Supporting Information for

Brown Carbon Aerosols from Fresh Wildfire Smoke: Associations with Low-Volatility Organics and Chemical Compound Groups

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S1 TOA-based Volatility distribution estimation

The organic carbon (OC) volatility distribution was estimated using a temperature profile based on the NIOSH870 protocol[1]. In this protocol, a filter punch is placed within the instrument and the temperature is systematically increased to obtain different OC bins. Heating is performed in helium where each sample is first heated to 310°C; OC emitted at this temperature is categorized as OC1. Subsequently, the temperature is ramped up to 475°C, 615°C, and 870°C, to obtained OC2, OC3, and OC4, respectively. A correction is performed to account for any OC charring that might have occurred during the heating process. Once the OC has been quantified, oxygen is added along with helium to the system to yield the elemental carbon (EC) remaining on the filter.

The maximum saturation vapor pressure of the organics emitted during each temperature ramp was estimated using the Clausius-Clapeyron equation as shown below:

$$P_{sat} = P_{atm} \cdot \exp\left(-\frac{\Delta H_{vap}}{R}\left(\frac{1}{T_{amb}} - \frac{1}{T_{peak}}\right)\right) \quad [S1]$$

Where P_{sat} is the saturation vapor pressure at an ambient temperature (T_{amb}) of 300 K, P_{atm} is atmospheric pressure, R is the gas constant (8.314 J/mol/K), T_{peak} is the temperature at which a given OC peak is obtained, and ΔH_{vap} is the heat of vaporization calculated according to Donahue et al.[2], such that the ΔH_{vap} value is 100 kJ/mol for a saturation concentration of 1 µg/m^3 and a subsequent factor of 10 spacing in saturation concentration results in a change of 5.8 kJ/mol. The P_{sat} value is converted to a saturation concentration (C*) based on the ideal gas law:

$$C^* = \frac{P_{sat} MW}{R T_{amb}} \quad [S2]$$

MW is the molecular weight of the compounds, estimated based on the distribution provided in Grieshop et al.[3]. The equations are iteratively solved to obtain the C* value for a given value of T_{peak}. Based on these calculations, we estimated that the temperatures corresponding to the four OC peaks would result in OC emissions with maximum C* values of 1.8E3, 0.38, 3E-4, and 6E-10 µg/m^3 for OC1, OC2, OC3, and OC4, respectively. These values resulted in our classification of OC1 as SVOC, OC2 and OC3 as LVOC, and OC4 as ELVOC based on the volatility distribution set provided by Donahue et al.[4]. These calculations do not provide exact measurements of the volatilities of the organics, but they give reliable estimates of the class of organics released during each temperature phase of the thermal-optical analysis. Ma et al.[5] correlated the OC peaks obtained using the IMPROVE_A protocol with organic volatility and found good agreement between the two

quantities, supporting the use of such calculations to estimate OC volatility from offline measurements.

S2 ToF-MS based volatility

Li et al.[6] have provided parametrizations to estimate the saturation mass concentration (C_0) for a given compound based on its elemental composition. The equation used to determine C_0 is:

$$log_{10}C_0 = (n_C^0 - n_C)b_C - n_O b_O - 2.\frac{n_C n_O}{n_C + n_O} b_{CO} - n_N b_N - n_S b_S \quad [S3]$$

Where n^0_C is the reference carbon number, n_C, n_O, n_N, and n_S denote the numbers of carbon, oxygen, nitrogen, and sulfur atoms, respectively. The values for n^0_C and the b coefficients are provided in Table S3. The C_0 values obtained for the different compounds was subsequently classified as IVOC, SVOC, LVOC, or ELVOC based on the volatility distributions set by Donahue et al.[4].

S3 Johnson-Neyman Analysis

The Johnson-Neyman analysis technique is used to explore the moderating effect of a continuous variable on the relationship between a predictor and response variable. The technique provides a range for the moderating variable over which the effect of the predictor variable on the response variable is statistically significant. We used a workbook developed by Carden et al.[7] to perform the Johnson-Neyman analysis. The EC and EL/LVOC concentrations were input as the predictor and moderating variables, respectively. The water-insoluble BrC Abs_{405} values were input as the response variable. A p-value of 0.01 was used to set the statistical significance of the results. The worksheet outputs the range for the moderating variable over which the effect of the change in the EC concentrations is significant on the WI BrC Abs_{405} values. The upper limit of this range was above the total EL/LVOC concentrations measured in our study, so only the lower bound was reported in this study. The values from the analysis are only indicative of the results observed in our study and might not be applicable to other combustion systems. Regardless, the Johnson-Neyman analysis provides a decent estimate for the significance of the moderating variable on observed relationships between two different parameters.

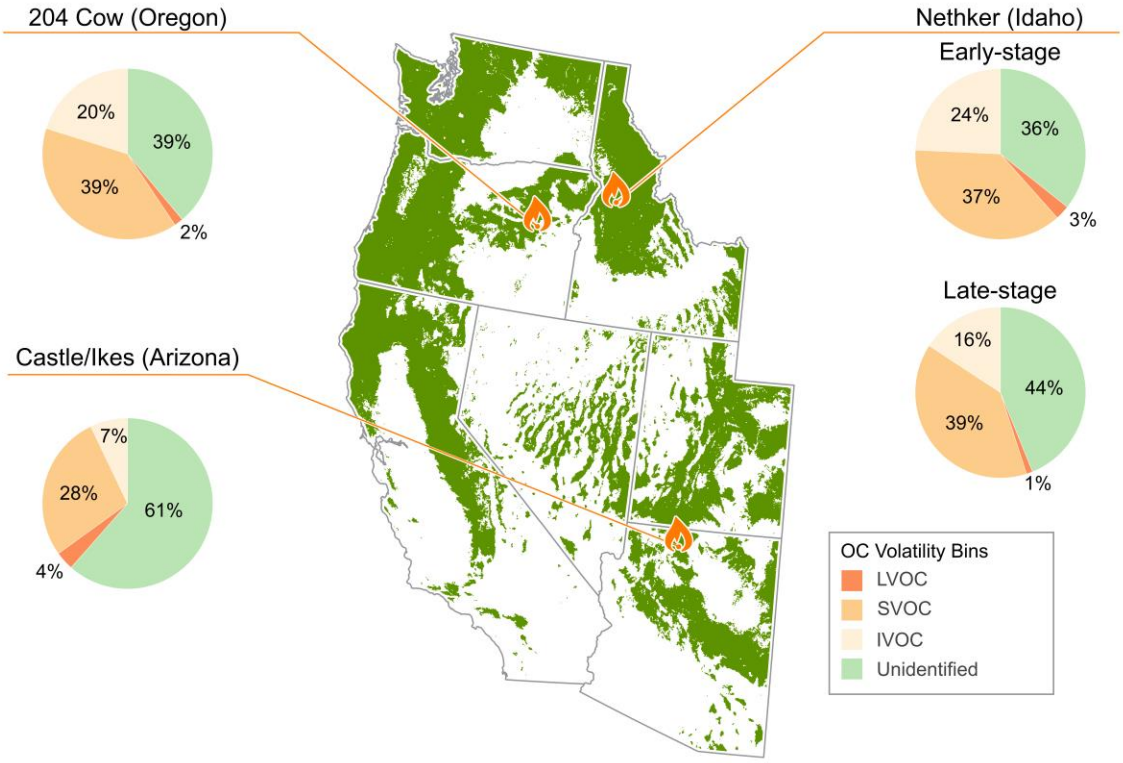

Figure S1: *OC volatility distributions estimated from the Tof-MS data for carbonaceous aerosols sampled across three wildfire locations in Idaho, Oregon, and Arizona. The results are similar to those presented in Figure 1 with SVOC and IVOC compounds placed in separate bins.*

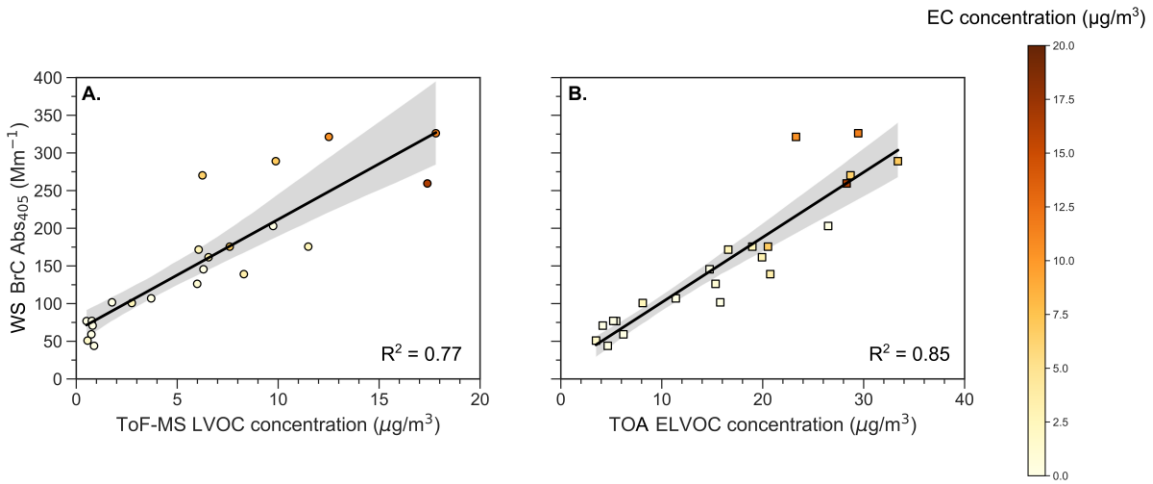

Figure S2: *Dependence of WS BrC Abs_{405} on A.) LVOC and B.) ELVOC concentrations determined using HR ToF-MS and TOA data, respectively. The solid black lines represent linear fits to the data with the shaded region representing 95% confidence intervals. The color bar depicts the EC concentration for each sample obtained from TOA.*

We observed a positive association between WS BrC and EL/LVOC concentrations. Samples with higher EC concentrations had slightly lower light absorption coefficients which indicates that the compounds co-emitted with greater EC concentrations might have low water-solubility.

Table S1. Information on the date and time of sampling for the different fires. It was hard to distinguish emissions from the Castle and Ikes fires, consequently, the samples from those fires are termed as Arizona fires here.

Filter #	Date	Sample Time Start (local time)	Sample Time Stop (local time)	Fire
1	8/9/2019	12:49	13:45	Nethker Fire
2	8/9/2019	13:58	14:58	Nethker Fire
3 (field blank)	8/9/2019	15:13	15:23	Nethker Fire
4	8/9/2019	16:54	17:54	Activity Barn
5	8/10/2019	21:34	22:34	Nethker Fire
6	8/11/2019	1:01	2:01	Nethker Fire
7	8/11/2019	4:29	5:29	Nethker Fire
8	8/11/2019	5:45	6:45	Nethker Fire
9	8/11/2019	7:35	8:35	Nethker Fire
10	8/11/2019	9:21	10:29	Nethker Fire
11	8/12/2019	12:51	13:51	Nethker Fire
12	8/12/2019	14:22	15:22	Nethker Fire
13	8/13/2019	21:39	22:39	Nethker Fire
14	8/13/2019	23:29	0:29	Nethker Fire
15	8/14/2019	1:38	2:38	Nethker Fire
16 (background)	8/15/2019	14:37	15:39	Nethker Fire
17 (background)	8/15/2019	19:56	20:54	Nethker Fire
18	8/15/2019	17:23	18:23	Nethker Fire
19	8/17/2019	0:07	1:07	Nethker Fire
20 (field blank)	8/17/2019	0:17	0:24	Nethker Fire
21	8/17/2019	1:29	2:29	Nethker Fire
22	8/20/2019	0:47	1:51	Arizona Castle Fire
23	8/20/2019	2:40	3:41	Arizona Ikes Fire
24	8/21/2019	0:14	1:14	Arizona Ikes Fire
25 (background)	8/21/2019	4:06	5:06	Arizona
26 (background)	8/21/2019	21:24	22:27	Arizona
27	8/22/2019	1:03	2:03	Arizona
28	8/22/2019	3:34	4:34	Arizona
29 (background)	8/25/2019	17:21	17:52	Oregon Cow Fire
30 (background)	8/25/2019	18:10	19:15	Oregon Cow Fire
31	8/25/2019	20:08	21:12	Oregon Cow Fire
32	8/26/2019	2:53	3:53	Oregon Cow Fire
33	8/26/2019	5:40	6:40	Oregon Cow Fire

Table S2. Compounds tentatively identified during chemical analysis along with the group they were classified in.

Compound Number	Compound Name	MW (w/o TMS)	Formula (w TMS)	Compound classification
1	Levoglucosan	162	C15H34O5Si3	Sugar
2	Dehydroabietic acid	300	C23H36O2Si	Terpenoid
3	Isopimaric acid	302	C23H38O2Si	Terpenoid
4	Didehydroabietic acid	298	C22H30O3Si	Terpenoid
5	UNK-4109	-	-	Sugar
6	Hexadecanoic acid	256	C19H40O2Si	Acid
7	Retene	234	C18H18	PAH
8	vanillic acid	168	C14H24O4Si2	Aromatic
9	UNK-3427	-	-	Sugar
10	Methyl dehydroabietate	314	C21H30O2	Terpenoid
11	Pimaric acid	302	C23H38O2Si	Terpenoid
12	Docosanoic acid	340	C25H52O2Si	Acid
13	7-Oxodehydroabietic acid	314	C23H34O3Si	Terpenoid
14	Palustric Acid	302	C23H38O2Si	Terpenoid
15	UNK-3423	-	-	Sugar
16	Eicosanoic acid	312	C23H48O2Si	Acid
17	Isopimaral	286	C20H30O	Terpenoid
18	Kaur-16-en-18-ol, (4-alpha)-	272	C23H40OSi	Terpenoid
19	Tetradecanoic acid	228	C17H36O2Si	Acid
20	3-Vanilpropanol	182	C16H30O3Si2	Aromatic
21	7-Oxodehydroabietic acid Methyl ester	328	C21H28O3	Aromatic
22	Q2-499	286	C20H30O	Terpenoid
23	UNK-3959	266	C20H26	PAH
24	8-Isopropyl-1,3-dimethylphenanthrene	248	C19H20	PAH
25	Tetracosanoic acid	368	C27H56O2Si	Acid
26	Q2-47	-	-	Sugar
27	UNK-4117	-	-	Sugar
28	UNK-4100	298	C22H30O3Si	Terpenoid
29	UNK-4083	254	C19H38O2Si	Acid
30	UNK-AHG-HZ-SOAZ-157	162	C15H34O5Si3	Sugar
31	Dehydroabietal	284	C20H28O	Terpenoid
32	Phenanthrene, 2,3,5-trimethyl-	220	C17H16	PAH
33	UNK-4115			Sugar
34	Octadecanoic acid	284	C21H44O2Si	Acid
35	Sandaracopimarinal	286	C20H30O	Terpenoid
36	UNK-1464	284	C20H28O	Terpenoid
37	UNK-3425	-	-	Sugar
38	UNK-4128	-	-	Sugar

39	UNK-4126	-	-	Sugar
40	Q2-287	-	-	Sugar
41	Deoxy-ribo-hexonic acid 1-4-lactone	162	C15H34O5Si3	Oxygenated
42	Sandaracopimaric acid	302	C23H38O2Si	Terpenoid
43	Heptadecanoic acid isomer	270	C20H42O2Si	Acid
44	10,18-Bisnorabieta-5,7,9(10),11,13-pentaene	238	C18H22	PAH
45	Phenanthrene, 1,7-dimethyl-	206	C16H14	PAH
46	Mannosan	162	C15H34O5Si3	Sugar
47	UNK-3730	296	C20H24O2	Terpenoid
48	Abietic acid	302	C23H38O2Si	Terpenoid
49	UNK-4033	-	-	Unknown
50	Coniferyl aldehyde	178	C13H18O3Si	Aromatic
51	Q2-608	-	-	Unknown
52	UNK-4108	-	-	Sugar
53	UNK-3871	270	C20H30	Terpenoid
54	Levopimaric acid	302	C23H38O2Si	Terpenoid
55	UNK-3885	-	-	Terpenoid
56	UNK-4047	232	C18H16	PAH
57	Isopimaric acid isomer	302	C23H38O2Si	Terpenoid
58	1-(10-Methylanthracen-9-yl)ethanone	234	C17H14O	PAH
59	4-nitrocatechol	155	C12H21O4Si2N	Nitrogen
60	1-Hexacosanol	382	C29H62OSi	Alcohol
61	Hexacosanoic acid	396	C29H60O2Si	Acid
62	UNK-4080	-	-	Triterpenoid
63	Azelaic acid	188	C15H32O4Si2	Oxygenated
64	Docosanol	326	C25H54OSi	Alcohol
65	Divanillyl	274	C22H34O4Si2	Aromatic
66	Dehydroabietinol	286	C23H38OSi	Terpenoid
67	6,4'-Dimethoxy-7-hydroxyisoflavone	298	C20H22O5Si	Terpenoid
68	Q2-479	-	-	Sugar
69	UNK-3976	-	-	Unknown
70	UNK-4119	-	-	Sugar
71	4,4-(Tetrahydrofuran-3,4-diyldimethanediyl)bis(2-methoxyphenol)	344	C26H40O5Si2	Aromatic
72	UNK-4089	288	C23H40OSi	Terpenoid
73	UNK-3669	-	-	Unknown
74	UNK-3708	-	-	Unknown
75	9-Tetradecenoic acid	226	C17H34O2Si	Acid
76	Juvibione	266	C16H26O3	Sesquiterpenoid
77	UNK-3942	-	-	Unknown
78	UNK-4075	-	-	Sugar

79	Hydroquinone	110	C12H22O2Si2	Aromatic
80	5-(hydroxymethyl)furfural	126	C9H14O3Si	Aromatic
81	Butanedioic acid	118	C10H22O4Si2	Oxygenated
82	UNK-2323	-	-	Sugar
83	UNK_3858	-	-	Oxygenated
84	Vanillin	152	C11H16O3Si	Aromatic
85	Q2-517	218	C17H26O2Si	Aromatic
86	Q2-558	-	-	Sugar
87	Pyrogallol	126	C15H30O3Si3	Aromatic
88	3-Hydroxybenzoic acid	138	C13H22O3Si2	Aromatic
89	4-Hydroxybenzoic acid	138	C13H22O3Si2	Aromatic
90	3,4-Dihydroxybenzaldehyde	138	C13H22O3Si2	Aromatic
91	Acetovanillone	166	C12H18O3Si	Aromatic
92	1,2,4-Benzenetriol	126	C15H30O3Si3	Aromatic
93	Galactosan	162	C15H34O5Si3	Sugar
94	UNK-3841	-	-	Sugar
95	3-(Methylthio)benzoic acid	168	C11H16O2SiS	Sulfur-containing
96	Methyl vanillate	182	C12H18O4Si	Aromatic
97	UNK-4105	180	C14H24O2Si	Aromatic
98	UNK-4094	-	-	Sugar
99	Q2-494	-	-	Sugar
100	UNK-4039	-	-	Sugar
101	Dodecanoic acid	200	C15H32O2Si	Acid
102	Q2-546	-	-	Sugar
103	Q2-560	-	-	Sugar
104	UNK-3784	-	-	Sugar
105	UNK-3940	-	-	Sugar
106	Q2-537	-	-	Sugar
107	Q2-593	-	-	Sugar
108	2, 4-Dihydroxyacetophenone	152	C14H24O3Si2	Aromatic
109	Suberic acid	174	C14H30O4Si2	Oxygenated
110	UNK-3176	162	C15H34O5Si3	Oxygenated
111	UNK-4054	164	C15H36O5Si3	Sugar
112	Adonitol	152	C20H52O5Si5	Sugar
113	(2R,3R,4R,5S)-Hexane-1,2,3,4,5,6-hexaol	182	C18H46O6Si4	Sugar
114	UNK-3931	-	-	Unknown
115	UNK-4081	-	-	Sugar
116	Veratric acid	182	C12H18O4Si	Aromatic
117	UNK-4002	194	C14H22O3Si	Aromatic
118	UNK-3440			Sugar
119	Methyl 3,4-dihydroxybenzoate	168	C14H24O4Si2	Aromatic
120	UNK-3860	-	-	Sugar

121	UNK-4113	-	-	Sugar
122	Hexonic acid, 3-deoxy-, gamma-lactone, 2,5,6-tris-OH	162	C15H34O5Si3	Oxygenated
123	4-methyl-5-nitrocatechol	169	C13H23O4Si2N	Nitrogen
124	3-methyl-5-nitrocatechol	169	C13H23O4Si2N	Nitrogen
125	Homovanillic Acid	182	C13H23O4SiN	Aromatic
126	Isophthalic acid	-	-	Aromatic
127	Tridecanoic acid	214	C16H34O2Si	Acid
128	3,4-Dihydroxybenzoic acid	154	C16H30O4Si3	Aromatic
129	UNK-3471	240	C18H32O5Si2	Aromatic
130	UNK-4098	261	C18H27O3SiN	Nitrogen
131	UNK-3929	243	C20H29O3Si2N	Nitrogen
132	Pentadecanoic acid isomer 1	242	C18H38O2Si	Acid
133	Pentadecanoic acid isomer 2	242	C18H38O2Si	Acid
134	Coniferyl alcohol	180	C16H28O3Si2	Aromatic
135	Q2-572	254	C16H30O2	Acid
136	Pentadecanoic acid	242	C18H38O2Si	Acid
137	Vanillyl glycol	198	C19H38O4Si3	Aromatic
138	UNK-446	266	C19H22O	Aromatic
139	1-Hexadecanol	242	C19H42OSi	Alcohol
140	4b,8-Dimethyl-2-isopropylphenanthrene, 4b,5,6,7,8,8a,9,10-octahydro-	256	C19H28	Terpenoid
141	UNK-4020	-	-	Sugar
142	UNK-4048	-	-	Unknown
143	C16 acid isomer 1	256	C19H40O2Si	Acid
144	UNK-4107	256	C19H28	Terpenoid
145	UNK-4045	260	C21H32O4Si2	Aromatic
146	7-Isopropyl-1,1,4a-trimethyl-1,2,3,4,4a,9,10,10a-octahydrophenanthrene	270	C20H30	Terpenoid
147	Manool	290	C20H34O	Terpenoid
148	9-Ethyl-10-methylanthracene	220	C17H16	PAH
149	Eicosane	282	C20H42	Alkane
150	Henicosane	296	C21H44	Alkane
151	Heneicosene	294	C21H42	Alkene
152	1-Octadecanol	270	C21H46OSi	Alcohol
153	Heptadecanoic acid	270	C20H42O2Si	Acid
154	Linoleic acid	280	C21H40O2Si	Acid
155	Octadecenoic acid	282	C21H42O2Si	Acid
156	Q2-3009	-	-	Aromatic
157	UNK-2182	194	C15H26OSi	Terpenoid

158	Q2-613	302	C22H34O3Si	Terpenoid
159	Tricosane	324	C23H48	Alkane
160	Nonadecanoic acid	298	C22H46O2Si	Acid
161	1-Eicosanol	298	C23H50OSi	Alcohol
162	Epiambrettolic acid	270	C22H46O3Si2	Oxygenated
163	1-Tetracosene	336	C24H48	Alkene
164	Tetracosane	338	C24H50	Alkane
165	(1R,4aS,7S)-7-Ethenyl-1,4a,7-trimethyl-9-oxo-2,3,4,5,6,8,10,10a-octahydrophenanthrene-1-carboxylic acid	316	C23H36O3Si	Terpenoid
166	1-Naphthalenecarboxylic acid, decahydro-5-(5-hydroxy-3-methylpentyl)-1,4a-dimethyl-6-methylene-, (1R,4aS,5R,8aS)-	322	C26H50O3Si2	Oxygenated
167	Docosanoic acid, methyl ester	354	C23H46O2	Ester
168	Heneicosanoic acid	326	C24H50O2Si	Acid
169	1-Monopalmitin	330	C25H54O4Si2	Oxygenated
170	Tetracosanoic acid, methyl ester	382	C25H50O2	Ester
171	Tricosanoic acid	354	C26H54O2Si	Acid
172	Tetracosanol	354	C27H58OSi	Alcohol
173	Nonacosane	408	C29H60	Alkane
174	Pentacosanoic acid	382	C28H58O2Si	Acid
175	Stigmasta-3,5-diene	396	C29H48	Triterpenoid
176	Nonacosan-10-ol	424	C32H68OSi	Alcohol
177	Hentriacontane	436	C31H64	Alkane
178	Matairesinol, (3R-E)-,	358	C26H38O6Si2	Aromatic
179	Beta-sitosterol	414	C32H58OSi	Triterpenoid
180	Stigmasta-3,5-dien-7-one	410	C29H46O	Triterpenoid
181	Gamma-Sitostenone	412	C29H48O	Triterpenoid
182	Octacosanoic acid	424	C31H64O2Si	Acid
183	Campesterol	400	C31H56OSi	Triterpenoid
184	Stigmastanol	416	C32H60OSi	Triterpenoid
185	UNK-AHG-LY-AMZ-140	-	-	Oxygenated
186	Q24-198	227	C14H25O4SiN	Nitrogen
187	Benzene, 1,3-dimethoxy-5-[(1E)-2-phenylethenyl]-	240	C16H16O2	Aromatic
188	3-Hydroxy-4-methyl-6H-benzo[c]chromen-6-one	226	C17H18O3Si	Aromatic
189	Naphthalene, 1,2,3,4-tetrahydro-6-(1-phenylethyl)-	236	C18H20	Aromatic
190	Isopimaric acid, methyl ester	316	C21H32O2	Terpenoid

191	Acetosyringone	196	C13H20O4Si	Aromatic
192	Pyroglutamic acid	129	C11H23O3Si2N	Nitrogen
193	Acenaphthene	154	C12H10	PAH
194	Bornyl acetate	196	C12H20O2	Terpenoid
195	16-Hydroxyhexadecanoic acid	272	C22H48O3Si2	Oxygenated
196	Q2-551	-	-	Sugar
197	UNK-3858	132	C12H28O3Si2	Oxygenated
198	Undecanedioic acid	304	C17H36O4Si2	Oxygenated

1. “UNK-” compounds are species observed but unidentified in the Fire Lab 2016 study[8].
2. “UNK-AHG-LY-AMZ-” compounds are species observed but unidentified in the GoAmazon study[9].
3. “UNK-AHG-HZ-SOAZ-” compounds are species observed but unidentified in the SOAS study[10].
4. "Qa-b” compounds are species observed but unidentified in this study[11] a is the filter number, and b is the compound ID in that filter sample.

Table S3. Composition classes and values of n^0_C and b values used from Li et al.

Classes	n^0_C	b_C	b_O	b_{CO}	b_N	b_S
CH	23.80	0.4861				
CHO	22.66	0.4481	1.656	-0.7790		
CHN	24.59	0.4066			0.9619	
CHON	24.13	0.3667	0.7732	-0.07790	1.114	
CHOS	24.06	0.3637	1.327	-0.3988		0.7579
CHONS	28.50	0.3848	1.011	0.2921	1.053	1.316

Table S4. Coefficients for the multivariate regression with NOCs transformed to square root values. The form of the equations is x BrC Abs_{405} = a + b.PAH + c.aromatic + d.($NOC^{0.5}$)

x	a	b	c	d	Adjusted R^2
MeS	56.2	6.7	8.6	290.4	0.86
WS	27.7	5.1	4.0	167.5	0.92

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