

1 **An Analysis of AERONET Aerosol Absorption Properties and Classifications**

2 **Representative of Aerosol Source Regions**

3  
4 D. M. Giles,<sup>1,2,3</sup> B. N. Holben,<sup>2</sup> T. F. Eck,<sup>2,4</sup> A. Sinyuk,<sup>1,2</sup> A. Smirnov,<sup>1,2</sup> I. Slutsker,<sup>1,2</sup> R. R.  
5 Dickerson,<sup>3</sup> A. M. Thompson,<sup>5</sup> and J. S. Schafer<sup>1,2</sup>

6  
7 1. Sigma Space Corporation, Lanham, MD 20706 USA

8 2. Biospheric Sciences Laboratory, NASA Goddard Space Flight Center, Greenbelt, MD 20771  
9 USA

10 3. Department of Atmospheric and Oceanic Science, The University of Maryland, College Park,  
11 MD 20742 USA

12 4. Universities Space Research Association, Columbia, MD 21044 USA

13 5. Department of Meteorology, The Pennsylvania State University, University Park, PA 16802  
14 USA

15  
16  
17  
18 Submitted to the Journal of Geophysical Research–Atmospheres

19 AGU Index Terms: 0305, 0345, 0360

20 **Abstract**

21           Partitioning of mineral dust, pollution, smoke, and mixtures using remote sensing  
22 techniques can help improve accuracy of satellite retrievals and assessments of the aerosol  
23 radiative impact on climate. Spectral aerosol optical depth ( $\tau$ ) and single scattering albedo ( $\omega_o$ )  
24 from Aerosol Robotic Network (AERONET) measurements are used to form absorption [i.e.,  $\omega_o$   
25 and absorption Ångström exponent ( $\alpha_{abs}$ )] and size [i.e., extinction Ångström exponent ( $\alpha_{ext}$ ) and  
26 fine mode fraction of  $\tau$ ] relationships to infer dominant aerosol types. Using the long-term  
27 AERONET data set (1999-2010), 19 sites are grouped by aerosol type based on known source  
28 regions to: (1) determine the average  $\omega_o$  and  $\alpha_{abs}$  at each site (expanding upon previous work);  
29 (2) perform a sensitivity study on  $\alpha_{abs}$  by varying the spectral  $\omega_o$ ; and (3) test the ability of each  
30 absorption and size relationship to distinguish aerosol types. The spectral  $\omega_o$  averages indicate  
31 slightly more aerosol absorption (i.e., a  $0.0 < \delta\omega_o \leq 0.02$  decrease) than in previous work and  
32 optical mixtures of pollution and smoke with dust show stronger absorption than dust alone.  
33 Frequency distributions of  $\alpha_{abs}$  show significant overlap among aerosol type categories and at  
34 least 10% of the  $\alpha_{abs}$  retrievals in each category are below 1.0. Perturbing the spectral  $\omega_o$  by  
35  $\pm 0.03$  induces significant  $\alpha_{abs}$  changes from the unperturbed value by at least  $\sim \pm 0.6$  for Dust,  
36  $\sim \pm 0.2$  for Mixed, and  $\sim \pm 0.1$  for Urban/Industrial and Biomass Burning. The  $\omega_{o440nm}$  and  $\alpha_{ext440-}$   
37  $_{870nm}$  relationship shows the best separation among aerosol type clusters, providing a simple  
38 technique for determining aerosol type from surface- and future space-based instrumentation.

39

## 40 1.0 Introduction

41 Particles suspended in the atmosphere are difficult to characterize both temporally and  
42 spatially due to their short lifetime and geographically diverse sources. Aerosol mixtures—  
43 whether dominated by dust, sulfate, carbon, sea salt, or mixtures of these particles—pose a  
44 challenge to satellite and sub-orbital remote sensing techniques when identifying aerosol type  
45 [Jeong and Li 2005; Levy et al., 2007; Kalapureddy et al., 2009; Lee et al., 2010; Kahn et al.,  
46 2010; Russell et al., 2010]. Remote sensing techniques can quantify the aerosol particle size  
47 using spectral aerosol optical properties, but inferring aerosol type requires knowledge of the  
48 source regions usually obtained through use of ancillary data sets (e.g., back trajectory models,  
49 satellite product, electron microscopy) to determine emission sources, transport mechanisms,  
50 composition, and morphology. The discrimination of aerosol types increases accuracy of the  
51 assessment of the aerosol radiative impact and therefore is important to climate modeling [Diner  
52 et al., 1999; Satheesh and Moorthy 2005]. Variations in spectral aerosol absorption magnitudes  
53 can enable partitioning among aerosols from various source regions, fuel types, or combustion  
54 phases. Aerosol absorption together with size can potentially determine dominant aerosol type  
55 from remote sensing and in situ measurements.

56 Various methods have been proposed using aerosol optical and microphysical properties  
57 to distinguish aerosol types. The magnitude of the aerosol optical depth (AOD,  $\tau_{\text{ext}}$ ) and the  
58 spectral dependence of AOD with respect to wavelength (i.e., Ångström exponent,  $\alpha_{\text{ext}}$ ) is  
59 commonly used in aerosol remote sensing to infer dominant aerosol types given knowledge of  
60 the source region or typical aerosol transport mechanisms [e.g., Kalapureddy et al., 2009, Boselli  
61 et al., 2012]. Other techniques using the derivative of the Ångström exponent or spectral  
62 difference of Ångström exponent wavelength pairs along with aerosol loading and particle

63 effective radius may provide further information on particle type with respect to size and growth  
64 of particles [Gobbi *et al.*, 2007; Basart *et al.*, 2009]. Although size varies among particle types,  
65 the spectral absorption also varies. Studies [Omar *et al.*, 2005, Levy *et al.*, 2007; Mielonen *et al.*,  
66 2009; Lee *et al.*, 2010; Russell *et al.*, 2010] have suggested relationships utilizing the aerosol  
67 absorption and size properties to determine the dominant aerosol type from Aerosol Robotic  
68 Network (AERONET) retrievals [Holben *et al.*, 1998; Dubovik *et al.*, 2000; Dubovik *et al.*, 2002;  
69 Dubovik *et al.*, 2006]. Information content from these relationships varies from generic  
70 identification of major aerosol particle types (e.g., dust, mixed, urban/industrial pollution, and  
71 biomass burning smoke) to specific degrees of absorbing aerosols. Recently, Russell *et al.*  
72 [2010] have proposed using the absorption Ångström exponent (AAE,  $\alpha_{\text{abs}}$ ), the spectral  
73 absorption aerosol optical depth dependence on wavelength, to further define aerosol type from  
74 AERONET retrievals. For comparison to the Cloud-Aerosol Lidar with Orthogonal Polarization  
75 (CALIOP) instrument, Mielonen *et al.* [2009] utilized the AERONET single scattering albedo  
76 ( $\omega_0$ ) difference between 440 and 1020 nm (as suggested by Bergstrom *et al.* [2002] and  
77 implemented by Derimian *et al.* [2008]) and  $\alpha_{\text{ext}}$  to estimate aerosol type. Further, Lee *et al.*  
78 [2010] modified this relationship to use only  $\omega_0$  from 440 nm and the fine mode fraction ( $\eta$ ) of  
79 the AOD at 550 nm to determine the particle size partitioning. Other techniques using spectral  
80 lidar ratios and multiple aerosol optical and microphysical properties retrieved from AERONET  
81 have been implemented to determine aerosol type categories for various applications [Cattrall *et*  
82 *al.*, 2005; Omar *et al.*, 2005; Qin and Mitchell 2009; Burton *et al.*, 2012].

83 In this study, for data available between 1999 and 2010, 19 AERONET sites were  
84 classified by dominant aerosol type [i.e., Dust, Mixed, Urban/Industrial (U/I), and Biomass  
85 Burning (BB)] based on previous literature. First, aerosol absorption parameters (i.e.,  $\omega_0$  and

86  $\alpha_{\text{abs}}$ ) were analyzed and compared to previous work. Second, sensitivity tests were performed on  
87 the  $\alpha_{\text{abs}}$  by perturbing  $\omega_o$  to determine variability within each dominant aerosol type category.  
88 Last, the absorption and size relationships were evaluated and compared to each other based on  
89 the dominant aerosol type categorizations.

## 90 **2.0 Instrumentation and Method**

91 The Aerosol Robotic Network is a ground-based network of standardized Cimel Sun and  
92 sky scanning radiometers measuring AOD at multiple wavelengths from 340 to 1640 nm and  
93 retrieving other columnar optically effective aerosol properties (e.g., volume size distribution,  
94 complex index of refraction, and single scattering albedo) from sky radiance measurements at  
95 four wavelengths: 440, 675, 870, and 1020 nm [Holben *et al.*, 1998]. The AOD estimated  
96 uncertainty varies spectrally from  $\pm 0.01$  to  $\pm 0.02$  with the highest error in the ultraviolet  
97 wavelengths [Holben *et al.*, 1998; Eck *et al.*, 1999] and calibrated sky radiance measurements  
98 typically have an uncertainty less than 5% [Holben *et al.*, 1998]. Further descriptions of the  
99 instrumentation, calibration, methodology, data processing, and data quality are described  
100 elsewhere [Holben *et al.*, 1998; 2006; Eck *et al.*, 1999; 2005; Smirnov *et al.*, 2000]. For all sky  
101 radiance wavelengths (440, 675, 870, and 1020 nm), the  $\omega_o$  uncertainty is expected to be  $\pm 0.03$   
102 based on Version 1 almucantar retrieval computations when  $\tau_{440\text{nm}} > 0.4$  [Holben *et al.*, 1998; Eck  
103 *et al.*, 1999; Dubovik *et al.*, 2002]. When compared to AERONET  $\omega_o$  retrievals, in situ  
104 measurements of  $\omega_o$  were within AERONET uncertainty estimates [Leahy *et al.*, 2007; Johnson  
105 *et al.*, 2009; Müller *et al.*, 2010; Toledano *et al.*, 2011].

106 In-depth discussions of the almucantar retrieval products are given by Dubovik and King  
107 [2000] and Dubovik *et al.* [2000; 2002; 2006] and quality criteria are discussed by Holben *et al.*

108 [2006]. *Dubovik et al.* [2002] provided averaged almucantar retrieval aerosol optical and  
109 microphysical properties based on aerosol types and source region using AERONET pre-Version  
110 1 data (i.e., data collected and analyzed prior to the release of quality assured Version 1 retrieval  
111 data set in 2003). These results have been used throughout the literature to define aerosol type  
112 based on the aerosol absorption characteristics [*Russell et al.*, 2010 and references therein].  
113 Notably, the Version 2 retrievals (i.e., released in 2006) utilized new input data sets (e.g., NCEP  
114 reanalysis, MODIS ecosystem type-based BRDF functions, and geographically and temporally  
115 varying black sky albedo), more dynamic calculations of the surface reflectance than the Version  
116 1 assumption of a green Earth surface reflectance, robust quality checks of the measured sky  
117 radiance inputs, and improved criteria for acceptable sky residual fits [*Holben et al.*, 2006; *Leahy*  
118 *et al.*, 2007; *Eck et al.*, 2008 and references therein]. For example, in the United Arab Emirates  
119 and Arabian Gulf, Version 2 improvements provided more consistent  $\omega_0$  magnitudes and spectra  
120 for coarse-mode dust aerosol over two vastly different surfaces (i.e., small island versus bright  
121 desert) with  $\omega_0$  differences of less than 0.01 compared to 0.03 for the Version 1 spheroid  
122 inversion model and with increased absorption at 440 nm, which typically occurs in iron-rich  
123 desert dust, rather than spectrally neutral  $\omega_0$  from Version 1 retrievals [*Eck et al.*, 2008].

124 Additional instrument checks were implemented to assess absorption properties from the  
125 Version 2 almucantar retrievals. To improve the quality of the sky radiance measurements used  
126 for almucantar retrievals, instrument collimator consistency checks were performed to remove  
127 potential artifacts (e.g., induced by spider webs in the tube, excessive dust, or contamination on  
128 the sensor head window due to moisture or dust) in the radiance measurements. The sky  
129 radiance measurements at  $\pm 6^\circ$  azimuth from solar zenith—using the solar aureole and sky gains  
130 for instruments with only Silicon detectors—were required to have a percent difference of less

131 than 10% spectrally from 440 to 1020 nm. For Silicon and InGaAs detector instruments (where  
 132 each detector measures in a different collimator tube), the temperature corrected Silicon and  
 133 InGaAs  $\tau_{1020\text{ nm}}$  difference ( $\Delta\tau$ ) must be less than  $\Delta\tau_{\text{limit}}$  of  $0.06/m$  (where  $m$  is the optical air  
 134 mass), which results in a  $\Delta\tau_{\text{limit}}$  of 0.03 when  $m$  equals 2 and 0.06 for the overhead sun ( $m=1$ ).  
 135 Collimator consistency checks provide an improved method to further quality assure the Level  
 136 2.0 almucantar retrieval data set.

137 Measured aerosol optical depth and computed almucantar retrieval products can be used  
 138 to derive additional aerosol properties. The extinction Ångström exponent ( $\alpha_{\text{ext}}$ ) was calculated  
 139 from the spectral dependence of AOD or  $\tau_{\text{ext}}$  with wavelength ( $\lambda$ ) using equation (1) [Ångström  
 140 1964]:

$$\alpha_{\text{ext}} = -d\ln[\tau_{\text{ext}}(\lambda)]/d\ln(\lambda) \quad (1)$$

141 For a wavelength range between 440 and 870 nm typically using 440, 500, 675, and 870 nm  
 142 AOD—and computed by linear regression of  $\ln \tau$  versus  $\ln \lambda$ —values near 0 indicate mainly  
 143 coarse mode (radius,  $r > 1 \mu\text{m}$ ) aerosol particles, while values near 2 indicate mainly fine or  
 144 accumulation mode ( $r < 1 \mu\text{m}$ ) aerosol particles [Holben *et al.*, 1991; Kaufman *et al.*, 1992; Eck *et*  
 145 *al.*, 1999; Reid *et al.*, 1999; Holben *et al.*, 2001]. The absorption AOD or  $\tau_{\text{abs}}$  is calculated for  
 146 each wavelength using equation (2):

$$\tau_{\text{abs}}(\lambda) = \tau_{\text{ext}}(\lambda) * [1 - \omega_o(\lambda)] \quad (2)$$

147 [Eck *et al.*, 2010; Russell *et al.*, 2010; Giles *et al.*, 2011a]. Similar to  $\alpha_{\text{ext}}$ , the spectral  
 148 dependence of  $\tau_{\text{abs}}$  with  $\lambda$  on logarithmic scale gives the absorption Ångström exponent or  $\alpha_{\text{abs}}$  in  
 149 equation (3):

$$\alpha_{\text{abs}} = -d\ln[\tau_{\text{abs}}(\lambda)]/d\ln(\lambda) \quad (3)$$

150 Assuming a spectrally constant index of refraction, very small spherical black carbon particles ( $r$   
151  $\sim 0.01 \mu\text{m}$ ) can have a  $\lambda^{-1}$  dependence or  $\alpha_{\text{abs}}$  of 1.0 [Bergstrom *et al.*, 2002], while larger  
152 optically effective black carbon particles (e.g.,  $r > 0.1 \mu\text{m}$ ) may have  $\alpha_{\text{abs}}$  values below 1.0 for  
153 large cores and up to 1.6 for various shell coatings [Lack and Cappia 2010]. Russell *et al.* [2010]  
154 analyzed AERONET pre-Version 1 almucantar retrievals from Dubovik *et al.* [2002] and showed  
155  $\alpha_{\text{abs}}$  values vary between  $\sim 1.2$  and 3.0 for Dust,  $\sim 0.75$  and 1.3 for U/I, and  $\sim 1.2$  and 2.0 for BB.  
156 Eck *et al.* [2010] analyzed AERONET Version 2 almucantar retrievals and showed sites  
157 dominated by optical mixtures of dust, smoke, and pollution had  $\alpha_{\text{abs}}$  values between  $\sim 1.2$  and  
158 1.8 for mixed size particles (i.e., fine mode fraction of the AOD at 675 nm ranged between  $\sim 0.35$   
159 and 0.65). In the present study, the fine mode AOD ( $\tau_f$ ) and coarse mode AOD ( $\tau_c$ ) from the  
160 almucantar retrieval—as inferred from the size distribution and refractive indices—were  
161 interpolated to 550 nm using the linear fit of the logarithms of  $\tau_f$ ,  $\tau$  (i.e.,  $\tau_f + \tau_c$ ) for 440, 675, and  
162 870 nm wavelengths to calculate the fine mode fraction of the AOD [i.e.,  $\eta = \tau_f / (\tau_f + \tau_c)$ ] at 550  
163 nm ( $\eta_{550\text{nm}}$ ).

164 Nineteen AERONET sites were selected for the analysis based on the availability of an  
165 extensive data record (i.e., greater than five data equivalent years of AOD measurements from  
166 1999 to 2010) and the geographic distribution among aerosol source regions (Figure 1). The  
167 sites were designated as one of four commonly used aerosol classifications: Dust, Mixed,  
168 Urban/Industrial (U/I), and Biomass Burning (BB). The classifications were established based  
169 on the source regions and known seasonal changes in aerosol type over these regions (see  
170 references in Table 1). Further, these selected sites should be subject to high aerosol loading  
171 (i.e.,  $\tau_{440\text{nm}} > 0.4$ ) to meet the Version 2, Level 2.0 almucantar retrieval sensitivity requirement for  
172 absorption parameters (e.g.,  $\omega_o$ ,  $\tau_{\text{abs}}$ ) [Dubovik *et al.*, 2000; Holben *et al.*, 2006]. Sea salt (as well



173 as biogenic) aerosols as a dominant particle type category were not considered in this study since  
174  $\tau_{500\text{nm}}$  is typically less than 0.1 for pure maritime environments [Smirnov *et al.*, 2002]; however,  
175 for maritime locations affected by aerosol plumes (e.g., Saharan dust transport over Cape Verde  
176 islands),  $\tau_{440\text{nm}} > 0.4$  can be satisfied [Smirnov *et al.*, 2009]. Hence, the  $\tau_{440\text{nm}} > 0.4$  criterion biases  
177 the data set only to high aerosol loading periods to ensure enough radiometric sensitivity to  
178 compute absorption reliably [Dubovik *et al.*, 2002]. Although Dust, U/I, and BB categories may  
179 represent the dominant aerosol type, episodic aerosol incursions outside of their classification  
180 category likely have occurred at any site during the analysis period (e.g., dust over Shirahama or  
181 Lake\_Argyle, biomass burning smoke over GSFC) [Sano *et al.*, 2003; Qin and Mitchell 2009;  
182 Eck *et al.*, 2003b; O'Neill *et al.*, 2005]. The Mixed aerosol category encompasses sites primarily  
183 affected by different mixtures of aerosol types (e.g., dust and pollution or dust and biomass  
184 burning smoke mixtures) on a seasonal basis, increasing the probability of at least an optical  
185 mixture state [Derimian *et al.*, 2006; Eck *et al.*, 2010]. Although no explicit seasonal  
186 partitioning is performed, the  $\tau_{440\text{nm}} > 0.4$  criterion captures mainly seasonal increases in aerosol  
187 loading at some sites (e.g., GSFC and Mongu) [Holben *et al.*, 2001].

## 188 **3.0 Results**

### 189 **3.1 Retrieved Absorption Properties by Dominant Aerosol Type**

190 Dust particles aggregated with varying combinations of clay, quartz, and hematite exhibit  
191 strong absorption in the blue wavelength region (e.g., 440 nm) with lower absorption in the  
192 visible and near infrared wavelengths (i.e.,  $\omega_0$  increasing with wavelength) [Sokolik and Toon  
193 1999]. For fine mode particles ( $r < 1.0 \mu\text{m}$  in the volume size distribution), hygroscopic aerosol  
194 particles (e.g., sulfates) have near neutral spectral dependence and high scattering efficiency  
195 [Dubovik *et al.*, 2002]. Black carbon (BC) particles have the strongest absorption in the near-

196 infrared ( $\omega_o$  decreasing with  $\lambda$  when the sole absorber), while aerosols composed of brown  
197 carbon (BrC) or organic carbon (OC) exhibit stronger absorption in ultraviolet and visible bands  
198 ( $\omega_o$  increasing with  $\lambda$  when the sole absorber) [Eck *et al.*, 2009]. Varying concentrations of BC  
199 particles optically mixed with dust, BrC, and/or OC can produce ambiguous  $\omega_o$  wavelength  
200 dependence (i.e., increasing, decreasing, or constant with  $\lambda$ ); however, the net effect is stronger  
201 absorption across the retrieved spectrum (e.g., 440 to 1020 nm) [Dubovik *et al.*, 2002; Giles *et*  
202 *al.*, 2011a].

203 The AERONET Version 2, Level 2.0 absorption properties at each site are presented in  
204 Figure 2 and Table 2 to provide an update to Dubovik *et al.* [2002] and Russell *et al.* [2010]. The  
205 spectral  $\omega_o$  behavior is similar to Dubovik *et al.* [2002] for most regions. For Solar Village  
206 (Dust), Capo Verde (Dust), GSFC (U/I), Mexico City (U/I), and Mongu (BB), the  $\omega_o$  differences  
207 between Dubovik *et al.* [2002] and Table 2 (i.e.,  $\omega_o$  Dubovik 2002 -  $\omega_o$  Table 2) showed an overall  
208 average decrease of 0.01 for these sites with the largest decrease of 0.02 spectrally for GSFC and  
209 Capo Verde and smallest decrease ranging from 0 to 0.01 for Mongu. Notably, the  $\omega_o$  standard  
210 deviations are significantly greater by 0.01 to 0.03 in the present study than Dubovik *et al.* [2002]  
211 for all five sites. Table 2 differs from Dubovik *et al.* [2002] due to utilizing different analysis  
212 criteria (e.g.,  $\tau_{440\text{nm}} > 0.4$  in Table 2 vs.  $\tau_{1020\text{nm}} \geq 0.3$  and  $\alpha_{\text{ext}} \leq 0.6$  for desert dust in Dubovik *et al.*  
213 [2002]), implementing improved surface characterization and inversion quality checks in  
214 Version 2 (as discussed in Section 2), and utilizing a larger data set (e.g., the number of  $\omega_o$   
215 retrievals at GSFC is four times larger than Dubovik *et al.* [2002]). For  $\omega_{o440\text{nm}}$  as a function of  
216  $\tau_{440\text{nm}}$ , the  $R^2$  values—calculated based on a second order fit—ranged from 0.0 to 0.16 for each  
217 site, indicating weak correlation and only up to 16% of  $\omega_{o440\text{nm}}$  variation was explained by  $\tau_{440\text{nm}}$ .  
218 Table 2 shows that the Dust category has the least variability among sites likely due to the

219 similar mineral composition, while the BB category has the largest variability likely due to  
220 various fuel types and fuel combustion phases resulting from different relative BC emissions  
221 [Eck *et al.*, 2003b]. The Mixed category ( $0.33 < \eta_{550\text{nm}} \leq 0.66$ )  $\omega_o$  average shows strong spectral  
222 absorption and dust-like  $\omega_o$  spectra with stronger absorption at 440 nm due to significant dust  
223 contribution to the optical mixture. Sokolik and Toon [1998] showed that varying hematite  
224 amounts in dust can lead to increased absorption spectrally from the blue to near-infrared  
225 wavelength region. Using  $\alpha_{\text{ext}} < 0.2$  to designate “pure dust” as suggested by Kim *et al.* [2011],  
226 the overall “pure dust” average of  $\omega_o$  for all Dust category sites is 0.91, 0.97, 0.97, 0.97 for the  
227 440, 675, 870, and 1020 nm wavelengths, respectively. These “pure dust”  $\omega_o$  values are lower  
228 by up to 0.02, spectrally, than those reported by Dubovik *et al.* [2002] for Dust sites and are  
229 lower by up to 0.01 for  $\omega_o$  at 550 nm (logarithmically interpolated between 440 nm and 675 nm)  
230 compared to similar sites analyzed by Kim *et al.* [2011]. Table 2 shows the Dust site averages  
231 are lower than “pure dust,” indicating possible incursions by other aerosols (e.g., biomass  
232 burning smoke). Eck *et al.* [2010] and Giles *et al.* [2011a] also showed increasing absorption  
233 with wavelength for decreasing  $\alpha_{\text{abs}}$ , indicating an optical mixture and possibly aggregation of  
234 dust and carbonaceous particles at Kanpur, India. We interpolated  $\eta$  to 550 nm using the linear  
235 fit of the logarithms of  $\tau_f$ ,  $\tau$  (i.e.,  $\tau_f + \tau_c$ ) and the 440, 675, and 870 nm wavelengths similar to Lee  
236 *et al.* [2010]. In Figure 3a, the Mixed category for the coarse mode particles ( $\eta_{550\text{nm}}$ : 0.0-0.33)  
237 resembles dust  $\omega_o$  spectra as shown in Figure 2a. In Figure 3c, for fine mode particles ( $\eta_{550\text{nm}}$ :  
238 0.66-1.0), the  $\omega_o$  magnitudes and variability are similar to U/I or BB particle types categories but  
239 with less  $\omega_o$  spectral dependence possibly due to varying amounts of BC, BrC, and OC  
240 [Derimian *et al.*, 2006; Eck *et al.*, 2009; 2010]. The average  $\omega_o$  for  $\alpha_{\text{abs}}$  binned between 1.5 and  
241 2.0 shown by Giles *et al.* [2011a] at Kanpur closely resembles the absorption magnitude and

242 spectral shape of mixed aerosol types for the Mixed category ( $0.33 < \eta_{550\text{nm}} \leq 0.66$ ) in Table 2 as  
243 well as Figure 2b and Figure 3b suggesting various mixtures of aerosol particles contributing to  
244 the absorption.

245 The  $\omega_o$  and  $\tau_{\text{ext}}$  are used to derive the  $\tau_{\text{abs}}$  from AERONET data.  $\tau_{\text{abs}}$  and  $\alpha_{\text{abs}}$  were  
246 averaged for each site based on dominant particle type category in Figure 4 and Table 2. For the  
247 five sites (Solar Village, Capo Verde, GSFC, Mexico City, and Mongu), a comparison of  
248 average  $\alpha_{\text{abs}}$  values in Table 2 with *Russell et al.* [2010] for the 440-870 nm range shows the  
249 largest difference in  $\alpha_{\text{abs}}$  (i.e.,  $\alpha_{\text{abs Russell 2010}} - \alpha_{\text{abs Table 2}}$ ) at GSFC (-0.25) and Capo Verde (+1.2).  
250 For the other three sites, the  $\alpha_{\text{abs}}$  averages in Figure 4 are comparable to those reported by *Russell*  
251 *et al.* [2010] and *Giles et al.* [2011a]. In Figure 5, the Mixed category was further stratified by  
252 the  $\eta_{550\text{nm}}$  as in Figure 3. The coarse particle range ( $\eta_{550\text{nm}}$ : 0.0-0.33) shows similar  $\alpha_{\text{abs}}$  (1.7-2.3)  
253 as the Dust category (which is expected for dust dominated cases) and the fine particle range  
254 ( $\eta_{550\text{nm}}$ : 0.66-1.0) shows an  $\alpha_{\text{abs}}$  (0.8-1.5) similar to BB and U/I categories. The mixed size  
255 particle range ( $\eta_{550\text{nm}}$ : 0.33-0.66) is nearly identical to the Mixed category  $\alpha_{\text{abs}}$  (1.2-1.7) in  
256 Figure 4b and similar to values reported by *Eck et al.* [2010]. As shown by *Bergstrom et al.*  
257 [2007] and *Russell et al.* [2010], the  $\alpha_{\text{abs}}$  may vary significantly when considering the aerosol  
258 particle size between fine and coarse modes; however, when considering U/I and BB aerosols  
259 within the fine particle range, significant overlap results in  $\alpha_{\text{abs}}$ . The sensitivity of  $\alpha_{\text{abs}}$  with  
260 respect to input parameters will be investigated in the next section.

### 261 **3.2 Absorption Ångström Exponent Sensitivity Study**

262 The retrieved values of  $\alpha_{\text{abs}}$  have a normal distribution (Figure 6) when calculating  $\alpha_{\text{abs}}$   
263 using three wavelengths (440-675-870 nm) for each dominant aerosol type. *Russell et al.* [2010]  
264 showed that the average  $\alpha_{\text{abs}}$  values generally decreased with increasing spectral range possibly

265 due in part to the crude surface reflectance assumption made in early AERONET analysis (as  
266 discussed in Section 3.1), while *Gyawali et al.* [2012] showed an increasing  $\alpha_{\text{abs}}$  values with  
267 increasing spectral range for clean and polluted days during winter in Reno, Nevada. However,  
268 increasing or decreasing trends of  $\alpha_{\text{abs}}$  depend on the wavelength interval [*Lack and Cappa*  
269 2010]. These  $\alpha_{\text{abs}}$  values computed from 440-675-870 nm wavelength range have large  
270 variability with standard deviations ranging from  $\pm 0.3$  to  $\pm 0.6$  ( $1.76 \pm 0.58$  for Dust;  $1.53 \pm 0.44$  for  
271 Mixed;  $1.21 \pm 0.37$  for U/I;  $1.35 \pm 0.35$  for BB). Individual  $\alpha_{\text{abs}}$  retrieval calculations [ $\alpha_{\text{abs}}$  (Dust):  
272  $\sim 0-4$ ;  $\alpha_{\text{abs}}$  (Mixed):  $\sim 0-3.5$ ;  $\alpha_{\text{abs}}$  (U/I):  $\sim 0-2$ ;  $\alpha_{\text{abs}}$  (BB):  $\sim 0-2.5$ ] are within the range of all  
273 dominant particle types; therefore,  $\alpha_{\text{abs}}$  should not be used alone to determine aerosol types  
274 without the use of other information (e.g., aerosol size). Further, Figure 6 shows a significant  
275 percentage of  $\alpha_{\text{abs}}$  below 1.0, which is the black carbon limit for very small particles [*Bergstrom*  
276 *et al.*, 2002]. However, *Lack and Cappa* [2010] suggested  $\alpha_{\text{abs}}$  (from 380-750 nm) values for  
277 larger optically effective BC particles may exist between  $\sim 0.2$  and 1.6 depending on the BC  
278 coating material. Nonetheless, the U/I category has over 22% of the  $\alpha_{\text{abs}}$  retrievals below 1.0,  
279 while the other categories have  $\sim 10\%$  of the data below  $\alpha_{\text{abs}}$  of 1.0 also possibly due to the  
280 uncertainty of the retrieved  $\omega_0$ .

281 A sensitivity study of  $\alpha_{\text{abs}}$  was performed to test the response of  $\alpha_{\text{abs}}$  by varying  $\omega_0$  for  
282 each wavelength (i.e., 440, 675, and 870 nm) and holding  $\tau_{\text{ext}}$  constant (i.e., AOD cannot be  
283 perturbed since it is used in the calculation of  $\omega_0$  retrieval). First,  $\omega_0$  was varied by  $\pm 0.03$ , the  
284 current AERONET  $\omega_0$  uncertainty estimate [*Dubovik et al.*, 2002]. Additional tests included  
285 varying  $\omega_0$  by  $\pm 0.01$ ,  $\pm 0.02$ , and  $\pm 0.04$  to show the variability of  $\alpha_{\text{abs}}$  with various degrees of  $\omega_0$   
286 uncertainty. Different spectral  $\omega_0$  inputs schemes were implemented to determine the  $\alpha_{\text{abs}}$   
287 response by varying  $\omega_0$  equally across all wavelengths, by perturbing  $\omega_0$  at only one end point in

288 the 440-675-870 nm wavelength set (i.e., 440 nm or 870 nm), and by perturbing  $\omega_o$  at 440 nm or  
289 870 nm in the 440-870 nm wavelength pair (i.e., excluding 675 nm). Positive  $\omega_o$  perturbation  
290 may approach values of 1.0 (i.e., absolute scattering) and can produce large positive or negative  
291  $\alpha_{\text{abs}}$  due to very low  $\tau_{\text{abs}}$ . To prevent such cases, the  $\omega_o$  magnitude was limited to less than 0.995  
292 for positive  $\omega_o$  perturbations for all wavelengths resulting in a reduced data subset.

293 Table 3 shows the sensitivity of  $\alpha_{\text{abs}}$  to perturbations in  $\omega_o$ . The perturbation of  $\pm 0.03 \omega_o$   
294 (the current estimated uncertainty) changed  $\alpha_{\text{abs}}$  by at least  $\sim \pm 0.6$  for Dust,  $\sim \pm 0.2$  for Mixed, and  
295  $\sim \pm 0.1$  for U/I and BB. The perturbations of  $\omega_o$  by  $\pm 0.02$  showed  $\sim 0.1$  smaller corresponding  
296 change in  $\alpha_{\text{abs}}$  with respect to  $\pm 0.03 \omega_o$  perturbations for Dust and less than 0.05-0.10 for the  
297 other categories. Perturbations of  $\omega_o$  by  $\pm 0.04$  showed large deviations from the unperturbed  
298 data set, indicating much greater uncertainty for  $\alpha_{\text{abs}}$  with increasing  $\omega_o$  uncertainty. The  
299 simulated overestimation of spectral  $\omega_o$  for U/I and BB (i.e.,  $\delta\omega_o = -0.03$ ) showed a higher  $\delta\alpha_{\text{abs}}$   
300 suggesting a possibility that the unperturbed  $\alpha_{\text{abs}}$  is underestimated and may possibly, at least  
301 partly, explain  $\alpha_{\text{abs}}$  below 1.0 in these categories. However, *Lack and Cappa* [2010] determined  
302 that the large  $\alpha_{\text{abs}}$  variation (-0.2 and 1.3 for the 380-750 nm wavelength range) for BC particles  
303 with coatings are possible and  $\alpha_{\text{abs}}$  values less than 1.0 may occur with larger BC particles (e.g.,  
304  $r_{\text{core}} > 0.1 \mu\text{m}$  and  $r_{\text{shell}} > 0.25 \mu\text{m}$ ). *Gyawali et al.* [2012] showed laboratory measurements of  
305 kerosene soot particles have  $\alpha_{\text{abs}}$  values of  $\sim 0.8$  for the 355-1020 nm range and in situ  
306 measurement values of  $\alpha_{\text{abs}}$  measured during the Reno, Nevada, winter period varied for clean  
307 days ( $\text{PM}_{2.5} < 40 \mu\text{g}/\text{m}^3$ ) between  $\sim 1.0$  and 1.4 and for polluted days ( $\text{PM}_{2.5} \geq 40 \mu\text{g}/\text{m}^3$ ) between  
308 0.9 and 1.2 for the 405-870 nm wavelength range. Although these model simulations and  
309 laboratory and in situ measurements suggest  $\alpha_{\text{abs}}$  values may occur below 1.0, AERONET  
310 remotely sensed values of  $\alpha_{\text{abs}}$  have not yet been compared to coincident column-effective in situ

311 measurements (e.g., measured by aircraft) but this analysis will be addressed in future work. In  
312 the present analysis, the simulated underestimation of spectral  $\omega_o$  (i.e.,  $\delta\omega_o=+0.03$ ) for Dust and  
313 Mixed indicates possible underestimation of the unperturbed  $\alpha_{abs}$ , which could also result in  $\alpha_{abs}$   
314 below 1.0. Table 3 also shows the Dust and Mixed categories change in the same direction as  
315 the  $\omega_o$  perturbation possibly due to weak spectral dependence of  $\tau_{ext}$ , while U/I and BB  
316 categories have the opposite response. Two additional tests were conducted by perturbing  $\omega_o$   
317 using the wavelength pair (440-870 nm) and only varying the end points of the 440-675-870 nm  
318 set and the differences between unperturbed  $\alpha_{abs}$  averages were minimal (not shown). However,  
319 perturbing one  $\omega_o$  end point for either the 440-870 nm wavelength pair (not shown) or the 440-  
320 675-870 nm set (Table 3) produced very large deviations in  $\alpha_{abs}$  by up to  $\sim 1.2$  for Dust,  $\sim 0.7$  for  
321 Mixed,  $\sim 1.0$  for U/I, and  $\sim 0.6$  for BB. The perturbation of end points simulates atypical behavior  
322 of the instrument while deployed in the field (e.g., anomalous filter degradation) showing  
323 potential issues in using real-time data products unless further screening is implemented, such as  
324 the instrument collimator consistency checks (stated in Section 2.0), which may be utilized to  
325 help remove  $\omega_o$  artifacts (i.e., collimator or sensor head window obstructions) and improve the  
326 reliability of  $\alpha_{abs}$  retrievals. These sensitivity tests quantified the effect of the reduction of  $\omega_o$   
327 uncertainty on improving estimates of  $\alpha_{abs}$ .

### 328 **3.3 Cluster Analysis by Dominant Aerosol Type**

329 Knowledge of aerosol particle spectral absorption provides insight to determine aerosol  
330 particle dominance of dust, carbonaceous matter, or hygroscopic aerosols (e.g., sulfates, nitrates,  
331 or sea salt). While the co-albedo (or  $1-\omega_o$ ) indicates the magnitude of absorption and  $\alpha_{abs}$   
332 provides some indication of the dominance of carbonaceous particles (e.g., BC, BrC, and OC) or  
333 iron oxides in dust, these parameters alone cannot fully describe the aerosol particle type.

334 Recent studies have suggested applying an aerosol particle size parameter (e.g.,  $\alpha_{\text{ext}}$  or  $\eta$ ) to  
335 separate larger dust particles from other aerosol types and mixtures [Lee *et al.*, 2010; Russell *et*  
336 *al.*, 2010; Giles *et al.*, 2010; 2011a; 2011b]. In this section, several years of AERONET  
337 retrievals of  $\omega_{0440\text{nm}}$ ,  $\alpha_{\text{abs}440-870\text{nm}}$ ,  $\alpha_{\text{ext}440-870\text{nm}}$ , and  $\eta_{550\text{nm}}$  (with wavelength subscripts removed  
338 hereafter) were analyzed for each dominant aerosol type category using a density based  
339 clustering utilizing the Voronoi tessellation [Voronoi 1908; Ishimoto *et al.*, 2010] to determine  
340 the relative concentration of points (density = 1/polygon area) for each absorption and size  
341 relationship. In these density plots (e.g., Figures 7-10), the high density represents the primary  
342 mode for the dominant aerosol particle type category. Various clustering techniques were  
343 attempted previously to categorize dominant aerosol particle type at AERONET sites [Cattrall *et*  
344 *al.*, 2005; Omar *et al.*, 2005; Levy *et al.*, 2007; Qin and Mitchell 2009; Russell *et al.*, 2010,  
345 Boselli *et al.*, 2012]. For each absorption and size relationship and aerosol type category in this  
346 study (Figure 11), dominant aerosol particle clusters were computed using averages weighted by  
347 density magnitudes normalized to a 64-level scale (corresponding to a 64-bit color scale).  
348 Although weighting reduces the bias introduced by outliers affecting the normal average,  
349 additional thresholds were applied to the aerosol size parameters. To further define weighted  
350 cluster averages, the  $\alpha_{\text{ext}}$  cluster averages utilized a 0.8 threshold, where  $>0.8$  indicates mainly  
351 small sub-micron radius particles and  $\leq 0.8$  is mainly large super-micron radius particles (where  
352  $\alpha_{\text{ext}440-870\text{nm}}=0.8$  is approximately equivalent to  $\eta_{500\text{nm}}=0.5$  as shown for example by Eck *et al.*  
353 [2005; 2010]). In addition, the  $\eta$  cluster averages were defined using thresholds of 0.0 to  $\leq 0.33$   
354 (for coarse mode dominated particles), 0.33 to  $\leq 0.66$  (for mixed size), and  $>0.66$  (for fine mode  
355 dominated particles). For the BB category (Figure 11c and Figure 11d), cluster separation was



356 imposed to calculate two additional clusters using a  $\omega_{0440\text{nm}}$  threshold of 0.90 based on the  
357 density cluster analyses shown in Figure 9 and Figure 10.

358 The relationships of aerosol absorption and size are analyzed with respect to the  
359 dominant aerosol type category. For Figures 7-10, the primary density clusters are clear  
360 (denoted by orange and red regions representing relative value levels of ~45 to 64). For  
361 example, the Dust category shows a cluster in the region with  $\alpha_{\text{ext}}$  of ~0.2-0.3 and  $\eta$  of ~0.2-0.3,  
362 indicative of domination by coarse mode particles. To provide a better assessment of the  
363 clusters, the weighted cluster average and one standard deviation was calculated for each  
364 parameter shown in Figure 11. In Figure 11a and Figure 11b, the primary Dust clusters show  
365 variation of the  $\alpha_{\text{abs}}$  mainly between 1.5 and 2.3, which are slightly lower values than reported by  
366 *Russell et al.* [2010]. In Figure 11c and Figure 11d,  $\omega_o$  also varies significantly in the primary  
367 Dust cluster from 0.89-0.93, possibly due to variation in mineral composition of dust [*Sokolik*  
368 *and Toon* 1999]. For  $\alpha_{\text{ext}}$  (Figure 11a and Figure 11c), the Mixed aerosol type category has two  
369 primary density clusters (1) “Mixed-Large Particle” cluster for mainly super-micron particles  
370 (centered at  $\alpha_{\text{ext}}\sim 0.4$ ) and (2) “Mixed-Small Particle” for mainly submicron particles (centered at  
371  $\alpha_{\text{ext}}\sim 1.25$ ). In comparison to the Dust cluster, the Mixed-Large Particle cluster tends to have a  
372 slightly smaller contribution to larger particles in the 0.3-0.6  $\alpha_{\text{ext}}$  range, while  $\eta$  relationships  
373 (Figure 11b and Figure 11d) show the Mixed-Large Particle cluster for coarse particles is nearly  
374 identical to the Dust cluster. The Mixed category for mixed sizes ( $0.33 < \eta_{550\text{nm}} \leq 0.66$ ) does not  
375 show high cluster density due to varying sizes and contributions of the aerosol particles  
376 containing dust with pollution or biomass burning smoke with strongly varying absorption [*Eck*  
377 *et al.*, 2010]. In Figure 11, the Mixed-Small Particle clusters ( $\alpha_{\text{ext}}\sim 1.0$  to 1.5; also  $\eta\sim 0.8$ -0.95)  
378 show significant variability likely due to variation in carbonaceous particle contribution

379 (primarily BC but also OC) with  $\alpha_{\text{abs}}$  between  $\sim 1.3$  and  $1.7$  similar to those observed in Kanpur  
380 for fine mode dominated cases by *Giles et al.* [2011a]. As indicated by *Russell et al.* [2010] and  
381 shown in Figure 6, the U/I and BB category types for the  $\alpha_{\text{abs}}$  and  $\alpha_{\text{ext}}$  relationship tend to overlap  
382 each other. For primary density clusters in these two categories, the  $\alpha_{\text{abs}}$  vary from  $\sim 1.1$  to  $1.8$ .  
383 Until the  $\omega_o$  uncertainty is known and constrained further (given the sensitivity results of Section  
384 3.2), the usefulness of  $\alpha_{\text{abs}}$  to determine various carbonaceous aerosol particles is doubtful except  
385 in separating cases dominated by BC from cases dominated by BrC or OC. A “region” of higher  
386  $\alpha_{\text{abs}}$  values from the density cluster analysis for BB (Figure 7d and Figure 8d) likely indicates  
387 aged smoke from primarily smoldering combustion containing higher concentrations of BrC or  
388 OC and relatively low BC [*Eck et al.*, 2009; *Moosmüller et al.*, 2009; 2011], especially above an  
389  $\alpha_{\text{abs}}$  of  $1.6$  for fine mode particles [*Lack and Cappa* 2010] and also supported by Figure 9d and  
390 Figure 10d with  $\omega_o$  above  $0.90$ . For example, according to *Eck et al.* [2009] and *Arola et al.*  
391 [2011], significant absorbing OC concentrations and high OC/BC ratios likely occurred at the  
392 Bonanza Creek site where Table 2 shows the spectral  $\omega_o$  average is  $\sim 0.95$  and averages of  $\alpha_{\text{abs}}$ ,  
393  $\alpha_{\text{ext}}$ , and  $\eta_{550\text{nm}}$  are  $1.8$ ,  $1.5$ , and  $0.96$ , respectively. However, the  $\omega_o$  relationships (Figure 11c  
394 and Figure 11d) show more cluster separation than  $\alpha_{\text{abs}}$  relationships (Figure 11a and Figure  
395 11b). In Figure 11d, the primary U/I cluster is centered above  $0.95$ , while the main BB cluster is  
396 centered on  $\sim 0.89$  with two BB sub-clusters centered on  $\sim 0.93$  and  $\sim 0.87$   $\omega_o$  (calculated by using  
397  $\omega_o$  threshold of  $0.90$ ); however, the BB clusters overlap with the Mixed-Small Particle cluster.  
398 The  $\omega_{o440\text{nm}}$  and  $\alpha_{\text{ext}440-870\text{nm}}$  relationship (Figure 11c) shows distinct high density clusters in all  
399 categories (i.e., between Dust and Mixed-Large Particle, and among Mixed-Small Particle-U/I-  
400 BB, and between U/I-BB clusters), while minimal overlap occurs with the U/I and the less  
401 absorbing ( $\omega_o > 0.90$ ) BB sub-cluster. Nonetheless, the analysis has shown that the  $\omega_{o440\text{nm}}$  and

402  $\alpha_{\text{ext}440-870 \text{ nm}}$  relationship demonstrates that the dominant particle type may be ascertained simply  
403 from commonly measured or retrieved absorption and size parameters.

#### 404 **4.0 Conclusions**

405 In this paper, absorption properties (i.e., single scattering albedo and absorption  
406 Ångström exponent) were averaged for 19 AERONET sites to show correspondence to  
407 representative aerosol source regions. Sensitivity tests on absorption Ångström exponent were  
408 performed by varying the single scattering albedo within plausible constraints based on  
409 uncertainty estimates. Lastly, the absorption and size relationships were evaluated and compared  
410 to each other based on the dominant aerosol type categorizations.

411 (1) A summary of aerosol absorption parameters from the AERONET Version 2, Level  
412 2.0 almucantar retrievals was presented to expand upon previous work using pre-Version 1  
413 retrievals. A comparison of five sites common to *Dubovik et al.* [2002] showed a 0.01 average  
414 spectral (from 440 to 1020 nm) decrease in single scattering albedo ( $\omega_o$ ) with the largest  
415 decreases spectrally of 0.02 at Capo Verde and GSFC AERONET sites. The average absorption  
416 Ångström exponent ( $\alpha_{\text{abs}440-870 \text{ nm}}$ ) computed from Version 2 retrievals was 1.2 lower for Capo  
417 Verde and 0.25 higher for GSFC than reported by *Russell et al.* [2010] computed from pre-  
418 Version 1 retrievals. Aerosol mixtures exhibited stronger spectral absorption (i.e., lower  $\omega_o$ ) and  
419 increased dominance of absorbing carbonaceous particles (i.e., lower  $\alpha_{\text{abs}440-870 \text{ nm}}$ ) than for dust  
420 alone, possibly due to an optical mixture state (e.g., dust and smoke or dust and pollution) or the  
421 aggregation of dust and carbonaceous particles.

422 (2) The  $\alpha_{\text{abs}440-870 \text{ nm}}$  calculated from AERONET data ranged from ~0 to 3.5 among  
423 dominant aerosol type categories. Frequency distributions of  $\alpha_{\text{abs}440-870 \text{ nm}}$  exhibited significant

424 overlap among aerosol types, while the Urban/Industrial and Biomass Burning distributions were  
425 nearly identical for  $\alpha_{\text{abs}440-870 \text{ nm}}$  values above 1.0. Further, frequency distributions showed  
426 approximately 10% of the  $\alpha_{\text{abs}}$  retrievals had values below 1.0 for most aerosol categories but as  
427 high as 22% for the Urban/Industrial category.

428 (3) A sensitivity study perturbing the  $\omega_o$  by the current AERONET uncertainty ( $\pm 0.03$ )  
429 showed  $\alpha_{\text{abs}}$  changes by at least  $\sim \pm 0.6$  for Dust,  $\sim \pm 0.2$  for Mixed, and  $\sim \pm 0.1$  for Urban/Industrial  
430 and Biomass Burning. The sensitivity study quantified the improvement in estimates of  $\alpha_{\text{abs}}$   
431 resulting from reducing the  $\omega_o$  uncertainty. Variations within the uncertainty of  $\omega_o$  retrievals  
432 may explain some of the observed  $\alpha_{\text{abs}}$  values below 1.0 in AERONET data although in situ  
433 measurements suggest that some of these  $\alpha_{\text{abs}}$  values may be real depending on the aerosol  
434 particle composition and size.

435 (4) Absorption and size relationships were examined using density cluster analysis for  
436 each dominant aerosol particle type. The  $\omega_{o440\text{nm}}$  vs.  $\alpha_{\text{ext}440-870 \text{ nm}}$  relationship showed at least five  
437 distinct aerosol type clusters [Dust, Mixed-Large Particle, Mixed-Small Particle,  
438 Urban/Industrial, and Biomass Burning (with two sub-clusters)], while the  $\alpha_{\text{abs}440-870 \text{ nm}}$  vs.  $\alpha_{\text{ext}440-870 \text{ nm}}$   
439 relationship had fewer distinct clusters due to less definition for mainly small aerosol  
440 particles ( $\alpha_{\text{ext}440-870 \text{ nm}} > 1.5$ ).

441 We showed the  $\omega_{o440\text{nm}}$  and  $\alpha_{\text{ext}440-870\text{nm}}$  relationship provided a better clustering  
442 relationship and it may be applied to measurements of aerosol absorption and size properties  
443 derived from surface- and potentially future space-based platforms. From  $\omega_{o440\text{nm}}$  and  $\alpha_{\text{ext}440-870 \text{ nm}}$   
444 clusters, at least, major dominant aerosols types and some mixtures can be identified using  
445 common aerosol absorption and size parameters without prior knowledge of aerosol transport or

446 source regions. Alternatively, when  $\omega_o$  is not available but  $\alpha_{\text{abs}440-870\text{nm}}$  is (e.g., using spectral  
447 absorption coefficients measured in situ to calculate  $\alpha_{\text{abs}}$ ),  $\alpha_{\text{abs}440-870\text{nm}}$  vs.  $\alpha_{\text{ext}440-870\text{nm}}$  may also  
448 provide a reasonable aerosol type classification. A combination of  $\omega_{o440\text{nm}}$  vs.  $\alpha_{\text{ext}440-870\text{nm}}$  and  
449  $\alpha_{\text{abs}440-870\text{nm}}$  vs.  $\alpha_{\text{ext}440-870\text{nm}}$  relationships could provide a more detailed classification of aerosol  
450 composition which will require further investigation.

## 451 **5.0 Acknowledgements**

452 The NASA AERONET project is supported by the NASA EOS project office, and the  
453 Radiation Sciences Program, NASA Headquarters. We would like to thank the following  
454 principal investigators and their staff for maintaining the following sites: Didier Tanre  
455 (Banizoumbou, Capo Verde, Dakar, and Ouagadougou), Naif Al-Abbadi (Solar Village), Rachel  
456 Pinker (Ilorin), Sachi Tripathi and Ramesh Singh (Kanpur), Arnon Karnieli (SEDE BOKER),  
457 Pucal Wang (XiangHe), Giuseppe Zibordi (Ispra), Amando L. Contreras (Mexico City),  
458 Alexander Aculinin (Moldova), Itaru Sano (Shirahama), Paulo Artaxo (Abracos Hill, Alta  
459 Floresta), John Vande Castle (Bonanza Creek), and Ross Mitchell (Lake Argyle). Finally, the  
460 authors thank the AERONET team for calibrating and maintaining instrumentation and  
461 processing these data.

462 **References**

- 463 Ångström, A. (1964), The parameters of atmospheric turbidity, *Tellus*, *16*(1), 64–75.
- 464 Arola, A., Schuster, G., Myhre, G., Kazadzis, S., Dey, S., and Tripathi, S. N. (2011), Inferring  
465 absorbing organic carbon content from AERONET data, *Atmos. Chem. Phys.*, *11*, 215-  
466 225, doi:10.5194/acp-11-215-2011.
- 467 Basart, S., Pérez, C., Cuevas, E., Baldasano, J. M., and Gobbi, G. P. (2009), Aerosol  
468 characterization in Northern Africa, Northeastern Atlantic, Mediterranean Basin and  
469 Middle East from direct-sun AERONET observations, *Atmos. Chem. Phys.*, *9*, 8265-  
470 8282, doi:10.5194/acp-9-8265-2009.
- 471 Bergstrom, R. W., P. B. Russell, and P. Hignett (2002), Wavelength dependence of the  
472 absorption of black carbon particles: Predictions and results from the TARFOX  
473 experiment and implications for the aerosol single scattering albedo, *J. Atmos. Sci.*, *59*,  
474 567–577, doi:10.1175/1520-0469(2002)059<0567:WDOTAO>2.0.CO;2.
- 475 Bergstrom, R. W., P. Pilewskie, P. B. Russell, J. Redemann, T. C. Bond, P. K. Quinn, and B.  
476 Sierau (2007), Spectral absorption properties of atmospheric aerosols, *Atmos. Chem.*  
477 *Phys.*, *7*, 5937–5943, doi:10.5194/acp-7-5937-2007.
- 478 Boselli A., R. Caggiano, C. Cornacchia, F. Madonna, L. Mona, M. Macchiato, G. Pappalardo,  
479 and S. Trippetta (2012), Multi year sun-photometer measurements for aerosol  
480 characterization in a Central Mediterranean site, *J. Atmos. Res.*, *104-105*, 98-110, doi:  
481 10.1016/j.atmosres.2011.08.002.
- 482 Burton, S. P., Ferrare, R. A., Hostetler, C. A., Hair, J. W., Rogers, R. R., Obland, M. D., Butler,  
483 C. F., Cook, A. L., Harper, D. B., and Froyd, K. D. (2012), Aerosol classification using  
484 airborne High Spectral Resolution Lidar measurements – methodology and examples,  
485 *Atmos. Meas. Tech.*, *5*, 73-98, doi:10.5194/amt-5-73-2012.
- 486 Cattrall, C., J. Reagan, K. Thome, and O. Dubovik (2005), Variability of aerosol and spectral  
487 lidar and backscatter and extinction ratios of key aerosol types derived from selected  
488 Aerosol Robotic Network locations, *J. Geophys. Res.*, *110*, D10S11,  
489 doi:10.1029/2004JD005124.
- 490 Derimian, Y., A. Karnieli, Y. J. Kaufman, M. O. Andreae, T. W. Andreae, O. Dubovik, W.  
491 Maenhaut, I. Koren, and B. N. Holben (2006), Dust and pollution aerosols over the  
492 Negev desert, Israel: Properties, transport, and radiative effect, *J. Geophys. Res.*, *111*,  
493 D05205, doi:10.1029/2005JD006549.
- 494 Derimian, Y., A. Karnieli, Y. J. Kaufman, M. O. Andreae, T. W. Andreae, O. Dubovik, W.  
495 Maenhaut, and I. Koren (2008), The role of iron and black carbon in aerosol light  
496 absorption, *Atmos. Chem. Phys.*, *8*, 3623–3637, doi:10.5194/acp-8-3623-2008.

- 497 Dey, S., S. N. Tripathi, R. P. Singh, and B. N. Holben (2004), Influence of dust storms on the  
498 aerosol optical properties over the Indo-Gangetic basin, *J. Geophys. Res.*, *109*, D20211,  
499 doi:10.1029/2004JD004924.
- 500 Diner, D. J., G. P. Asner, R. Davies, Y. Knyazikhin, J.-P. Muller, A. W. Nolin, B. Pinty, C. B.  
501 Schaaf, and J. Stroeve (1999), New Directions in Earth Observing: Scientific  
502 Applications of Multiangle Remote Sensing, *Bull. Amer. Meteor. Soc.*, *80*, 2209–2228,  
503 doi: [http://dx.doi.org/10.1175/1520-0477\(1999\)080<2209:NDIEOS>2.0.CO;2](http://dx.doi.org/10.1175/1520-0477(1999)080<2209:NDIEOS>2.0.CO;2).
- 504 Dubovik, O., and M. D. King (2000), A flexible inversion algorithm for retrieval of aerosol  
505 optical properties from Sun and sky radiance measurements, *J. Geophys. Res.*, *105*,  
506 20,673–20,696, doi:10.1029/2000JD900282.
- 507 Dubovik, O., A. Smirnov, B. N. Holben, M. D. King, Y. J. Kaufman, T. F. Eck, and I. Slutsker  
508 (2000), Accuracy assessments of aerosol optical properties retrieved from AERONET  
509 Sun and sky-radiance measurements, *J. Geophys. Res.*, *105*, 9791–9806,  
510 doi:10.1029/2000JD900040.
- 511 Dubovik, O., B. N. Holben, T. F. Eck, A. Smirnov, Y. J. Kaufman, M. D. King, D. Tanre, and I.  
512 Slutsker (2002), Variability of absorption and optical properties of key aerosol types  
513 observed in worldwide locations, *J. Atmos. Sci.*, *59*, 590–608, doi:10.1175/1520-  
514 0469(2002)059<0590:VOAAOP>2.0.CO;2.
- 515 Dubovik, O., et al. (2006), Application of spheroid models to account for aerosol particle  
516 nonsphericity in remote sensing of desert dust, *J. Geophys. Res.*, *111*, D11208,  
517 doi:10.1029/2005JD006619.
- 518 Eck, T. F., B. N. Holben, J. S. Reid, O. Dubovik, A. Smirnov, N. T. O’Neill, I. Slutsker, and S.  
519 Kinne (1999), Wavelength dependence of the optical depth of biomass burning, urban,  
520 and desert dust aerosols, *J. Geophys. Res.*, *104*(D24), 31,333–31,349,  
521 doi:10.1029/1999JD900923.
- 522 Eck, T. F., et al. (2003a), Variability of biomass burning aerosol optical characteristics in  
523 southern Africa during the SAFARI 2000 dry season campaign and a comparison of  
524 single scattering albedo estimates from radiometric measurements, *J. Geophys.*  
525 *Res.*, *108*(D13), 8477, doi:10.1029/2002JD002321.
- 526 Eck, T. F., B. N. Holben, J. S. Reid, N. T. O’Neill, J. S. Schafer, O. Dubovik, A. Smirnov, M. A.  
527 Yamasoe, and P. Artaxo (2003b), High aerosol optical depth biomass burning events: A  
528 comparison of optical properties for different source regions, *Geophys. Res. Lett.*, *30*(20),  
529 2035, doi:10.1029/2003GL017861.
- 530 Eck, T. F., et al. (2005), Columnar aerosol optical properties at AERONET sites in central  
531 eastern Asia and aerosol transport to the tropical mid-Pacific, *J. Geophys. Res.*, *110*,  
532 D06202, doi:10.1029/2004JD005274.
- 533 Eck, T. F., et al. (2008), Spatial and temporal variability of column-integrated aerosol optical  
534 properties in the southern Arabian Gulf and United Arab Emirates in summer, *J.*  
535 *Geophys. Res.*, *113*, D01204, doi:10.1029/2007JD008944.

- 536 Eck, T. F., et al. (2009), Optical properties of boreal region biomass burning aerosols in central  
537 Alaska and seasonal variation of aerosol optical depth at an Arctic coastal site, *J.*  
538 *Geophys. Res.*, *114*, D11201, doi:10.1029/2008JD010870.
- 539 Eck, T. F., et al. (2010), Climatological aspects of the optical properties of fine/coarse mode  
540 aerosol mixtures, *J. Geophys. Res.*, *115*, D19205, doi:10.1029/2010JD014002.
- 541 Giles, D. M., B. Hoblen, T. F. Eck, A. Sinyuk, R.R. Dickerson, A. M. Thompson, I. Slutsker, Z.  
542 Li, S. N. Tripathi, R. Singh, and G. Zibordi (2010), Identifying Aerosol Type/Mixture  
543 from Aerosol Absorption Properties Using AERONET, *Eos Trans. AGU*, *91*(26), West.  
544 Pac. Geophys. Meet. Suppl., Abstract A33D-05.
- 545 Giles, D. M., et al. (2011a), Aerosol properties over the Indo-Gangetic Plain: A mesoscale  
546 perspective from the TIGERZ experiment, *J. Geophys. Res.*, *116*, D18203,  
547 doi:10.1029/2011JD015809.
- 548 Giles, D. M., B. N. Holben, T. F. Eck, A. Sinyuk, A. Smirnov, I. Slutsker, R. R. Dickerson, A.  
549 M. Thompson, and J. S. Schafer (2011b), Dominant Aerosol Particle Type/Mixture  
550 Identification at Worldwide Locations Using the Aerosol Robotic Network (AERONET),  
551 Abstract A14E-07 presented at 2011 Fall Meeting, AGU, San Francisco, Calif., 5-9 Dec.
- 552 Gobbi, G. P., Kaufman, Y. J., Koren, I., and Eck, T. F. (2007), Classification of aerosol  
553 properties derived from AERONET direct sun data, *Atmos. Chem. Phys.*, *7*, 453-458,  
554 doi:10.5194/acp-7-453-2007.
- 555 Gyawali, M., et al. (2012), Photoacoustic optical properties at UV, VIS, and near IR wavelengths  
556 for laboratory generated and winter time ambient urban aerosols, *Atmos. Chem. Phys.*, *12*,  
557 2587-2601, doi:10.5194/acp-12-2587-2012.
- 558 Holben, B. N., T. F. Eck, and R. S. Fraser (1991), Temporal and spatial variability of aerosol  
559 optical depth in the Sahel region in relation to vegetation remote sensing, *Int. J. Remote*  
560 *Sens.*, *12*(6), 1147–1163, doi:10.1080/01431169108929719.
- 561 Holben, B. N., et al. (1998), AERONET—A federated instrument network and data archive for  
562 aerosol characterization, *Remote Sens. Environ.*, *66*, 1–16, doi:10.1016/S0034-  
563 4257(98)00031-5.
- 564 Holben, B. N., et al. (2001), An emerging ground-based aerosol climatology: Aerosol optical  
565 depth from AERONET, *J. Geophys. Res.*, *106*(D11), 12,067–12,097,  
566 doi:10.1029/2001JD900014.
- 567 Holben, B. N., T. F. Eck, I. Slutsker, A. Smirnov, A. Sinyuk, J. Schafer, D. Giles, and O.  
568 Dubovik (2006), AERONET’s Version 2.0 quality assurance criteria, in Remote Sensing  
569 of the Atmosphere and Clouds, edited by S.-C. Tsay et al., *Proc. SPIE*, *6408*, 64080Q,  
570 doi:10.1117/12.706524.
- 571 Ishimoto, H., Y. Zaizen, A. Uchiyama, K. Masuda, Y. Mano (2010), Shape modeling of mineral  
572 dust particles for light-scattering calculations using the spatial Poisson–Voronoi  
573 tessellation, *J. of Quant. Spect. and Rad. Trans.*, *111*, 16, doi:10.1016/j.jqsrt.2010.06.018.



- 574 Jeong, M.-J., and Z. Li (2005), Quality, compatibility, and synergy analyses of global aerosol  
575 products derived from the advanced very high resolution radiometer and Total Ozone  
576 Mapping Spectrometer, *J. Geophys. Res.*, *110*, D10S08, doi:10.1029/2004JD004647.
- 577 Johnson, B. T., S. Christopher, J. M. Haywood, S. R. Osborne, S. McFarlane, C. Hsu, C.  
578 Salustro, and R. Kahn (2009), Measurements of aerosol properties from aircraft and  
579 ground-based remote sensing: A case-study from the Dust and Biomass-burning  
580 Experiment (DABEX), *Q. J. R. Met. Soc.*, *135*, doi:10.1002/qj.420.
- 581 Kahn, R. A., B. J. Gaitley, M. J. Garay, D. J. Diner, T. F. Eck, A. Smirnov, and B. N. Holben  
582 (2010), Multiangle Imaging Spectroradiometer global aerosol product assessment by  
583 comparison with the Aerosol Robotic Network, *J. Geophys. Res.*, *115*, D23209,  
584 doi:10.1029/2010JD014601.
- 585 Kalapureddy, M. C. R., D. G. Kaskaoutis, P. Ernest Raj, P. C. S. Devara, H. D. Kambezidis, P.  
586 G. Kosmopoulos, and P. T. Nastos (2009), Identification of aerosol type over the Arabian  
587 Sea in the premonsoon season during the Integrated Campaign for Aerosols, Gases and  
588 Radiation Budget (ICARB), *J. Geophys. Res.*, *114*, D17203, doi:10.1029/2009JD011826.
- 589 Kaufman, Y. J., A. Setzer, D. Ward, D. Tanre, B. N. Holben, P. Menzel, M. C. Pereira, and R.  
590 Rasmussen (1992), Biomass Burning Airborne and Spaceborne Experiment in the  
591 Amazonas (BASE-A), *J. Geophys. Res.*, *97*(D13), 14,581–14,599,  
592 doi:10.1029/92JD00275.
- 593 Kim, D., Chin, M., Yu, H., Eck, T. F., Sinyuk, A., Smirnov, A., and Holben, B. N. (2011), Dust  
594 optical properties over North Africa and Arabian Peninsula derived from the AERONET  
595 dataset, *Atmos. Chem. Phys.*, *11*, 10733-10741, doi:10.5194/acp-11-10733-2011.
- 596 Lack, D. A. and Cappa, C. D. (2010), Impact of brown and clear carbon on light absorption  
597 enhancement, single scatter albedo and absorption wavelength dependence of black  
598 carbon, *Atmos. Chem. Phys.*, *10*, 4207-4220, doi:10.5194/acp-10-4207-2010.
- 599 Leahy, L. V., T. L. Anderson, T. F. Eck, and R. W. Bergstrom (2007), A synthesis of single  
600 scattering albedo of biomass burning aerosol over southern Africa during SAFARI 2000,  
601 *Geophys. Res. Lett.*, *34*, L12814, doi:10.1029/2007GL029697.
- 602 Lee J., J. Kim, C.H. Song, S.B. Kim, Y. Chun, B.J. Sohn, and B.N. Holben (2010),  
603 Characteristics of aerosol types from AERONET sunphotometer measurements, *Atmos.*  
604 *Environ.*, *44*, 26, doi: 10.1016/j.atmosenv.2010.05.035.
- 605 Levy, R. C., L. A. Remer, and O. Dubovik (2007), Global aerosol optical properties and  
606 application to Moderate Resolution Imaging Spectroradiometer aerosol retrieval over  
607 land, *J. Geophys. Res.*, *112*, D13210, doi:10.1029/2006JD007815.
- 608 Mélin, F., and G. Zibordi (2005), Aerosol variability in the Po Valley analyzed from automated  
609 optical measurements, *Geophys. Res. Lett.*, *32*, L03810, doi:10.1029/2004GL021787.
- 610 Mielonen, T., A. Arola, M. Komppula, J. Kukkonen, J. Koskinen, G. de Leeuw, and K. E. J.  
611 Lehtinen (2009), Comparison of CALIOP level 2 aerosol subtypes to aerosol types

612 derived from AERONET inversion data, *Geophys. Res. Lett.*, *36*, L18804,  
613 doi:10.1029/2009GL039609.

614 Mitchell, R. M., D. M. O'Brien, and S. K. Campbell (2006), Characteristics and radiative impact  
615 of the aerosol generated by the Canberra firestorm of January 2003, *J. Geophys. Res.*,  
616 *111*, D02204, doi:10.1029/2005JD006304.

617 Moosmüller H., R. K. Chakrabarty, W. P. Arnott (2009), Aerosol light absorption and its  
618 measurement: A review, *J. of Quant. Spect. and Rad. Trans.*, *110*, 11,  
619 doi:10.1016/j.jqsrt.2009.02.035.

620 Moosmüller, H., Chakrabarty, R. K., Ehlers, K. M., and Arnott, W. P. (2011), Absorption  
621 Ångström coefficient, brown carbon, and aerosols: basic concepts, bulk matter, and  
622 spherical particles, *Atmos. Chem. Phys.*, *11*, 1217-1225, doi:10.5194/acp-11-1217-2011.

623 Müller, D., et al. (2010), Mineral dust observed with AERONET Sun photometer, Raman lidar,  
624 and in situ instruments during SAMUM 2006: Shape-independent particle properties, *J.*  
625 *Geophys. Res.*, *115*, D07202, doi:10.1029/2009JD012520.

626 Omar, A. H., J.-G. Won, D. M. Winker, S.-C. Yoon, O. Dubovik, and M. P. McCormick (2005),  
627 Development of global aerosol models using cluster analysis of Aerosol Robotic Network  
628 (AERONET) measurements, *J. Geophys. Res.*, *110*, D10S14,  
629 doi:10.1029/2004JD004874.

630 O'Neill, N. T., S. Thulasiraman, T. F. Eck, and J. S. Reid (2005), Robust optical features of fine  
631 mode size distributions: Application to the Québec smoke event of 2002, *J. Geophys.*  
632 *Res.*, *110*, D11207, doi:10.1029/2004JD005157.

633 Prasad, A. K., and R. P. Singh (2007), Changes in aerosol parameters during major dust storm  
634 events (2001–2005) over the Indo-Gangetic Plains using AERONET and MODIS data, *J.*  
635 *Geophys. Res.*, *112*, D09208, doi:10.1029/2006JD007778.

636 Qin, Y. and Mitchell, R. M. (2009), Characterisation of episodic aerosol types over the  
637 Australian continent, *Atmos. Chem. Phys.*, *9*, 1943-1956, doi:10.5194/acp-9-1943-2009.

638 Reid, J. S., T. F. Eck, S. A. Christopher, P. V. Hobbs, and B. Holben (1999), Use of the  
639 Ångström exponent to estimate the variability of optical and physical properties of aging  
640 smoke particles in Brazil, *J. Geophys. Res.*, *104*(D22), 27,473–27,489,  
641 doi:10.1029/1999JD900833.

642 Reid, J. S., et al (2003)., Analysis of measurements of Saharan dust by airborne and ground-  
643 based remote sensing methods during the Puerto Rico Dust Experiment (PRIDE), *J.*  
644 *Geophys. Res.*, *108*(D19), 8586, doi:10.1029/2002JD002493.

645 Russell, P. B., R. W. Bergstrom, Y. Shinozuka, A. D. Clarke, P. F. DeCarlo, J. L. Jimenez, J. M.  
646 Livingston, J. Redemann, O. Dubovik, and A. Strawa (2010), Absorption Ångström  
647 Exponent in AERONET and related data as an indicator of aerosol composition, *Atmos.*  
648 *Chem. Phys.*, *10*, 1155–1169, doi:10.5194/acp-10-1155-2010.

- 649 Sano, I., S. Mukai, Y. Okada, B. N. Holben, S. Ohta, and T. Takamura (2003), Optical properties  
650 of aerosols during APEX and ACE-Asia experiments, *J. Geophys. Res.*, *108*(D23), 8649,  
651 doi:10.1029/2002JD003263.
- 652 Satheesh, S. K., K. K. Moorthy (2005), Radiative effects of natural aerosols: A review, *Atmos.*  
653 *Environ.*, *39*, 11, 2089-2110, doi:10.1016/j.atmosenv.2004.12.029.
- 654 Schafer, J. S., T. F. Eck, B. N. Holben, P. Artaxo, and A. F. Duarte (2008), Characterization of  
655 the optical properties of atmospheric aerosols in Amazônia from long-term AERONET  
656 monitoring (1993–1995 and 1999–2006), *J. Geophys. Res.*, *113*, D04204,  
657 doi:10.1029/2007JD009319.
- 658 Singh, R. P., S. Dey, S. N. Tripathi, V. Tare, and B. Holben (2004), Variability of aerosol  
659 parameters over Kanpur, northern India, *J. Geophys. Res.*, *109*, D23206,  
660 doi:10.1029/2004JD004966.
- 661 Smirnov, A., B. N. Holben, T. F. Eck, O. Dubovik, and I. Slutsker (2000), Cloud-screening and  
662 quality control algorithms for the AERONET database, *Remote Sens. Environ.*, *73*, 337–  
663 349, doi:10.1016/S0034-4257(00) 00109-7.
- 664 Smirnov, A., B. N. Holben, Y. J. Kaufman, O. Dubovik, T. F. Eck, I. Slutsker, C. Pietras, R. N.  
665 Halthore (2002), Optical Properties of Atmospheric Aerosol in Maritime Environments.  
666 *J. Atmos. Sci.*, *59*, doi: 10.1175/1520-0469(2002)059<0501:OPOAAI>2.0.CO;2.
- 667 Smirnov, A., et al. (2009), Maritime Aerosol Network as a component of Aerosol Robotic  
668 Network, *J. Geophys. Res.*, *114*, D06204, doi:10.1029/2008JD011257.
- 669 Sokolik, I. N., and O. B. Toon (1999), Incorporation of mineralogical composition into models  
670 of the radiative properties of mineral aerosol from UV to IR wavelengths, *J. Geophys.*  
671 *Res.*, *104*(D8), 9423–9444, doi:10.1029/1998JD200048.
- 672 Tanré, D., Y. J. Kaufman, B. N. Holben, B. Chatenet, A. Karnieli, F. Lavenu, L. Blarel, O.  
673 Dubovik, L. A. Remer, and A. Smirnov (2001), Climatology of dust aerosol size  
674 distribution and optical properties derived from remotely sensed data in the solar  
675 spectrum, *J. Geophys. Res.*, *106*, 18,205–18,217, doi:10.1029/2000JD900663.
- 676 Toledano, C., et al. (2011), Optical properties of aerosol mixtures derived from sun-sky  
677 radiometry during SAMUM-2, *Tellus*, Ser. B, *63*, 635–648, doi:10.1111/j.1600-  
678 0889.2011.00573.x.
- 679 Voronoi, G. F. (1908). Nouvelles applications des parametres continus a la theorie de forms  
680 quadratiques, *J. Reine Agnew. Math.*, *134*, 198-287.
- 681 Yang, M., Howell, S. G., Zhuang, J., and Huebert, B. J. (2009), Attribution of aerosol light  
682 absorption to black carbon, brown carbon, and dust in China – interpretations of  
683 atmospheric measurements during EAST-AIRE, *Atmos. Chem. Phys.*, *9*, doi:10.5194/acp-  
684 9-2035-2009.

685

686 **Table 1.** Previous studies identifying regional aerosol sources affecting AERONET sites

Aerosol Type Source Regions	Affected AERONET Sites	Selected References
Most Regions with Various Types	Most Sites	<i>Holben et al.</i> [2001]
Dust - African	Banizoumbou, Capo Verde, Dakar, Ouagadougou,	<i>Tanre et al.</i> [2001]; <i>Reid et al.</i> [2003]
Dust - Asian	XiangHe, Shirahama	<i>Eck et al.</i> [2005]
Smoke -Amazonia	Abracos Hill, Alta Floresta	<i>Eck et al.</i> [2003b]; <i>Schafer et al.</i> [2008]
Smoke - Australian	Lake_Argyle	<i>Mitchell et al.</i> [2006]
Smoke - Boreal	Bonanza Creek	<i>Eck et al.</i> [2009]
Smoke - African	Mongu	<i>Eck et al.</i> [2003a; 2003b]
Pollution - Europe	Ispira	<i>Melin and Zibordi</i> [2005]
Mixed - Asia	XiangHe, SEDE_BOKER	<i>Derimian et al.</i> [2006]; <i>Eck et al.</i> [2010]; <i>Yang et al.</i> [2009]
Mixed - India	Kanpur	<i>Dey et al.</i> [2004]; <i>Singh et al.</i> [2004]; <i>Prasad et al.</i> [2007]; <i>Giles et al.</i> [2011a]
Mixed - Africa	Ilorin	<i>Eck et al.</i> [2010]

687 **Table 2.** Average aerosol absorption and size properties by aerosol type category from AERONET Version 2 almucantar retrievals<sup>a</sup>

Site	Date Range	$\omega_o$	$\alpha_{\text{abs}440-870\text{nm}}$	$\alpha_{\text{ext}440-870\text{nm}}$	$\eta_{550\text{nm}}$	N
		440/675/870/1020 nm				
Dust						
Banizoumbou	1999-2010	0.91/0.95/0.96/0.96 0.04/0.04/0.04/0.04	1.7±0.6	0.3±0.2	0.28±0.20	2901
Capo_Verde	1999-2010	0.91/0.96/0.97/0.97 0.03/0.03/0.03/0.03	2.0±0.6	0.2±0.2	0.24±0.16	1202
Dakar	2000-2010	0.89/0.95/0.96/0.96 0.03/0.04/0.04/0.03	1.9±0.6	0.3±0.2	0.28±0.23	2250
Ouagadougou	1999-2007	0.90/0.94/0.95/0.95 0.04/0.04/0.04/0.03	1.6±0.5	0.3±0.2	0.30±0.21	1497
Solar_Village	1999-2010	0.91/0.95/0.96/0.96 0.02/0.02/0.02/0.02	1.8±0.6	0.3±0.3	0.28±0.25	3029
Mixed (for $0.33 < \eta_{550\text{nm}} \leq 0.66$ )						
Ilorin	1999-2009	0.86/0.90/0.92/0.92 0.05/0.05/0.04/0.04	1.6±0.4	0.7±0.2	0.47±0.23	798
Kanpur	2001-2010	0.87/0.90/0.92/0.93 0.03/0.03/0.03/0.03	1.4±0.4	0.7±0.2	0.48±0.22	963
SEDE_BOKER	1999-2010	0.91/0.93/0.93/0.94 0.02/0.02/0.03/0.03	1.2±0.5	0.7±0.2	0.48±0.20	170
XiangHe	2001, 2004-2010	0.88/0.92/0.93/0.93 0.03/0.03/0.03/0.03	1.8±0.4	0.8±0.2	0.53±0.22	446

688 <sup>a</sup> Aerosol optical depth (AOD) at 440 nm is greater than 0.4 for Version 2, Level 2.0 almucantar retrievals. The spectral single  
689 scattering albedo ( $\omega_o$ ) averages are listed first followed by their standard deviations. The absorption and extinction Ångström  
690 exponents ( $\alpha_{\text{abs}}$  and  $\alpha_{\text{ext}}$ ) and are computed using the 440-675-870 nm wavelength interval. The fine mode fraction of the AOD  
691 ( $\eta_{550\text{nm}}$ ) is interpolated to 550 nm as discussed in Section 2.

692

**Table 2.** (Continued)

Site	Date Range	$\omega_o$ 440/675/870/1020 nm	$\alpha_{\text{abs}440-870\text{nm}}$	$\alpha_{\text{ext}440-870\text{nm}}$	$\eta_{550\text{nm}}$	N
Urban/Industrial						
GSFC	1999–2010	0.96/0.95/0.94/0.93 0.02/0.02/0.03/0.03	1.1±0.2	1.8±0.2	0.94±0.20	882
Ispra	1999-2010	0.93/0.93/0.92/0.91 0.03/0.04/0.04/0.04	1.4±0.4	1.6±0.2	0.92±0.24	583
Mexico_City	1999-2010	0.89/0.88/0.86/0.85 0.04/0.04/0.05/0.06	1.3±0.3	1.6±0.2	0.87±0.18	540
Moldova	1999-2010	0.93/0.92/0.90/0.89 0.03/0.04/0.05/0.05	1.2±0.3	1.6±0.3	0.87±0.28	558
Shirahama	2000-2010	0.94/0.93/0.92/0.92 0.03/0.03/0.04/0.05	1.1±0.5	1.3±0.3	0.81±0.35	726
Biomass Burning						
Abracos_Hill	1999-2005	0.93/0.91/0.90/0.88 0.02/0.03/0.04/0.05	1.3±0.4	2.0±0.1	0.95±0.14	342
Alta_Floresta	1999-2010	0.93/0.92/0.90/0.89 0.02/0.03/0.04/0.05	1.5±0.4	1.9±0.2	0.92±0.18	593
Bonanaza Creek	1999-2005, 2008- 2010	0.95/0.96/0.96/0.95 0.03/0.03/0.04/0.04	1.8±0.5	1.5±0.2	0.96±0.22	144
Lake_Argyle	2002-2006, 2009-2010	0.85/0.83/0.82/0.81 0.04/0.05/0.06/0.07	1.4±0.3	1.5±0.4	0.79±0.36	176
Mongu	1999-2007, 2009	0.87/0.83/0.80/0.77 0.03/0.04/0.04/0.05	1.2±0.2	1.9±0.1	0.92±0.10	1411

695 **Table 3.** Sensitivity of the absorption Ångström exponent ( $\alpha_{\text{abs}}$ ) to perturbations of single  
696 scattering albedo ( $\omega_o$ ) for each dominant aerosol particle type

Type	$\lambda$ (nm)	$\alpha_{\text{abs}}$ <sup>a</sup>		$\delta\alpha_{\text{abs}}$ <sup>b</sup>			N
		$\delta\omega_o=0.0$	$\delta\omega_o$	All $\tau(\lambda)$	$\tau_{440\text{nm}}$	$\tau_{870\text{nm}}$	
Dust	440-675-870	1.76±0.58	-0.01	-0.27			10879
		1.67±0.52	+0.01 <sup>c</sup>	+0.40			9807
		1.76±0.58	-0.02	-0.45			10879
		1.49±0.42	+0.02 <sup>c</sup>	+0.67			7290
		1.76±0.58	-0.03 <sup>d</sup>	-0.57	+0.47	-0.90	10879
		1.33±0.38	+0.03 <sup>cd</sup>	+0.79	-0.54	+1.16	4898
		1.76±0.58	-0.04	-0.67			10879
		1.23±0.36	+0.04 <sup>c</sup>	+0.85			3342
Mixed	440-675-870	1.53±0.44	-0.01	-0.09			7199
		1.52±0.42	+0.01 <sup>c</sup>	+0.13			7051
		1.53±0.44	-0.02	-0.16			7199
		1.47±0.38	+0.02 <sup>c</sup>	+0.23			6623
		1.53±0.44	-0.03 <sup>d</sup>	-0.21	+0.40	-0.53	7199
		1.43±0.35	+0.03 <sup>cd</sup>	+0.30	-0.51	+0.71	6060
		1.53±0.44	-0.04	-0.25			7199
		1.40±0.33	+0.04 <sup>c</sup>	+0.35			5479
Urban/ Industrial	440-675-870	1.21±0.37	-0.01	+0.05			3289
		1.20±0.36	+0.01 <sup>c</sup>	-0.10			3174
		1.21±0.37	-0.02	+0.09			3289
		1.19±0.35	+0.02 <sup>c</sup>	-0.21			2874
		1.21±0.37	-0.03 <sup>d</sup>	+0.12	+0.74	-0.52	3289
		1.18±0.34	+0.03 <sup>cd</sup>	-0.31	-1.02	+0.58	2428
		1.21±0.37	-0.04	+0.14			3289
		1.18±0.34	+0.04 <sup>c</sup>	-0.40			2027
Biomass Burning	440-675-870	1.35±0.35	-0.01	+0.03			2666
		1.34±0.34	+0.01 <sup>c</sup>	-0.04			2639
		1.35±0.35	-0.02	+0.06			2666
		1.33±0.32	+0.02 <sup>c</sup>	-0.10			2598
		1.35±0.35	-0.03 <sup>d</sup>	+0.08	+0.45	-0.31	2666
		1.32±0.31	+0.03 <sup>cd</sup>	-0.19	-0.62	+0.35	2512
		1.35±0.35	-0.04	+0.11			2666
		1.31±0.29	+0.04 <sup>c</sup>	-0.29			2421

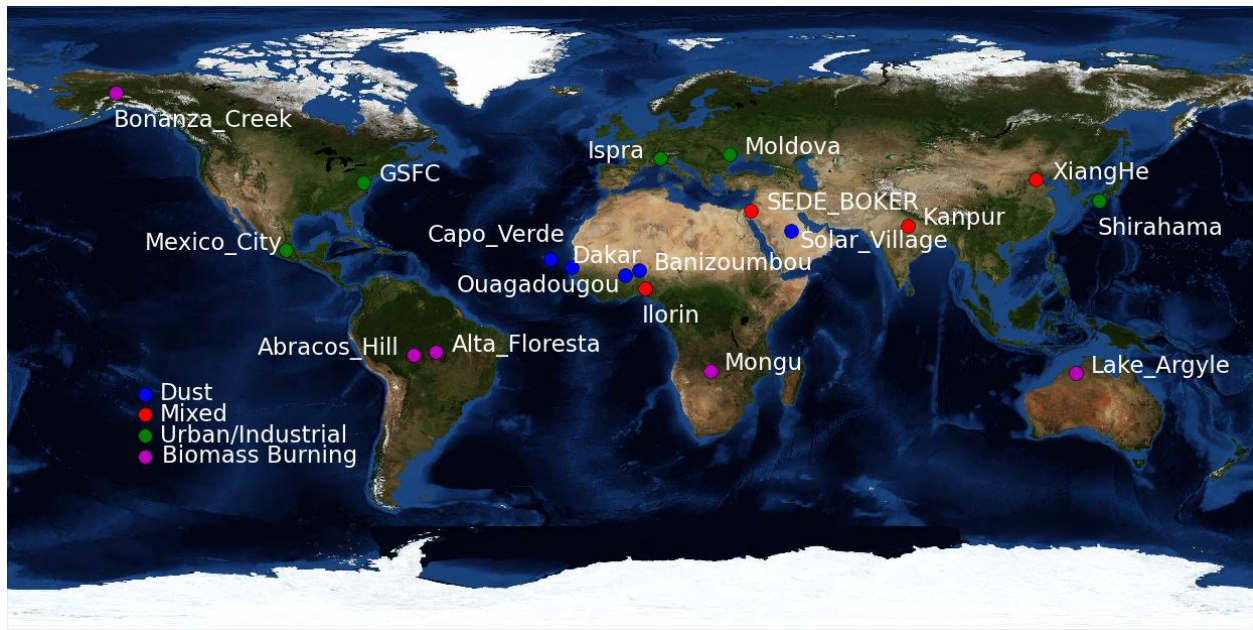
697 <sup>a</sup> indicates the unperturbed  $\alpha_{\text{abs}}$  average is recalculated based on available  $\omega_o$ .

698 <sup>b</sup> indicates wavelength(s) used in perturbation of  $\omega_o$ .

699 <sup>c</sup> indicates positive perturbation of  $\omega_o$  must be less than 0.995 for any wavelength.

700 <sup>d</sup> indicates these criteria are the current uncertainty estimates based on *Dubovik et al.* [2000].

701

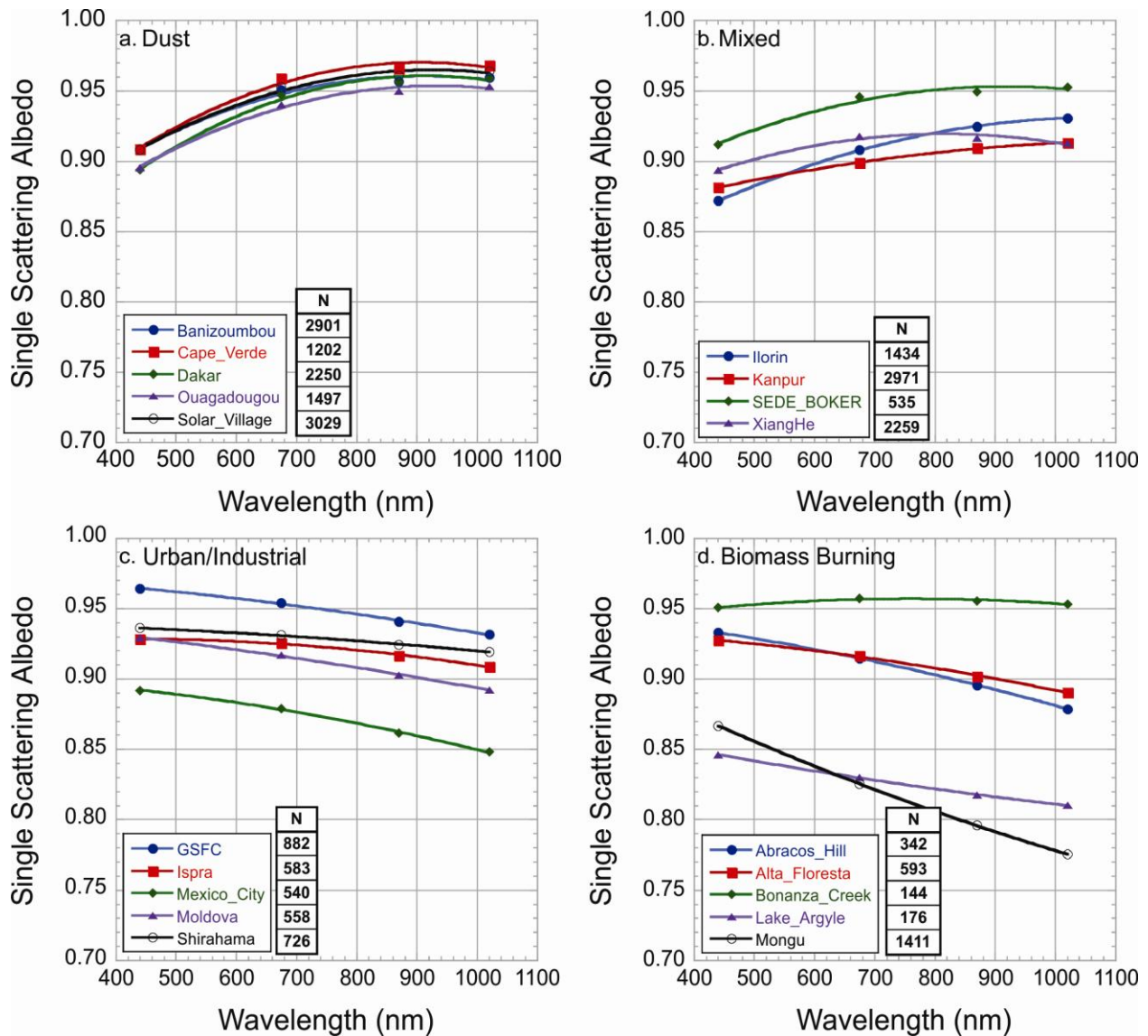


702

703 **Figure 1.** Distribution of the AERONET sites based on the dominant particle type. Sites were  
 704 selected based on data volume, geographic location, and primary aerosol source region. Other  
 705 dominant particle types (e.g., sea salt and biogenic aerosols) were not considered due to low  
 706 aerosol loading conditions ( $\tau_{440\text{nm}} \leq 0.4$ ), which was a limiting threshold for AERONET Version  
 707 2, Level 2.0 aerosol absorption retrievals [Dubovik *et al.*, 2002; Holben *et al.*, 2006].

708

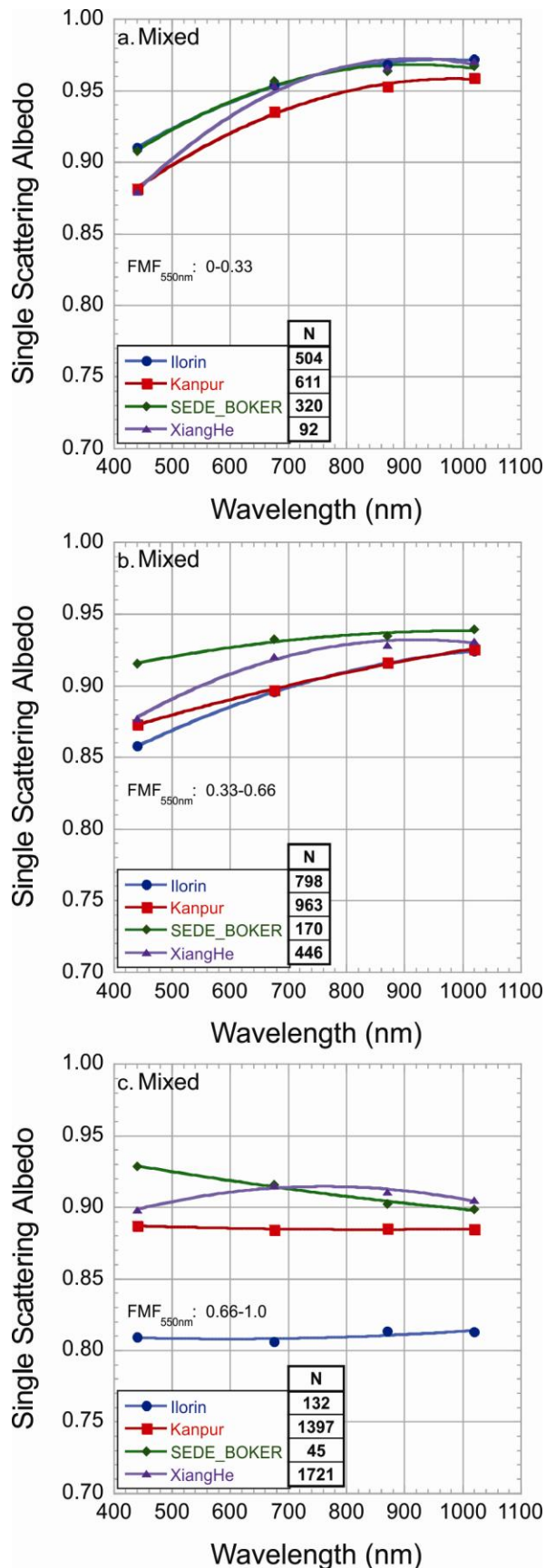




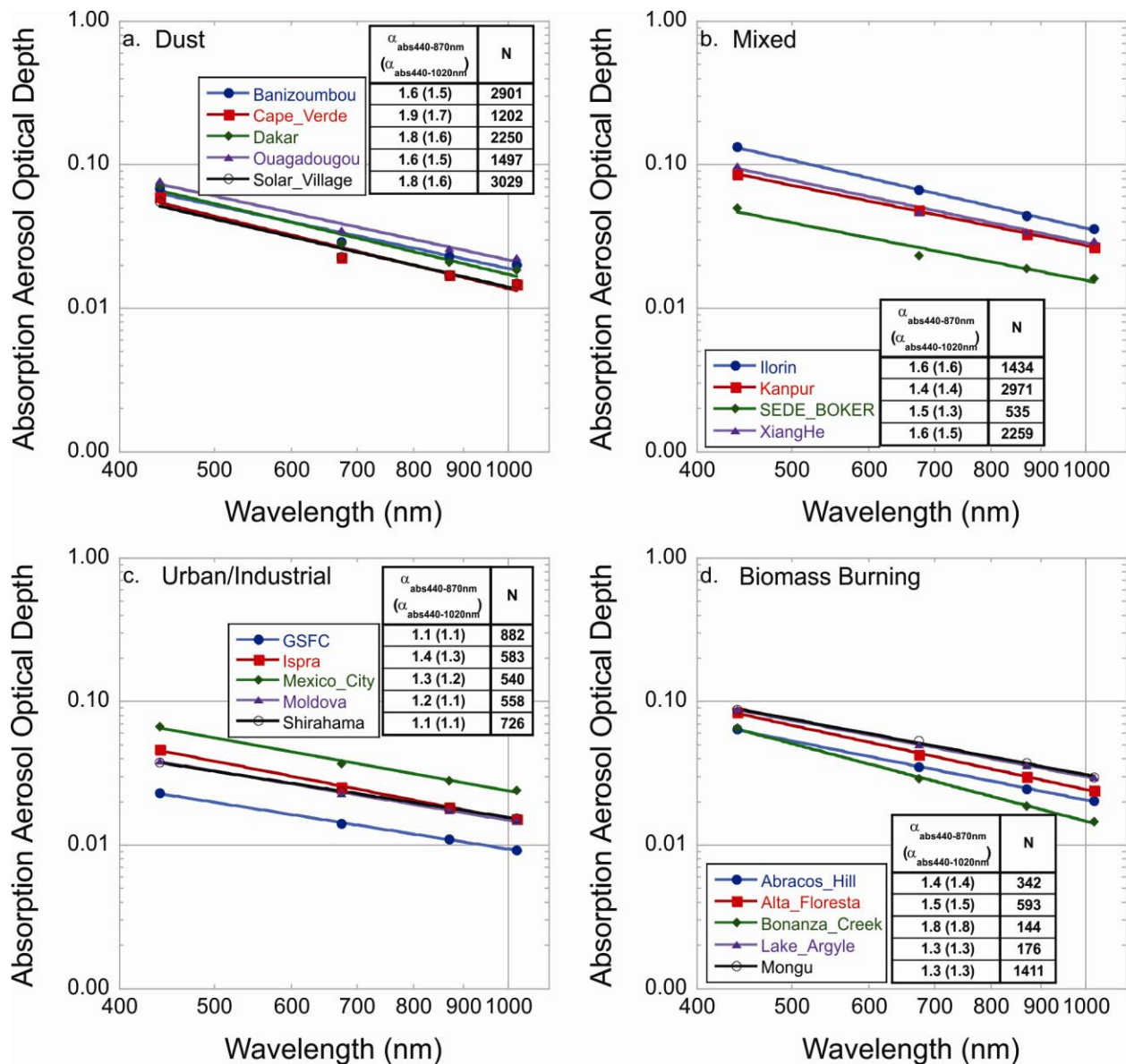
709

710 **Figure 2.** Spectral single scattering albedo averages were grouped by dominant aerosol particle  
 711 category for  $\tau_{440\text{nm}} > 0.4$  using AERONET Version 2, Level 2.0 data. The plots utilize second  
 712 order polynomial fit.

713



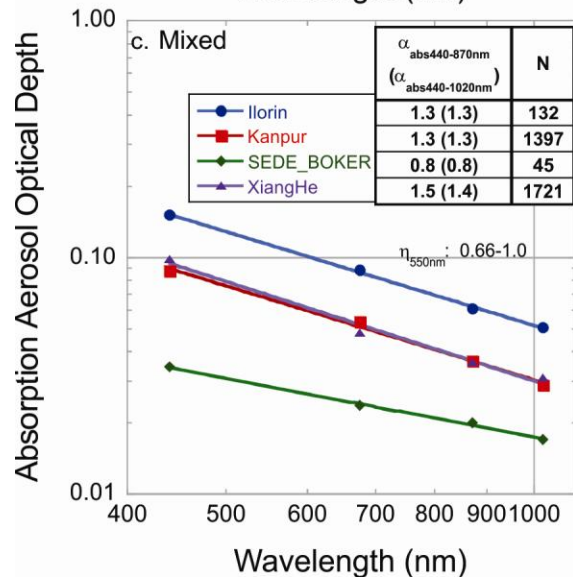
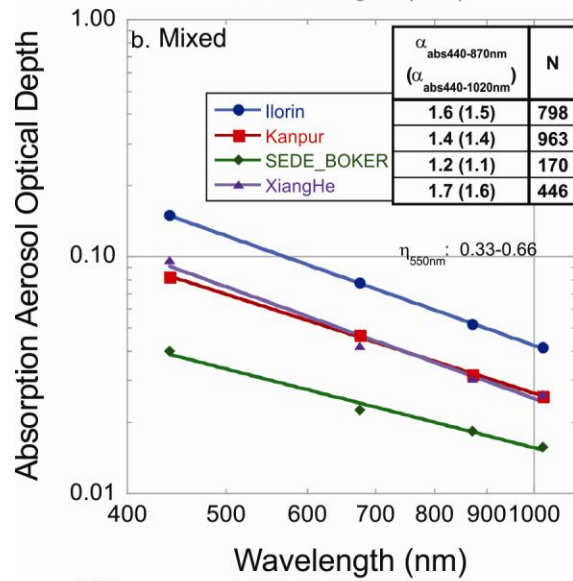
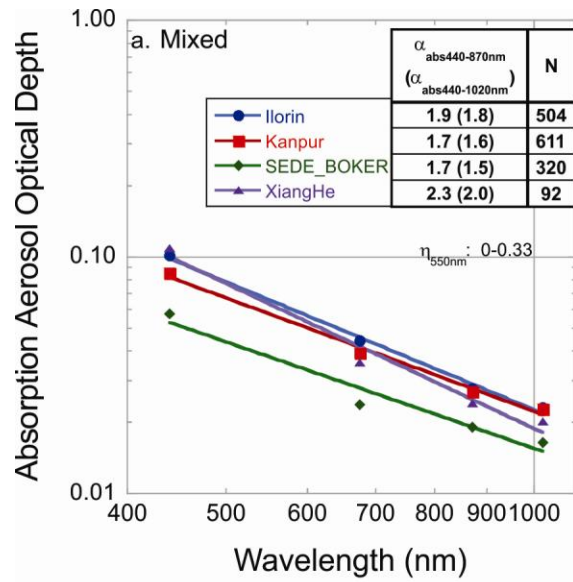
**Figure 3.** Similar to Figure 2, except the spectral single scattering albedo averages for the Mixed category were grouped by fine mode fraction of AOD ( $\eta_{550\text{nm}}$ ) using the ranges 0.0-0.33 for coarse mode dominated particles (a), 0.33-0.66 for mixed size particles (b), and 0.66-1.0 for fine mode dominated particles (c).



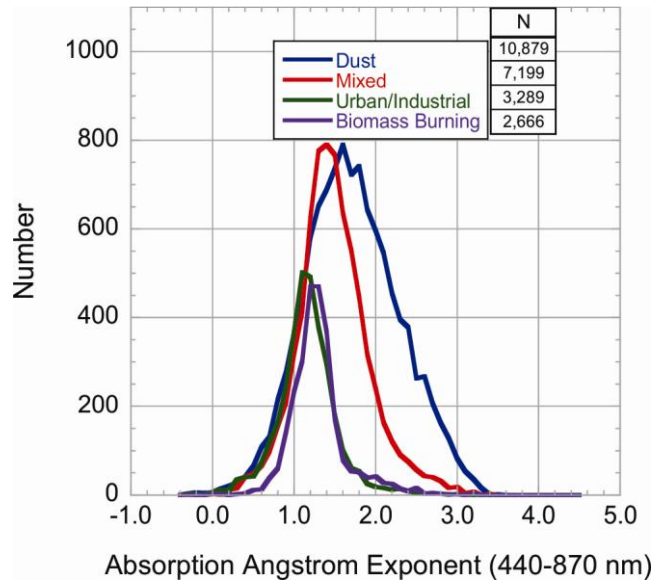
723

724 **Figure 4.** Absorption aerosol optical depth ( $\tau_{\text{abs}}$ ) and absorption Ångström exponent ( $\alpha_{\text{abs}}$ )  
 725 averages were grouped by dominant aerosol particle category for  $\tau_{440\text{nm}} > 0.4$  using AERONET  
 726 Version 2, Level 2.0 data. The plots use the power law fit and slopes of these lines are the  $\alpha_{\text{abs}}$   
 727 (440-870 nm or 440-1020 nm) listed adjacent to the legend in each plot.

728



**Figure 5.** Similar to Figure 3, except  $\tau_{\text{abs}}$  and  $\alpha_{\text{abs}}$  averages for the Mixed category were grouped by fine mode fraction of the AOD ( $\eta_{550\text{nm}}$ ) using ranges of 0.0-0.33 for coarse mode dominated particles (a), 0.33-0.66 for mixed size particles (b), and 0.66-1.0 for fine mode particles (c).

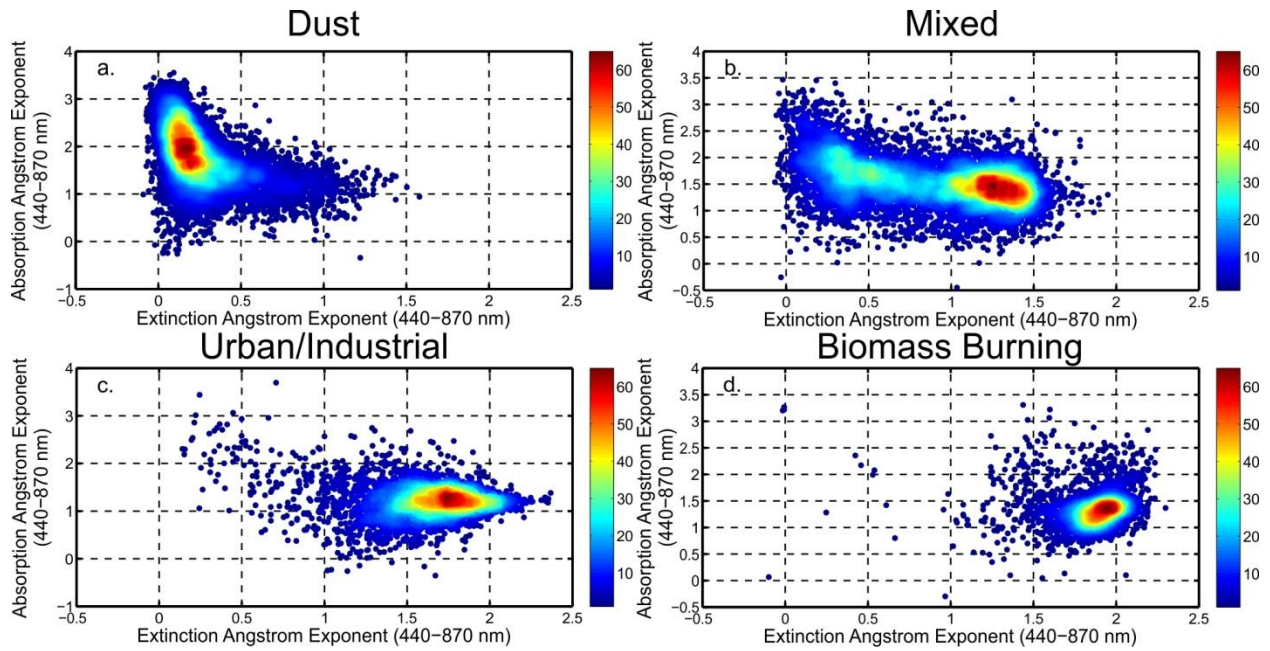


737

738 **Figure 6.** Absorption Ångström exponent ( $\alpha_{\text{abs}}$ ) frequency distribution for each dominant  
 739 aerosol particle category using AERONET Version 2, Level 2.0 data. Approximately 10% of the  
 740  $\alpha_{\text{abs}}$  retrievals (22% for Urban/Industrial) were below 1.0 or  $\lambda^{-1}$  dependence.

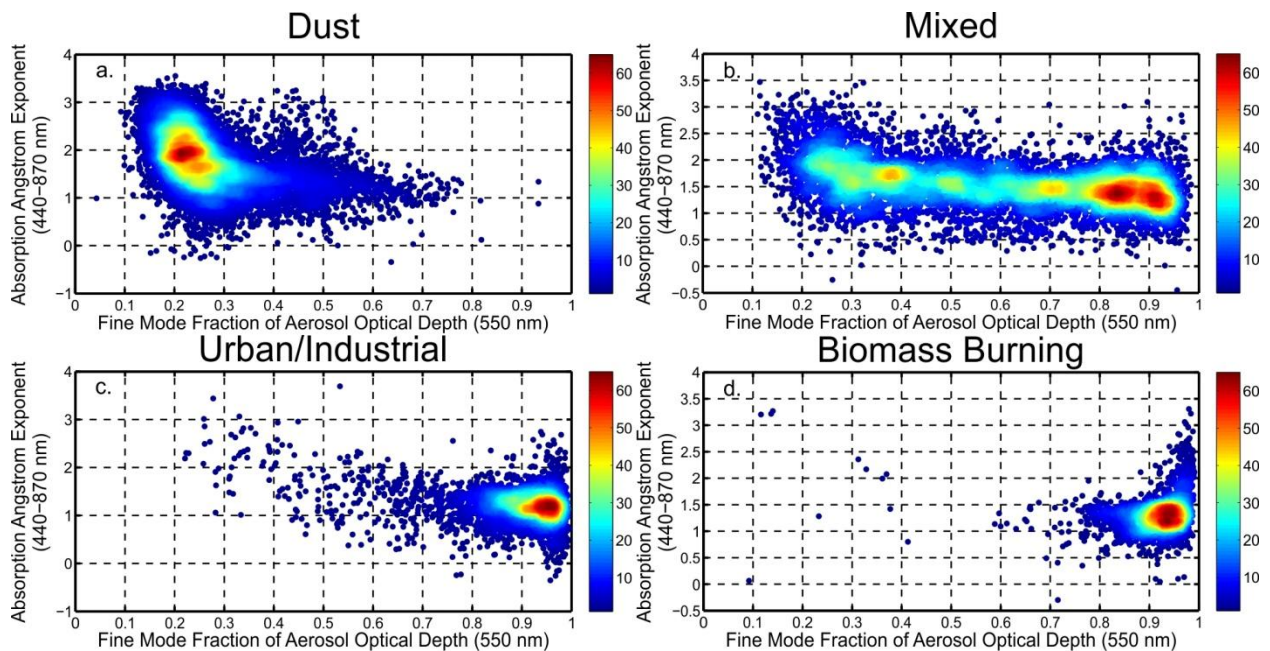
741





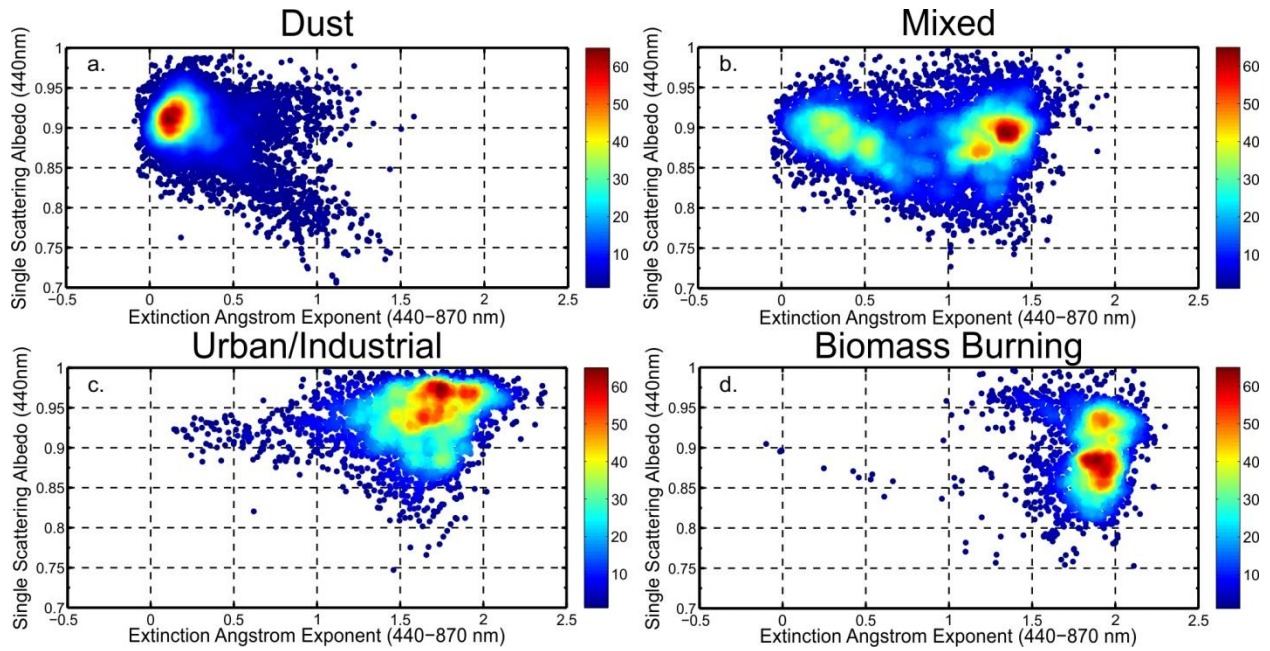
742

743 **Figure 7.** Relative number density plots for the absorption Ångström exponent (440-870 nm)  
 744 and extinction Ångström exponent (440-870 nm) relationship based on dominant aerosol type  
 745 using AERONET Version 2, Level 2.0 data. The color scale represents the relative density of  
 746 points in each aerosol type partitioned data set, where orange to red colors (levels ~45-64)  
 747 indicate the highest number density based on the Voronoi tessellation.



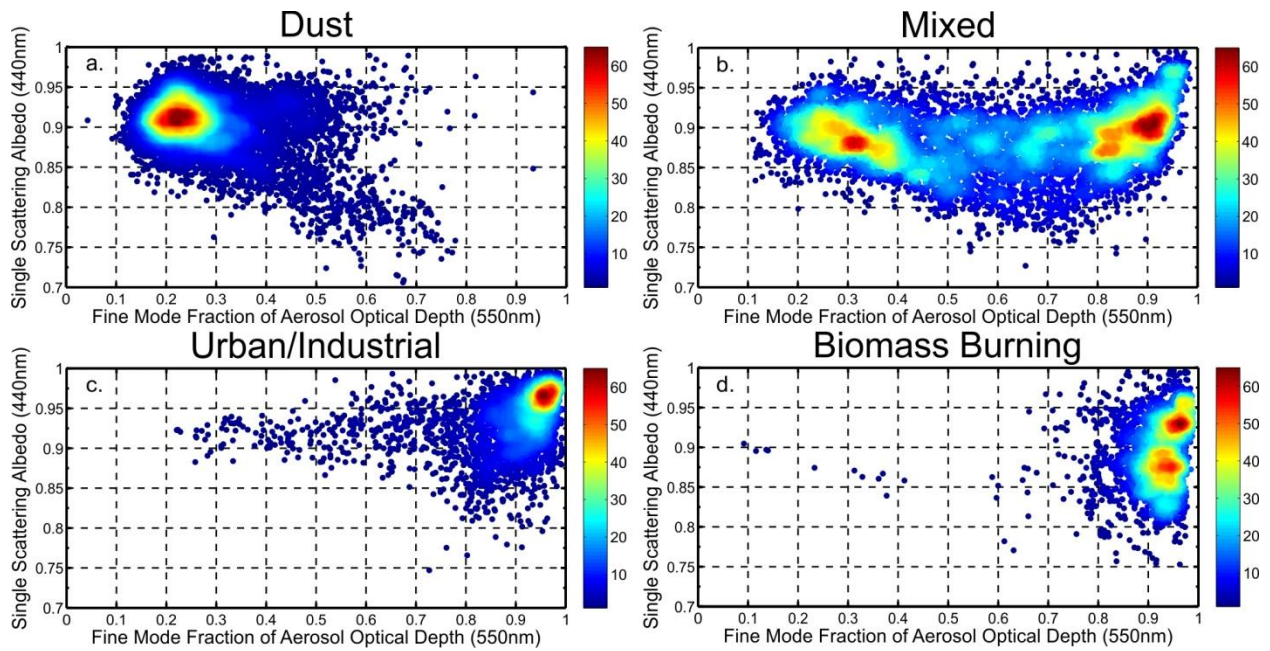
748

749 **Figure 8.** Similar to Figure 7, except for the absorption Ångström exponent (440-870 nm) and  
 750 fine mode fraction of the aerosol optical depth (550 nm) relationship.



751

752 **Figure 9.** Similar to Figure 7, except for the single scattering albedo (440 nm) and the extinction  
 753 Ångström exponent (440-870 nm) relationship.

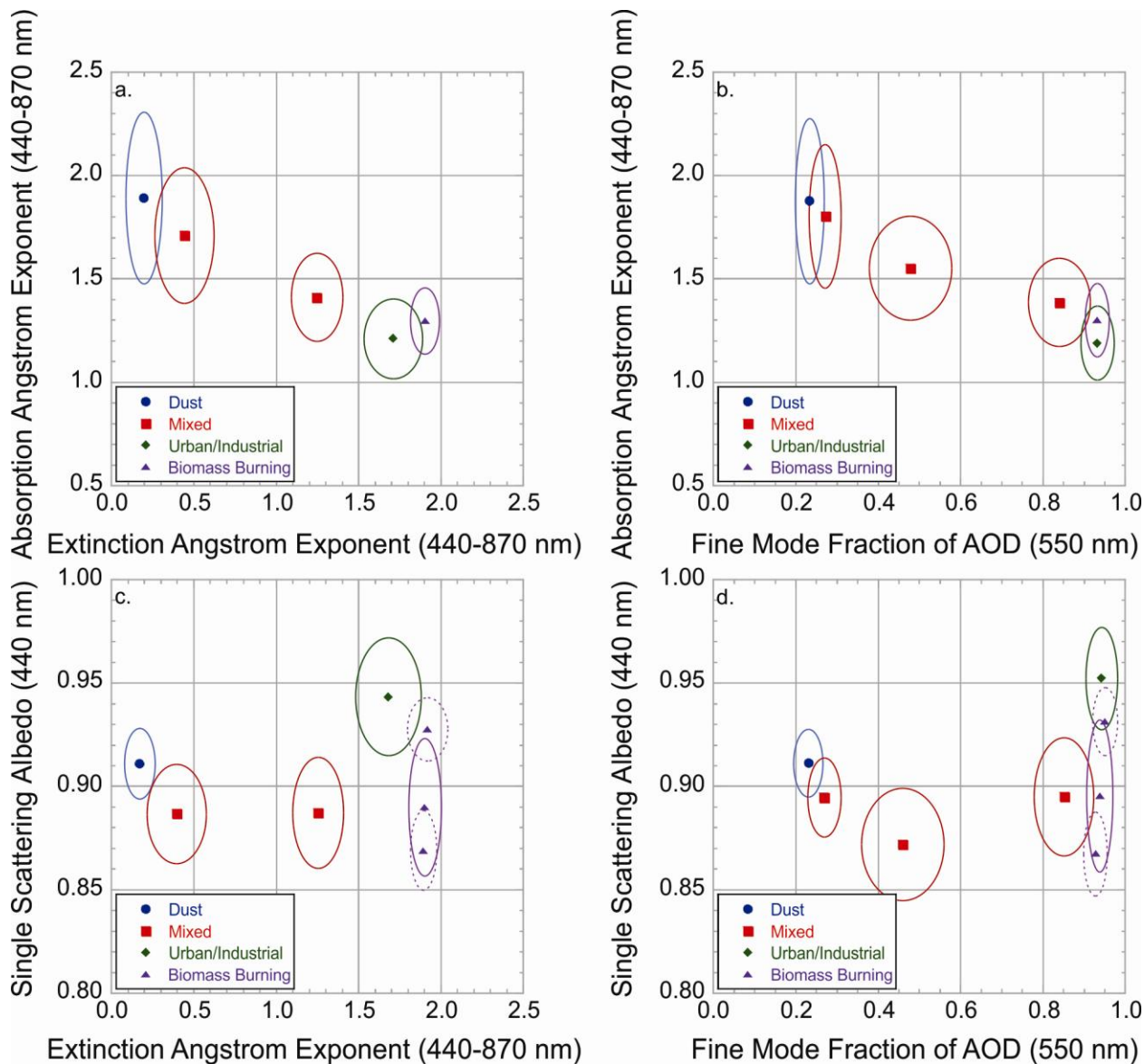


754

755 **Figure 10.** Similar to Figure 7, except for the single scattering albedo (440 nm) and fine mode  
 756 fraction of the aerosol optical depth (550 nm) relationship.

757





758

759 **Figure 11.** Weighted cluster averages were grouped for each aerosol type category and  
 760 relationship using AERONET Version 2, Level 2.0 data. The Mixed category averages were  
 761 calculated using a 0.8 extinction Ångström exponent threshold between mainly small and mainly  
 762 large particles. For the fine mode fraction of AOD, the Mixed category averages were calculated  
 763 based on the 0.0-0.33, 0.33-0.66, and 0.66-1.0 ranges. For single scattering albedo plots, the  
 764 Biomass Burning category was further partitioned by calculating averages using a single  
 765 scattering albedo threshold of 0.90 to produce two sub-clusters (dashed ellipses) observed in  
 766 Figure 9 and Figure 10.