Atmospheric Environment 150 (2017) 395-406

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Emissions from prescribed burning of timber slash piles in Oregon



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HIGHLIGHTS

G R A P H I C A L A B S T R A C T

- Dry biomass piles burned with higher combustion efficiency than wet piles.
- Piles that had been covered with polyethylene had lower emissions than wet piles.
- Burning the polyethylene cover on the pile had no distinctive effect on emissions.

A R T I C L E I N F O

Article history: Received 16 August 2016 Received in revised form 10 November 2016 Accepted 11 November 2016 Available online 12 November 2016

Keywords: Emission factors Timber slash Pile cover Moisture Polyethylene Biomass burning



ABSTRACT

Emissions from burning piles of post-harvest timber slash (Douglas-fir) in Grande Ronde, Oregon were sampled using an instrument platform lofted into the plume using a tether-controlled aerostat or balloon. Emissions of carbon monoxide, carbon dioxide, methane, particulate matter (PM_{2.5}), black carbon, ultraviolet absorbing PM, elemental/organic carbon, filter-based metals, polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzodioxins/dibenzofurans (PCDD/PCDF), and volatile organic compounds (VOCs) were sampled to determine emission factors, the amount of pollutant formed per amount of biomass burned. The effect on emissions from covering the piles with polyethylene (PE) sheets to prevent fuel wetting versus uncovered piles was also determined. Results showed that the uncovered ("wet") piles burned with lower combustion efficiency and higher emission factors for VOCs, PM_{2.5}, PCDD/PCDF, and PAHs. Removal of the PE prior to ignition, variation of PE size, and changing PE thickness resulted in no statistical distinction between emissions. Results suggest that dry piles, whether covered with PE or not, exhibited statistically significant lower emissions than wet piles due to better combustion efficiency.

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1. Introduction

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http://dx.doi.org/10.1016/j.atmosenv.2016.11.034 1352-2310/Published by Elsevier Ltd. To reduce wildfire risk and to improve timber forest productivity and health, woody biomass fuels from selective thinning and timber harvests are mechanically treated and piled for burning (Cross et al., 2013; Trofymow et al., 2014). This practice is becoming more prevalent, particularly in the Pacific Northwest, as prescribed fire complexity and risk associated with elevated fuel levels





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(proximity to the wildland/urban interface, smoke effects on air quality and respiratory health) limit the use of broadcast prescribed burning (Wright et al., 2010). Pile burning mitigates concerns about fire safety and air quality as it allows managers to burn under optimal weather conditions and with reduced staffing levels (Wright et al., 2010). Biomass pile burns are often the most economical way to dispose or utilize the biomass due to collection. transportation, and end-product processing costs (Springsteen et al., 2011). Depending on the season and rainfall history, burn piles can smolder for days after they are lit resulting in significant quantities of air pollution (Springsteen et al., 2011). To promote pile combustion, the biomass is preferably dry, resulting in faster, hotter, and more efficient burns, presumably with less pollutants. Common practice involves covering these large piles with polyethylene (PE) film until burn conditions are optimal to prevent moisture saturation during the rainy season. This has raised some questions about emissions from the burning plastic film. The Oregon Department of Forestry (ODF) has used small amounts of PE film sheeting (9.3 m^2) per pile through administrative rulemaking (OAR 629-048-0210) (Oregon Department of Forestry (2014)). Often this is not enough to keep piles dry for efficient consumption after significant rainfall. Because of this limitation, ODF is seeking data to determine whether or not larger and thicker coverings of PE have deleterious effects on burn emissions.

Only a few studies (Hardy, 1996) have investigated pile burn emissions in the field and often the number of pollutants characterized was limited (Hardy, 1996; Ward et al., 1989). Laboratory burns of *pinus ponderosa* slash (twigs, needles, and small branches) by Yokelson et al. (1996) characterized emissions from burn piles (1 m \times 2 m) using FTIR analysis. Their work determined emission factors for smoldering/flaming phase as partitioned by modified combustion efficiency. Other work (Hosseini et al., 2014) examined emissions from 2 kg mixtures of manzanita wood (*Arctostaphylos* sp.) with 0, 5, and 50 g of shredded low density PE but found no statistical effect of increase PE content on over 190 compounds.

To complement the laboratory scale work previously done on assessing potential contribution of PE to biomass emissions, this work aimed to characterize and compare emissions from burning woody biomass piles, including dried PE-covered piles and wetted piles, in a large-scale field application.

2. Methods

2.1. Biomass piles

Tests were conducted during mid-October in western Oregon, on a timber-harvested Douglas fir (Pseudotsuga menziesii) stand (45° 0' 44.14'' N, -123° 41' 6.49'' W) located about 8 km southwest of Grand Ronde, Oregon and 30 km east of the Pacific coast. The site was at 880 m elevation on a ridge top with an about 10 m change in elevation in the test area. After timber harvesting, the piled material was primarily small branches and limbs of size less than 20 cm in diameter.

Biomass piles approximately 2.5 m high and 5 m in diameter and spaced at least 15 m apart were constructed by the landowner (Fig. 1). Three pile types were tested nominally: Dry, Wet, and Dry Polyethylene (PE) covered. Polyethylene sheeting covered eight of the piles throughout the summer to comprise the Dry and PEcovered test piles for the October tests. The PE was removed from four piles prior to testing and were designated Dry piles. The remaining four covered piles were left with the PE in place and were designated Dry PE piles. PE-covered piles had two film thicknesses, 0.10 mm (4 mil) and 0.15 mm (6 mil), and two area sizes, 3.0 m by 3.0 m (10 ft by 10 ft), and 6.1 m by 6.1 m (20 ft by 20 ft) (Table 1). The remaining four piles were uncovered



Fig. 1. Typical burn pile, uncovered.

Table 1 Test order and type

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Test day	Test order, Type, PE size ^a (if applicable)
Day 1	Burn 1: WET 01
	Burn 2: DRY, PE 6.1 \times 6.1 m, 0.15 mm
Day 2	Burn 3: WET 02
-	Burn 4: DRY, uncovered
	Burn 5: DRY, PE 3 \times 3 m, 0.15 mm
Day 3	Burn 6: WET 03
-	Burn 7: DRY, uncovered
	Burn 8: DRY, PE 3 \times 3 m, 0.10 mm
	Burn 9: DRY, uncovered
Day 4	Burn 10: DRY, PE 6.1 × 6.1 m, 0.15 mm
-	Burn 11: DRY, PE 3 × 3 m, 0.15 mm
	Ambient background

^a PE = Polyethylene, area in m x m, thickness in mm.

throughout the summer and designated as Wet piles. Air emissions were only collected from three of these Wet piles, the fourth pile was used to check plume height for best collection efficiency prior to emission sampling.

Terrain constraints to pile access, a desire to prevent the emissions from upwind smoldering fires from impinging on new burn piles, and effects of week-long meteorological conditions prohibited true random pile testing. The resultant "ordered" testing affects randomness and may have introduced bias into the measurements as a result of dynamic meteorological variables (conditions present at the end of the testing may be different than those at the beginning) confounding the comparisons. Four days of sampling were conducted in later October. Meteorological data for these dates are reported in Supporting Information (SI). The order and notation for the tests are presented in Table 1.

2.2. Sampling method

Fires were initiated by drip torch immediately after which emissions were sampled using an aerostat-lofted sampler system (Fig. 2) detailed more fully elsewhere (Aurell and Gullett, 2013; Aurell et al., 2011). Briefly, the system consists of a 5 m diameter, helium-filled aerostat, connected with two tethers to all-terrain vehicle (ATV)-mounted winches, upon which is mounted a sampler/sensor system termed the "Flyer". The Flyer was



Fig. 2. Aerostat with Flyer (Left) and Flyer close up (Right).

maneuvered into the burn pile plume by controlling tether length and the location of the ATV-mounted tether winches. Sampling periods consisted of both active flaming and smoldering emissions.

2.3. Instrumentation on the Flyer

Emission samples were analyzed for carbon monoxide (CO), methane (CH₄), carbon dioxide (CO₂), particulate matter equal to or less than 2.5 μ m (PM_{2.5}), black carbon (BC), ultraviolet absorbing (UVPM), elemental/organic/total carbon (EC, OC, TC), polyaromatic hydrocarbons (PAHs), polychlorinated dibenzodioxins/dibenzofurans (PCDDs/PCDFs), filter-based metals, and volatile organic compounds (VOCs). Targeted emission constituents and their sampling methods are listed in Table 2.

The Flyer was equipped with a data acquisition and control program allowing emission samplers to be turned on and off at CO_2 levels above ambient levels (trigger settings: 800 ppm for VOCs and 450 ppm CO_2 for all other emission samplers). The control program data were also transmitted to the ground permitting the operator full control of the emission samplers.

The CO₂ analyzer and the CO sensor were calibrated daily in accordance with EPA Method 3A (1989). A precision gas divider Model 821S (Signal Instrument Co. Ltd., England) was used to dilute the high-level span gases for acquiring the mid-point concentrations for CO₂ analyzer and CO sensor calibration curves. The precision gas divider was evaluated in the field as specified in U.S. EPA

Table 2

Target pollutants and sampling methods.

Method 205 (2014). The PM_{2.5} and EC/OC/TC sample pumps as well as the AE51/AE52 were calibrated with a Gilibrator Air Flow Calibration System (Sensidyne LP, USA) before and after the field campaign. SUMMA canisters were equipped with a manual valve, metal filter (frit), pressure gauge, pressure transducer, and an electronic solenoid valve which enabled the SUMMA to be opened remotely by the ground-based software to maximize desired sample collection and minimize sampling of ambient air.

PCDD/PCDF samples were cleaned and analyzed using an isotope dilution method based on U.S. EPA Method 23 (1991). Concentrations were determined using high resolution gas chromatography/high resolution mass spectrometry (HRGC/HRMS) with a Hewlett-Packard gas chromatograph 6890 Series coupled to a Micromass Premier mass spectrometer (Waters Corp., Milford, MA, USA) with an RTX-Dioxin 2, 60 m \times 0.25 mm \times 0.25 μm film thickness column (Restek Corp., Bellefonte, PA, USA). For analysis of tetra- through octa-CDDs/Fs, Method 8290A (U.S. EPA Method 8290A, 2007) was followed. The standard used for PCDD/PCDF identification and quantification is a mixture of standards containing tetra- to octa-PCDD/F native and 13C-labeled congeners designed for modified U.S. EPA Method 23 (1991). Not all of the seventeen PCDD/PCDF toxic equivalent factor (TEF) weighted congeners were detected in all samples. The congeners that were not detected (ND) were set to zero in the text, however SI Tables S6–S9 show values both ND = 0 and ND = limit of detection (LOD). The PCDD/PCDF toxic equivalent (TEQ) emission factors were

Analyte	Method/Instrument	Frequency	Method reference
CO ₂	NDIR LICOR-820 ^a	Continuous 1 Hz	(U.S. EPA Method 10A)
CO	Electrochemical cell e2V EC4-500-CO ^b	Continuous 1 Hz	(U.S. EPA Method 10A)
PM _{2.5}	SKC Impactor, 47 mm filter 2 µm pore size/	Batch — 10 L/min ^c constant	40 CFR Part 50 (1987)
	gravimetric	flow	
PM _{2.5}	DustTrak 8520 ^d	Continuous 1 Hz	Laser optical, factory calibration
PCDD/PCDF/PAHs	Quartz filter/PUF/XAD/PUF ^e	Batch – 650 L/min nominal	U.S. EPA Compendium Method TO-9A (1999)
		flow ^f	
VOCs	6 L SUMMA canister	30-60 min integrated sample	U.S. EPA Compendium Method TO-15 (1999)
CO, CO_2, CH_4	6 L SUMMA canister	30-60 min integrated sample	(U.S. EPA Method 25C)
Black carbon	Aethalometer, AE51 ^g /AE52 ^g	Continuous 1 Hz/0.1 Hz	880 nm by light absorption, factory calibration
UVPM	Aethalometer, AE52 ^g	Continuous 0.1 Hz	370 nm by light absorption, factory calibration
Elemental, organic and Total	SKC Impactor, 47 mm quartz filter	Batch – 10 L/min ^c constant	Modified NIOSH Method 5040 (Khan et al.,
carbon		flow	2012)

^a LI-COR Biosciences, USA.

^b SGX Sensortech, United Kingdom.

^c Leland Legacy sample pump, SKC Inc., USA.

d TSI Inc., USA.

- ^e Filter size 20.3 \times 25.4 cm, Polyurethane foam (PUF) size 7.6 \times 3.8 cm.
- ^f Windjammer brushless direct current blower AMETEK Inc., USA.

g AethLabs, USA.

determined using the World Health Organization (WHO) 2005 toxic equivalent factors (TEF) (Van den Berg et al., 2006). Only four PCDD/PCDF congeners were detected in all samples; (1,2,3,4,6,7,8 -HpCDD, 1,2,3,4,6,7,8,9 - OCDD, 2,3,7,8 - TCDF, 1,2,3,4,6,7,8 -HpCDF) these emission factors were used for intercomparison purposes. These emission factors represent the low end of the absolute emission factor but are the most reliable for intercomparison.

A portion of the methylene chloride extract from the PCDD/ PCDF/PAH sample was used for the PAH analysis using a modified EPA Method 8270D (2007). Labeled standards for PAHs were added to the XAD-2 trap before the sample was collected and internal standards were added before mass analysis. The PAHs TEQ emission factors were determined using TEFs by Larsen and Larsen (1998).

Ambient air background samples were collected for each of the target pollutants. Only the VOC emissions were background corrected. PCDD/PCDF, PAH and PM burn samples had over 20, 170, and 200 times higher concentrations than the ambient air background sample, respectively.

2.4. Calculations

Emission factors, expressed as mass of pollutant per mass of biomass burned, were based on the carbon balance method (Nelson, 1982). This method concurrently measures the target analyte along with the amount of fuel burned, the latter assumed to be determined by the ΔCO_{2} measurements and assuming a 50% carbon concentration in the biomass fuel. The minor carbon mass emitted as hydrocarbons and PM is ignored without significant effect on the emission factor. The resultant emission factors are expressed as mass of pollutant per mass of biomass consumed (B_c) .

The modified combustion efficiency (MCE), $\Delta CO_2/(\Delta CO_2 + \Delta$ $CO+\Delta CH_4$) (with CH_4 included in VOC samples only), was calculated for each of the emission samples.

Custom photometric calibration factors were derived for each burn conducted for the DustTrak 8520 by simultaneous collection

Ta	bl	e	3
Re	sι	ılt	s.

of PM _{2.5} mass on a filter (averaged	continuous	PM _{2.5}	concentration
divided by PM _{2.5} by filter	r mass).			

Single factor one-way analysis of variance (ANOVA) with a level of significance $\alpha = 0.05$ was used to determine any differences in air pollution emissions between PE covered and uncovered biomass piles. To establish significant difference the ANOVA-returned p value (significant value) has to be less than level of significance (0.05) and the F/F_{crit} value has to be greater than 1.0.

3. Results and discussion

Eleven pile burns were sampled over a five day period with emission factor results summarized in Table 3. The plumes were sampled with the aerostat/Flyer in close proximity to the fires to maximize the sample collection mass without placing operators or instruments at risk. Typical aerostat heights above the pile burn were 20-70 m. Pile emission sampling averaged 45 min. Ambient temperatures ranged from 2 to 13 °C, winds 0-32 km/h, and humidity ranged from 100% for the first two days of testing to 35–40% on the last two days. Additional meteorological data are presented in the Supporting Information.

The potential effect of day-of-testing on the results due to, for example meteorological condition changes through the week, were examined by the chronological examination of the emission factors for all targeted pollutants. This analysis is of limited utility due to the non-random order in which the tests were run. Nonetheless, no effects related to testing date, or time of day were found on the Wet/Dry PM_{2.5}, PAH, and PCDD/PCDF emission factors were found including the Dry PE PCDD/PCDF results. However, an effect of the testing date was found for Dry PE on the PM_{2.5} emission factors and was inconclusive on the PAH results.

3.1. CO, CH_4 , and CO_2

Typical concentration results throughout the duration of a Dry and Wet burns are shown in Fig. 3. Fluctuations in the

$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	Pollutant	Unit	WET	DRY	DRY PE	DRY PE	DRY PE
$\begin{array}{cccccc} CO_2^{e} & g/kg \ B_c & 1689 \ (36\%)^b & 1785 \ (3.1\%)^c & 1,758^d & 1,795^d & 1,756^d \\ CO^e & g/kg \ B_c & 82 \ (20\%)^b & 29 \ (112\%)^c & 43^d & 22^d & 46^d \\ CH_4^e & g/kg \ B_c & 5.7 \ (2.1\%)^b & 1.1 \ (135\%)^c & 2.6^d & 1.5^d & 2.0^d \\ PM_{2.5} & g/kg \ B_c & 18 \ (58\%)^b & 4.5 \ (9.5\%)^b & 6.0 \ (78\%)^c & 5.2 \ (69\%)^c & 3.4^d \\ BC & g/kg \ B_c & 0.47 \ (12\%)^c & 0.24 \ (5.7\%)^b & 0.27 \ (38\%)^c & 0.28 \ (29\%)^c & 0.28^d \\ UVPM & g/kg \ B_c & 0.50^d & 0.24 \ (6.9\%)^c & NS & 0.30^d & NS \\ EC & g/kg \ B_c & 0.18 \ (10\%)^c & 0.12 \ (18\%)^b & 0.10 \ (12\%)^c & 0.14 \ (16\%)^c & 0.13^d \\ OC & g/kg \ B_c & 8.3 \ (9.5\%)^c & 2.6 \ (21\%)^b & 3.6 \ (110\%)^c & 2.7 \ (73\%)^c & 1.9^d \\ OC & g/kg \ B_c & 8.5 \ (9.5\%)^c & 2.6 \ (21\%)^b & 3.6 \ (110\%)^c & 2.7 \ (73\%)^c & 1.9^d \\ OC/EC & Ratio & 0.043 \ (60\%)^c & 0.053 \ (9.2\%)^b & 0.045 \ (1.2\%)^c & 0.066 \ (94\%)^c & 0.081^d \\ EC/PM_{2.5} & Ratio & 0.015 \ (39\%)^c & 0.027 \ (22\%)^b & 0.019 \ (67\%)^c & 0.030 \ (55\%)^c & 0.038^d \\ EVCs^d & mg/kg \ B_c & 88 \ (10\%)^b & 15 \ (27\%)^b & 26 \ (118\%)^c & 24 \ (109\%)^c & 1.255^d \\ \Sigma \ PAH_{16} & mg/kg \ B_c & 88 \ (10\%)^b & 15 \ (27\%)^b & 26 \ (118\%)^c & 24 \ (10\%)^c & 14^d \\ \end{array}$				Uncovered	6.1 × 6.1 m 0.15 mm	3 × 3 m 0.15 mm	$3 \times 3 \text{ m}$ 0.10 mm
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	$\begin{array}{c} CO_2^{e} \\ CO_2^{e} \\ CO_2^{e} \\ CD_2^{e} \\ PM_{2.5} \\ BC \\ UVPM \\ EC \\ OC \\ TC \\ OC/EC \\ BC/PM_{2.5} \\ EC/PM_{2.5} \\ EC/PM_{2.5} \\ \Sigma \ VOCs^{f} \\ \Sigma \ PAH_{16} \\ \Sigma \ PAH - TEQ \\ \Sigma \ PCDD/PCDF \\ \Sigma \ PCDD/PCDF \\ EQ^{g} \\ \end{array}$	g/kg B _c g/kg B _c Ratio Ratio Ratio Ratio Ratio mg/kg B _c mg/kg B _c mg B [a]P _{eq} /kg B _c ng/kg B _c ng TEQ/kg B _c	$\begin{array}{c} 1689\ (36\%)^{\rm b}\\ 82\ (20\%)^{\rm b}\\ 5.7\ (2.1\%)^{\rm b}\\ 18\ (58\%)^{\rm b}\\ 0.47\ (12\%)^{\rm c}\\ 0.50^{\rm d}\\ 0.18\ (10\%)^{\rm c}\\ 8.3\ (9.5\%)^{\rm c}\\ 45\ (0.6\%)^{\rm c}\\ 0.043\ (60\%)^{\rm c}\\ 0.015\ (39\%)^{\rm c}\\ 4106\ (50\%)\\ 88\ (10\%)^{\rm b}\\ 2.7\ (11\%)^{\rm b}\\ 15\ (37\%)^{\rm b}\\ 0.18\ (11\%)^{\rm b}\\ 0.18\ (11\%)^{\rm b}\\ \end{array}$	$\begin{array}{c} 1785 \ (3.1\%)^c\\ 29 \ (112\%)^c\\ 1.1 \ (135\%)^c\\ 4.5 \ (9.5\%)^b\\ 0.24 \ (5.7\%)^b\\ 0.24 \ (6.9\%)^c\\ 0.12 \ (18\%)^b\\ 2.5 \ (22\%)^b\\ 2.6 \ (21\%)^b\\ 2.1 \ (32\%)^b\\ 0.053 \ (9.2\%)^b\\ 0.027 \ (22\%)^b\\ 612 \ (95\%)^c\\ 15 \ (27\%)^b\\ 0.27 \ (32\%)^b\\ 0.27 \ (32\%)^b\\ 0.077 \ (32\%)^b\\ 0.077 \ (59\%)^b\\ \end{array}$	$\begin{array}{c} 1,758^{\rm d} \\ 43^{\rm d} \\ 2.6^{\rm d} \\ 6.0 (78\%)^{\rm c} \\ 0.27 (38\%)^{\rm c} \\ NS \\ 0.10 (12\%)^{\rm c} \\ 3.5 (112\%)^{\rm c} \\ 3.6 (110\%)^{\rm c} \\ 34 (104\%)^{\rm c} \\ 0.045 (1.2\%)^{\rm c} \\ 0.019 (67\%)^{\rm c} \\ 1.266^{\rm d} \\ 26 (118\%)^{\rm c} \\ 0.48 (123\%)^{\rm c} \\ 0.48 (123\%)^{\rm c} \\ 8.0 (137\%)^{\rm c} \\ 0.14 (192\%)^{\rm c} \end{array}$	$\begin{array}{c} 1.795^{\rm d} \\ 22^{\rm d} \\ 1.5^{\rm d} \\ 5.2 \ (69\%)^{\rm c} \\ 0.28 \ (29\%)^{\rm c} \\ 0.30^{\rm d} \\ 0.14 \ (16\%)^{\rm c} \\ 2.5 \ (76\%)^{\rm c} \\ 2.7 \ (73\%)^{\rm c} \\ 17 \ (62\%)^{\rm c} \\ 0.066 \ (94\%)^{\rm c} \\ 0.030 \ (55\%)^{\rm c} \\ 1.036^{\rm d} \\ 24 \ (109\%)^{\rm c} \\ 0.55 \ (100\%)^{\rm c} \\ 7.6 \ (145\%)^{\rm c} \\ 0.066 \ (189\%)^{\rm c} \end{array}$	$\begin{array}{c} 1.756^{d} \\ 46^{d} \\ 2.0^{d} \\ 3.4^{d} \\ 0.28^{d} \\ NS \\ 0.13^{d} \\ 1.8^{d} \\ 1.9^{d} \\ 1.9^{d} \\ 14^{d} \\ 0.081^{d} \\ 0.038^{d} \\ 1.255^{d} \\ 14^{d} \\ 0.24^{d} \\ 5.1^{d} \\ 0.057^{d} \\ \end{array}$

^a Units in mass of pollutant per mass of biomass consumed (B_c). NS = No sample. Relative standard deviation (RSD) and relative percent difference (RPD) within parentheses. ^b RSD.

^c RPD.

^d Single sample. Derived from SUMMA Canisters.

^f Sum of 74 VOCs analyzed via U.S. EPA Compendium Method TO-15 (1999).

^g Not detected congeners set to zero, results for each congener and homologue is presented in SI Tables S5–S10.

^h For intercomparison purpose only, PCDD/PCDF congeners detected in all samples: 1,2,3,4,6,7,8 – HpCDD, 1,2,3,4,6,7,8,9 – OCDD, 2,3,7,8 – TCDF, 1,2,3,4,6,7,8 – HpCDF.



Fig. 3. Typical concentration traces of CO₂, CO, BC, PM_{2.5} and modified combustion efficiency (MCE) for Dry and Wet burns. Traces displayed in 60 s moving average.

concentrations are typical and reflect wind shifts moving the Flyer in an out of the plume. The CO and CH₄ emission factors were almost twice as high for the wet piles as the dry (Table 3). Hardy (1996) estimated 1.64 and 5.52 g/kg for CH₄ from flaming and smoldering, respectively. Our work resulted in whole-burn values of 1.1 g/kg (DRY) to 5.7 g/kg (WET). The CO₂, CO and CH₄ emission factors in this study were also in the same range as found in the literature of open burning of Douglas-fir 1601–1772 g/kg, 74–138 g/ kg, 0.3–7.9 g/kg^{26, 27}, respectively.

3.2. PM_{2.5}

The PM_{2.5} results show a statistically significant (F = 2.7, p = 0.004) increase in the Wet (18 ± 11 g/kg B_c) versus the Dry uncovered + Dry PE covered (4.9 ± 1.8 g/kg) emission factor (Fig. 4 Inset). Individual emission factors (Fig. 4) show no distinction between the Dry uncovered and Dry PE covered piles. The PM_{2.5} emission factors compare with a value of 6.75 g/kg consumed estimated from hand-pile biomass burns by Wright et al. (2010). The Wet emission factor (18 ± 11 g/kg B_c) derived at a MCE of 0.839 ± 0.057 is in the same range as found in the literature of open burning of Douglas-fir, 15.7 ± 5.2 g/kg dry fuel consumed (Urbanski et al., 2009) at a MCE of 0.916 ± 0.016.

Examination of the relationship between $PM_{2.5}$ and the MCE showed that lower combustion efficiencies were correlated with higher $PM_{2.5}$ loads. Fig. 5 shows that comparison of same-day WET and DRY samples (Day 2 and Day 3) continues to verify the

distinction with the passage of time, suggesting that the nonrandom testing did not affect the conclusions. The distinction in the PM_{2.5} emission factors occurs in the initial half of the burns. Fig. 6 shows that the early portion of the WET pile burns when the fire is getting started is responsible for the high PM_{2.5} emissions. This distinction with the DRY burns persists until the second half of the burn when smoldering was more prevalent.

3.3. Black carbon, UVPM, elemental/organic carbon

BC, EC, OC, and TC values were all higher for the WET burns as compared to all of the DRY and PE burns (Fig. 7). No statistical distinctions in these values (BC, EC, OC, and TC emission factors) were observed for the varying sizes and thicknesses of PE. BC showed approximately a factor of two higher values than EC and they did not correlate strongly with each other (r^2 of 0.49, SI Fig. S1) which may be due to the low number of data points. The EC emission factor, 0.10–0.18 g/kg B_c, is in the same range as found in the literature, 0.19 \pm 0.41 g/kg dry fuel, from laboratory burns of Douglas-fir (McMeeking et al., 2009). The relationship between EC and BC emission factors and MCE is shown in Fig. 8.

The OC/EC values, a surrogate for comparison of optical reflectance/warming properties, indicates values ranging between 14 and 45, the latter being the WET burns (Table 3). Values greater than unity are common and anticipated for biomass burns. These values are the opposite of what is observed with, for example, crude oil combustion (Gullett et al., 2016), where the OC/EC ratio is about 1/15.



Fig. 4. PM_{2.5} results. Inset chart shows Wet versus DRY (PE-covered and uncovered). Error bars represents 1 standard deviation if nothing else stated.



Fig. 5. The relationship between PM_{2.5} emission factor and combustion quality (modified combustion efficiency, MCE).



Fig. 6. Comparison of PM_{2.5} emission factors at 4 min intervals throughout the burn durations, comparing the combined WET and combined DRY results.



Fig. 7. BC, EC, UVPM, OC and TC results. Inset chart shows Wet versus DRY (PE-covered and uncovered). Error bars represents absolute difference if nothing else stated.



Fig. 8. BC and EC in relationship to modified combustion efficiency (MCE).

3.4. Volatile organic compounds (VOCs)

VOC results for the most concentrated species are shown in Table 4. The full set of VOC emission factors are summarized in Supporting Information, Tables S11–S13. ANOVA analysis (Fig. 9) of acrolein, benzene, styrene and 1,3-butadiene showed statistical differences between WET and DRY piles, (Benzene F = 1.6, p = 0.0208; Acrolein F = 3.3, p = 0.004; Styrene F = 1.9, p = 0.015; 1,3-Butadiene F = 1.4, p = 0.026). Benzene is a common VOC associated with incomplete combustion. Acrolein is a toxic, irritant, 3-C carbonyl and is not listed as a carcinogen on EPA or international lists. 1,3-butadiene is listed as a human carcinogen. Styrene is "reasonably anticipated to be a human carcinogen" (The U.S. Department of Health and Human Services Public Health Service (2011)). The relationship between emission factors for these select VOCs and MCE is shown in Fig. 10.

3.5. PCDD/PCDF

Results for PCDD/PCDF emission factors for Dry, Wet, and PE are summarized in Table 3. Fig. 11 presents data for four of the 17 congeners that comprise the PCDD/PCDF TEQ value (Van den Berg et al., 2006) that were present in all 11 samples (complete data are shown in SI Tables S5–S10). As such, these emission factors represent the low end of the absolute emission factor but are the most reliable in terms of intercomparisons. Wet PCDD/PCDF values are higher than Dry uncovered piles [F = 2.0, p = 0.017]. Dry and PE values show no statistical difference between them [F = 0.01, p = 0.814]. Within the PE grouping, no distinction was observed between the PE sheet size and thickness, although the limited number of tests limits the statistical power of this test.

Fig. 12 examines the effect of combustion quality as measured by MCE on the PCDD/PCDF emission factors. Three distinct groupings of emission factors for Dry, Wet, and PE are indicated. While Wet results show no apparent trend with MCE, PE results suggest that PCDD/PCDF emission factors decline with increased MCE ($r^2 = 0.93$). This is similar to observations for both PM_{2.5} and select VOCs. Evaluation of the whole data set shows an $r^2 = 0.82$ with declining emission factor and MCE. Additional data are necessary to verify these MCE indications, although this trend is consistent with historical observations that equate improved combustion conditions with decreased PCDD/PCDF emissions.

These four-congener PCDD/PCDF emission factors are approximately ten times lower than four-congener literature values of 0.11-0.22 ng TEQ/kg B_c from open burning of pine savannas (Aurell and Gullett, 2013; Aurell et al., 2015).

Table 4	
VOC result	

Compound	WET ^a	DRY uncovered ^b	DRY PE 3 \times 3 m	DRY PE 3 \times 3 m	DRY PE 6.1×6.1
			0.10 mm	0.15 mm	0.15 mm
	mg/kg biomass c	ronsumed			
Benzene ^c	757 ± 416	115 (74%)	216	289	222
Propene	682 ± 373	119 (107%)	252	199	250
Acetone	668 ± 280	32	163	78	ND
Acrolein ^c	463 ± 168	97 (101%)	134	99	180
Vinyl Acetate ^c	309 ± 133	52 (116%)	78	51	134
Toluene ^c	297 ± 172	52 (109%)	100	98	116
1,3-Butadiene	231 ± 136	31 (100%)	78	71	74
2-Butanone (MEK)	156 ± 76	27 (137%)	49	21	72
Styrene ^c	111 ± 59	16 (104%)	25	33	35
Acetonitrile	69 ± 40	17 (119%)	34	12	38
m,p-Xylenes ^c	68 ± 41	13 (136%)	22	15	27
Ethylbenzene	43 ± 26	7.5 (107%)	14	12	15
alpha-Pinene	41 ± 31	8.7 (120%)	17	17	14
D-Limonene	31 ± 21	6.7 (117%)	8.7	12	13
Acrylonitrile ^c	27 ± 14	6.0 (50%)	12	7.0	11
o-Xylene ^c	23 ± 14	4.4 (145%)	8.0	4.5	9.1
1,2,4-Trimethylbenzene	12 ± 5.8	2.4 (143%)	3.8	1.9	4.2
1.3.5-Trimethylbenzene	3.5 + 1.6	1.2	1.2	0.49	1.2

^a Range of data equal one standard deviation.

^b Range of data equals relative percent difference.

^c On U.S. EPA's list of hazardous air pollutants (2008). The VOCs shown here were selected based on the number of samples detectable above three times the detection limit and their relevance to the EPA's list of hazardous air pollutants list and their role as greenhouse gas/ozone precursors. Full list of the 74 analyzed VOCs and their emission factors are presented in SI Tables S11–S12.



Fig. 9. VOC results. Error bars represent one standard deviation for WET burns and DRY combined burns, and absolute difference for DRY uncovered burns. * = On U.S EPA's list of hazardous air pollutants.

3.6. PAHs

Individual PAH emission factors (for the 16 EPA PAHs) are shown in Table 5 and Sum of the 16 EPA PAHs are shown in Fig. 13. Similar to observations of PM_{2.5}, select VOCs, and PCDD/PCDF, Wet piles resulted in greater emissions (statistically significant, F = 14.3, p < 0.0001), by a factor of 4–5. No distinction was observed, however, between any of the Dry (cover and uncovered) PAH emission factors. These emission factors compared to a value of 28 mg/kg burning Douglas-fir in a laboratory setting (Jenkins et al., 1996).

The PAH measurements reflect both gas phase and particlebound PAH compounds. The relationship between the emission factors for $PM_{2.5}$ and PAHs were examined in Fig. 14. Predictably higher $PM_{2.5}$ is associated with higher PAHs.

The relationship between PAHs and combustion quality (MCE) is shown in Fig. 15. As with previous emissions, lower combustion quality (MCE) is associated with higher PAH emissions. All of the Wet results have the lowest MCE and highest PAH levels.

4. Comparison with others' data

Comparison of our results with previously compiled data on open pile burning of woody biomass from twelve sources (Springsteen et al., 2011) places our data within the range of reported results. Literature values for PM (total) ranged from 3 to



Fig. 10. The effect of modified combustion efficiency (MCE) on select VOC emission factors.



Fig. 11. PCDD/PCDF emission factors in ng TEQ/kg biomass consumed. Error bars represent 1 standard deviation if nothing else stated.



Fig. 12. PCDD/PCDF emission factors in ng TEQ/kg biomass consumed by group versus MCE.

22 g/kg dry biomass burned whereas our results were 3-18 g/kg B_c (these units are similar but derived differently). Likewise, reported

CO emission factors were 17-164 g/kg in comparison to our results of $22-82 \text{ g/kg } B_c$. CH₄ values were reported at 0.9-11 g/kg versus

Table 5

PAH	emission	factors
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PAHs	WET ^a	DRY ^a	DRY PE ^b	DRY PE ^b	DRY PE ^c
		Uncovered	6.1 × 6.1, 6 mm	3×3 , 6 mm	$3 \times 3, 4 \text{ mm}$
	mg/kg biomass coi	nsumed			
Naphthalene	17 (3.4%)	4.4 (37%)	8.1 (101%)	7.4 (109%)	5.0
Acenaphthylene	16 (14%)	2.5 (24%)	4.6 (129%)	4.1 (106%)	2.3
Acenaphthene	1.6 (21%)	0.34 (24%)	0.60 (135%)	0.46 (117%)	0.27
Fluorene	6.4 (35%)	0.97 (27%)	1.7 (132%)	1.5 (122%)	0.75
Phenanthrene	19 (20%)	3.3 (26%)	4.8 (128%)	4.5 (113%)	2.5
Anthracene	4.1 (15%)	0.65 (28%)	1.0 (127%)	0.98 (113%)	0.50
Fluoranthene	6.9 (3.4%)	0.90 (30%)	1.4 (117%)	1.6 (107%)	0.76
Pyrene	6.2 (10%)	0.78 (31%)	1.3 (118%)	1.5 (102%)	0.68
Benzo(a)anthracene	2.1 (10%)	0.24 (28%)	0.43 (128%)	0.44 (109%)	0.20
Chrysene	2.5 (10%)	0.38 (24%)	0.61 (123%)	0.58 (111%)	0.30
Benzo(b)fluoranthene	1.3 (14%)	0.13 (28%)	0.24 (123%)	0.25 (102%)	0.11
Benzo(k)fluoranthene	1.7 (6.9%)	0.16 (35%)	0.29 (121%)	0.34 (94%)	0.15
Benzo(<i>a</i>)pyrene	1.7 (12%)	0.16 (33%)	0.29 (124%)	0.34 (98%)	0.14
Indeno (1,2,3-cd)pyrene	0.84 (12%)	0.073 (38%)	0.13 (119%)	0.17 (93%)	0.067
Dibenz (a,h)anthracene	0.20 (14%)	0.021 (28%)	0.037 (126%)	0.041 (102%)	0.022
Benzo (ghi)perylene	0.98 (14%)	0.086 (38%)	0.15 (117%)	0.21 (90%)	0.079
SUM 16-EPA PAH	88 (11%)	15 (27%)	26 (118%)	24 (109%)	13.8

^a Range of data within parentheses equals relative standard deviation.

^b Range of data within parentheses equals relative percent difference.

^c Single sample.



Fig. 13. Average PAH emission factors for each category. Error bars represent 1 standard deviation if nothing else stated.

ours at 1-6 g/kg B_c. Few other pollutants for field pile burns are characterized in the literature.

A laboratory study by Hosseini et al. (2014) looked at emissions from burning forest debris (manzanita) with and without PE addition, showing no effect of the added PE on emissions. Our dry pile results for PM emissions (5.2 \pm 2.4 g/kg B_c) spanned theirs (4.5 \pm 0.43 g/kg biomass). Our OC and benzene results were slightly higher (2.6 \pm 1.3 g/kg B_c and 192 \pm 81 mg/kg B_c, respectively) versus those in the laboratory study (1.7 \pm 0.06 g/kg biomass and 174 \pm 21 mg/kg biomass, respectively). More extensive comparisons are limited by differences in biomass type and MCE (the laboratory burns state a MCE of 0.98–0.99 versus the fields' MCE of 0.86–0.95).

5. Conclusion

Field sampling of eleven biomass pile burns determined emission factors for a wide range of pollutants. Comparison of piles that were naturally wetted versus those that were dry showed statistically higher emission factors for PM_{2.5}, PAHs, VOCs, and PCDD/ PCDF for the wet piles. Emission levels were negatively correlated with combustion quality as represented by MCE. Variation of PE cover size and thickness showed no statistically significant difference in emission factor for any of the pollutants suggesting that the PE was not contributing significantly to any of the measured pollutants. Time-resolved PM_{2.5} emissions were highest at the beginning of the burns; for the Dry pile tests, this startup period lasted



Fig. 15. Comparison of PAH emission factors with modified combustion efficiency (MCE).

for less than 4 min; for the Wet pile tests, it was four times longer, about 16 min. For the Wet pile tests, $PM_{2.5}$ emission factors were higher than those of the Dry pile tests for at least half of the burn durations, after which they were similar. These tests suggest that use of PE as a biomass pile cover results in lower emission factors than those from piles exposed to moisture, reducing pollutant levels during slash pile burns. These emission factors, together with estimates of burn pile numbers, size, and fuel consumption, can be used by management and regulatory communities to minimize smoke impacts while limiting the potential hazard of biomass fuel loading.

Acknowledgements

The authors appreciate the site access and cooperation of Jerry Anderson, the test site manager for Hancock Timber Resources. Jeff Classen and Gail Culbertson, both of the Dallas Unit of Western Oregon ODF District, provided fire duties, transportation, and logistical support. Sean Riordan and Paul Davies (ATA Aerospace) along with Tracy Gerber, US Air Force Research Laboratory (Kirtland AFB) provided aerostat flight operations. Sue MacMillan, Brian Finneran, and Anthony Barnack of the Oregon Department of Environmental Quality provided technical support on toxics and emissions. David Weise (Pacific SW Research Station), Roger Ottmar (Pacific NW Research Station), Shawn Urbanski (Missoula Fire Laboratory), and Harold Merritt (Plum Creek Timber) provided technical support. This work was funded by the Oregon Department of Forestry and the U.S. EPA Office of Research and Development through a Cooperative Research and Development Agreement, #868-15.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2016.11.034.

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