

1 **Aerosol Optical Properties in the Southeastern United**
2 **States in Summer-Part 2: Sensitivity of Aerosol Optical**
3 **Depth to Relative Humidity and Aerosol Parameters**

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23

24 **Abstract**

25 Aircraft observations of meteorological, trace gas, and aerosol properties were made between
26 May and September 2013 in the southeastern United States (U.S.). Regionally representative
27 aggregate vertical profiles of median and interdecile ranges of the measured parameters were
28 constructed from 37 individual aircraft profiles made in the afternoon when a well-mixed

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1 boundary layer with typical fair-weather cumulus was present (Wagner et al., 2015). We use
2 these 0-4 km aggregate profiles and a simple model to calculate the sensitivity of aerosol
3 optical depth (AOD) to changes in dry aerosol mass, relative humidity, mixed layer height,
4 the central diameter and width of the particle size distribution, hygroscopicity, and dry and
5 wet refractive index, while holding the other parameters constant. The calculated sensitivity is
6 a result of both the intrinsic sensitivity and the observed range of variation of these
7 parameters. These observationally based sensitivity studies indicate that the relationship
8 between AOD and dry aerosol mass in these conditions in the southeastern U.S. can be highly
9 variable and is especially sensitive to relative humidity (RH). For example, calculated AOD
10 ranged from 0.137 to 0.305 as the RH was varied between the 10th and 90th percentile
11 profiles with dry aerosol mass held constant. Calculated AOD was somewhat less sensitive to
12 aerosol hygroscopicity, mean size, and geometric standard deviation, σ_g . However, some
13 chemistry-climate models prescribe values of σ_g substantially larger than we or others
14 observe, leading to potential high biases in model-calculated AOD of ~25%. Finally, AOD
15 was least sensitive to observed variations in dry and wet aerosol refractive index and to
16 changes in the height of the well-mixed surface layer. We expect these findings to be
17 applicable to other moderately polluted and background continental airmasses in which an
18 accumulation mode between 0.1-0.5 μm diameter dominates aerosol extinction.

19

20 **1 Introduction**

21 Aerosols in the atmosphere scatter and absorb solar radiation and alter the earth's energy
22 balance. The magnitude and variation of this aerosol direct radiative effect has large
23 uncertainties that are being addressed by numerous observational and modeling studies. Key
24 to these investigations, measurements of AOD, the vertically integrated aerosol extinction
25 coefficient (σ_{ext}), provide information on the spatial and temporal distribution of the
26 atmospheric aerosol. Long-term remote-sensing measurements of AOD made by ground-
27 based networks such as the AERONET sunphotometers (Holben et al., 2001) and by space-
28 based instruments (Kahn, 2011), have produced a global record showing the spatial and
29 temporal variation of AOD. These observations are often used to evaluate and constrain earth
30 system models that simulate aerosol emissions, formation, processing, and removal. For such
31 comparisons, the models must convert their simulated aerosol parameters to optical
32 extinction, then vertically integrate to compare with measured AOD values. These models

1 typically track dry aerosol mass using bulk, modal, or binned microphysical schemes, then
2 calculate ambient extinction based on assumed or simulated particle diameter, width of the
3 size distribution, and the hygroscopic uptake of water (Liu et al., 2012). Conversely, there are
4 active efforts to assimilate AOD measurements for use by air quality models that predict dry
5 aerosol mass (usually $PM_{2.5}$, aerosol mass of particles smaller than 2.5 μm diameter;
6 Benedetti et al., 2009; Saide et al., 2014). In these cases the remotely sensed AOD
7 measurements must be converted to an in situ dry mass concentration at a specific altitude,
8 usually using prescribed aerosol characteristics based on limited prior in situ measurements.

9 Aerosol optical depth is dependent upon several aerosol characteristics in addition to mass,
10 the parameter that is often of interest. Many particles are composed of compounds that can
11 take up water with increasing atmospheric relative humidity (RH). This hygroscopic water
12 uptake changes particle size and refractive index and can lead to dramatic changes in the
13 extinction as a function of RH, even when dry aerosol mass is constant. Since atmospheric
14 RH is highly variable temporally, horizontally, and especially vertically, aerosol water plays
15 an important role in establishing the relationship between ambient extinction (or AOD) and
16 dry aerosol mass. As van Donkelaar et al. (2015) succinctly state, "the relationship between
17 AOD and [ground-level] $PM_{2.5}$ depends on aerosol vertical distribution, humidity, and aerosol
18 composition, which are impacted by changes in meteorology and emissions." Perhaps less
19 recognized by some researchers, aerosol extinction is also quite sensitive to particle diameter
20 because the extinction cross-section increases sharply with increasing diameter. Similarly, the
21 width of the size distribution (usually described by the geometric standard deviation, σ_g)
22 describes how particle concentration varies as a function of diameter, and thus affects the
23 optical extinction for a given aerosol mass concentration. The refractive index of the particles,
24 influenced by the aerosol water content, also affects the amount of extinction produced by a
25 particle of a given total mass.

26 Globally averaged, dust, sea-salt, biomass burning, and anthropogenic aerosols dominate
27 AOD (e.g., Boucher et al., 2013; Jacobson, 2001). Between and within each of these aerosol
28 categories there are substantial variations in particle diameter and shape, hygroscopicity, size
29 distribution width, mixing state, and refractive index, as well as in the vertical distribution of
30 these properties. Because of these confounding influences, the relationship between AOD and
31 dry aerosol mass is expected to vary in different regions and seasons.

1 Several studies have examined the relationship between detailed aerosol characteristics and
2 the direct aerosol radiative effect, ambient extinction, or AOD. Hegg et al. (1993) examined
3 the sensitivity of ambient extinction to particle diameter and refractive index. They found that
4 extinction was particularly sensitive to the initial dry size of the aerosol prior to hygroscopic
5 growth. McComiskey et al. (2008) evaluated in detail how aerosol intensive properties
6 affected the top-of-atmosphere and surface radiation for a wide range of aerosol types, finding
7 the greatest sensitivity to the aerosol single scatter albedo. Magi et al. (2005) used airborne in
8 situ measurements in the eastern U.S. to estimate the contribution of dry particulate
9 constituents and aerosol water to AOD. Koloutsou-Vakakis et al. (1998) found that aerosol
10 composition and hygroscopicity were important in relating aerosol mass concentration
11 measurements to ambient scattering. Using airborne and remote sensing measurements,
12 Crumeyrolle et al. (2014) showed a strong relationship between AOD and surface and in situ
13 aerosol mass concentrations in the eastern U.S. Ziemba et al. (2013) report good closure
14 between remotely sensed profiles of aerosol extinction and in situ measurements taken in the
15 eastern U.S. when aerosol hygroscopic growth was taken into account. Esteve et al. (2012)
16 found that uncertainty in hygroscopic growth was likely the largest contributor of
17 discrepancies between AOD determined from remote sensing and from in situ measurements.
18 Esteve et al. (2016) used measurements and a radiative transfer model to determined that the
19 aerosol direct radiative effect in western Europe in spring was moderately sensitive to the size
20 distribution of the aerosol and less so to the refractive index of the particles. Several global
21 modeling studies have found strong sensitivities of the direct aerosol radiative effect to
22 particle size, composition, and hygroscopicity (e.g., Adams et al., 2001; Boucher and
23 Anderson, 1995; Nemesure et al., 1995; Pilinis et al., 1995). Adams et al. (2001) used global
24 model simulations to demonstrate that the water content of the aerosol, especially for
25 $RH > 90\%$, plays an important role in altering the aerosol direct radiative effect, and that
26 hygroscopicity and the RH field must be well described in climate models. More detailed
27 studies using both measurements and modeling suggest that high RH near clouds can
28 substantially enhance the aerosol extinction at spatial scales that are unresolved by climate
29 models and some remote sensing measurements (e.g., Bar-Or et al., 2012; Haywood et al.,
30 1997; Koren et al., 2007; Twohy et al., 2009).

31 In this study we focus on the relationship between measured aerosol properties and calculated
32 AOD for a specific aerosol type, the submicron-dominated mixed organic-sulfate aerosol
33 typical of moderately polluted and background continental air. This type of aerosol is found in

1 several regions globally, including southern Africa, Eurasia, and South America (e.g.,
2 Vakkari et al., 2013; A companion paper (Part 1; Brock et al., 2015) uses detailed in situ
3 airborne measurements of dry aerosol composition, dry size distribution and change in optical
4 extinction as a function of relative humidity, $f(rh)$, to examine the hygroscopicity of the
5 aerosol in this environment. In Brock et al. (2015) it was found that observed $f(rh)$ could be
6 described accurately using a physically based, single-parameter function. The fitted
7 parameter, κ_{ext} , is related to but not identical with the chemically determined κ_{chem} from the κ -
8 Köhler theory of Petters and Kreidenweiss (2007). In Brock et al. (2015) we found that the
9 value of κ_{chem} for the dominant organic component must have been <0.10 to be consistent
10 with the observed $f(RH)$ for $>75\%$ of the cases examined.

11 In this analysis (Part 2), the κ_{ext} parameterization developed in Brock et al. (2015) is used to
12 determine ambient extinction. Vertical profiles of this ambient extinction are then integrated
13 to calculate the AOD from the surface to the top of the profile, and the effects of aerosol
14 mass, hygroscopicity, size distribution, refractive index, and vertical distribution on the AOD
15 are evaluated. The purpose of this effort is to identify which parameters must be well
16 simulated or observed to relate AOD to dry aerosol mass in this and similar environments.
17 Similar studies are needed in regions with other aerosol types to develop a comprehensive
18 understanding of the relationship between AOD, aerosol composition, shape, and size, and
19 atmospheric RH to reduce uncertainty in aerosol radiative effects (Kahn, 2011).

20

21 **2 Methods**

22 **2.1 Instrumentation**

23 We analyze vertical profiles derived from airborne, in situ measurements from the May-July
24 2013 Southeastern Nexus of Air Quality and Climate (SENEX) and the portions of the
25 August-September 2013 Study of Emissions and Atmospheric Composition, Clouds, and
26 Climate Coupling by Regional Surveys (SEAC⁴RS) projects that were made in the
27 southeastern U.S. Details of the instruments, measurements, and methodology for generating
28 regionally representative vertical profiles of aerosol, gas-phase, and meteorological
29 parameters are given by Wagner et al. (2015) and may also be found in Brock et al. (2015).
30 Measurements included the composition of the sub- $0.7\mu\text{m}$ non-refractory composition, the
31 dry particle size distribution from ~ 0.004 to $1.0\mu\text{m}$, and aerosol extinction at 532nm

1 wavelength and three relative humidities (~15%, ~70%, and ~90%) on the humidified branch
2 of deliquescence/efflorescence curve. As described in Brock et al. (2015), the contribution to
3 extinction due to particles with diameters $>0.7\mu\text{m}$ was found to be small and is ignored in this
4 work. All values presented here, except for ambient extinction, have been corrected to
5 standard temperature and pressure (STP) conditions, defined as 1 atmosphere and 273.15K.

6 **2.2 Creating aggregate vertical profiles**

7 Measurements were made in summer during periods when the NASA DC-8 (SEAC⁴RS) and
8 NOAA WP-3D (SENEX) aircraft were sampling the fully developed planetary boundary
9 layer in fair weather cumulus conditions. Such conditions are representative of the
10 summertime lower troposphere in daytime in the southeastern U.S. (Warren et al., 2007). As
11 described in more detail by Wagner et al. (2015), individual profiles made over Mississippi,
12 Alabama, and Georgia in the afternoon between 12:00 and 18:30 Central Daylight Time
13 (CDT) were aggregated into 150m vertical bins. Only 37 profiles (of 74 total) that showed a
14 distinct and easily characterized vertical structure and that were made in the presence of fair-
15 weather cumulus clouds were chosen for analysis. Three layers with distinct aerosol and gas-
16 phase chemical characteristics were evident in the analyzed profiles: 1) a well-mixed layer
17 extending from the surface to the vicinity of cloud base in which short-lived gas-phase
18 species were nearly homogeneously distributed; 2) the free troposphere in the upper portion
19 of the profile, with low mixing ratios of short-lived species and generally lower abundances
20 of pollutants; and 3) a cloud layer, or transition layer, between the well-mixed layer and the
21 free troposphere, displaying intermediate chemical lifetimes and mixing ratios that are a result
22 of mixing between the well-mixed layer and the free troposphere.

23 From this complex vertical structure we wish to calculate representative vertical profiles of
24 aerosol and meteorological parameters. However, direct altitude-based averaging of the
25 individual profiles would combine air from the well-mixed layer, the transition layer, and the
26 free troposphere because the heights of these layers varied from profile to profile. To avoid
27 this problem, Wagner et al. (2015) defined a normalized altitude, h_{norm} , for each profile such
28 that the top of the mixed layer, h_{ML} , is assigned a normalized altitude of 1, and the top of the
29 transition layer, h_{TL} , is assigned a normalized altitude of 2:

30 $0 < h < h_{ML}, h_{norm} = h / h_{ML}$

$$1 \quad h_{ML} < h < h_{TL}, \quad h_{norm} = 1 + (h - h_{ML}) / (h_{TL} - h_{ML}) \quad (1)$$

$$2 \quad h > h_{TL}, \quad h_{norm} = 1 + h / h_{TL}$$

3 For each profile h_{ML} was defined as the highest altitude at which the virtual potential
 4 temperature was constant and above which there was a rapid reduction in the isoprene
 5 concentration. The value of h_{TL} was defined by a temperature inversion and a rapid decrease
 6 in the CO mixing ratio. The individual altitude-normalized profiles were averaged to produce
 7 an aggregate profile for each parameter of interest, with 10 normalized altitude bins in each
 8 layer. These aggregate profiles include median values, as well as the 10th, 25th, 75th, and
 9 90th percentiles of each normalized altitude bin to describe atmospheric variability. To
 10 calculate AOD, the normalized altitudes of the aggregate profiles in Wagner et al. were
 11 converted back to average altitude-based profiles using the median values $h_{ML}=1132$ m and
 12 $h_{TL}=2137$ m above ground level (AGL). The altitude bins of this aggregate profile are in
 13 increments of 113.2 m for the well-mixed layer, 100.5 m in the transition layer, and 213.7 m
 14 in the free troposphere. The resulting aggregate profiles are representative of the summertime,
 15 cumulus-topped fair-weather planetary boundary layer and lower free troposphere of the
 16 southeastern U.S. when the daytime boundary layer is fully developed.

17 2.3 Determining ambient extinction

18 Ambient extinction must be estimated from measurements that are made inside the aircraft
 19 cabin under different thermodynamic conditions than the atmosphere. As described in Brock
 20 et al. (2016), the hygroscopic growth parameter $f(RH)$ is the ratio of ambient extinction $\sigma(RH)$
 21 to extinction measured at the dry ($RH_0 \sim 15\%$) condition, $\sigma(RH_0)$. The value of $f(RH)$ was
 22 calculated for each data point in three different ways. In the first method, κ -Köhler theory was
 23 applied to measurements of aerosol size distribution and composition to predict particle
 24 diameter as a function of RH. Mie theory was then used to predict the ambient extinction
 25 from the deliquesced particle size distribution. In the second method, the observed 3-point
 26 $f(RH)$ values were used to fit a curve of the form

$$27 \quad \frac{\sigma(RH)}{\sigma(RH_0)} \equiv f(RH) = \left[\frac{(100 - RH_0)}{(100 - RH)} \right]^Y \quad (2)$$

28 and the extinction at ambient RH was calculated using the fitted coefficient. Finally, a new
 29 parameterization of the form

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1 $f(RH) = 1 + \kappa_{ext} \frac{RH}{100-RH}$ (3)

2 ~~was fitted to the observed 3-point $f(RH)$ values and the extinction at ambient RH calculated,~~
3 The γ parameterization, Eq. (2), has been widely used in previous studies (e.g., Attwood et al.,
4 2014; Doherty et al, 2005; Kasten, 1969; Massoli et al., 2009; Quinn et al., 2005; Ziemba et
5 al., 2013). However, the γ parameterization did not fit the observed dependence of extinction
6 with RH in the southeastern U.S. as well as did the κ_{ext} parameterization, Eq. (3), which was
7 developed in Brock et al. (2016). In Section 3.3 we examine the sensitivity of calculated AOD
8 to whether γ or κ_{ext} is chosen to parameterize $f(RH)$.

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10 3 Results and analysis

11 3.1 Vertical profiles

12 The 37 individual profiles meeting the criteria described in Sect. 2.2 were combined into an
13 aggregate profile following Eq. (1), from which 10th percentile, median (50th percentile), and
14 90th percentile values ~~were calculated. Because the distribution of most parameters was not~~
15 ~~Gaussian, percentile values are used to represent the range of observed variability.~~ Median
16 values of STP-corrected dry aerosol extinction decreased from $\sim 60 \text{ Mm}^{-1}$ at the bottom of the
17 aggregate profile to $\sim 40 \text{ Mm}^{-1}$ at the top of the transition layer $\sim 2100 \text{ m}$ above ground level
18 (Fig. 1a), with an abrupt decrease to $\sim 10 \text{ Mm}^{-1}$ in the free troposphere. Wagner et al. (2015)
19 used gas-phase and aerosol tracers to show that this profile was the result of a well-mixed
20 layer below cloud base at $\sim 1100 \text{ m}$, a cloud or transition layer between ~ 1100 and $\sim 2100 \text{ m}$,
21 and the free troposphere above $\sim 2100 \text{ m}$. Within the transition layer, Wagner et al. found a
22 small but statistically significant increase of $\sim 15\%$ in aerosol mass above the values expected
23 from mixing alone. This enhancement was composed of roughly equal amounts of sulfate and
24 organic mass and resulted in a higher sulfate mass fraction in this layer compared to the well-
25 mixed layer below. Relative humidity increased from $\sim 60\%$ at the lowest altitudes to a
26 median value of $\sim 80\%$ in the transition layer (Fig. 1b), with lower median RH but greater
27 variability in the free troposphere. Ambient extinction (Fig. 1c) reached a maximum in the
28 transition layer where RH was highest, with 90th percentile values ~ 3 times greater than the
29 median values.

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1 Aerosol optical depth was calculated between the surface and the top of the profile by
2 integrating ambient extinction from the surface upward (Fig. 1d). The extinction within the
3 well-mixed layer was extrapolated to the surface for each individual profile. Wagner et al.
4 (2015) show that measurements of extinction at the Centreville, Alabama surface site during
5 the SENEX time period agreed with values measured at the lowest altitude of the aircraft,
6 supporting such extrapolation. The median AOD at 532 nm was 0.19. This value is similar to
7 values of AOD at 532nm of 0.19 and 0.17 at the AERONET (Holben et al., 2001) locations of
8 Centreville, Alabama (n=268) and Atlanta, Georgia (n=48), respectively. These mean Aeronet
9 AOD values were made between 12:00 and 18:30 local time on the days included in this
10 analysis, and the AOD at 532 nm was logarithmically interpolated using the Ångström
11 exponent from measurements made at 500 and 675 nm. This consistency between the AOD
12 derived from the aircraft in situ measurements and that measured at the AERONET sites
13 indicates that there were not significant aerosol layers above 4 km in most of the profiles
14 measured, that the aggregated profiles are regionally representative, and that lower
15 tropospheric extinction dominated regional AOD. Placed in the context of a multiyear AOD
16 record from the Atlanta AERONET site, the data analyzed here are typical of the summertime
17 maximum in AOD found in the southeastern U.S. (Fig. 2).

18 3.2 Contribution of the well-mixed and transition layers to total AOD

19 As discussed in Wagner et al. (2015), air in the transition, or cloud, layer is depleted in short-
20 lived gas-phase tracers such as isoprene. This depletion in isoprene suggests that air parcel
21 transport between the surface and the transition layer is slow and/or intermittent, and is
22 probably associated with cloud outflow. The transition layer is likely composed of a
23 combination of a residual well-mixed layer from the previous day, air that has been lifted
24 through cloud convection above the current day's well-mixed layer, and free-tropospheric air
25 mixed from above. Because of this relative isolation, the aerosol in the transition layer aloft
26 may be different than that measured at the surface. In cases where the contribution of the
27 transition layer aerosol extinction to AOD is substantial, this segregation between the
28 transition layer and the surface adds uncertainty to efforts to directly relate remotely sensed
29 AOD measurements to surface values, for example for epidemiological studies that use
30 satellite-based AOD measurements as proxies for surface aerosol concentration (e.g.,
31 Crumeyrolle et al., 2014; Engel-Cox et al., 2004; Kim et al., 2015; Kloog et al., 2011; van
32 Donkelaar et al, 2015). Ultimately the transition layer and well-mixed layer aerosols are

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1 coupled through dry and moist convection, but the observed isoprene depletion in the
2 transition layer suggests a substantial temporal lag in the response of that layer to changes to
3 the aerosol in the well-mixed layer.

4 To evaluate the importance of the transition layer to AOD, the contribution of it and the well-
5 mixed layer to total column AOD was examined for each altitude-normalized profile that
6 penetrated both layers. The AOD within the well-mixed and transition layers was then
7 calculated and compared with the total integrated AOD from the profile. The fractional
8 contribution of the free troposphere layer to the total AOD was not calculated because only 5
9 of the 37 profiles penetrated far enough into the free troposphere to reasonably estimate the
10 AOD from this layer. Histograms of the total AOD and the fractional contribution of the well-
11 mixed and transition layers (Fig. 3) show that both layers contributed substantially to the
12 column AOD. The mean fractional contributions of the well-mixed and transition layers to
13 total AOD were 0.56 and 0.43, respectively, while the median fractional contributions were
14 0.54 and 0.43, respectively. These results demonstrate that the transition layer, which is not in
15 immediate contact with the surface, contributed nearly half of the integrated AOD in the
16 southeastern U.S. during the SENEX and SEAC⁴RS measurements. The substantial fraction
17 AOD provided from this layer aloft may affect correlations between surface aerosol
18 concentrations and satellite-derived AOD, and should be investigated more systematically.

19 3.3 Sensitivity of AOD to measured parameters

20 As described in Sect. 3.1, the aggregation of individual vertical profiles results in a single
21 vertical profile and interdecile range that represents typical mid-day conditions in the
22 summertime in the southeastern U.S. This aggregate profile and variability range is used to
23 estimate the sensitivity of the relationship between AOD and dry mass to changes in
24 measured parameters that affect AOD. These sensitivity calculations indicate which
25 parameters are most important to accurately relate AOD and non-water aerosol mass in this
26 region and season.

27 For the sensitivity calculations we use a single-mode lognormal model to describe the size
28 distribution of the optically active accumulation mode aerosol. The geometric mean diameter
29 D_g , geometric standard deviation σ_g , and total particle number concentration N for this
30 lognormal model were calculated from the measured size distributions following Hinds
31 (1999). Prior to calculating these values, the size distributions were corrected using the

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1 refractive index based on the aerosol composition measurements, the composition model of
2 Zaveri et al. (2005), and the simulated response of the UHSAS instrument as a function of
3 refractive index as described in detail in Brock et al. (2015). Mie theory for homogeneous
4 spheres (Bohren and Huffman, 1998) was used to calculate the ambient extinction from the
5 lognormal model distribution. For each sensitivity case the ambient extinction profile was
6 determined using the median profiles of RH, κ_{ext} , D_g , and σ_g and N . To determine the
7 sensitivity of AOD to a particular parameter, the 10th and 90th percentile profiles of the
8 tested parameter were used to recalculate ambient extinction, which was then integrated to
9 determine the 10th and 90th percentile AOD value. All other dry parameters were maintained
10 at the median profile while the one tested parameter was varied. As RH was varied, ambient
11 particle diameter and refractive index were allowed to change due to water uptake and loss
12 using κ -Köhler theory and the κ_{chem} determined from the aerosol composition measurements
13 as described in Brock et al. (2015). The ambient extinction profile was then calculated using
14 Mie theory and the calculated ambient particle size distribution and refractive index. Finally,
15 AOD for that sensitivity case was determined by integrating the vertical profile of calculated
16 ambient extinction.

17 To evaluate the sensitivity of AOD to dry aerosol mass, the AMS mass concentration profiles
18 were calculated and the number of particles in the model size distribution were varied to
19 match the mass concentration. Since D_g and σ_g were held at their median profile values, this
20 simply changed the number concentration of particles, which should produce a linearly
21 proportional change in AOD with dry aerosol mass.

22 Note that these sensitivity tests do not account for co-variance of parameters that might be
23 expected in the atmosphere. For example, larger dry particle diameters might be associated
24 with a more sulfate-rich, more hygroscopic aerosol. The sensitivity evaluations simply
25 describe the first-order response of AOD to changes in the interdecile range of a single
26 parameter, with all other dry parameters being held constant using the median profile for
27 each. More sophisticated model simulations, for example using a large eddy simulation model
28 with aerosol input parameters constrained by observations, could be used to further
29 investigate these sensitivities and the couplings between parameters.

30 The median AOD calculated from the lognormal size distribution profile was 0.18, similar to
31 the value of 0.19 directly determined from the in situ measurements of aerosol extinction. As
32 expected, AOD was linearly sensitive to variations in aerosol mass (Fig. 4, Table 1). Aerosol

1 optical depth was also highly sensitive to RH as it varied between the 10th and 90th percentile
2 profile, with a variation in AOD of +72% and -23% relative to the median value. This strong
3 response in AOD to RH occurred while the altitude-averaged mean value of the extinction-
4 weighted RH varied from 59% RH to 88% RH for the 10th and 90th percentile profiles. These
5 results show that variability in RH is large, which propagates nonlinearly to aerosol water. As
6 has been previously found (e.g., Adams et al., 2001; Haywood et al., 1997), aerosol water is
7 an important and variable contributor to aerosol extinction that has a strong effect on the
8 relationship between dry particle mass concentration, AOD and direct radiative forcing.

9 Aerosol optical depth was less sensitive to D_g and σ_g (+21%/-19% and +15/-20%,
10 respectively) as they were varied between their 10th and 90th percentile profiles, largely
11 because these parameters did not vary much in our data. For comparison with more diverse
12 literature values, the symbol in Fig. 4 shows the AOD calculated by assuming $\sigma_g=1.8$
13 prescribed by the modal aerosol model (MAM) as incorporated into the CAM-Chem earth
14 system model (Liu et al., 2012). The AOD calculated using this σ_g value is higher than the
15 observed median AOD by 27%. Values of σ_g of 2.0 are commonly used in global simulations
16 of aerosol radiative effects (e.g., Adams et al., 2001), although it has been pointed out by
17 Nemesure et al (1995) that such σ_g values are probably unrealistically high and do not
18 represent most observations. Given the sensitivity of AOD to the particle size distribution, it
19 is clearly important that both models and retrieval algorithms use values that are constrained
20 by in situ observations for the aerosol type being investigated. In moderately polluted and
21 background conditions (excepting cases dominated by dust and seasalt), σ_g values larger than
22 ~ 1.6 for the accumulation mode aerosol generally are not supported by observations (e.g.,
23 Brock et al., 2011; Kotchenruther et al., 1999; Nemesure et al., 1995; Rissler et al., 2006;
24 Vaccari et al., 2013).

25 Variation of the ambient refractive index profile, which is dominated by the addition of water,
26 had a smaller effect on AOD, as did the variation in the hygroscopicity parameter κ_{ext} . The
27 calculated AOD was not sensitive to variation in the dry real refractive index of the aerosol
28 because of the very small range observed in this parameter for the organic-dominated aerosol
29 encountered in the southeastern U.S. Similarly, the change in AOD associated with the
30 observed range of profiles of the imaginary component of the refractive index was
31 insignificant due to the low concentrations of black carbon observed.

1 An additional calculation was made to evaluate the change in AOD due to the choice of
2 hygroscopicity model, e.g., γ (Eq. (1)) vs. κ_{ext} (Eq. (2)). In this sensitivity test, the AOD was
3 determined from ambient extinction first using the median profile of γ , and then the median
4 profile of κ_{ext} , and the difference between these AODs was calculated. The choice of
5 hygroscopicity model produced a change in calculated AOD about half that from measured
6 variability in D_g and σ_g (Fig. 4, Table 1). The γ parameterization produced on average more
7 hygroscopic growth and a larger AOD than did the κ_{ext} parameterization. This larger AOD is
8 due to an overprediction of aerosol water content and related extinction between ~60-90% RH
9 by the γ parameterization (Brock et al., 2015).

10 A final test was made of the sensitivity of AOD to variations in the thickness of the well-
11 mixed layer under conditions of total columnar aerosol mass loading (i.e., constant sources
12 and sinks). This test was made because regional-scale models often have difficulty simulating
13 the height of the well-mixed layer (e.g., Kim et al., 2015; Scarino et al., 2014). If the aerosol
14 were dry, variations in boundary layer height would not affect AOD much, because the
15 increasing height of well-mixed layer would be compensated by dilution of the aerosol
16 (assuming the air being mixed in during mixed-layer growth does not contribute to extinction
17 within the layer). However, as the well-mixed layer increases in height, the temperature in the
18 upper part of the layer decreases with the lapse rate, causing an increase in RH. Thus for the
19 same columnar dry aerosol mass loading, a growing well-mixed layer might increase AOD.
20 Compensating this increased aerosol water is a reduction in ambient aerosol concentration,
21 hence extinction, due to decreasing mean air density as the layer grows in altitude.

22 We simulate this effect with a simple model constrained by our observations. An aerosol was
23 assumed to be perfectly mixed within the well-mixed layer, with a resulting dry extinction
24 that decreased as atmospheric density decreased with altitude. The dry extinction at the
25 bottom of the well-mixed layer was the median value at the lowest layer of the aggregate
26 profile (Fig. 1a). Ambient extinction at each level in the well-mixed layer was calculated
27 using Eq. (2), a fixed value of κ_{ext} of 0.082, and the median profile of RH (Fig. 1b). The height
28 of the mixed layer was allowed to vary from 113 to 1433m, while the AOD of the transition
29 layer was assumed to remain constant at the mean value of 0.081. The contribution of the
30 aerosol in the free troposphere to AOD was ignored. The AOD integrated through the depth
31 of the well-mixed layer varied from 0.082 (most shallow layer) to 0.079 (deepest layer). The
32 decrease in ambient concentration with height more than compensated for the increased

1 extinction due to higher RH as the height of the well-mixed layer increased. Compared to the
2 total AOD, the resulting variability in AOD due to the change in height of the well mixed
3 layer was +/- 1%. Thus, despite the increase in RH with altitude, the effect of variability in
4 the height of the well-mixed layer on total AOD was negligible.

5

6 **4 Discussion and Conclusions**

7 There has been considerable research on the effects of aerosol optical, microphysical, and
8 chemical properties on aerosol extinction and AOD based on in situ measurements, laboratory
9 studies, and modeling. However, few studies have systematically investigated the sensitivity
10 of AOD to variations in the aerosol and meteorological parameters such as RH. Hegg et al.
11 (1993) examined the sensitivity of ambient extinction to particle diameter and refractive
12 index. Hegg et al. found that, as the dry aerosol humidified and grew, variations in the dry
13 mass median diameter relative to the extinction efficiency curve produced substantial $f(RH)$
14 variability. Decreasing refractive index due to water uptake was a secondary contributor.
15 Koloutsou-Vakakis et al. (1998) found that insoluble (presumably organic) material played an
16 important role in both dry and ambient extinction, and that the difference between
17 efflorescence and deliquescence branches of the hygroscopicity curves was important to
18 consider when relating aerosol mass concentration measurements to ambient scattering. Magi
19 et al. (2005) used airborne in situ measurements in the eastern U.S. to estimate the
20 contribution of dry particulate constituents and aerosol water to AOD. They found that
21 aerosol water contributed between $38\pm 8\%$ and $55\pm 15\%$ of the total AOD, depending upon the
22 hygroscopic growth model used. These numbers can be compared to our observations, which
23 show an enhancement in AOD of 54% and 85% above the dry AOD when aerosol water
24 content is included using our median profiles and the κ_{ext} and γ parameterizations,
25 respectively.

26 Analysis of data from NASA's Deriving Information on Surface conditions from Column and
27 Vertically Resolved Observations Relevant to Air Quality (DISCOVER-AQ) airborne
28 program have shown a strong relationship between AOD and surface and in situ aerosol mass
29 concentrations in the eastern U.S. (Crumeyrolle et al., 2014). Ziemba et al. (2013) found that
30 aerosol water (using the γ hygroscopic growth parameterization) was an important component
31 of the extinction profile measured by lidar and in situ measurements in the eastern U.S. In
32 contrast to these studies, we have focused on the *sensitivity* of AOD to RH and to aerosol

1 properties. Our analysis suggests that it is critical to properly account for RH and its vertical
2 distribution to quantitatively relate remotely sensed AOD to in situ aerosol properties such as
3 mass. Within the range of variability observed during the SENEX and SEAC⁴RS projects, the
4 geometric mean diameter and standard deviation were roughly equal contributors to AOD
5 variability. However, in some numerical models (e.g., Adams et al., 2001; Liu et al., 2012)
6 the prescribed choices for the width of the aerosol size distribution fall outside the range of
7 our observations, leading to potential biases in AOD that exceed 25% (Fig. 4). The AOD-
8 weighted values of σ_g ranged from 1.35 to 1.61 in our measurements, consistent with those
9 reported in the eastern U.S. by Magi et al. (2005) and with other recent literature (e.g., Brock
10 et al., 2011; Rissler et al., 2006; Vaccari et al., 2013). Substantially larger values of σ_g may
11 not be appropriate for the southeastern U.S. or other moderately polluted midlatitude and
12 background continental environments, and may bias the AOD-dry mass relationship and lead
13 to errors in the calculated radiative balance and associated feedbacks.

14 The sensitivities of AOD to RH, to the mean diameter and width of the size distribution, and
15 to the hygroscopicity model, indicate the need for a more systematic investigation. Numerical
16 models that incorporate aerosol radiative forcing need to be constrained by observations
17 similar to those reported here in other types of environments, especially the dust, sea-salt,
18 biomass burning, and heavily polluted cases that globally dominate aerosol direct radiative
19 effects (Jacobsen, 2001; Kahn, 2011). One effort, Systematic Aircraft Measurements to
20 Characterize Aerosol Air Masses (SAM-CAAM), has been proposed to make repeated
21 measurements of critical in situ and remotely sensed parameters in a wide range of air mass
22 types across the globe (Kahn, 2013). A comprehensive observational program such as SAM-
23 CAAM could help disentangle the relationship between in situ aerosol and meteorological
24 properties and AOD in different air masses, and, coupled with model and measurement
25 refinement, reduce uncertainty in direct aerosol radiative effects.

26 **Author contribution**

27 All authors contributed measurements and/or analyses for this manuscript. CB prepared the
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1 Table 1. AOD calculated from sensitivity tests.

Parameter varied	Extinction-weighted ¹ parameter value			Calculated AOD percentile values		
	10th	50th	90th	10th	50th	90th
dry aerosol mass ($\mu\text{g m}^{-3}$)	6.2	12.6	17.4	0.082	0.177	0.253
relative humidity (%)	58.8	74.7	87.9	0.137	0.177	0.305
number geometric mean diameter (μm)	0.125	0.146	0.171	0.144	0.177	0.214
geometric standard deviation	1.42	1.51	1.60	0.141	0.177	0.204
dry refractive index:	real	1.545	1.549	0.173	0.175 ²	0.178
	imaginary	0.004	0.007			
ambient refractive index:	real	1.409	1.450	0.149	0.177	0.194
	imaginary	0.004	0.007			
κ_{ext}	0.077	0.116	0.185	0.168	0.177	0.200
mixed layer height ³ (m)	113	1132	1433	0.161	0.163	0.164

2 ¹Weighted as $\sum_{i=0}^n X_i \sigma_{ext,i} / \sum_{i=0}^n \sigma_{ext,i}$ where X_i is the parameter being examined in each altitude bin i .

1 ²50th percentile AOD does not match other sensitivity cases because the wet refractive index profile calculated from the median dry
2 refractive index, RH and κ_{ext} profiles is not identical to the median profile of wet refractive index calculated from instantaneous RH
3 and κ_{ext} values.

4 ³Not weighted by extinction. Units for mixed layer height are meters above the surface and show the range of values modeled rather
5 than percentiles. Values of AOD are for the range in mixed layer height described in the text rather than for specific percentiles.

6

1 Figure 1. Aggregate vertical profiles of a) dry extinction at standard temperature and pressure,
2 b) relative humidity, c) extinction at ambient RH, pressure and temperature, d) ambient
3 aerosol optical depth integrated from the surface upward to the indicated altitude, and e)
4 number geometric median diameter and f) geometric standard deviation for a single-mode
5 lognormal size distribution calculated from the measured dry particle number size
6 distribution. Light shading shows the interdecile (10% to 90%) range, dark shading the
7 interquartile (25% to 75%) range, and the solid line the median value. The horizontal solid
8 and dashed lines show the tops of the well-mixed and transition layers, respectively. The solid
9 vertical line in (c) shows the extrapolation of ambient extinction to ground level to calculate
10 AOD. Note the scale difference between (a) and (c).

11

12 Figure 2. Atmospheric AOD measurements from the Atlanta, Georgia and Centreville,
13 Alabama AERONET sunphotometer network sites using Level 2.0 data (Holben et al., 2001).
14 The median and interquartile range for the SENEX and SEAC⁴RS data used in this analysis
15 are shown by the symbol and vertical error bars, respectively.

16

17 Figure 3. a) Histogram of the AOD values integrated from the surface upward from individual
18 profiles of aerosol extinction. b) Histogram of the fractional contribution of the well-mixed
19 layer to the total AOD from each case in (a). c) As in (b), but for the fraction contribution
20 from the transition layer.

21

22 Figure 4. Range in AOD at a wavelength of 532nm due to variations in measured parameters.
23 AOD values integrated from profiles using a model aerosol size distribution and the 10th and
24 90th percentile range of observed aerosol and meteorological parameters (bars) and from the
25 median profile (black circles). Numerical values show the extinction-weighted 10th-90th
26 percentile range of the indicated parameter. The " κ_{ext} vs. γ " bar describes the change in AOD
27 associated with choice of hygroscopicity model, centered on the median AOD. The " Δ Mixed
28 Layer Height" numerical values show the range in the simulated height of the well-mixed
29 layer. The blue symbol on the " Δ Standard Deviation" line shows calculated AOD using a
30 fixed geometric standard deviation of 1.8 as prescribed in the Modal Aerosol Model used in
31 the CAM-Chem earth system model (Liu et al., 2012).

Response to Reviewer 1

Manuscript Number: ACP-2015-821

Manuscript Title: Aerosol optical properties in the southeastern United States in summer – Part 2: Sensitivity of Aerosol Optical Depth to Relative Humidity and Aerosol Parameters

The discussion below includes the complete text from the reviewer, along with our responses and corresponding changes made to the revised manuscript. The authors thank the reviewer for useful comments that have improved the manuscript.

Comment: The manuscript presents aircraft observations of aerosols and calculated aerosol optical depth. The focus is on the sensitivity of the AOD to relative humidity and aerosol properties. While the research subject and the conclusions are not particularly original, this paper will be a nice addition to the existing literature thanks to the careful analysis (e.g. on the UHSAS response, the kappa/gamma parameterizations) and discussion (e.g. on the transition layer, the need for more systematic investigation, the co-variance of parameters, the mixed-layer height). The manuscript refers to Brock et al. (2015) in an important manner. Provided that that discussion paper is accepted, I recommend publication of the present manuscript. My suggestions are as follows.

Comment: Page 31480, line 16, “were calculated” should be either replaced with “we calculated” or moved to the end of the sentence.

Response: Corrected.

Also, a brief explanation would be nice for the choice of 10th and 90th percentile values instead of, say, a one standard deviation range (68%).

Response: We now add the following sentence in Section 3.1, "Because the distribution of most parameters was not Gaussian, percentile values are used to represent the range of observed variability."

Page 31481, line 10, state how the surface ambient extinction was determined. One way to do so is to move here the two full sentences in line 16-19 of page 31482.

Response: We have moved the sentences and changed them to read, "The extinction within the well-mixed layer was extrapolated to the surface for each individual profile. Wagner et al. (2015) show that measurements of extinction at the Centreville, Alabama surface site during the SENEX time period agreed with values measured at the lowest altitude of the aircraft, supporting such extrapolation."

Page 31493, bring Rissler et al. above Saide et al.

Response: Corrected.

In addition to the changes noted above, in the Introduction we have added mention of two relevant papers, one of which recently appeared in ACPD.

Response to Reviewer 2

Manuscript Number: ACP-2015-821

Manuscript Title: Aerosol optical properties in the southeastern United States in summer – Part 2: Sensitivity of Aerosol Optical Depth to Relative Humidity and Aerosol Parameters

The discussion below includes the complete text from the reviewer, along with our responses and corresponding changes made to the revised manuscript. The authors thank the reviewer for useful comments that have improved the manuscript.

Comment: The manuscript analyses the sensitivity of aerosol optical depth to relative humidity and to different aerosol parameters. The work uses the parameterization of hygroscopic growth developed in the companion paper (Aerosol optical properties in the southeastern United States in summer – Part 1: Hygroscopic growth). The paper is well written and the results are appropriately discussed. In this sense, the manuscript is suitable for publication in ACP provided that the companion paper is accepted and after minor revisions suggested below.

Comment: Along the manuscript, the authors present some numerical results with an excessive number of significant figures. See for example the comment on AOD at page 31487 lines 1-2. This fact reflects the need of strengthen the discussion on the uncertainties of the derived variables, having in mind that the experimental measurements and the models developed present uncertainties that in some way affect the result of the study.

Response: We have evaluated the precision of the figures presented throughout the manuscript and stand by the values shown. In the example that concerns the reviewer, this is a model sensitivity test where the height of the well-mixed layer is allowed to vary over the observed range of heights. There are only slight differences in the AOD, which are due to changes in the humidity distribution. Certainly we could not measure AOD to this level of precision, but as a model sensitivity study, the small change (requiring three significant figures to show a difference) makes the point; the AOD is not sensitive to this parameter. Regarding how uncertainties affect the overall results, we feel the two manuscripts Brock et al. 2016a ("Part 1") and the current manuscript "Part 2" together robustly explore the uncertainties in the data and analysis, including two Monte Carlo simulations of propagated uncertainties and a numerical model of the response of the UHSAS optical particle counter. The current manuscript (Part 2) focuses on simulations by a simple model of the sensitivity of AOD to the range of observed variability of various parameters, and not to experimental uncertainties.

Comment: The description on determining ambient extinction must be improved (refer to page 31480 lines 2-6). Although the procedure for deriving $f(\text{RH})$ is described in the companion paper, the wording of these sentences on two of the

higroscopic growth models considered must be improved. In its present form is a little bit confusing, please state first that you derive the model and then that you apply it to the ambient conditions during the experimental campaign.

Response: We have improved the description the three methods we used to calculate the ambient $f(RH)$ value while still keeping the description succinct. This section now reads:

"The value of $f(RH)$ was calculated for each data point in three different ways. In the first method, κ -Köhler theory was applied to measurements of aerosol size distribution and composition to predict particle diameter as a function of RH. Mie theory was then used to predict the ambient extinction from the deliquesced particle size distribution. In the second method, the observed 3-point $f(RH)$ values were used to fit a curve of the form (Eqn 2) and the extinction at ambient RH was calculated using the fitted coefficient. Finally, a new parameterization of the form (Eqn 3) was fitted to the observed 3-point $f(RH)$ values and the extinction at ambient RH calculated."

In addition to the changes noted above, in the Introduction we have added mention of two relevant papers, one of which recently appeared in ACPD.