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*Supplement of*

## **The diverse chemical mixing state of aerosol particles in the southeastern United States**

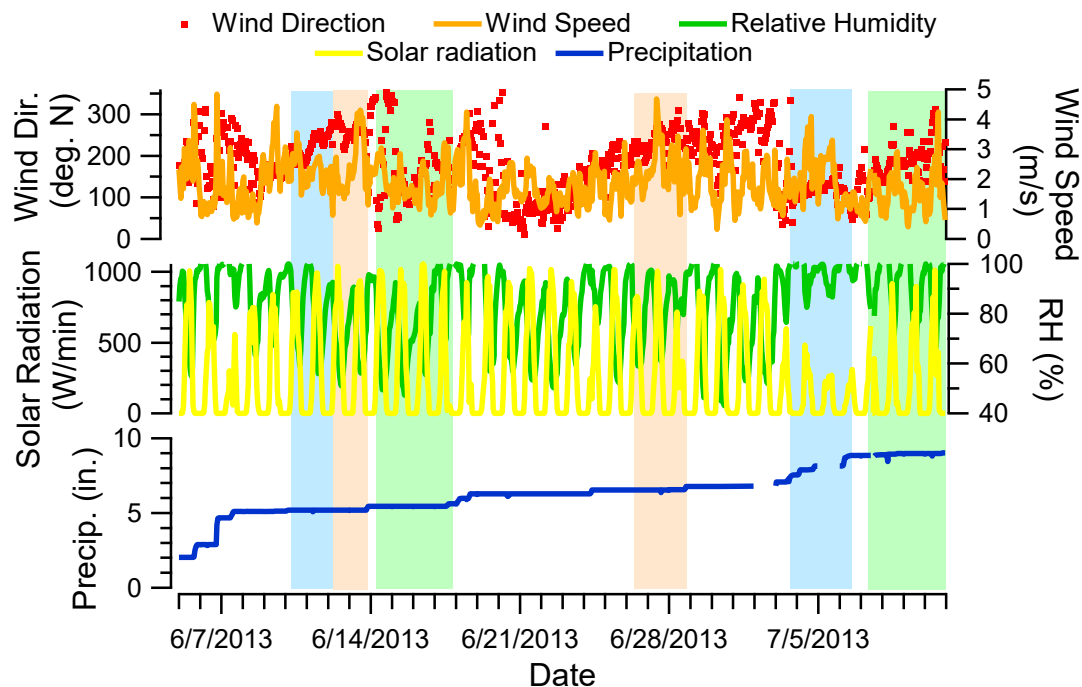
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15 **1 SEARCH network PM<sub>2.5</sub> and meteorological measurements**

16 The SouthEastern Aerosol Research and Characterization Network (SEARCH) is a multi-pollutant  
17 network designed to address regulatory and scientific questions related to ozone and its precursors,  
18 particulate matter mass and composition, and atmospheric visibility in addition to other research  
19 concerns. Active since 1992, Centreville, AL is one of a handful of sites that is part of SEARCH.  
20 During SOAS, meteorological conditions including wind direction, wind speed, solar radiation,  
21 precipitation, and relative humidity were monitored from the SEARCH network, plotted in Figure  
22 S1. Additionally, the network also collected and determined mass concentrations of EC, NH<sub>4</sub><sup>+</sup>,  
23 NO<sub>3</sub><sup>-</sup>, OC, SO<sub>4</sub><sup>2-</sup>, and PM<sub>2.5</sub> (using a tapered element oscillating microbalance, TEOM) shown in  
24 Figure 1. The box green boxes overlaid on the meteorological data in Figure S1 identify the two  
25 SOA-rich time periods. Similarly, tan boxes indicate the dust-rich time periods and blue boxes the  
26 SSA-rich time periods which were analyzed in this study.



27  
28 **Figure S1.** SEARCH meteorological data for Centreville, AL during SOAS with green boxes overlaid for  
29 the two SOA-rich time periods, tan boxes for the dust-rich time periods, and blue boxes overlaid for the  
30 SSA-rich time periods.

31

32 **2 CCSEM-EDX analysis**

33 SOAS intensive time periods, selected based on meteorological conditions, had shorter MOUDI  
34 collection times (3 hours rather than 11 hours.) The intensive dates are shown in Table S1 below,  
35 with the sample times highlighted in blue showing periods where CCSEM-EDX was run.

36 **Table S1.** Intensive sample collection times. Highlighted samples were analyzed using CCSEM-EDX.

<b>Intensive Date</b>	<b>Time (CST)</b>
6/10/13	8:00-11:00
6/10/13	12:00-15:00
6/10/13	16:00-19:00
6/10/13	20:00-7:00
6/11/13	8:00-11:00
6/11/13	12:00-15:00
6/11/13	16:00-19:00
6/11/13	20:00-7:00
6/12/13	8:00-11:00
6/12/13	12:00-15:00
6/12/13	16:00-19:00
6/12/13	20:00-7:00
6/14/13	8:00-11:00
6/14/13	12:00-15:00
6/14/13	16:00-19:00
6/14/13	20:00-7:00
6/15/13	8:00-11:00
6/15/13	12:00-15:00
6/15/13	16:00-19:00
6/15/13	20:00-7:00
6/16/13	8:00-11:00
6/16/13	12:00-15:00
6/16/13	16:00-19:00
6/16/13	20:00-7:00
6/29/13	8:00-11:00
6/29/13	12:00-15:00
6/29/13	16:00-19:00
6/29/13	20:00-7:00
6/30/13	8:00-11:00
6/30/13	12:00-15:00
6/30/13	16:00-19:00
6/30/13	20:00-7:00
7/1/13	8:00-11:00
7/1/13	12:00-15:00
7/1/13	16:00-19:00
7/1/13	20:00-7:00
7/9/13	8:00-11:00
7/9/13	12:00-15:00
7/9/13	16:00-19:00
7/9/13	20:00-7:00

37 All sampling periods that were analyzed using CCSEM-EDX are indicated in Table S2 below. The  
38 MOUDI stage(s) that were analyzed, in addition to the number of particles per sample, are also  
39 indicated. The aerodynamic diameter 50% cut points, detailed in Marple et al (1991), are as  
40 follows: stage 5 (1.8  $\mu\text{m}$ ), stage 6 (1.00  $\mu\text{m}$ ), stage 7 (0.56  $\mu\text{m}$ ), stage 8 (0.32  $\mu\text{m}$ ), stage 9 (0.18  
41  $\mu\text{m}$ ), stage 10 (0.100  $\mu\text{m}$ ), and stage 11 (0.056  $\mu\text{m}$ ). Not every sample collected was analyzed due  
42 to time/funding constraints and damaged substrates.

43

44 **Table S2.** Sampling times of all CCSEM-analyzed MOUDI samples and the number of particles analyzed  
 45 per stage.

Sample Date	Time (CST)	Stage(s)	# of particles
6/5/13	10:00-19:00	7	145
6/6/13	20:00-7:00	6 / 7	23 / 43
6/7/13	8:00-19:00	7	305
6/7/13	20:00-7:00	6	222
6/8/13	8:00-19:00	6	167
6/10/13	8:00-11:00	6 / 8	395 / 580
6/10/13	12:00-15:00	6 / 8	518 / 497
6/10/13	16:00-19:00	8	281
6/10/13	20:00-7:00	7	567
6/11/13	8:00-11:00	7	431
6/11/13	16:00-19:00	7	450
6/11/13	20:00-7:00	7	553
6/12/13	8:00-11:00	6 / 8	305 / 151
6/12/13	12:00-15:00	5 / 7 / 8	129 / 474 / 1314
6/12/13	16:00-19:00	6 / 7	365 / 220
6/12/13	20:00-7:00	7 / 9	581 / 2313
6/13/13	8:00-19:00	5 / 8 / 10	462 / 653 / 688
6/13/13	20:00-7:00	7	122
6/14/13	8:00-11:00	6 / 7	101 / 355
6/14/13	12:00-15:00	6 / 7 / 8	22 / 343 / 402
6/14/13	16:00-19:00	8	512
6/14/13	20:00-7:00	6	100
6/15/13	8:00-11:00	7 / 8	384 / 380
6/15/13	20:00-7:00	6 / 7 / 8	84 / 532 / 2304
6/16/13	8:00-11:00	7	239
6/16/13	16:00-19:00	7	338
6/16/13	20:00-7:00	6 / 8	514 / 791
6/17/13	8:00-19:00	7	2707
6/20/13	8:00-19:00	6 / 7	134 / 938
6/26/13	20:00-7:00	6 / 7	295 / 539
6/28/13	20:00-7:00	7	95
7/1/13	12:00-15:00	7	392
7/3/13	20:00-7:00	7	711
7/4/13	8:00-19:00	7	1826
7/5/13	8:00-19:00	7	448
7/6/13	8:00-19:00	6 / 10 / 11	369 / 64 / 342
7/7/13	8:00-19:00	7 / 10	209 / 690
7/7/13	20:00-7:00	9	153
7/8/13	8:00-19:00	6 / 9 / 11	137 / 755 / 1246
7/8/13	20:00-7:00	9	260
7/9/13	16:00-19:00	5 / 6 / 10 / 11	527 / 446 / 846 / 879
7/11/13	8:00-19:00	9	1262

46

47

### 48 3 Calculation of particle volume equivalent diameters

49 To describe the impact of size on aerosol chemical diversity during SOAS, projected area  
50 diameters ( $D_{pa}$ ) measured using CCSEM-EDX were converted to volume equivalent diameters  
51 ( $D_{ve}$ ) using a conversion factor determined from atomic force microscopy (AFM) height images  
52 of organic particles from SOAS collected on silicon substrates (Ted Pella Inc.). As particles can  
53 undergo spreading upon impaction on substrates,  $D_{ve}$  represents the size of particles before  
54 impaction. AFM images from organic aerosol particles collected during SOAS on June 14, 2013  
55 were used in the subsequent analysis to calculate a conversion factor between  $D_{pa}$  and  $D_{ve}$  for SOA  
56 and biomass burning particles. As SSA during SOAS was predominately aged by  $\text{HNO}_3$  leading  
57 to  $\text{NaNO}_3$  in the particle phase (Bondy et al., 2017b), a SSA spreading conversion factor was  
58 calculated using laboratory-generated  $\text{NaNO}_3$  (Bondy et al., 2017a). Though organic aerosol  
59 particles and SSA are expected to spread upon impaction as they are generally liquids at the  
60 temperatures and relative humidities presented, mineral dust, fly ash and primary biological  
61 particles are not expected to spread as they are solid. Thus,  $D_{pa}$  is equivalent to  $D_{ve}$  for mineral  
62 dust, fly ash and biological particles.

63 AFM was performed on a nanoIR2 system (Anasys Instruments). AFM height/deflection  
64 images were collected in contact mode (IR power 21.27%, filter in) at a scan rate of 1 Hz using a  
65 gold-coated contact mode silicon nitride probe (Anasys Instruments,  $13 \pm 4$  kHz resonant  
66 frequency, 0.07-0.4 N/m spring constant). Volumes of particles were measured using SPIP  
67 software (v6.2.6, Image Metrology, Hørsholm, Denmark), and from these volumes,  $D_{ve}$  was  
68 calculated for each particle. Table S3 shows the measured height and diameter, calculated volume,  
69 and calculated  $D_{ve}$  for select organic particles from SOAS (~100 particles were actually used to  
70 calculate the conversion factor). From these results, SOA and biomass burning particles were  
71 multiplied by a conversion factor of 0.49 to convert  $D_{pa}$  to  $D_{ve}$ .

72  
73



82 **Table S4.** AFM-measured and volume-calculated diameters of NaNO<sub>3</sub> particles impacted on silicon  
 83 substrates, representative of SSA.

Particle #	$D_{pa}$ (nm)	Height (nm)	Volume (nm <sup>3</sup> )	$D_{ve}$ (nm)
1	448	84	8981224	258
2	68	17	102174	58
3	171	29	821752	116
4	136	19	435493	94
5	100	13	205804	73
6	105	12	228540	76
7	250	58	2467066	168
8	258	77	2776954	174
9	153	29	648760	107
10	284	72	3454101	188
11	251	51	2288227	164
12	217	24	1243357	133
13	265	57	2718737	173
14	89	15	169364	69
15	320	67	4208101	200
16	745	108	24432611	360
17	273	55	2854158	176
18	217	40	1542059	143
19	458	79	8608666	254
20	199	33	1133683	129
21	319	69	4500131	205
22	383	64	5131053	214
23	187	9	676804	109
24	250	50	2184633	161

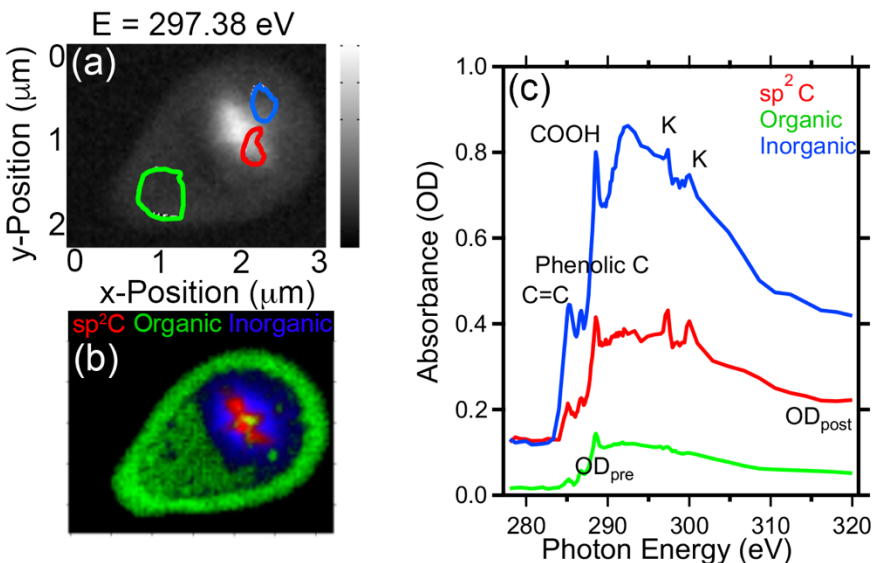
84

#### 85 **4 STXM-NEXAFS of a biological particle**

86 STXM-NEXAFS was used to investigate the electronic properties and bonding nature of chemical  
 87 species within carbon-containing particles since these particles were so prevalent at SOAS. The  
 88 carbon K-edge was probed using this technique, and high spatial resolution information was  
 89 obtained regarding carbonaceous particles. As SOA will be examined in detail using STXM-  
 90 NEXAFS, only a primary biological particle is shown in Figure S2. From the NEXAFS spectrum  
 91 (c), this particle contains C=C (285 eV), phenolic C (286.7 eV), and COOH (288.5 eV) functional  
 92 groups (Moffet et al., 2010b;Moffet et al., 2010a;Solomon et al., 2005). The inorganic component  
 93 was identified based on the absorbance of the pre-edge from 278 to 283 eV, which is offset  
 94 compared to the post-edge region at 320 eV (Moffet et al., 2010a). The NEXAFS spectrum  
 95 contains two sharp peaks at 297.1 and 299.7 eV characteristic of potassium. These features, along



96 with the oblong morphology, were used to identify the particle as biological (Moffet et al., 2010b).  
97 Although this particle also contains an  $sp^2$  C inclusion, rather than soot the  $sp^2$  C in the center of  
98 the particle is likely due to chlorophyll, a chelating porphyrin present within plant cells (Legall et  
99 al., 2007; Mauzerall, 1977).



100

101 **Figure S2.** STXM-NEXAFS data of a primary biological particle. STXM images of the particles are shown  
102 in (a). Singular value decomposition maps combining  $sp^2$  carbon (red), organic (green), and inorganic (blue)  
103 are depicted in (b). The NEXAFS spectrum for the particle at the carbon K-edge is shown in (c). Inorganic  
104 material was identified in both particles according to the pre-edge/post-edge offset.

## 105 5 Fresh soot calculation

106 Soot was difficult to detect using CCSEM-EDX due to interference from the Formvar B coating  
107 on the TEM grid. Therefore, the size distribution for soot was manually calculated from SEM  
108 images using samples on various stages from one day, and then a correction factor was applied to  
109 each sampling period based on the SEARCH network mass concentrations of elemental carbon  
110 (EC). This method likely overestimates the contribution of fresh soot since organic  
111 carbon/elemental carbon (OC/EC) SEARCH measurements include both fresh and aged soot,  
112 however the EC mass was used as an approximation of soot's contribution. To calculate the size  
113 distribution of soot particles during SOAS, all SEM images from July 9, 2013 4pm-7pm CST were

114 inspected for soot agglomerates. This sample was chosen for analysis because stages 7-11 of the  
 115 MOUDI (0.056-0.56  $\mu\text{m}$  50% size cut; relevant sizes for soot) were available for imaging. Once a  
 116 soot particle was identified, the particle was traced in ImageJ software to calculate the area. From  
 117 this area, similar to the CCSEM software output, the projected area diameter ( $D_{pa}$ ) was calculated  
 118 and a size distribution using all soot  $D_{pa}$  was generated, shown below.

119 **Table S5.** Size distribution for fresh soot calculated for July 9, 2013 stages 7-11.

<i>Projected Area Diameter (<math>\mu\text{m}</math>)</i>	<i>Frequency</i>	<i>Fraction of soot per bin</i>
0.133352	0	0
0.177828	0	0
0.237137	5	0.065789474
0.316228	14	0.184210526
0.421697	27	0.355263158
0.562341	18	0.236842105
0.749894	11	0.144736842
1	0	0
1.333521	0	0
1.778279	1	0.013157895
2.371374	0	0
3.162278	0	0
4.216965	0	0
5.623413	0	0
7.498942	0	0
10	0	0
<b>Total</b>	76	

120  
 121 The fraction of soot per size bin was then used with the SEARCH network EC mass  
 122 concentrations, measured using oxidative combustion, to calculate an approximate number of soot  
 123 particles within each size range. First, the average EC mass was calculated for each time period of  
 124 interest during SOAS. Then, the average  $\text{PM}_{2.5}$  mass, measured using a tapered element oscillating  
 125 microbalance (TEOM), was calculated for each time period. A scaling factor was generated by  
 126 dividing EC mass/TEOM mass. The scaling factor calculated for each time period is: SOA (0.022),  
 127 mineral dust (0.018), and SSA (0.029). This scaling factor was then multiplied by the size  
 128 distributions in Table S5, giving results for the fraction of soot within the designated size bins for  
 129 each of the three time periods.

130 **6 Mass calculations and mixing state parameters**

131 To calculate the mixing state parameters, atomic percentages were converted to mass fractions as  
 132 described in the experimental section. To calculate elemental diversity, the mass of each element  
 133 was used in the mixing state calculations shown below. To calculate mixing state due to aging  
 134 during the SOA-rich, dust-rich, and SSA-rich time periods, elemental masses were assigned to  
 135 specific source-based particle classes with the compositions described below. The elemental mass  
 136 fractions for each of the three time periods are shown in Figure S3 depicting each element's  
 137 contribution as a function of size. For the source-based elemental assignments, SOA particles  
 138 consisted of solely S, biomass burning particles contained K and Cl, fly ash particles consisted of  
 139 Si and Al, dust particles contained Na, Mg, Al, Si, K, Ca, Ti, and Fe, SSA particles contained Na,  
 140 Mg, Cl, K, and Ca, and biological particles consisted of P, Cl, and K. Secondary species in this  
 141 study were represented by S in every particle class, since organic (C, O) and nitrogen-containing  
 142 species (N) are not quantitative using SEM-EDX (Laskin et al., 2006).

143 In addition to mass calculations, mixing state parameters were calculated for each  
 144 elemental class to quantify diversity, and for the SOA, dust, and SSA time periods to quantify  
 145 aging. The experimental section describes the equations used to calculate the entropy and mixing  
 146 state index. Below are the definitions and equations for aerosol mass and mass fraction used to  
 147 calculate the entropy along with equations for particle, species, and bulk population diversity  
 148 (Riemer and West, 2013). The mass of species  $a$  in particle  $i$  is termed  $\mu^a_i$  where  $a = 1, \dots, A$  and  $i$   
 149  $= 1, \dots, N$ . The total mass of particle  $i$  ( $\mu_i$ ) is given by

$$150 \quad \mu_i = \sum_{a=1}^A \mu_i^a \quad (2)$$

151 The total mass of species  $a$  in the population ( $\mu^a$ ) is given by

$$152 \quad \mu^a = \sum_{i=1}^N \mu_i^a \quad (3)$$

153 The total mass of the population ( $\mu$ ) is given by

$$154 \quad \mu = \sum_{i=1}^N \mu_i \quad (4)$$

155 The mass fraction of species  $a$  in particle  $i$  ( $p^a_i$ ) is given by

$$156 \quad p^a_i = \frac{\mu_i^a}{\mu_i} \quad (5)$$

157 The mass fraction of particle  $i$  in the population ( $p_i$ ) is given by

$$158 \quad p_i = \frac{\mu_i}{\mu} \quad (6)$$

159 The mass fraction of species  $a$  in the population ( $p^a$ ) is given by

$$160 \quad p^a = \frac{\mu^a}{\mu} \quad (7)$$

161 The particle diversity of particle  $i$  ( $D_i$ ) is given by

$$162 \quad D_i = e^{H_i} \quad (8)$$

163 where  $H_i$  is the mixing entropy of particle  $i$ . The average particle species diversity ( $D_\alpha$ ) is given by

$$164 \quad D_\alpha = e^{H_\alpha} \quad (9)$$

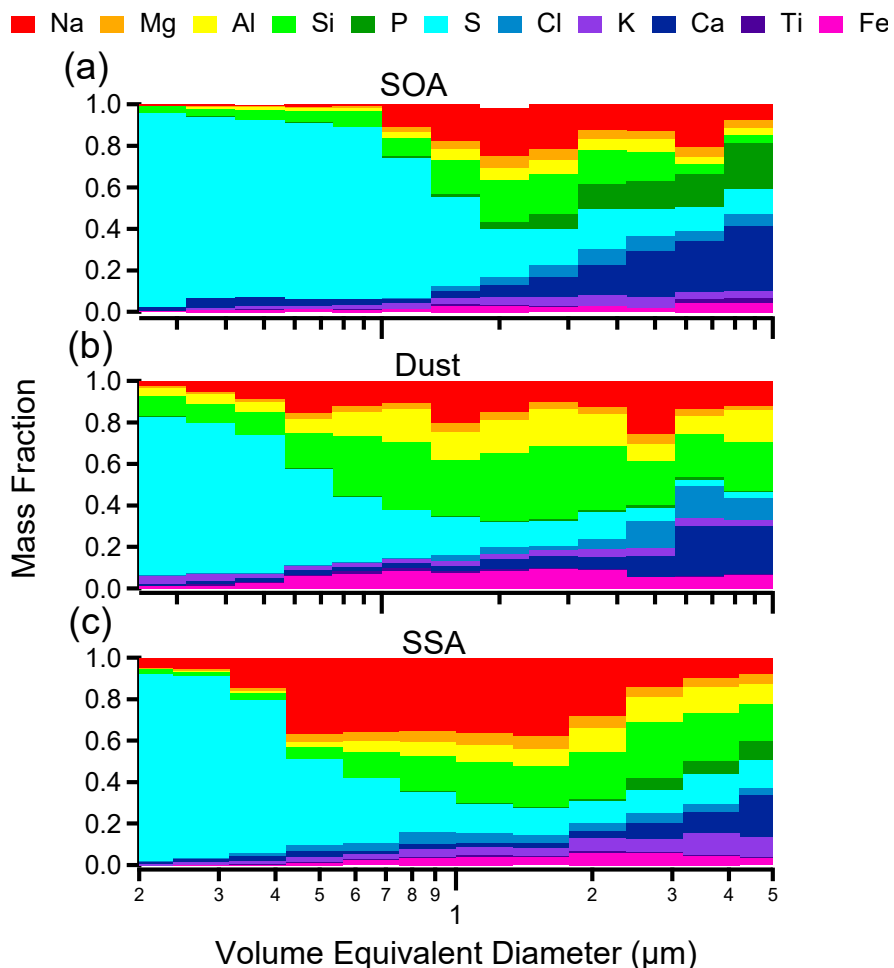
165 where  $H_\alpha$  is the average particle mixing entropy. The bulk population species diversity ( $D_\gamma$ ) is  
166 given by

$$167 \quad D_\gamma = e^{H_\gamma} \quad (10)$$

168 where  $H_\gamma$  is the population bulk mixing entropy.  $D_\alpha$  was used as a quantitative measure of  
169 elemental diversity for each particle class during SOAS (SOA, biomass burning particles, fly ash,  
170 dust, SSA, and biological particles.) However to quantify particle aging due to S during SOAS,  
171 the mixing state index ( $\chi$ ), a ratio between the average particle species diversity and bulk  
172 population species diversity, was calculated. While  $D_\alpha$  is a useful metric to quantify elemental  
173 diversity,  $\chi$  quantifies the degree of internal versus external mixing present within particle  
174 populations.

175

176



177  
 178 **Figure S3.** Mass fractions as a function of volume equivalent diameter for particle-rich time periods: (a)  
 179 SOA-rich periods (June 14-17 and July 7-11, 2013), (b) dust-rich periods (June 12-13 and June 26-28,  
 180 2013), and (c) SSA-rich periods (June 10-11 and July 3-6, 2013.) C and O are not included in element  
 181 quantification due to substrate interferences. \*Only particles with a diameter between 0.2 - 5 μm are shown  
 182 due to too few particles present at larger sizes for quantitative analysis.

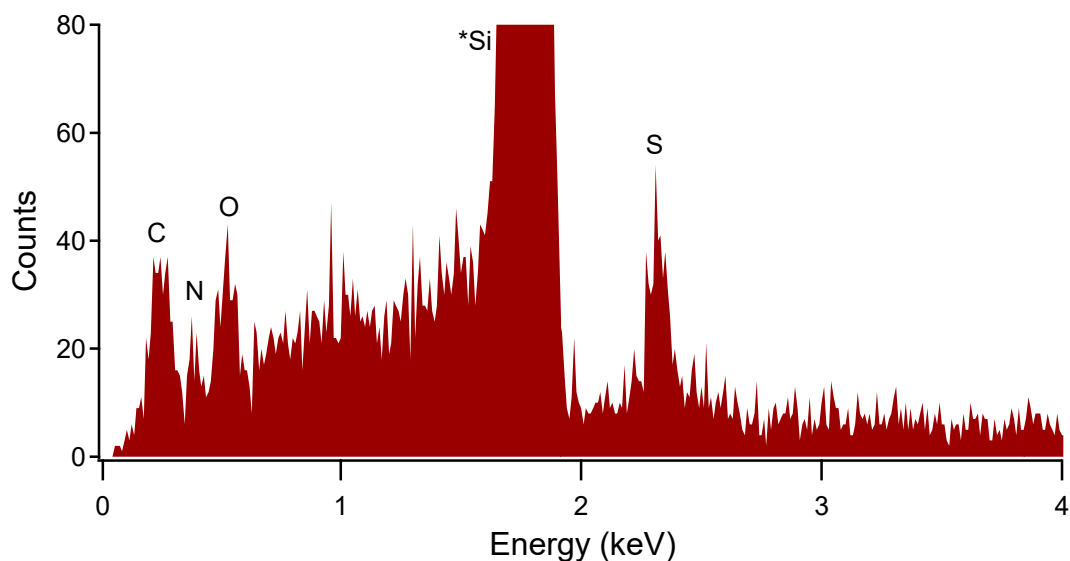
183 **7 STXM-NEXAFS soot identification**

184 Two samples, June 10 and July 7, 2013, were analyzed using STXM-NEXAFS. In order to  
 185 calculate the number fraction of particles from each sample that contained sp<sup>2</sup> C in the form of  
 186 soot, matlab was used to visualize every particle in the sample. Each particle was screened on a  
 187 per pixel basis for regions of high C=C content (> 35% C=C). If a pixel contained > 35 % C=C,  
 188 then the script rendered a red pixel, stating that it was safe to call that region soot. The same was  
 189 performed for the other colors (i.e. blue=inorganic, green=organic). Using this data, 6.9 % of

190 particles by number contained soot collected June 10, 2013 and 9.9 % of particles collected July  
191 7, 2013 contained soot.

## 192 **8 EDX of SOA on silicon**

193 Most of the CCSEM-EDX analysis in this study was conducted on Formvar-coated TEM grids.  
194 However, since Formvar (a polymer) interferes with particle carbon and oxygen X-ray signals,  
195 additional EDX spectra of SOA particles were collected on Si substrates. EDX spectra from 61  
196 particles (June 15, 2013 8pm-7am St. 8 sample) were collected and quantified with respect to C,  
197 N, O, and S. Figure S4 shows an example spectrum of an SOA/sulfate particle collected on Si.  
198 Note, the signal for Si extend beyond 80 counts, however the y-axis range shown here was selected  
199 to view the elements of interest (C, N, O, S). The average weight % of elements within SOA/sulfate  
200 from this analysis was 40 % C, 11 % N, 28 % O, and 20 % S. Though SOA/sulfate only contained  
201 20% sulfur from this analysis, the mixing state indices for aerosol populations were calculated  
202 based on SOA/sulfate containing only sulfur, since CCSEM-EDX is not quantitative for C, N, and  
203 O.



204  
205 **Figure S4.** Example EDX spectrum of an SOA/sulfate particle collected on a silicon substrate.

206

## 207 **9 Circularity equation**

208 To determine the average circularity for SOA, biomass burning aerosol, and fly ash classes, the  
209 mode circularity was averaged across the k-means clusters assigned to that class in Equation 1:

$$210 \quad C = 4\pi A/p^2 \quad (11)$$

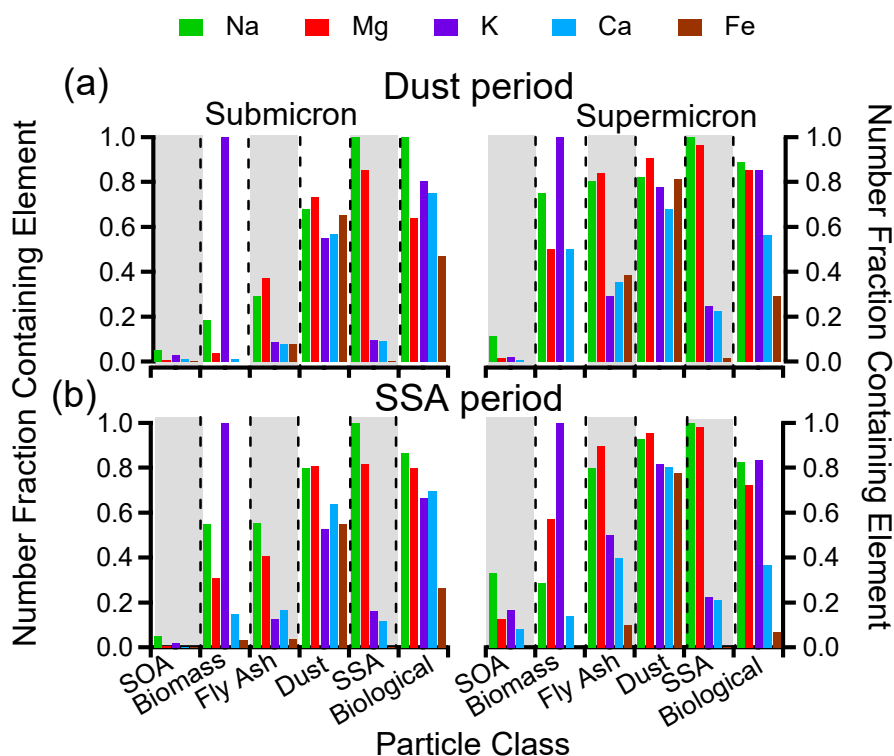
211 where  $C$  is circularity,  $A$  is area of the particle, and  $p$  is the particle perimeter.

212

## 213 **10 Nonvolatile cations**

214 The number fraction of particles containing nonvolatile cations (Na, Mg, K, Ca, Fe) in sub- and  
215 supermicron sizes is shown in Figure S5 for the dust and SSA periods. In general, the number  
216 fraction of metal-containing particles is consistent for each class across the different periods  
217 (although there are minor differences between the sub- and super micron size ranges), suggesting  
218 that nonvolatile cations don't vary with processing, but are inherent to each class.

219



221

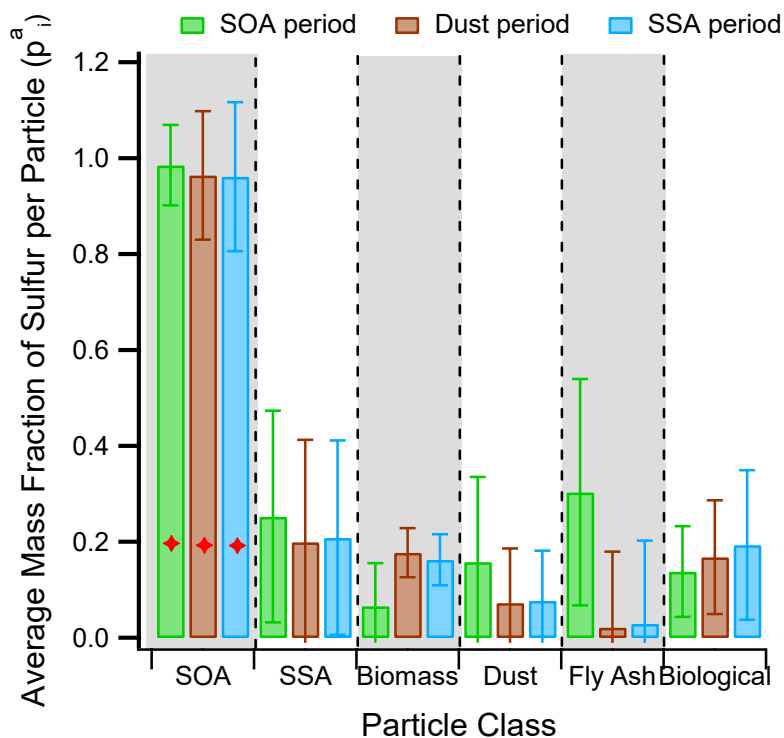
222 **Figure S5.** Size-resolved particle class compositions indicate the number fraction of particles in each  
 223 class containing non-volatile cations Na, Mg, K, Ca, and Fe during the (a) dust period and (b) SSA period  
 224 in the submicron and supermicron size range.

## 225 11 Significance of aging by sulfur during three events

226 The degree of secondary processing for each particle class was calculated as the average mass  
 227 fraction of sulfur per particle (Figure S6). This parameter was used to calculate average sulfur  
 228 diversity and along with the bulk population diversity, mixing state indices could be quantified.  
 229 However, the mass fraction calculations here exclude C, N, and O since low Z elements are only  
 230 semiquantitative with CCSEM. Excluding C, N, and O, SOA/sulfate have average sulfur mass  
 231 fractions of  $0.98 \pm 0.08$ ,  $0.96 \pm 0.13$ ,  $0.96 \pm 0.16$ , during the SOA-influenced, dust-influenced, and  
 232 SSA-influenced events, respectively. However, analysis of SOA on a non-carbonaceous substrate,  
 233 which allowed C, N, and O to be quantified, demonstrated that the average mass fraction of sulfur



234 in SOA was actually  $0.20 \pm 0.04$ . This “actual” mass value for SOA was used to scale the average  
 235 mass fraction of sulfur for each period and is portrayed in Figure S6 by red markers. Using this  
 236 scaled mass fraction of sulfur, the “actual” mass of sulfur was 0.197 during the SOA-influenced  
 237 events, 0.193 during the dust periods, and 0.192 during the SSA periods.



238  
 239 **Figure S6.** The secondary processing of particles by sulfate was calculated for each class during the three  
 240 time periods of interest as the average mass fraction of sulfur per particle. Red markers indicate the scaled  
 241 “actual” mass fraction of sulfur for SOA including mass contributions from C, N, and O.

242  
 243 A student’s t-test was used to compare the average mass fraction of sulfur per particle for six main  
 244 particle classes (SOA/sulfate, biomass burning aerosol, fly ash, dust, SSA, and biological) during  
 245 the SOA events, dust events, and SSA events. The standard deviation,  $S_{pooled}$ , was calculated using  
 246 the following equation,

247

$$S_{pooled} = \sqrt{\frac{s_1^2(n_1-1) + s_2^2(n_2-1)}{n_1+n_2-2}} \quad (12)$$

248 where  $s_1$  and  $s_2$  are the standard deviations from the two samples, and  $n_1$  and  $n_2$  are the number of  
 249 samples in each category. Then the student's t-test was calculated,

$$250 \quad t_{\text{calculated}} = \frac{x_1 - x_2}{S_{\text{pooled}}} \sqrt{\frac{n_1 n_2}{n_1 + n_2}} \quad (13)$$

251 where  $x_1$  and  $x_2$  are the mean mass fraction of sulfur per particle class. The results of the student t-  
 252 tests are located in Table S6, Table S7, and Table S8. The student's t-test was calculated to  
 253 compare the aging of particle classes during the SOA vs. dust events, SOA vs. SSA events, and  
 254 dust vs. SSA events. The difference in average sulfur mass fractions for all particle classes was  
 255 found to be significant for the SOA vs. dust events and SOA vs. SSA events, but the difference  
 256 was not statistically significant for the dust vs. SSA events at the 95% confidence interval.

257  
 258 **Table S6.** Student's t-test comparing average sulfur mass fractions among particle classes for the SOA vs.  
 259 dust events

	SOA	Biomass	Fly Ash	Dust	SSA	Biological
$x_1$	0.985	0.260	0.070	0.169	0.306	0.140
$x_2$	0.964	0.177	0.022	0.072	0.199	0.168
$s_1$	0.081	0.221	0.091	0.179	0.235	0.096
$s_2$	0.134	0.214	0.051	0.114	0.156	0.119
$S_{\text{pooled}}$	0.095	0.048	0.006	0.128	0.194	0.099
<i>degrees of freedom</i>	120	120	120	120	120	120
$t_{\text{calculated}}$	10.97	3.95	5.67	19.82	13.49	2.50
95% CI $t_{\text{table}}$	1.98	1.98	1.98	1.98	1.98	1.98
<i>significantly different?</i>	yes	yes	yes	yes	yes	yes

260  
 261 **Table S7.** Student's t-test comparing average sulfur mass fractions among particle classes for the SOA vs.  
 262 SSA events

	SOA	Biomass	Fly Ash	Dust	SSA	Biological
$x_1$	0.985	0.260	0.070	0.169	0.306	0.140
$x_2$	0.962	0.163	0.029	0.077	0.209	0.193
$s_1$	0.081	0.221	0.091	0.179	0.235	0.096
$s_2$	0.155	0.202	0.053	0.104	0.173	0.156
$S_{\text{pooled}}$	0.098	0.047	0.083	0.132	0.193	0.108
<i>degrees of freedom</i>	120	120	120	120	120	120
$t_{\text{calculated}}$	10.71	3.98	3.50	16.56	13.62	5.04
95% CI $t_{\text{table}}$	1.98	1.98	1.98	1.98	1.98	1.98
<i>significantly different?</i>	yes	yes	yes	yes	yes	yes

264 **Table S8.** Student's t-test comparing average sulfur mass fractions among particle classes for the dust vs.  
 265 SSA events

	<b>SOA</b>	<b>Biomass</b>	<b>Fly Ash</b>	<b>Dust</b>	<b>SSA</b>	<b>Biological</b>
$x_1$	0.985	0.177	0.022	0.072	0.199	0.168
$x_2$	0.964	0.163	0.029	0.077	0.209	0.193
$s_1$	0.081	0.214	0.051	0.114	0.158	0.119
$s_2$	0.134	0.202	0.053	0.104	0.173	0.156
$s_{pooled}$	0.144	0.209	0.052	0.111	0.168	0.142
<i>degrees of freedom</i>	120	120	120	120	120	120
$t_{calculated}$	0.568	0.57	0.94	1.63	1.78	1.29
<i>95% CI <math>t_{table}</math></i>	1.98	1.98	1.98	1.98	1.98	1.98
<i>significantly different?</i>	no	no	no	no	no	no

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