

1 Emissions from Forest and Grass Burns:
2 Comparison of Aerial and Ground Field
3 Measurements with Laboratory Burns

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13
14 **ABSTRACT**

15 Emissions from prescribed burns of forest and grass stands in western Florida were measured by
16 simultaneous aerial and ground sampling. Results were compared with biomass gathered from the
17 same stands and tested in an open burn laboratory test facility. Measurements included polycyclic
18 aromatic hydrocarbons (PAHs), volatile organic compounds (VOCs), particulate matter (PM_{2.5}),
19 elemental carbon (EC), organic carbon (OC), black carbon (BC), brown carbon (BrC),
20 polychlorinated dibenzo-p-dioxins (PCDDs), and polychlorinated dibenzofurans (PCDFs). The

21 flaming phase (high modified combustion efficiency) was characterized by high levels of BC and
22 BrC yet low levels of VOCs. In general, ground based measurements of PM_{2.5}, BC, and BrC
23 reported marginally higher emission factors than those measured by aerostat-lofted instruments in
24 the plume. The optically-determined BC emission factor was approximately ten times higher than
25 many previously reported results. Simultaneous BC and EC measurements showed that EC values
26 were, on average, 42% lower than the BC values, lending uncertainty to the common use of EC
27 measurements as a BC surrogate. PAH emission factors were indistinguishable across the
28 sampling scenarios, while PCDDs/PCDFs saw a significant decline in the laboratory testing.

29

30 **KEYWORDS:** Black carbon, PAH, Brown Carbon, PCDD, PCDF, VOCs, PM_{2.5}

31 **1. INTRODUCTION**

32 Wildfires and prescribed forest burns generate a variety of emissions that can cause adverse
33 health effects for humans, contribute to climate change, and give rise to decreased visibility.
34 Prescribed forest burns are carried out during the cooler, wetter months to reduce fuel buildup,
35 improve the habitat for animal and plant species, minimize spread of disease, and reduce the risk
36 of wildfires during the warmer months.

37 Measuring air emissions and deriving emission factors from prescribed forest burns are
38 important because it will provide data to better understand effects on air quality, as the emission
39 factors are used in national emission inventory calculations, air climate change models, and risk
40 assessments. Forest burns are one of the largest sources of particulate matter in the U.S.A (U.S.
41 EPA, 2012). Light-absorbing carbonaceous aerosols known as black carbon (BC) are considered
42 to be one of the major contributors to the global climate change (Ramanathan and Carmichael,
43 2008). Elemental carbon (EC), sometimes used interchangeably with BC despite measurement by
44 different means (thermal-optical versus optical/aethalometer), is an indicator of combustion extent.
45 The light-absorbing organic matter aerosols referred to as brown carbon (BrC) is a possible global
46 warming agent (Alexander et al., 2008; Andreae and Gelencser, 2006; Kirchstetter et al., 2004)
47 that is starting to get more attention. Volatile organic compounds (VOCs) from combustion such
48 as benzene, and semi-volatile organic compounds such as polycyclic aromatic hydrocarbons
49 (PAHs), polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans
50 (PCDFs), include pollutants with properties that are toxic, carcinogenic, bioaccumulative, and
51 persistent in the environment.

52 Measuring air emissions from actual field burns is challenging due to variation in plume height,
53 changes in wind direction, and safety for sampling equipment and personnel, all of which may

54 limit the time the sampling equipment can spend in the plume. Emissions can be measured in the
55 field by aerial or ground based sampling platforms or, alternatively, in laboratory test facilities.
56 Aerial measurements are important when plumes are lofted and when ground-based sampling may
57 create a bias toward the safer, smoldering phase of the burn. Laboratory measurements are utilized
58 when field measurements are impractical or costly and when species-, phase-, and condition-
59 specific burn data are desired.

60 Emission sampling is typically separated into flaming (good combustion) and smoldering
61 (poor combustion) phases, as each has reasonably distinctive emission factors. Samples further
62 afield from large burns may be a mix of both flaming and smoldering combustion phases as the
63 flame front and post-flame smoldering emissions are mixed and entrained into the plume.
64 Measurements at heights above the fire are more likely to generate a composite sample of flaming
65 and smoldering emissions than would be produced from point source measurements on the ground.
66 Collection of ground based samples may be biased toward the smoldering phase because they are
67 more amenable to safe sampling and equipment survival than intense flaming phases. During
68 ground-based sampling, mixing of the plume is limited, the pollutants have less aging, heavier
69 particles are less likely entrained, and pollutants are less likely to have been affected by
70 atmospheric chemistry such as formation of secondary organic aerosols (Cubison et al., 2011).
71 Evidence for differences between emissions sampled from airborne and ground-based platforms
72 was observed for hydrocarbons and oxygenated organic species by Burling et al. (2011). Methods
73 for aerial and ground measurements were demonstrated during three southeastern U.S. prescribed
74 burns (Aurell and Gullett, 2013). Initial reporting of concurrent application of these aerial- and
75 ground-based measurements (Strand et al., 2014) showed little difference in a variety of pollutants,
76 but only single aerial samples were available for comparison.

77 Due to their small size and controlled combustion conditions, laboratory tests can have the
78 advantage of allowing both species-specific studies and separate measurements of different
79 combustion phases. Comparability between laboratory and field emission data has been
80 demonstrated for African fires (Christian et al., 2003) and extensively covered by Yokelson et al.
81 (2012). Differences observed in laboratory versus field emission factors have been attributed to
82 higher modified combustion efficiency (MCE) values in laboratory burns (Christian et al., 2003)
83 of African savanna fuels. When they account for MCE, laboratory-based emission data predict
84 emission factors within 15% of the field values.

85 Work reported here, part of a comprehensive study to develop a fire and fuels dataset (Ottmar
86 et al., 2013), presents multipollutant emission factors derived through concurrent aerial and ground
87 sampling and accompanied by laboratory combustion tests on the same biomass.

88

89 **2. EXPERIMENTAL SECTION**

90 **2.1 Sampling Method and Instrumentation**

91 A light-weight instrument package termed the “Flyer” was attached to a helium filled, tethered
92 aerostat (4.3 m in diameter) mounted to an all terrain vehicle (ATV). The use of the aerostat and
93 Flyer instrumentation with the ATV has been described in detail elsewhere (Aurell and Gullett,
94 2013; Aurell et al., 2011). In summary, the instrumentation on the Flyer consisted of: non-
95 dispersive infrared for continuous measurement of carbon dioxide (CO₂) (LICOR 820, LICOR
96 Biosciences U.S.A), light-scattering photometers for continuous PM_{2.5} (DustTrak 8520, DustTrak
97 DRX, TSI Inc. U.S.A), micro aethlometers for BC and BrC (AE51, AE52, Aethlabs, U.S.A),
98 SUMMA canisters for VOCs, CO₂, and carbon monoxide (CO) (Columbia Analytical Services –
99 CAS, U.S.A.), batch sampling of PM_{2.5} onto a 47 mm in diameter Teflon filter (pore size 2 μm),

100 batch sampling onto quartz filters for EC, OC, and TC measurements, and batch sampling of
101 PCDDs/PCDFs onto a quartz filter and polyurethane foam sorbent. The Flyer's position and
102 altitude were tracked using a global positioning system (MTi-G, Xsens, Netherlands). The ground
103 based ATV was also equipped with a smaller sampling package for measurements of PAHs using
104 a quartz filter, polyurethane foam, and XAD-2 sorbent, described in detail in Aurell et al. (2011).

105 **2.2 Field Burn Description**

106 Simultaneous aerial and ground sampling was conducted during prescribed burns of one
107 grass/forb field (126.5 ha) and one managed longleaf pine (*Pinus palustris*) forest (151 ha) at Eglin
108 Air Force Base, Florida, during two days in November of 2012 (Ottmar et al., 2014; Ottmar et al.,
109 2013). This site was selected to complement previous testing done during the Prescribed Fire
110 Combustion and Atmospheric Dynamic Research (RxCADRE) research campaign at Eglin Air
111 Force Base (<http://www.firelab.org/document/rxcadre-overview>). The burns were ignited by drip
112 torch from all terrain vehicles (ATVs) in separate parabola shaped lines, described in detail
113 elsewhere (Ottmar et al., 2014). The aerostat sampler collected emissions downwind of the grass
114 field and forest area at an altitude of 50-110 m and 45-85 m, respectively. Forest biomass was
115 gathered on an adjacent, unburned area and transported for subsequent laboratory burn testing four
116 days later.

117 **2.3 Open burn test facility**

118 The U.S. EPA's 70 m³ open burn test facility (OBTF) was used for laboratory simulation of
119 prescribed burns of forest litter (no grass burns were conducted), (Aurell and Gullett, 2013). The
120 OBTF (described in detail elsewhere (Grandesso et al., 2011)) is an enclosed facility ventilated
121 with a high-volume blower that pulls in ambient air and smaller, interior fans that together ensure
122 complete mixing and oxygen concentration close to ambient conditions. Twelve biomass burns

123 were conducted atop an aluminum foil-covered (replaced between each burn) sand layer supported
124 by a steel plate (0.93 m× 0.93 m). The forest biomass was divided by a quartering procedure
125 (Aurell and Gullett, 2013) and placed on the foil at a charge size (1.4 kg) closely mimicking the
126 field area density (1.1 kg/m² (Ottmar et al., 2014)). For consistency, the OBTF sampling used the
127 same instrumentation (the Flyer) as in the field tests with the addition of a bench top CO monitor
128 (California Analytical Instruments Model 200, 0-1,000 ppm, U.S.A.). Ambient air background
129 samples were collected after the burns by sampling inside the OBTF for approximately 9 hours.
130 An ultimate analysis and PCDD/PCDF analysis was conducted on the raw forest biomass. For the
131 latter, three raw biomass samples (48±2 g) were Soxhlet-extracted using U.S. EPA Method 3540C
132 (1996) and analyzed for PCDDs/PCDFs by high resolution gas chromatography/high resolution
133 mass spectrometry (HRGC/HRMS).

134 **2.4 Data and Sample Analyses**

135 Emission factors were derived using the carbon mass balance approach (Laursen et al., 1992),
136 which assumes that all combusted carbon in the biomass is emitted to the atmosphere as CO₂, CO,
137 CH₄, and total hydrocarbons (THCs). Due to instrumentation size and weight considerations, only
138 CO₂ was measured in the field, while both CO₂ and CO were measured in the OBTF. The derived
139 emission factors (all except VOCs) in the OBTF were 6.7% ± 2.7 lower using CO₂ and CO as a
140 carbon source rather than only CO₂, which is within the total error of the method and likely the
141 reproducibility of the event. The VOC emission factors were 2.3% ± 0.16 lower using CO₂, CO,
142 and CH₄ than only CO₂ as a carbon source. Emission factors are expressed in terms of pollutant
143 mass per mass of biomass consumed by the burn using a biomass carbon fraction value of 0.5
144 (Strand et al., 2014).

145 PAHs were cleaned up according to U.S. EPA Method TO-13 (1999b) and analyzed using a
146 Hewlett-Packard gas chromatograph 5890 Series II with a DB-5 60 m × 0.25 mm × 0.25 μm column
147 (Agilent/J&W Scientific) connected to a Hewlett Packard 5972 mass spectrometer. PAH emission
148 factors were evaluated using toxic equivalent factors (TEFs) relative to the benzo-[a]-pyrene
149 toxicity equivalent (B[a]P-TEQs) (Larsen and Larsen, 1998). PCDD/PCDF samples were cleaned
150 up via a modified U.S. EPA Method TO-9A (1999a) and analyzed with a HRGC/HRMS using a
151 Hewlett-Packard gas chromatograph 6890 Series coupled to a Micromass Premier (Waters Inc.,
152 UK) with a Dioxin-2 60 m × 0.25 mm × 0.25 μm column (Restek). The PCDD/PCDF TEQ
153 emission factors were determined by using the 2005 World Health Organization (WHO) TEFs
154 (Van den Berg et al., 2006). Non-detect (ND) congeners were noted for some of the seventeen
155 PCDD/PCDF TEF congeners. The ND congeners were set to zero as well as to the limit of
156 detection (SI Table S2).

157 EC, TC, and OC were analyzed via a modified, thermal-optical analysis (TOA) using NIOSH
158 Method 5040 (1999c) (Khan et al., 2012). Custom correction factors were calculated for the
159 DustTrak, as per the manufacturer's recommendations, by dividing the average continuous PM_{2.5}
160 concentration by the PM_{2.5} batch filter concentration collected during the same time period. The
161 average correction factor was 2.4±0.61 for the OBTF measurements. The Modified Combustion
162 Efficiency (MCE), ($\Delta\text{CO}_2/(\Delta\text{CO}+\Delta\text{CO}_2)$), was calculated using the CO and CO₂ concentrations
163 collected by the SUMMA canisters and OBTF test data. The BC and BrC were divided into
164 categories of MCE > 0.95 (good combustion), >0.90 and <0.95, and <0.90 (poor combustion) for
165 analysis.

166

167 3. RESULTS AND DISCUSSION

168 Simultaneous aerial and ground-based samples were taken with the exception of the forest burn
169 where duplicate ground-based samples were possible due to the burn duration. The forest
170 biomass underwent multiple burns in the OBTF, resulting in three, composite-burn samples of
171 trace pollutants (e.g., PCDD/PCDF) and multiple replicates of filter-based pollutants. Results are
172 presented in Table 1.

173 **3.1 PM_{2.5}, BC, BrC, EC, OC, TC**

174 *3.1.1 Data Comparison*

175 The PM_{2.5} emission factors from field forest burns, 20-23 g/kg (Table 1), were slightly higher
176 than previously published values derived from the same location (13-14 g/kg) (Aurell and Gullett,
177 2013), as well as from wildfires from Portugal and Mexico 4.5-12 g/kg (Alves et al., 2011;
178 Yokelson et al., 2011). Both the field and OBTF forest burn emission factors (12-23 g/kg) were in
179 the upper range of previously published emission factors from prescribed burning and laboratory
180 burn tests of US biomass species 0.90-24 g/kg (Burling et al., 2011; Lee et al., 2005) and 1.6-34
181 g/kg (Hays et al., 2002; Hosseini et al., 2013), respectively. The PM_{2.5} emission factors from the
182 grass burns (14-18 g/kg) were similar or in the upper range of previously published data of different
183 grass species (savanna, saw grass, wire grass, Kentucky bluegrass) 4.0-15 g/kg (Dhammapala et
184 al., 2006; Urbanski et al., 2009; Yokelson et al., 2011). The BrC emission factors in this study
185 (0.68-1.8 g/kg) were in the same range as previously measured (0.9-2.1 g/kg (Aurell and Gullett,
186 2013)) in the southeast U.S.

187 The EC emission factors found in this study (0.39-0.62 g/kg) are in the same range as found in
188 the literature 0.27-1.3 g/kg (Alves et al., 2011; Andreae et al., 1998; Ferek et al., 1998; Hays et al.,
189 2002) (analyzed using thermal measurements) from prescribed burning, wildfires, and laboratory
190 burns of different biomass types. Thermal EC measurements, both current and historical, have

191 often been equated with BC, which is an optically measured property. Few BC emission factors
192 determined by light absorption measurements from biomass burns exist in the literature. Kondo et
193 al. (Kondo et al., 2011) reported an emission factor of 180 ng/m³ BC per ppm CO₂, or 0.11 g
194 BC/kg (assuming a biomass carbon fraction of 0.50), which is lower than found in this study (0.85-
195 1.4 g/kg). In a previous study we found BC emission factors of 1.0-2.7 g/kg (Aurell and Gullett,
196 2013) from prescribed burns of biomass from the southeast US, which are similar to those found
197 in this study. Seven of nine EC emission factors (0.39-0.62 g/kg) we measured were 42%±13%
198 lower than the simultaneously sampled BC emission factors (0.85-1.4 g/kg) but the concentrations
199 for all of the data points correlated well (correlation coefficient of 0.85, SI Figure S1). Importantly,
200 it shows that historical use of EC values to infer BC emission factors may have significantly
201 underestimated BC levels. Moderate to good correlations between BC and EC measurements have
202 previously been found in inter-comparison studies (r^2 0.44-0.88) (Ahmed et al., 2009; Jeong et al.,
203 2004; Kamboures et al., 2013; Ram et al., 2010). In a recent study using the same measurement
204 methods in this work, BC values using an aethalometer (AE51) were compared to EC
205 measurements (NIOSH method) and found to be 39% higher or a BC/EC ratio of 1.64 (Yelverton
206 et al., 2014), in agreement with our findings.

207 The OC emission factors for forest field burns, 11-15 g/kg, were double those from wildfires
208 in Portugal 6.0±2.9 g/kg (Alves et al., 2011) and forest burns in Brazil 6.8 g/kg (estimated Fc 0.5)
209 (Ferek et al., 1998) while the grass burn OC emission factors of 6.5-7.0 g/kg were similar to grass
210 burns in Brazil at 5.1 g/kg (estimated Fc 0.5) (Ferek et al., 1998).

211

212 Table 1. Results.

Compound	Unit	Grass burn	Grass burn	Forest burn	Forest burn	Forest burn
		Ground	Aerostat	Ground	Aerostat	OBTF ^h
Filter PM _{2.5}	g/kg biomass	18 ^g	14 ^g	23±1.8 ^{d,g}	20 ^g	12.2±6.9 ^d (11.4±6.4)
Continuous PM _{2.5}	g/kg biomass	20 ^g	15 ^g	25 ^g	24 ^g	11±4.6 (10±4.1 ^d)
Continuous BC ^a	g/kg biomass	1.1 ^g	0.91 ^g	0.89 ^g	1.4 ^g	1.3 ±0.40 ^d (1.2±0.38 ^d)
Continuous BrC ^a	g/kg biomass	1.8 ^g	NS ^g	NS ^g	0.92 ^g	0.68±0.37 ^d (0.58±0.31 ^d)
Continuous BC ^b	g/kg biomass	1.2	1.0	0.85±0.20 ^d	1.4	1.3±0.31 (1.2±0.26)
Continuous BrC ^b	g/kg biomass	1.8	NS	NS	1.0	0.66±0.27 (0.58±0.23)
Filter EC	g/kg biomass	0.62 ^g	0.56 ^g	0.39±0.16 ^{d,g}	0.46 ^g	0.89±0.29 ^d (0.84±0.28)
Filter OC	g/kg biomass	7.0 ^g	6.5 ^g	15±1.8 ^{d,g}	11 ^g	6.7±6.5 ^d (6.3±6.0)
Filter TC	g/kg biomass	7.6 ^g	7.1 ^g	16±1.9 ^{d,g}	12 ^g	7.6±6.4 ^d (7.1±5.9)
BC/PM _{2.5} ^c	mass ratio %	6.8 ^g	7.0 ^g	3.6±0.67 ^{d,g}	7.0 ^g	12.8±5.3 ^d
EC/PM _{2.5}	mass ratio %	3.5 ^g	3.9 ^g	1.6±0.54 ^{d,g}	2.3 ^g	9.6±6.9 ^d
BrC/PM _{2.5} ^c	mass ratio %	9.8 ^g	NS ^g	NS	5.0 ^g	6.1±2.6 ^d
OC/BC	ratio	5.3	6.6	18.4±4.4	7.9	5.1±4.1
OC/EC	ratio	11.2	11.6	42.5±13.2	24.4	9.6±11.4
PCDD/PCDF	ng WHO TEQ/kg biomass	1.9 [2.2] ^{e,f}		0.80	1.2 [1.5] ^e	0.092±0.10 ^e (0.086±0.10) [0.18±0.076 ^d (0.17±0.069)] ^e
PCDD/PCDF	Total ng/kg biomass	247 ^f		65	113	6.9±3.6 (6.5±3.3)
PCDD/PCDF	mass ratio	7.7 ^f		4.0	6.2	1.0±0.17
SUM 16 EPA PAHs	mg/kg biomass	35	NS	23	NS	27±3.7 ^d (25±3.6)

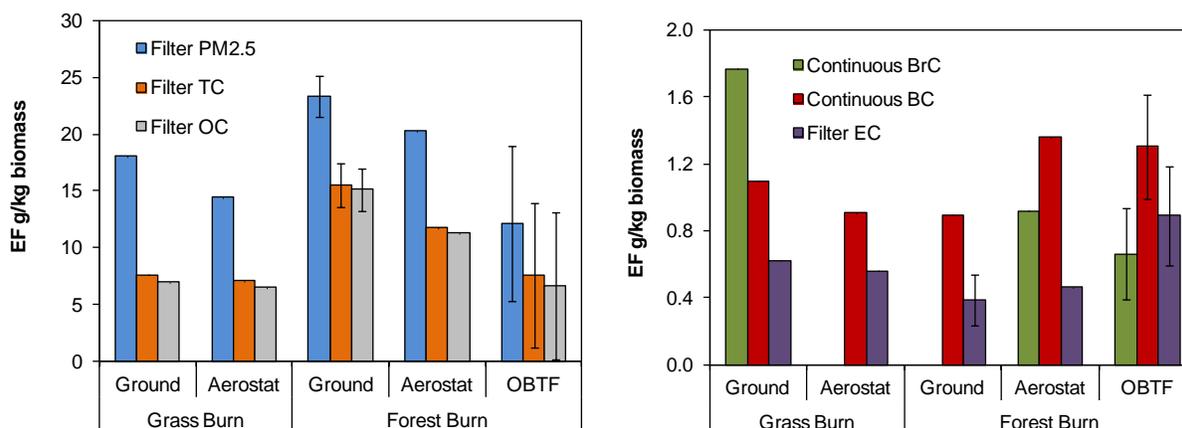
213 ^a sampled for a longer time interval than the batch filter. ^b sampled at the same time interval as the batch filter ^c batch filter and BC simultaneously
214 sampled. ^d 1 STDV ^e None detectable congeners = limit of detection within brackets. ^f Combined sample from Aerostat and Ground measurements.
215 ^g from Strand et al. 2014. ^h Emission factors calculated using carbon from both CO₂ and CO within parentheses.

216

217 3.1.2 Aerial versus Ground Sampling

218 The PM_{2.5}, TC, and OC emission factors were higher on the ground than in the air for both
219 forest and grass burns (Figure 1 and SI Figure S2), although the minimal sample size limits the
220 statistical difference.

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223 Figure 1. Particle and carbon type emission factors. Error bars denote ± 1 STDV. Ground and
224 aerostat data from this current study, see also (Strand et al., 2014).

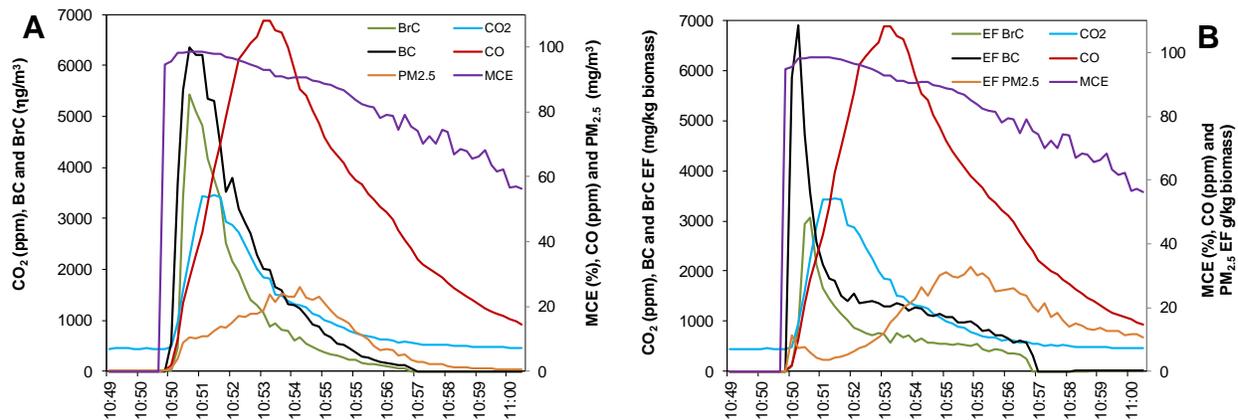
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226 3.1.3 Grass versus Forest Burns

227 The PM carbon analysis showed the grass burn had lower emission factors for TC and OC than
228 the forest burns for both ground and aerostat-based measurements (Figure 1).

229 3.1.4 Laboratory versus Field Sampling

230 The OBTF average $PM_{2.5}$, TC, and OC forest burn emission factors were similar to those
231 derived in the field. A typical concentration and emission factor plot from the OBTF tests is shown
232 in Figures 2A and 2B, respectively. The initial flaming region with high MCE is quickly followed
233 by a rapid spike in BC and BrC concentration that then falls off, replaced by increasing CO and
234 CO₂, followed by $PM_{2.5}$. BC and BrC emission factors, as well as the BC to $PM_{2.5}$ mass ratio,
235 increase at higher MCE values (Figure 3 and SI Figure S3) suggesting that the OBTF tests may
236 have been conducted under higher average combustion efficiencies than in the field (Table 1).
237 The difference in BC and EC concentrations resulted in the BC/ $PM_{2.5}$ ratio being 1.3-3.0 times
238 higher than the EC/ $PM_{2.5}$ ratio.

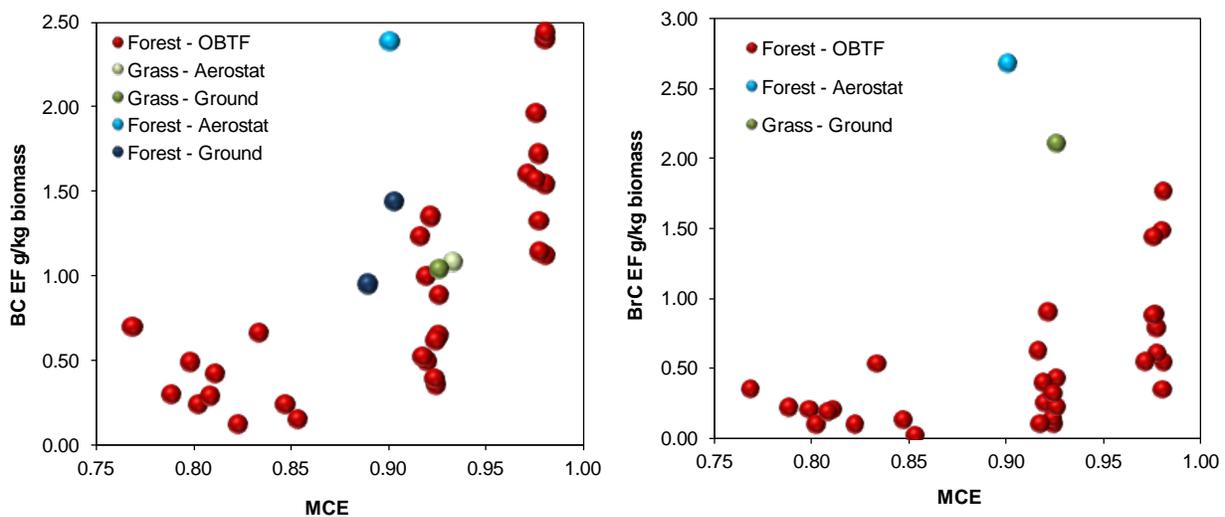


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241 Figure 2. Typical OBTF burn of forest biomass. Concentrations (A) and emission factors (B).

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245 Figure 3. BC and BrC versus modified combustion efficiency. Aerostat and Ground measurements
 246 from this work; see also Strand et al. (Strand et al., 2014).

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251 **3.2 Volatile Organic Compounds**

252 *3.2.1 Data Comparison*

253 The benzene emission factors for the forest/field burns (277 ± 24 and 283 mg/kg) were in the
254 same range as previous burns of understory forest with the same MCE (276 ± 79 to 526 ± 262 mg/kg)
255 (Aurell and Gullett, 2013) and in both flaming (211 ± 73 mg/kg) and smoldering (422 ± 244 mg/kg)
256 stages of pine dominated forest in Georgia (Lee et al., 2005). The OBTF benzene emission factor
257 (92 ± 20 mg/kg) was lower than previous laboratory forest litter burns (148-284 mg/kg) (Aurell and
258 Gullett, 2013; Hays et al., 2002). The benzene emission factors from the grass burns (192 ± 63 and
259 253 mg/kg) were in the same range as Brazilian grass and slash piles (168 mg/kg) (Yokelson et
260 al., 2007).

261 *3.2.2 Aerial versus Ground Sampling*

262 No correlation between altitude (aerostat-sampled versus ground-sampled) and VOC emission
263 factor levels was found, although more samples would be needed to confirm this finding.

264 *3.2.3 Grass versus Forest Burns*

265 VOCs for grass burns were lower than those from forest burns. Grass burn measurements,
266 whether by aerostat or ground sampling, had similar MCE values, about 0.92 to 0.94. These values
267 were higher than those from forest burns (0.88 to 0.90). The VOC emission factors decreased with
268 increased MCEs (Table 2, Figure 4), influenced more by the combustion quality than whether the
269 biomass was forest or grass. The higher MCEs in the OBTF consistently led to lower VOC
270 emission factors.

271 3.2.4 Laboratory versus Field Sampling

272 Consistent with the grass versus forest VOC/MCE relationship above, the VOC emission
 273 factors in the OBTF decreased with the higher MCE (Table 2, Figure 4) of the OBTF.

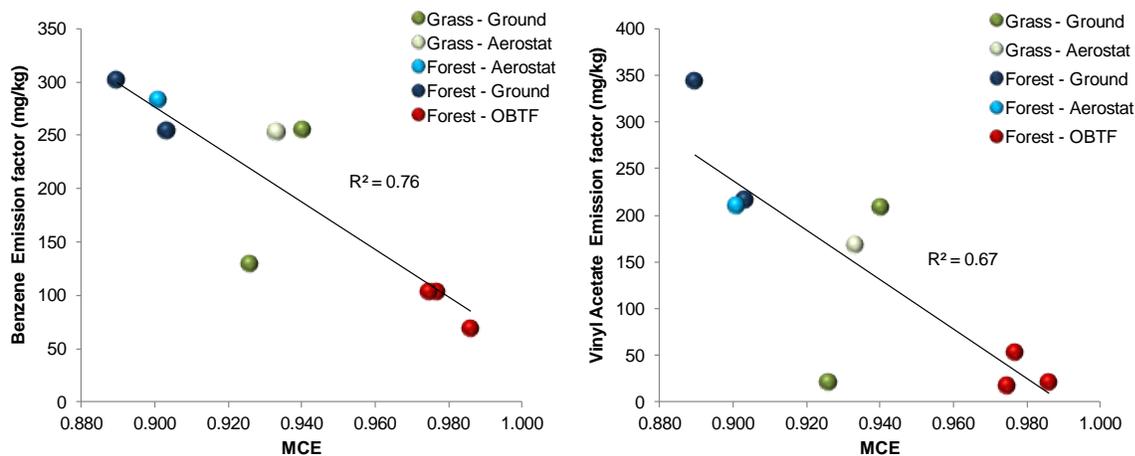
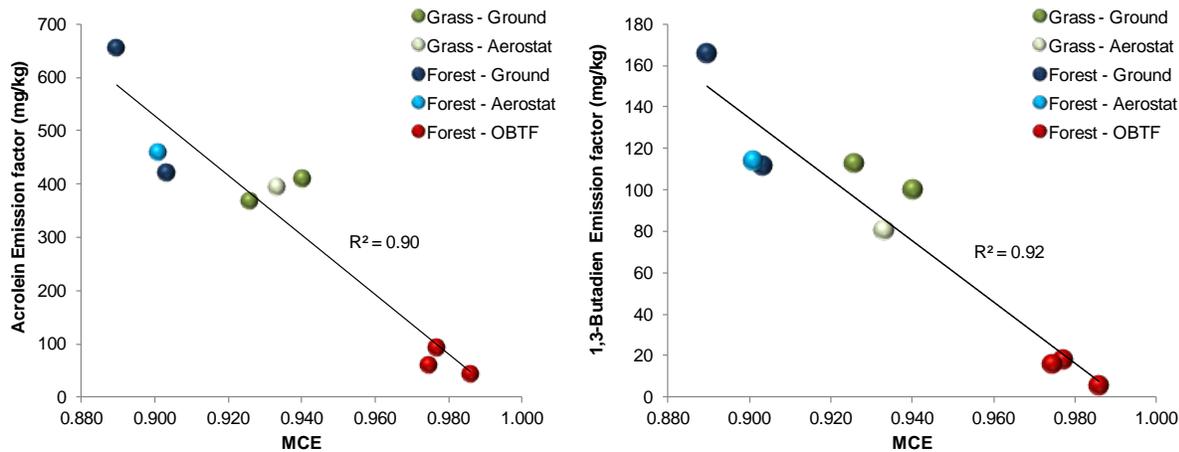
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275 Table 2. VOC, CO₂, CO, and CH₄ emission factors^d.

	Grass burn Ground mg/kg	Grass burn Aerostat mg/kg	Forest burn Ground mg/kg	Forest burn Aerostat mg/kg	Forest burn OBTF mg/kg
Sampling Altitude (m)	2	82	2	48	NA
Modified combustion efficiency	0.933±0.007 ^b	0.933	0.896±0.007 ^b	0.901	0.979±0.006 ^c
CO ₂	1711±13	1711	1656	1652	1793±12
CO	78±8.3	78	113	116	24±7.0
Methane (CH ₄)	NA	NA	NA	NA	0.89±0.27
Propene	261±22 ^b	204	329±14 ^b	314	84±31 ^c
Chloromethane ^a	15±2.5 ^b	15	10±1.1 ^b	11	9.9±5.4 ^c
1,3-Butadiene ^a	106±6.5 ^b	80	138±27 ^b	114	13±6.8 ^c
Acetonitrile	112±20 ^b	95	156±18 ^b	144	14±6.6 ^c
Acrolein ^a	388±21 ^b	394	537±118 ^b	458	65±26 ^c
Acrylonitrile ^a	39±3.8 ^b	36	37±3.9 ^b	42	6.3±2.4 ^c
Vinyl Acetate ^a	115±94 ^b	168	280±63 ^b	209	30±20 ^c
2-Butanone (MEK)	40±3.1 ^b	34	98±14 ^b	69	12±4.9 ^c
Benzene ^a	192±63 ^b	253	277±24 ^b	283	92±20 ^c
Toluene ^a	89	84	141±26 ^b	111	27±12 ^c
Ethylbenzene ^a	17	15	21±3.8 ^b	18	3.3±2.8 ^c
m,p-Xylene	26	23	47±9.4 ^b	35	5.9±5.7 ^c
Styrene ^a	41	35	50±10 ^b	43	9.2±4.2 ^b
o-Xylene ^a	7.0	7.0	15±3.0 ^b	12	2.2±0.76 ^b
Cumene ^a	1.2	ND	1.8±0.32 ^b	ND	0.49±0.21 ^b
alpha-Pinene	5.4	5.4	34±2.8 ^b	44	6.9±1.78 ^b
1,3,5-Trimethylbenzene	ND	ND	1.9±0.32 ^b	ND	0.37
1,2,4-Trimethylbenzene	4.0	4.6	8.3±1.7 ^b	5.9	1.1±0.40 ^b
d-Limonene	29	23	31±10 ^b	20	6.7±3.3
Naphthalene	40	42	46±7.2 ^b	44	16±3.0 ^b
n-Hexane	1.4±0.62 ^b	2.7	8.7±2.0 ^b	3.7	0.75±0.30 ^b
n-Heptane	0.48	0.44	6.9±2.4 ^b	0.79	0.46±0.087 ^b
Ethanol	21±7.9 ^b	13	22±6.6 ^b	6.5	0.90

276 ^a Included in the EPA list of hazardous air pollutants. ^b Range of data. ^c 1 STDV. ^d Emission factors
 277 calculated with CO and CO₂, except for CO, CO₂, and CH₄ for OBTF which included carbon from CH₄.
 278 ND = not detected, DNA = does not apply, NA = not analyzed.

279



282

Figure 4. VOCs vs MCE.

283

284 3.3 PAHs.

285 3.3.1 Data Comparison

286 Comparison of PAH emission factors against literature data is cumbersome as different PAHs
287 may be analyzed along with the use of different sampling methods (different configurations of
288 XAD, PUF, and quartz filter). In some cases, a toxicity measure allows inter-comparisons. The
289 B[a]P-TEQ emission factors presented from the literature in this paper have been derived using

290 the reported literature PAH values and the TEF values from Larsen and Larsen (1998) and, in some
291 cases, the carbon fraction of the fuel.

292 The emission factors for phenanthrene from forest burns in this study (3.1-3.4 mg/kg, Table 3)
293 were in the mid-range of literature data obtained from laboratory tests 0.11-11.52 mg/kg (Hays et
294 al., 2002; Hosseini et al., 2013; Jenkins et al., 1996; Singh et al., 2013) and was similar when
295 sampled using the same sampling method (filter and XAD sorbent) 2.6-3.9 mg/kg (Jenkins et al.,
296 1996). The phenanthrene emission factor from the grass burn, 3.7 mg/kg, was also similar to
297 previous laboratory tests of Kentucky bluegrass, 4.0 mg/kg (Dhammapala et al., 2007). The
298 emission factor from the field grass burn was approximately two times lower than laboratory burns
299 of Kentucky bluegrass reported in the literature: 0.31 mg B[a]P-TEQ/kg and 0.58 mg B[a]P-
300 TEQ/kg (Dhammapala et al., 2007), respectively. The forest field burn emission factors were
301 similar to laboratory burns of U.S. fir, pine, chaparral and oak 0.12-0.50 mg B[a]P-TEQ/kg
302 (Hosseini et al., 2013; Jenkins et al., 1996) but much lower than laboratory burned fuel woods
303 from India, 3.2 mg B[a]P-TEQ/kg (Singh et al., 2013). The B[a]P-TEQ emission factors from
304 prescribed burning of forest litter and grass presented in this paper are low in comparison to
305 conventional outdoor residential wood-fired hydronic heaters 6.1 mg B[a]P-TEQ/kg (Aurell et al.,
306 2012a) (white pine) and 2.2 mg B[a]P-TEQ/kg (Aurell et al., 2012a) (red oak), and open burns of
307 municipal waste 2.2 mg B[a]P-TEQ/kg (Aurell et al., 2012b), whereas they are in the same range
308 as a modern three stage combustion hydronic heaters (red oak) 0.31 mg B[a]P-TEQ/kg (Aurell et
309 al., 2012a), and higher than a pellet based hydronic heaters 0.063 mg B[a]P-TEQ/kg (Aurell et al.,
310 2012a)

311 3.3.2 *Grass versus Forest Burns*

312 Total PAHs (sum of 16 EPA PAHs) and B[a]P-TEQ emission factors from the forest and grass
313 fuels are presented in Table 3 and SI Table S1, respectively. The most abundant PAHs were
314 naphthalene, acenaphthylene, and phenanthrene. Naphthalene contributed to 56% of the total PAH
315 emission factors, on average (SI Figure S4). The total PAH emission factor for the grass burn (35
316 mg/kg) was higher than for the forest burn (23 and 27 mg/kg) (Table 3), although this was largely
317 due to the difference in the naphthalene emission factor. When naphthalene was excluded the total
318 PAH emission factors were similar for grass field and forest field at 14 mg/kg and 11 mg/kg,
319 respectively.

320 3.3.3 *Laboratory versus Field Sampling*

321 The total PAH emission factor for forest fuel from the OBTF burns was similar to the field data,
322 at 27 ± 3.7 mg/kg and 23 mg/kg, respectively. The benzo(a)pyrene contributed to 62% (on average)
323 of the B[a]P-TEQ emission factor, and fluoroanthene and benzo(b)fluoranthene contributed 17% and
324 5%, respectively. The B[a]P-TEQ emission factor from the field forest burn was lower than from
325 the OBTF, 0.14 mg B[a]P-TEQ/kg and 0.33 mg B[a]P-TEQ/kg, respectively.

326 The naphthalene emission factors from the field burns were slightly lower using the quartz
327 filter/XAD method rather than the SUMMA canister method, although some of this difference
328 may be due to different sampling intervals. The SUMMA canisters samples were collected during
329 a short time interval, 1-2 min, representing a segment of the burn process, while the two quartz
330 filter/XAD samples were collected for 40 and 160 min representing the entire burn process. The
331 OBTF naphthalene emission factors were similar for both methods even though the MCE was
332 lower (0.865) during the quartz filter/XAD method sampling duration than during the SUMMA
333 canister method sampling (0.979).

334 Table 3. Results 16 EPA PAHs.

PAH	Grass burn	Forest burn	Forest burn ^a
	Ground	Ground	OBTF
	mg/kg biomass	mg/kg biomass	mg/kg biomass
Naphthalene	22	13	14±2.3 (13±2.3)
Acenaphthylene	4.6	2.9	4.3±0.50 (4.0±0.50)
Acenaphthene	0.34	0.38	0.41±0.087 (0.39±0.081)
Fluorene	1.1	1.4	1.4±0.37 (1.3±0.34)
Phenanthrene	3.7	3.4	3.1±0.57 (2.9±0.52)
Anthracene	0.60	0.63	0.65±0.12 (0.61±0.11)
Fluoranthene	1.1	0.73	0.95±0.13 (0.89±0.12)
Pyrene	0.94	0.62	0.94±0.11 (0.87±0.11)
Benzo(a)anthracene	0.20	0.1	0.29±0.042 (0.28±0.041)
Chrysene	0.27	0.2	0.31±0.042 (0.29±0.039)
Benzo(b)fluoranthene	0.20	0.081	0.16±0.022 (0.15±0.021)
Benzo(k)fluoranthene	0.13	0.052	0.16±0.0046 (0.15±0.0055)
Benzo(a)pyrene	0.20	0.071	0.21±0.022 (0.20±0.021)
Indeno(1,2,3-cd)pyrene	0.13	0.052	0.13±0.011 (0.12±0.011)
Dibenz(a,h)anthracene	ND	0.0048	0.013±0.0033 (0.012±0.0030)
Benzo(g,h,i)perylene	0.13	0.033	0.11±0.0082 (0.10±0.0084)
SUM 16-EPA PAH	35	23	27±3.7 (25±3.6)
SUM PAH B[a]P	3.1E-01	1.4E-01	3.3E-01±3.5E-02

335 ^a 1 STDV. Emission factors derived using carbon from both CO and CO₂ within parentheses.336
337338 **3.4 PCDDs/PCDFs.**339 *3.4.1 Data Comparison*

340 The aerial forest emission factor for PCDD/PCDF (1.2 ng TEQ/kg, Table 1) was higher than
341 that previously measured from the same location/area with the same method (0.63 ng TEQ/kg)
342 (Aurell and Gullett, 2013). This may be due to seasonal differences in composition of the biomass
343 as the previous measurements were in February and these are in November. With 17 assigned
344 toxic equivalent factors (TEFs) the four collected PCDD/PCDF samples had 10-17 detected TEF
345 congeners (SI Table S2). The plume emissions exceeded the raw biomass emissions indicating that
346 the emissions are formed rather than evaporated from the raw biomass.

347 *3.4.2 Aerial versus Ground Sampling*

348 The aerial collected forest burn PCDD/PCDF emission factor was slightly higher than the
349 ground collected sample (1.2 ng TEQ/kg versus 0.80 ng/kg). If this difference is real, it may be
350 due to a higher MCE at the aerial collection point than on the ground, as indicated by the BC/PM_{2.5}
351 ratio discussed above.

352 *3.4.3 Grass versus Forest Burns*

353 The grass burn had a higher PCDD/PCDF emission factor than the forest burn, 1.9 ng TEQ/kg
354 compared to 1.0 ng TEQ/kg (on average), despite higher MCE values. This is contrary to typical
355 results, where increased PCDD/PCDF levels are linked to poor combustion. Differences in the
356 chemical composition of the species may be responsible for this destruction.

357 *3.4.4 Laboratory versus Field Sampling*

358 The OBTF forest burn PCDD/PCDF emission factor was much lower (0.092 ng TEQ/kg) than
359 from the field (1.2 ng TEQ/kg and 0.80 ng TEQ/kg, Table 1), contrary to previous results (Aurell
360 and Gullett, 2013) from this area. The limited number of samples show little MCE differences;
361 differences may be due to burn intensity but this remains to be explored further.

362

363 **4. CONCLUSION**

364 Sampling of forest and grass prescribed burn emissions was accomplished with both aerial-
365 and ground-based measurements. Field-gathered forest biomass was collected, burned, and
366 sampled for emissions in a laboratory test facility.

367 Emission factors for a comprehensive array of pollutants were determined. PM_{2.5} emission
368 factors from the forest burns were higher than previous values from the same site, higher than at

369 least two other reported wildfires, and in the upper range of published field and laboratory tests.
370 PM_{2.5} results for the grass field burns were consistent with measurements of other species.
371 While optically-measured BC values correlated well with thermally/optically-measured EC, the
372 former were about 1.4 times higher, suggesting caution when using historical EC values to
373 represent BC. TC and OC measurements appeared to be lower in aerial measurements than ground
374 measurements, suggesting further reactions above ground-level samplers or a bias in the gases
375 measured at ground vs air levels.

376 Laboratory burns of forest biomass in the OBTF showed some emission factor differences
377 from the field measurements which are not necessarily explained by differences in the MCE
378 values. High BC values, associated with flaming combustion, are noted for the OBTF, consistent
379 with the higher MCE values. BrC concentrations, too, were associated with flaming combustion
380 and CO₂ evolution. As the combustion intensity declines and CO rises, BC and BrC decrease,
381 followed by a lagging peak of PM_{2.5}. Laboratory measurements yielded lower PM_{2.5} and OC
382 values than field measurements, whereas the reverse was true for EC. Lower VOC concentrations
383 were noted in the laboratory burns; concentrations between field and lab measurements indicated
384 good inverse correlations (e.g., $r^2 = 0.7 - 0.9$) with MCE.

385 Comparison of aerial- versus ground-based sampling showed differences in carbon
386 measurements; aerostat-based sampling yielded about twice the BC and EC concentrations of
387 ground sampling. This was true only for the forest burns and not the grass burns, however.
388 Whether this is due to a mechanistic difference or a sampling bias is not clear and will require
389 additional measurements.

390 Little difference was apparent in the PAH data between forest and grass, aerial and ground,
391 and field and laboratory burns. Naphthalene was the most prevalent PAH observed. PCDD/PCDF

392 emission factors were similar for grass and forest burns and aerial and ground sampling but
393 significantly lower for the OBTF. This is likely related to the higher MCEs observed in the OBTF,
394 as PCDDs/PCDFs are linked to marginal combustion conditions.

395

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407

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