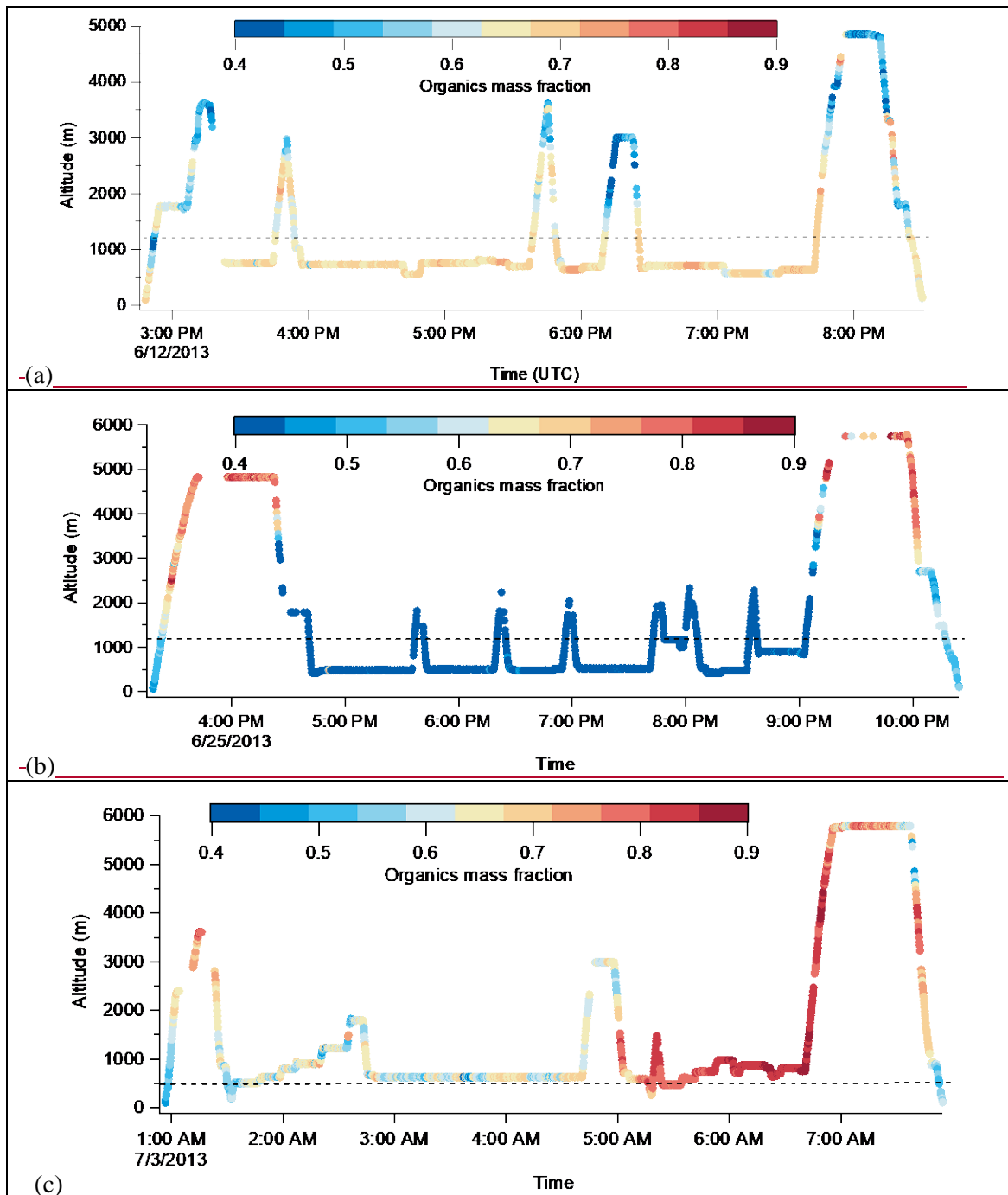
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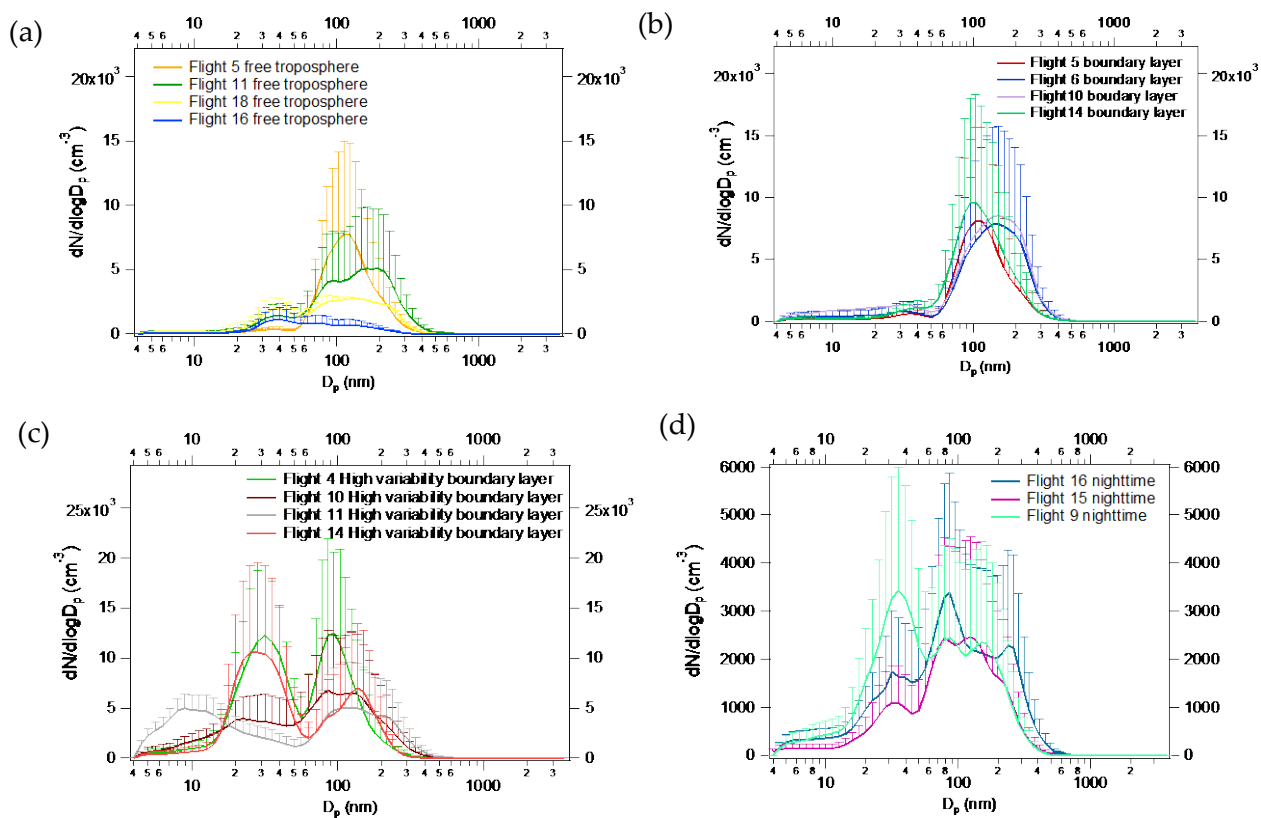
576 **Table 4:** Derived S_{max} , N_d , σ_w for all research flights along with the estimated contribution of each
577 parameter to the variability of the droplet number. The symbol “☼” next to each flight number refers to
578 daytime flight, and “☾” refers to a nighttime flight.

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%
Average					0.334	4%	75.2%	20.6%

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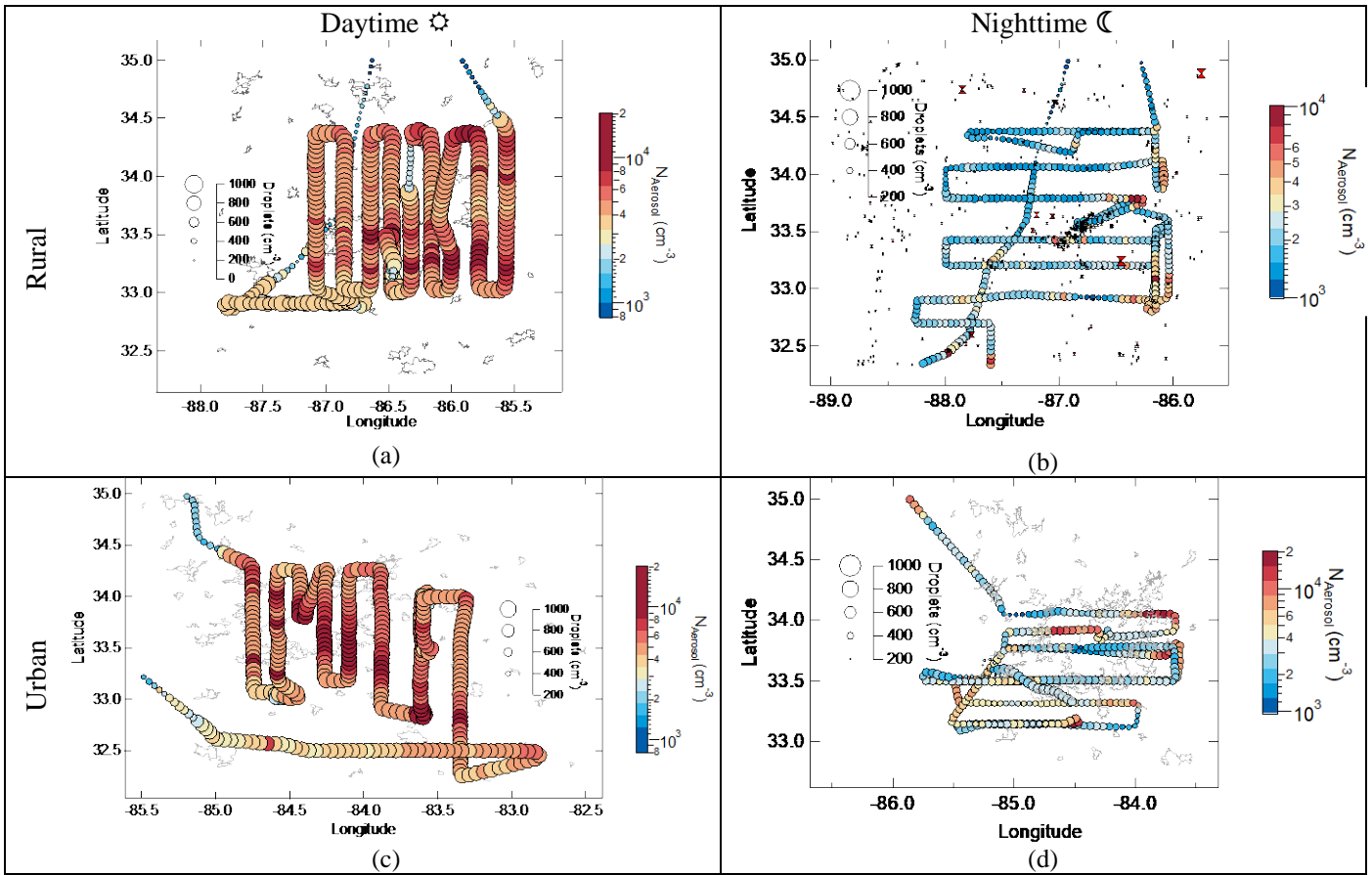
580 **Figure 1:** Altitude as a function of time (UTC) colored by organic mass fraction. Spatial and vertical
581 distribution of the organics mass fraction (a) for Flight 6, (b) for Flight 12 and (c) for Flight 16, denoting
582 the difference in chemical composition, which in turn, may influence cloud droplet number concentration.
583 The dashed line represents the boundary layer height, [derived from Wagner et al., \(2015\) for daytime \(1200](#)
584 [m\) and from Seidel et al., \(2012\) for nighttime \(500 m\).](#)



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

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

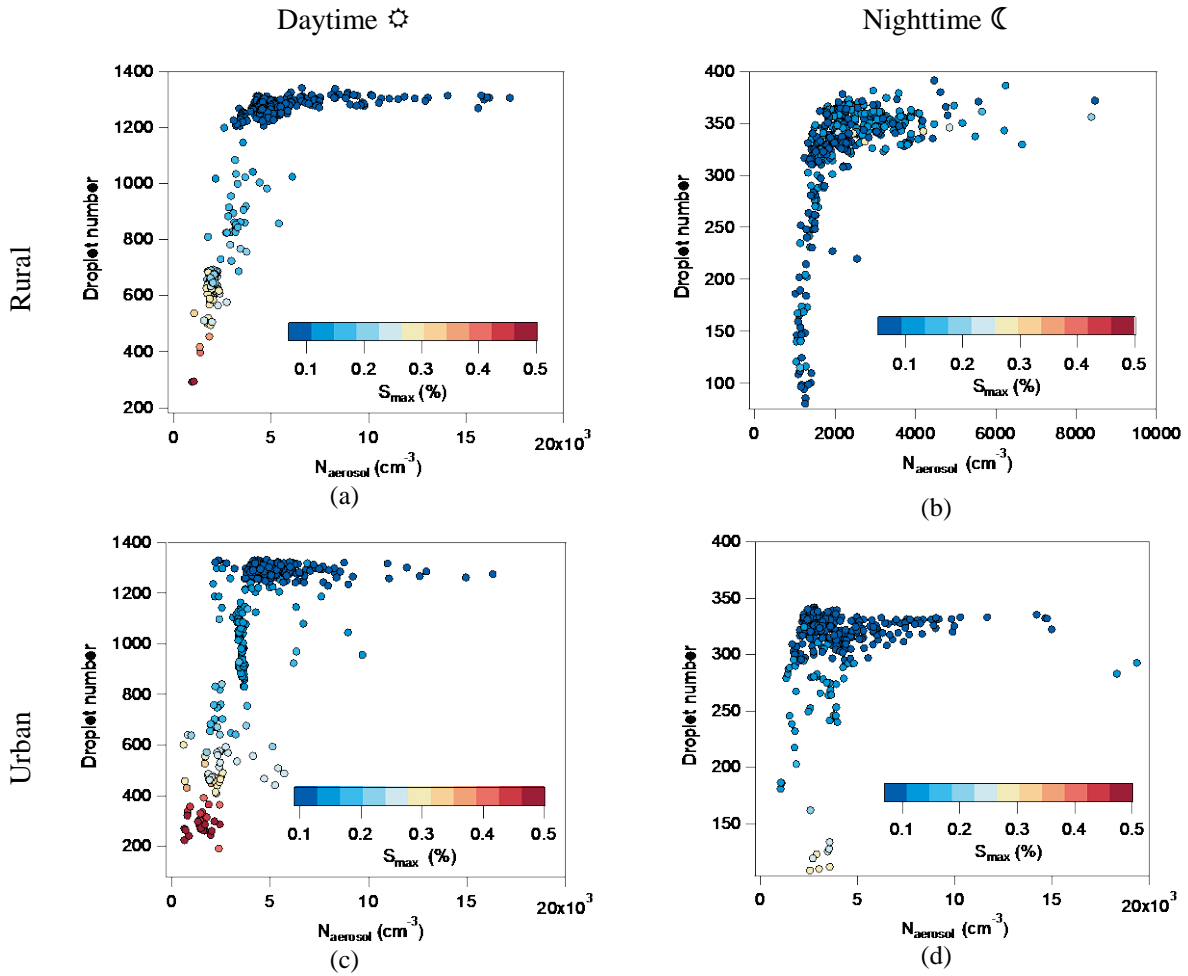
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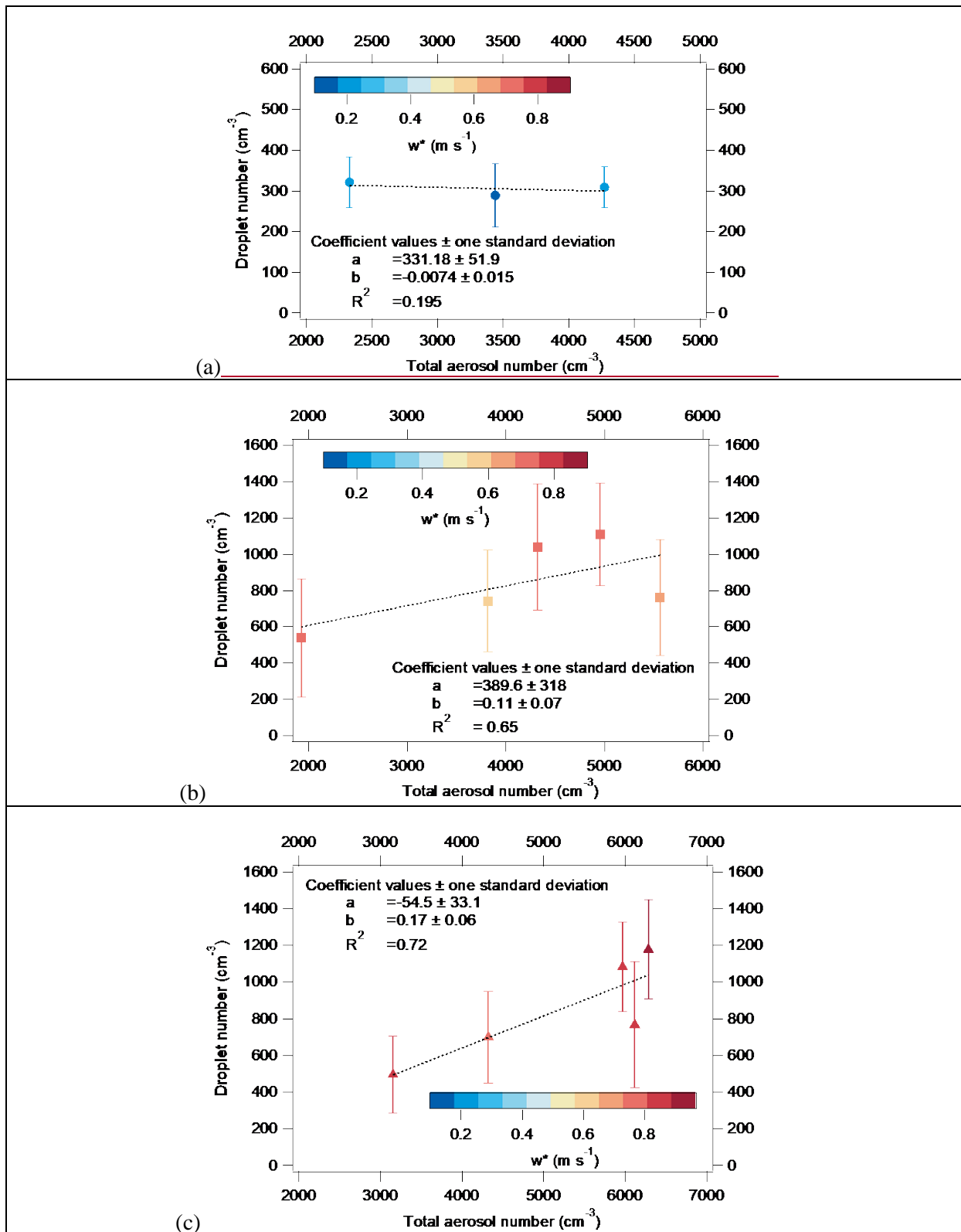
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602 **Figure 4:** Cloud droplet number vs. total aerosol number for the derived characteristic vertical velocity
 603 (w^*) of each flight (Table 4). (a) for the rural sector during daytime (Flight 5) and (b) nighttime (Flight 15).
 604 (c) for urban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

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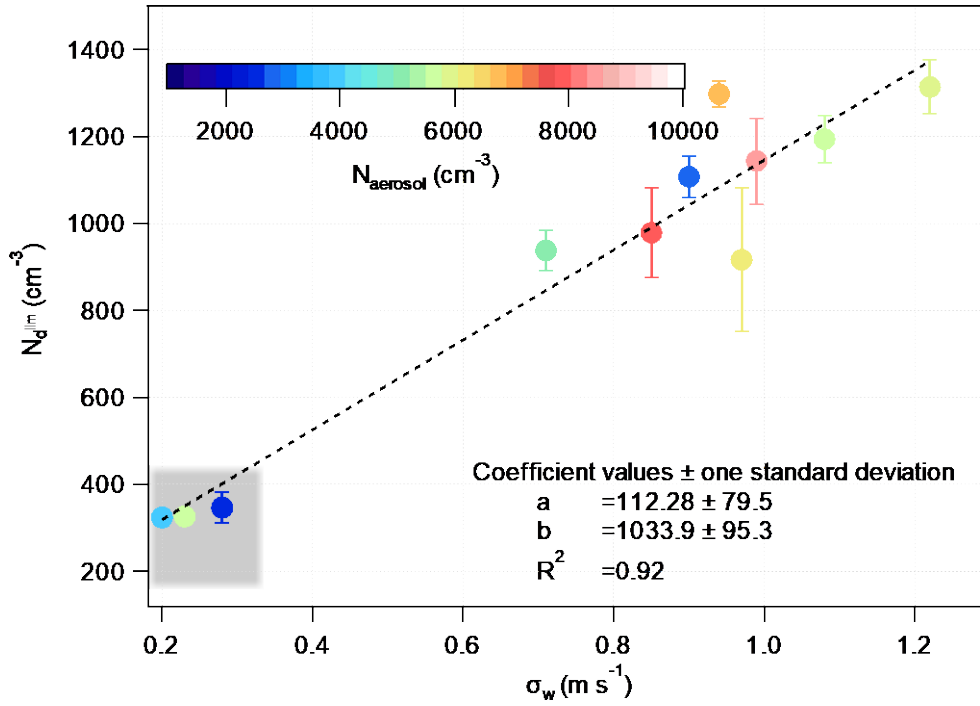
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610 **Figure 5:** Average cloud droplet number vs. total aerosol number, colored by characteristic velocity w^*

611 for each flight. Error bars represent the standard deviation of cloud droplet number during each flight.



612

613 **Figure 6:** Limiting droplet number vs. standard deviation of vertical velocity during flights where a
 614 velocity-limited regime is reached (all except Flights 4, 12). The shaded area represents the segments of the
 615 flights conducted during nighttime while color scale denotes total aerosol number levels.

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