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Characterization of occupational smoke exposure among wildland firefighters in the midwestern United States

Chieh-Ming Wu [a], Chi (Chuck) Song [b], Ryan Chartier [c], Jacob Kremer [d], Luke Naeher [d], Olorunfemi Adetona [a,*]

[a] Division of Environmental Health Sciences, College of Public Health, The Ohio State University, Columbus, OH, United States
[b] Division of Biostatistics, College of Public Health, The Ohio State University, Columbus, OH, United States
[c] RTI International, Research Triangle Park, NC, United States
[d] Department of Environmental Health Science, College of Public Health, University of Georgia, Athens, GA, United States

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ABSTRACT

Wildland firefighters are repeatedly exposed to elevated levels of wildland fire smoke (WFS) while protecting lives and properties from wildland fires. Studies reporting personal exposure concentrations of air pollutants in WFS during fire suppression or prescribed burn activities have been geographically limited to the western and southeastern United States. The objective of this study is to characterize exposure concentrations of air pollutants in WFS emissions among wildland firefighters who conducted prescribed burns in the Midwest. Between 2016 and 2019, a total of 35 firefighters (31 males and 4 females, age of 35.63 ± 9.31 years) were recruited to participate in this study. Personal particulate matter 2.5 ($PM_{2.5}$) and carbon monoxide (CO) exposure concentrations were measured during prescribed burns. The level of black carbon (BC) in WFS particulates was determined using the light transmission technique, while trace metal composition was analyzed using inductively coupled plasma mass spectrometry (ICP-MS). The results showed geometric means for $PM_{2.5}$, CO, and BC concentrations were 1.43 ± 0.13 mg/m^3, 7.02 ± 0.69 ppm, and 58.79 ± 5.46 μg/m^3, respectively. Although no occupational exposure limits (OELs) were exceeded by 8-h time-weighted average (TWA) exposure concentration observed in the firefighters, a total of 28 personal CO exposure concentrations were above the National Institute for Occupational Safety and Health (NIOSH) Recommended Exposure Limit (REL) Ceiling (200 ppm) for CO. $PM_{2.5}$ and CO concentrations were about 2–7 times higher in the Midwest than the other regions. Firefighters who performed holding had higher CO exposure concentrations compared to firefighters who performed lighting ($p < 0.01$), while lighters were exposed to higher level of BC in the smoke particulates ($p < 0.01$), possibly due to the domination of exposure by different combustion sources and stages. The levels of trace metals in WFS particulates were well below the corresponding OELs and no task-related difference was observed except for manganese. Our results suggest that wildland firefighters in the midwestern region have higher WFS exposures while working at prescribed burns compared to those western and southeastern United States.

1. Introduction

In the United States, an average of 6,846,857 acres was burned annually by a corresponding average of 64,072 wildfires between 2008 and 2019 (NIFC, 2019a). According to estimates from the United States Environmental Protection Agency (USEPA), wildland fire smoke (WFS), including wildfires and prescribed burns, contributed approximately 17% and 16% of total annual United States emissions of particulate matter with an aerodynamic diameter less than 2.5 μm ($PM_{2.5}$) and carbon monoxide (CO), respectively, into ambient air in the last decade (USEPA, 2019a). These WFS emissions substantially worsen ambient air quality, potentially result in adverse clinical outcomes, and negatively impact public health (Black et al., 2017; Cascio, 2018; Reid Colleen et al., 2016).

Ironically, the need for the use of prescribed burns for land management has increased as the number and intensity of wildfires have also increased due to climate change (Abatzoglou and Williams, 2016; Johnston et al., 2012; Westerling et al., 2006), past forest conservation

* Corresponding author. Cunz Hall, 1841 Neil Avenue, Columbus, OH, 43210, United States.
E-mail address: adetona.1@osu.edu (O. Adetona).

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practices (Stephens and Ruth, 2005), and the growth of wildland-urban interface (Radeloff et al., 2018). Unlike firefighting for episodic wildfires, wildland firefighters working at prescribed burns intentionally set fires in predetermined forest areas to reduce vegetative fuel load on the forest floor, and thereby lower the probability and severity of wildfires during fire-prone seasons. In the last decade, the annual average number of prescribed burns in the United States increased from 16,882 to 181,542 with a concurrent increase in the number of acres burned from 2,423,862 to 6,058,508 acres (NIFC, 2019b).

Although prescribed burns are thoroughly planned, wildland firefighters are still exposed to elevated levels of WFS, which consists of various pollutants including particulate matter (PM), CO, black carbon (BC), trace metals, and many other compounds that are hazardous to health (Adetona et al., 2013, 2015; Reinhardt and Ottmar, 2004). PM has been recognized as the major inhalation hazard in combustion emissions, including WFS (Naeher et al., 2007). Personal PM exposure concentrations in previous WFS studies are approximately 7–19 fold higher than the 24-hr US National Ambient Air Quality Standard (NAAQS) (35 μg/m^3) and sometimes exceed the Occupational Safety and Health Administration (OSHA) Permissible Exposure Limit (PEL) for respirable PM (5 mg/m^3) (Reinhardt and Ottmar, 2004). In addition, the average length of work shifts reported in the literature is between 4.5 and 12 h (Adetona et al., 2011, 2017; Gaughan et al., 2014; Henn et al., 2019; Slaughter et al., 2004). Their inhalation exposure is potentially worsened by high physical exertion, increased ventilation rate, and lack of suitable respiratory protection while working at prescribed burns (Adetona et al., 2016).

Although data for occupational exposure of wildland firefighters to air pollutants in WFS is available in the United States, such information is limited to western and southeastern regions (Adetona et al., 2011; Balachandran et al., 2013; Gaughan et al., 2014; Henn et al., 2019; Lee et al., 2005; Reinhardt and Ottmar, 2004). Different geographic areas might present unique WFS exposure scenarios due to fuel variability (e.g. tree species), different soil characteristics (e.g. organic and inorganic materials), and dissimilar fire conditions (e.g. acres and duration) (Dobraca et al., 2015; Hartzell, 1996). Furthermore, there is limited data about particulate composition (e.g. BC and trace metals) of WFS exposures of wildland firefighters. Apart from the personal monitoring for silica (Broyles, 2013; Reinhardt and Broyles, 2019), only environmental monitoring data for smoke particle components from prescribed burn emissions in the western and southeastern United States exist (Aurell and Gullett, 2013; Balachandran et al., 2013; Lee et al., 2005; Mazzoleni et al., 2007; Robinson et al., 2004, 2011; Zhang et al., 2013).

Consequently, the objectives of the present study were to: 1) assess wildland firefighter personal exposure levels of $PM_{2.5}$ and CO during prescribed burns in midwestern United States forests, 2) determine the levels of BC and trace metals in WFS particulates, and 3) investigate the effect of work tasks at prescribed burns on occupational exposure levels. Information to bridge the knowledge gap will support a more accurate and comprehensive assessment of occupational risks among wildland firefighters across the United States.

2. Material and methods

2.1. Study location and wildland firefighters

The Wayne National Forest (USFS–WNF), which is managed by the United States Forest Service (USFS) is the only national forest in Ohio. It is located in southeastern Ohio and covers approximately 854,025 acres with three administrative units (i.e. in Athens, Ironton, and Marietta) (USDA, 2019). The Ohio Department of Natural Resources – Division of Forestry (ODNR–DF) oversees the 21 state forests covering approximately 200,000 acres. Occupational exposure to WFS was assessed during prescribed burns that were mostly conducted around Athens and Ironton in the USFS–WNF (N = 7) and at Zaleski state forest (N = 1) in the southeastern area of the state. The forest in this area is primarily comprised of oak (63%), maple (21%), and cottonwood (9%) (Albright, 2018).

Occupational exposure was assessed during 8 prescribed burns occurring mostly in early spring season (March and April). Firefighters were briefed about the study's purpose, design, and procedure at their annual refresher course or prior to prescribed burns. They were given adequate information to allow them to make an informed and voluntary decision to participate in the study. Informed consent was then obtained from each participant. A baseline questionnaire was administered to each participating firefighter on entrance into the study to obtain personal information such as demographics, personal work history, relevant health status, etc. Depending on each prescribed burn shift, the number of wildland firefighters involved in burning activities was about 20 and 30 for burns by WNF (N = 7) and ODNR–DF (N = 1), respectively. The proportion of the firefighters that were available and that participated in this study is approximately 40% in WNF and 50% in ODNR–DF. This study was reviewed and approved by The Ohio State University Institutional Review Board (2017H0075).

2.2. Personal exposure assessment for $PM_{2.5}$ and CO

Lightweight MicroPEM aerosol sensor (240 g) (RTI International, RTP, NC) was worn by wildland firefighters in the breathing zone to measure $PM_{2.5}$ exposures to WFS emissions across entire prescribed burns. The flow rate of the MicroPEM was set at 0.4 lpm. The on-board 25-mm Teflon filter (3.0 μm porosity) (Pall Teflo®, Port Washington, NY) was used to determine time integrated $PM_{2.5}$ concentration by gravimetric analysis using Mettler Toledo UMX2 Ultra-Microbalance (Mettler Toledo, Columbus, OH). $PM_{2.5}$ mass was calculated as the difference between the pre- and post-weight of each filter using the microbalance equipped with a static neutralizer and then adjusted by the average weight change of the field blank filters (1.85 μg). Mass concentration was determined by dividing the adjusted net weight (mg) by the sampling volume (cubic meter). Each MicroPEM was properly calibrated using a TSI primary calibrator (Model 4146, TSI, MN) in accordance with the manufacturer's instruction manual before and after each prescribed burn day. PAC7000 single gas detector (Draeger, PA) was also carried by the firefighters in the breathing zone to measure time-integrated CO concentrations in WFS across work shift. The CO detectors were calibrated using a standard calibration gas (100 ppm CO, Draeger, PA) in the laboratory once per year, and fresh air calibration was performed right before each use.

2.3. Quantification of BC level

The level of BC in the particulates deposited on the Teflon filters was determined using SootScan™ Model OT21 Optical Transmissometer (Magee Scientific, CA). The transmissometer measures the attenuation factor (ATN) for blank and sample filter at 880 nm in the infrared spectrum by the following equation:

$$ATN = \ln(I_0 / I) \times 100$$

where I_0 and I represent light transmission through a blank filter and a sample filter, respectively. Since light absorption is directly related to the light attenuation (b_{att}) due to PM deposition on the filter surface (7.85×10^{-5} m^2), the attenuation can be calculated using the following equation:

$$b_{att} = \frac{Filter\ Collection\ Area(m^2)}{Sample\ Volume(m^3)} \times \ln(I_0 / I) \times 10^6$$

BC concentration (μg/m^3) was then determined based on mass absorption efficiency ($\sigma_{att}[\lambda]$ (m^2/g)) using the following equation:

$$BC(\mu g / m^3) = \frac{b_{att}(Mm^{-1})}{\sigma_{att}[\lambda]\ (m^2/g)}$$

where σ_{att} is the slope of the least-squares regression analysis (attenuation coefficient) for elemental carbon by thermal optical analysis. An attenuation coefficient of 13.7 $cm^2/\mu g$ was used. This value was determined in a previous study investigating BC emissions from different wood-fueled cookstoves (Garland et al., 2017).

2.4. Quantification of trace metals

Each Teflon filter was rolled loosely into a 1.5 mL microcentrifuge tube and mixed with 1 mL acid mixture (100 mL nitric acid, 10 mL hydrofluoric acid, and 1 mL 1000 ppm $AuCl_3$). The samples were then floated in a boiling water bath for 2h. Some samples were moved to SCP Science DigiPREP MS digestion block to more fully immerse the filters in the acid mixture if the samples were not well digested. Afterwards, the SCP digestion tubes were cleaned by adding 10 mL 50% v/v ultrapure nitric acid, capped and heated in the digestion block for 2h at 95 °C. The tubes were cooled and rinsed 3 times with deionized water. Using Teflon coated tweezers, the filters were pulled out of the microcentrifuge tube. The samples were transferred to a cleaned 50 mL SCP digestion tube. Acid from 1.5 mL microcentrifuge tubes was added into the 50 mL tube, and transfer was completed by rinsing the original vessel with 1 mL of acid mixture. The samples were then heated in the digestion block, with caps on, at 95 °C for ~16h (overnight). Afterwards, they were removed from the heat, allowed to cool, and diluted to the 10 mL mark with deionized water spiked with 11.11 ppb of the trace metal standards to give a concentration of 10 ppb in the final solution. Samples were swirled to ensure even mixing. Twenty-two metals (Ag, Al, As, Ba, Be, Cd, Co, Cr, Cu, Hg, K, Mn, Mo, Ni, Pb, Sb, Se, Th, Tl, U, V, and Zn) were then quantified using Thermo Finnigan Element 2 Inductively Coupled Plasma Sector Field Mass Spectrometer (ICP-MS). The limit of detection (LOD) and the number of samples above its LOD for each trace metal are presented in Table A.3.

2.5. Statistics

Descriptive statistics were performed on the concentrations of $PM_{2.5}$ (mg/m^3), CO (ppm), BC ($\mu g/m^3$), mass percentage of BC to $PM_{2.5}$ (BC/PM, %), and trace metals ($\mu g/g$ and $\mu g/m^3$) and presented as geometric mean and standard error (GM ± SE) as well as arithmetic mean and standard deviation (Mean ± SD). Trace metals with less than 60% of the samples having measurements above the LOD were excluded from analysis. Concentrations of trace metals less than the LODs were substituted using maximum likelihood estimation (MLE) (Ganser and Hewett, 2010). This situation only occurred for nickel. Since exposure concentration for comparison with the occupational exposure limits (OELs) is based on an 8-h exposure, the 8-h time-weighted average (TWA) concentration was calculated for the air pollutants in WFS emissions. All prescribed burn shifts observed in study are shorter than 8 h, a conservative zero-exposure concentration, therefore, was used for the remainder of the shift. The calculated TWA exposure concentration was compared to its corresponding OELs directly. $PM_{2.5}$, CO, BC, BC/PM, trace metals had a skewed distribution, therefore the data were log transformed to achieve a normal distribution for the following statistical analyses.

Work tasks (holding, lighting, and others) assigned to each firefighter were based on information provided in the task questionnaire administered after the completion of prescribed burn shifts. Wildland firefighters selected holding task if they spent more than half of the duration of the prescribed burn keeping the fires within the planned burn area. They selected lighting if they spent more than 50% of the duration performing ignition of biomass fuels on the forest floor using a drip torch with a 1:3 gasoline to diesel mixture. Firefighters who were burn bosses or indicated that they were involved in both holding and lighting tasks equally were categorized as performing "other" tasks. Personal exposure to $PM_{2.5}$, CO, BC, BC/PM, and trace metals across the different work tasks at the prescribed burns were determined using linear mixed effect models (LMMs) and included firefighters and date as random effects to account for longitudinal within-subject correlation and for possible heterogeneity from day to day in the data that are inherent in the repeated measurement design of the study. The model estimates were then back transformed (exponential of the estimate) to obtain the geometric mean concentration estimates with the 95% confidence interval by work task during prescribed burns. The associations between $PM_{2.5}$, CO, BC, and BC/PM (log-transformed value) were determined using the Pearson correlation coefficient (r). All statistical analyses were performed using the SAS version 9.4 (Cary, NC), and estimates were considered significant if $p < 0.05$.

Table 1
Characteristics of wildland firefighters (N = 35) and prescribed burn shifts.

Characteristics	Mean ± SD or #
Age (yr)	35.94 ± 9.45
Gender	Male: 31, Female: 4
Career length (yr)	9.53 ± 8.01
Work shift duration (hr)[a]	5.09 ± 1.57
Size of area burned (acre)[b]	308.88 ± 193.13

[a] The average of burn shift durations of wildland firefighters.
[b] The average of burn sizes of prescribed burn shifts.

Table 2
Personal exposure concentration and its corresponding time-weighted average (TWA) concentration of air pollutants in WFS emissions during prescribed burns.

	N	GE ± SE	Mean ± SD	95% CI	Range
Personal Exposure Concentration					
$PM_{2.5}$ (mg/m^3)	53	1.43 ± 0.13	1.75 ± 1.20	1.42–2.08	0.35–5.25
CO (ppm)	66	7.02 ± 0.69	9.38 ± 6.78	7.71–11.04	1.10–24.14
BC ($\mu g/m^3$)	51	58.79 ± 5.46	72.29 ± 48.79	58.57–86.01	7.35–250.16
BC/PM (%)	51	3.98 ± 0.43	5.28 ± 4.22	4.10–6.47	0.67–16.59
TWA Exposure Concentration					
$PM_{2.5}$ (mg/m^3)	53	0.76 ± 0.06	0.89 ± 0.49	0.76–1.03	0.16–2.10
CO (ppm)	66	4.09 ± 0.43	5.70 ± 4.70	4.54–6.87	0.56–18.52
BC ($\mu g/m^3$)	51	31.37 ± 2.93	38.19 ± 23.83	31.49–44.89	3.32–95.11

GE ± SE: geometric mean ± standard error; Mean ± SD: arithmetic mean ± standard deviation. 95% CI: 95% confidence interval.

3. Results

A total of 53 personal $PM_{2.5}$ and 66 personal CO exposure concentration measurements were obtained from wildland firefighters on prescribed burn days. A small hole was found in two Teflon filters in the area outside of the particle deposition and therefore BC was measured in 51 filters. Characteristics of the wildland firefighters and prescribed burn work shifts are in Table 1. Burn shift duration for each firefighter was determined using the sampling time recorded by the personal exposure monitors, MicroPEM and/or PAC7000 single gas detector. The burned acreage was recorded for each prescribed burn shift. The average personal exposure concentration and its corresponding TWA exposure concentration for the air pollutants in WFS emissions during prescribed burns are presented in Table 2.

The difference in the exposure concentration due to work tasks is shown in Fig. 1. Firefighters who performed the holding task during majority of their work shifts were exposed to 1.47 mg/m^3 (95% CI: 1.16–1.87 mg/m^3) of $PM_{2.5}$, 8.50 ppm (95% CI: 6.70–10.77 ppm) of CO,

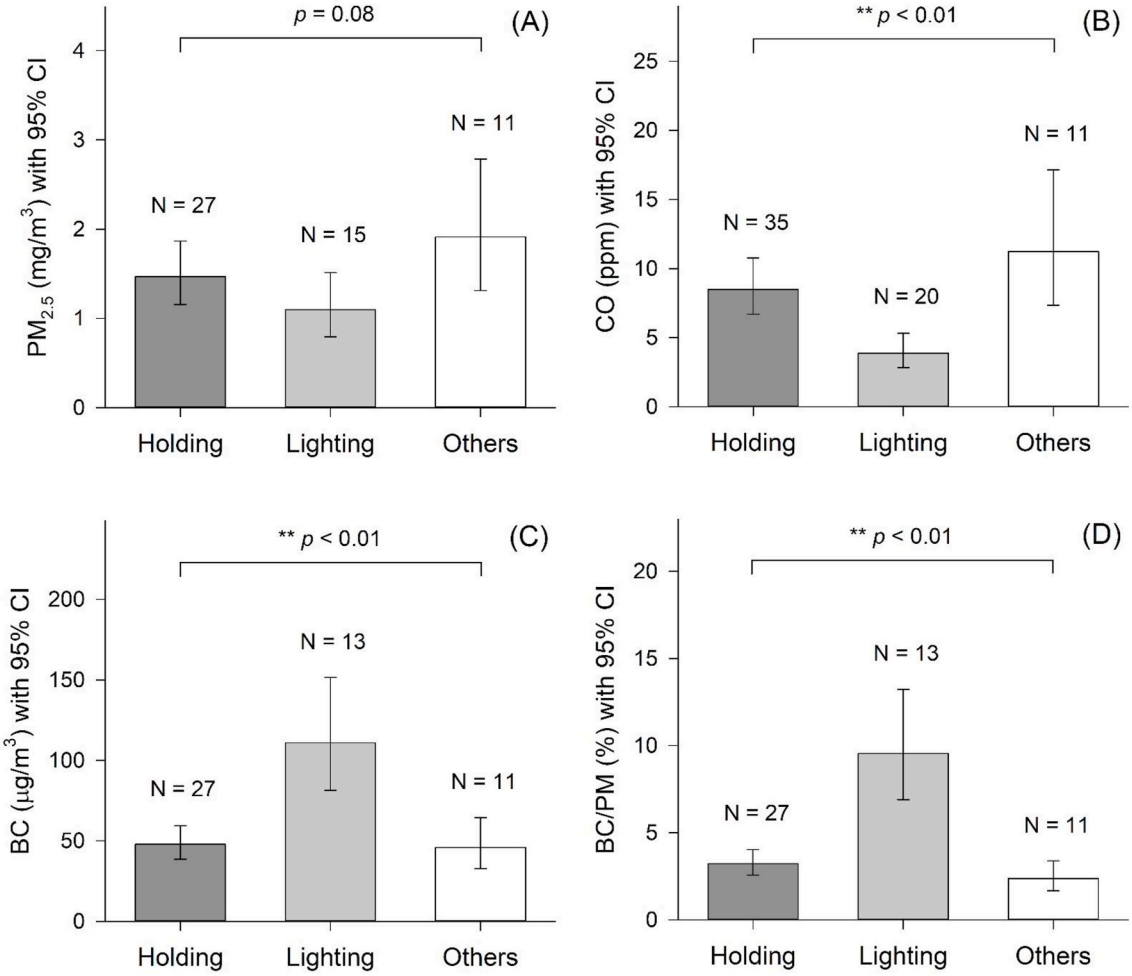

Fig. 1. Personal exposure concentration (estimate with 95% CI) of air pollutants: (A) $PM_{2.5}$, (B) CO, (C) BC, and (D) BC/PM in wildland fire smoke emissions by work tasks during prescribed burns. **: Difference across tasks is significant at $p = 0.01$. N = the number of person-day.

47.85 μg/m^3 (95% CI: 38.57–59.37 μg/m^3) of BC, and 3.22% (95% CI: 2.57–4.03%) of BC/PM, while those who performed lighting for the majority of the shifts were exposed to 1.10 mg/m^3 (95% CI: 0.80–1.51 mg/m^3) of $PM_{2.5}$, 3.88 ppm (95% CI: 2.84–5.31 ppm) of CO, 111.05 μg/m^3 (95% CI: 81.38–151.55 μg/m^3) of BC, and 9.55% (95% CI: 6.90–13.22%) of BC/PM during prescribed burn shifts. The firefighters who were involved in "Others" tasks had 1.91 mg/m^3 (95% CI: 1.31–2.79 mg/m^3) of $PM_{2.5}$, 11.23 ppm (95% CI: 7.36–17.14 ppm) of CO, 45.95 μg/m^3 (95% CI: 32.77–64.43 μg/m^3) of BC, and 2.37% (95% CI: 1.67–3.38%) of BC/PM at prescribed burns. There is no difference in personal $PM_{2.5}$ exposure concentration across the tasks ($p = 0.08$). However, significant task-related differences were observed for CO, BC, and BC/PM ($p < 0.01$). Compared to those who were assigned to holding and those who were deemed to have performed "Others" tasks, firefighters who performed lighting had a lower CO exposure concentration ($p < 0.01$) but a higher exposure level of BC and BC/PM ($p < 0.01$).

Trace metal composition (μg/g and μg/m^3) in WFS smoke particulates collected during prescribed burn is presented in Table 3 and the corresponding TWA exposure level is shown in Table A.4. The most abundant metal was potassium (8891.83 ± 533.61 μg/g, 12.7089 ± 0.8044 μg/m^3) followed by aluminum (2533.28 ± 323.55 μg/g, 3.6208 ± 0.3787 μg/m^3), chromium (634.78 ± 78.94 μg/g, 0.9073 ± 0.0956 μg/m^3), and zinc (368.15 ± 49.59 μg/g, 0.5262 ± 0.0582 μg/m^3). Silver, arsenic, beryllium, cobalt, mercury, selenium, thallium, and uranium were barely detected (# of samples below LOD > 40%) in the particulates from WFS emissions. A significant task-related difference was observed in manganese (μg/m^3) ($p = 0.02$, Table A.6). Firefighters who performed lighting had a lower manganese exposure concentration compared to the firefighters assigned to holding ($p = 0.02$) and others ($p = 0.02$).

Associations between air pollutants in WFS during prescribed burn shifts are presented in Table 4. Personal exposure concentration of $PM_{2.5}$ was positively associated with CO and BC exposure levels ($r = 0.83$ and 0.28 for CO and BC, respectively, $p < 0.05$) but negatively correlated with BC/PM ($r = -0.58$, $p < 0.01$). BC/PM was associated negatively with CO concentration ($r = -0.64$; $p < 0.01$) and positively correlated with BC concentration ($r = 0.62$; $p < 0.01$).

4. Discussion

Between 2008 and 2017, nearly 420,000 tons of $PM_{2.5}$ were emitted to the ambient air in Ohio due to forest prescribed burn activities, accounting for ~21% of total $PM_{2.5}$ emission in the state (USEPA, 2019b). Wildland firefighters, who are primarily responsible for performing prescribed burns between wildfire seasons, are directly exposed to elevated levels of WFS compared to the general population. Studies of WFS exposure assessment for wildland firefighters have mostly been done in the southeastern and western United States. No information for their occupational exposure during either wildfire suppression or prescribed burns is available in the Midwest where weather and fuel

Table 3
Trace metal composition (geometric mean ± standard error) in WFS particulates during prescribed burns (N = 53).

Metals	GM ± SE (μg/g)	Range (μg/g)	GM ± SE (μg/m^3)	Range (μg/m^3)
Ag	BDL	BDL	BDL	BDL
Al	2533.28 ± 323.55	258.58–15354.72	3.6208 ± 0.3787	0.7251–48.1848
As	BDL	BDL	BDL	BDL
Ba	207.18 ± 23.67	38.75–3679.83	0.2961 ± 0.0296	0.0741–1.7575
Be	BDL	BDL	BDL	BDL
Cd	6.27 ± 0.30	2.63–19.29	0.0090 ± 0.0009	0.0022–0.0371
Co	BDL	BDL	BDL	BDL
Cr	634.78 ± 78.94	112.34–6026.24	0.9073 ± 0.0956	0.1688–9.6972
Cu	175.60 ± 26.62	2.68–1068.32	0.2510 ± 0.0304	0.0051–1.3705
Hg	BDL	BDL	BDL	BDL
K	8891.83 ± 533.61	2265.78–22326.95	12.7089 ± 0.8044	4.4114–33.6714
Mn	128.48 ± 18.62	9.05–1410.86	0.1836 ± 0.0271	0.0093–1.8884
Mo	5.27 ± 0.61	1.13–34.81	0.0075 ± 0.0008	0.0009–0.0473
Ni	12.17 ± 1.62	2.53–118.78	0.0174 ± 0.0023	0.0033–0.1402
Pb	112.43 ± 17.97	14.41–8259.01	0.1607 ± 0.0240	0.0260–5.9699
Sb	5.46 ± 0.76	1.25–212.78	0.0078 ± 0.0009	0.0017–0.3381
Se	BDL	BDL	BDL	BDL
Th	1.35 ± 0.12	0.39–8.94	0.0019 ± 0.0001	0.0011–0.0065
Tl	BDL	BDL	BDL	BDL
U	BDL	BDL	BDL	BDL
V	5.40 ± 0.90	0.65–140.35	0.0077 ± 0.0012	0.0010–0.4120
Zn	368.15 ± 49.59	34.13–2310.18	0.5262 ± 0.0582	0.1052–4.0092

BDL: number of samples below detection limit > 40%.

Table 4
Correlation between air pollutants in WFS emissions.

	$PM_{2.5}$	CO	BC	BC/PM
$PM_{2.5}$		0.83**	0.28*	−0.58**
CO			0.06	−0.64**
BC				0.62**
BC/PM				

*p less than 0.05; **p less than 0.01.

conditions could be quite different.

4.1. $PM_{2.5}$ and CO exposure concentrations

Although, the OEL defined for respirable particulates is for PM with an aerodynamic diameter <3.5 or 4 μm ($PM_{3.5}$ or PM_4) and many previous studies monitored PM exposure for this particulate size (Gaughan et al., 2014; Reinhardt and Ottmar, 2004; Slaughter et al., 2004), particle size distribution in vegetative biomass smoke is mostly < 2.5 μm (Chakrabarty et al., 2006; Iinuma et al., 2007; Kim Yong et al., 2018). Therefore, personal exposure concentration of $PM_{2.5}$ was monitored among wildland firefighters during prescribed burns in this study.

A 12% difference in weight concentration between $PM_{2.3}$ and $PM_{3.5}$ was reported in a series of small open-burning experiments, in which a 10-mm nylon cyclone was used to collect biomass smoke in an 85 m^3 greenhouse (McMahon and Bush, 1992). Therefore, $PM_{2.5}$ concentrations observed in this study were increased by 12% to approximate $PM_{3.5}$ concentrations in case of comparisons with OELs. The geometric and arithmetic means for estimated $PM_{3.5}$ concentration were 1.60 ± 0.14 and 1.96 ± 1.34 mg/m^3 (95% CI: 1.59–2.33 mg/m^3), respectively. The geometric and arithmetic means for their corresponding TWA exposure concentrations were 0.85 ± 0.07 and 1.00 ± 0.55 mg/m^3 (95% CI: 0.18–2.35 mg/m^3).

In comparison with the currently existing OELs, no personal $PM_{3.5}$ exposure concentrations were above the American Conference of Governmental Industrial Hygienists (ACGIH) Threshold Limit Value (TLV) recommended for workplace exposures to respirable particulates (3 mg/m^3) and the Occupational Safety and Health Administration (OSHA) Permissible Exposure Limit (PEL) for respirable particulates not otherwise regulated (PNOR) (5 mg/m^3). However, because of the reactive nature of WFS particulates, the regulatory PNOR PEL is not recommended for WFS exposure that wildland firefighters experience during fire suppression and prescribed burning activities (Adetona et al., 2016; Reinhardt and Broyles, 2019). Reinhardt and Broyles (2019) have recently suggested to use 1.0 mg/m^3 as a fireline-average OEL for the PNOR respirable fraction in WFS emissions. This exposure limit is further reduced to 0.7 mg/m^3 for a recommended OEL guideline for 8-h shift-average PM_4 exposure. Under this criterion, a total of 33 personal exposure concentrations (~62%) of the estimated $PM_{3.5}$ exceeded this recommended 8-h shift-average OEL.

Compared to previous studies, the average respirable particulate concentration observed in this study was 1.8–3.2 times higher than the concentrations reported at wildland fires and prescribed burns in Western United States (Gaughan et al., 2014; Reinhardt and Ottmar, 2004; Slaughter et al., 2004). Wildland firefighters in this study also had a 2.7–5.4 times higher average $PM_{2.5}$ exposure concentration compared to those who worked at prescribed burns in the southeastern region of the country (Adetona et al., 2011, 2013, 2017).

Although none of the personal CO exposure concentrations were above the OSHA PEL (50 ppm), the National Institute for Occupational Safety and Health (NIOSH) Recommended Exposure Limit (REL) (35 ppm), and the ACGIH TLV (25 ppm), a total of 28 personal exposure measurements had instances when the NIOSH REL Ceiling for CO (200 ppm) was exceeded. The ceiling exceedance for CO was observed in 7 out of 8 prescribed burn shifts (88%). In addition, we observed 26 personal CO exposure concentrations, ranging from 10.08 to 24.14 ppm that were above the 8-hr NAAQS for CO (9 ppm). The average CO concentration observed in this study was 1.7–4.1 and 2.3–7.0 times higher, respectively, than those reported in previous occupational exposure studies for the firefighters in the western and southeastern regions (Adetona et al., 2011, 2013, 2017; Gaughan et al., 2014; Henn et al., 2019; Reinhardt and Ottmar, 2004).

Compared to wildland firefighters in the other regions, we observed a relatively shorter length of WFS exposure and fewer acres of burned area but substantially higher exposure levels of $PM_{2.5}$ and CO in WFS emissions. We hypothesize that the differences between results in our study and those from other regions were due to different types of vegetation. In Ohio, the most dominant forest type group is oak/hickory (~60%) (Albright, 2018), whereas loblolly/shortleaf pine group (~40%) and Douglas-fir group (~40%) are the major forest types in the Southeast and West, respectively (USDA, 2008). $PM_{2.5}$ mass emission factors from laboratory combustions of different fuel types have been reported in previous studies, indicating particle mass emission is ~1.8-fold higher in oak than Douglas fir and loblolly pine during fireplace combustion (Fine et al., 2002, 2004a). Similarly, wood stove combustion of oak emitted approximately 2.5- and 1.4-fold fine particulates, respectively, compared to Douglas fir and loblolly pine (Fine et al., 2004b). However, a recent study investigating smoke composition from combustions of various biomass fuels using a quartz-tube furnace system showed $PM_{2.5}$ and CO emission factors of red oak and pine at either flaming or smoldering combustion stage were comparable (Kim Yong et al., 2018). Therefore, the higher exposure levels observed in this study could be in part due to different forest type groups in Ohio compared to the other locations.

In this study, the average ambient air temperature and relative

humidity at the beginning of prescribed burn activities (~30 min before the initial ignition of fire) were 65.9 ± 6.9 °F and 39.5 ± 7.9%, respectively. Both parameters were close to a general 60:40 guideline of weather conditions for conducting a prescribed burn. This guideline can be expressed as either to proceed with prescribed burns when both air temperature and relative humidity measures are between the values of 40 and 60 or when air temperature is less than 60 °F and relative humidity is above 40% (Weir, 2009). In the United States, prescribed burn is mostly conducted in the Southeast (64%) followed by the West (33%) and the Northeast (3%) region due to weather concerns (Melvin, 2018), especially due to conditions that are favorable for good smoke dispersion and low risk of unstable fire behaviors. Although, there are no integrated data for forest fuel loads in different geographical regions, vegetative fuels accumulated on the forest floor in this study are expected to be higher than those in the other studies due to limited burning windows in the Midwest.

4.2. BC level in WFS particulates

In this study, BC level in the particulates was determined using the optical transmission technique in which light attenuation was converted to the concentration using an attenuation coefficient (13.7 $cm^2/\mu g$). This coefficient is based on the slope of a regression line of elemental carbon (EC) against light attenuation in a previous experimental study (Garland et al., 2017). EC, which is commonly measured using thermal-optical method, was often reported in prior WFS studies (Balachandran et al., 2013; Lee et al., 2005; Mazzoleni et al., 2007; Robinson et al., 2011; Zhang et al., 2013). The difference between these two carbonaceous particles in PM is operationally different due to the analytical methods (transmissometry vs reflectometry) used for quantification (Andreae and Gelencsér, 2006; Bond and Bergstrom, 2006).

No personal exposure data for carbonaceous aerosols in WFS has been reported, but our results (GE ± SE for BC: 58.79 ± 5.46 $\mu g/m^3$; for BC/PM: 3.98 ± 0.43%) are similar to several environmental monitoring studies with air samplers placed at or near the fireline of prescribed burns. The average BC/$PM_{2.5}$ mass ratio at ground level (~2.5m above) during prescribed burn in the Southeastern United States was reported to be 3.4% (Aurell and Gullett, 2013). Similarly, the average EC concentration from a two-day sampling for $PM_{2.5}$ within 50m of prescribed fires in the Western United States was 55 $\mu g/m^3$ with a range of 8–239 $\mu g/m^3$ (Robinson et al., 2011). In a different study in the Western United States, the mean percentage of EC to $PM_{2.5}$ mass during flaming ~10–30m from the prescribed burn area and smoldering within the burn area was ~3.7% (Robinson et al., 2004). An average EC concentration of 70 $\mu g/m^3$ was measured at 20–200m from prescribed mixed pile and understory burns in another study in the western region (Zhang et al., 2013).

Neither OSHA nor NIOSH has established OELs for BC or EC. The ACGIH has proposed a TLV of 20 $\mu g/m^3$ for EC exposure. It should be noted that this exposure limit is for PM in diesel exhaust, which is fundamentally different from PM in WFS emissions. Nonetheless, it is likely that this exposure limit for EC was exceeded in the current study as 37 out of the 51 BC measurements had TWA concentrations in excess of 20 $\mu g/m^3$. In addition to PM, BC has been suggested as an additional indicator for air quality management and linked to many adverse effects (Janssen et al., 2011). BC levels, ranging from ~1 to 24 $\mu g/m^3$, that were reported in previous studies investigating the association between biomass smoke exposure and health outcomes were well below the average BC level observed in this study (Baumgartner et al., 2014; Jacobson Lda et al., 2014; Rajkumar et al., 2019; Ravindra, 2019; Shan et al., 2014).

4.3. Trace metal composition in WFS particulates

To our knowledge, this study is the first to characterize personal exposure to trace metals in WFS emissions during prescribed burns. The exposure concentrations were well below the corresponding OELs for the metals that are regulated. However, it should be noted that the levels of trace elements in WFS particulates might depend on particle size fractions. In a controlled biomass combustion study, the highest concentration of the elements (especially for potassium and aluminum) was detected in coarse (>2.54 μm) and fine fractions (<0.96 μm), and the levels of the elements in intermediate size range (0.96–2.54 μm) were much lower (Ordou and Agranovski, 2017). Therefore, the total exposure levels of the metals in this study might actually be higher.

Trace elements reported in environmental monitoring studies for WFS emissions were quantified using X-ray fluorescence spectroscopy (XRF) (Balachandran et al., 2013; Lee et al., 2005; Robinson et al., 2004, 2011). Although, a good agreement between two analytical techniques, ICP-MS and XRF, was observed in previous comparison studies (Niu et al., 2010; Yatkin et al., 2012), the trace elements were measured in the particulates collected from residential/industrial areas, not from WFS emissions. In addition, the average $PM_{2.5}$ mass obtained in this study was ~10–30% of those reported in previous environmental exposure assessments (Balachandran et al., 2013; Robinson et al., 2004, 2011). Finally, the sampling devices used in the southeastern studies investigating WFS composition were far away from the combustion sources (about 5, 13, and 16 miles) (Balachandran et al., 2013; Lee et al., 2005), whereas trace elements reported in the western studies were measured inside and outside (about 10–50 m) of prescribed burn area (Robinson et al., 2004, 2011).

Despite differences in the analytical method, particle mass, and sampling distance, we still observe some similarities between this study and the others. The essential elements for tree growth such as copper and zinc were reported at quantifiable levels in the smoke particulates in all regions (Balachandran et al., 2013; Lee et al., 2005; Robinson et al., 2004, 2011). Aluminum, manganese, lead, and selenium were also observed in the Southeast prescribed burn studies (Balachandran et al., 2013; Lee et al., 2005), whereas chromium was detected in the West (Robinson et al., 2004). Furthermore, the most dominant metal in the particulates collected during prescribed burns in this study was potassium, which is in accordance with the studies in the southeastern and western regions (Balachandran et al., 2013; Lee et al., 2005; Robinson et al., 2004, 2011). Potassium, along with chloride, has been recognized as a useful tracer species in wood-smoke particulates due to its higher concentration in vegetation compared to the other elements (Reid et al., 2005).

4.4. Task-related difference in exposure concentrations

Similar to a previous study in the southeastern region (Adetona et al., 2017), we observed wildland firefighters who performed holding had higher exposure levels of $PM_{2.5}$ and CO compared to the firefighters who conducted lighting (Fig. 1). We further investigated peak CO exposure concentration by work tasks and observed that 16 out of 35 person-day measurement (46%) for the holding task had peak CO concentration that was above NIOSH REL Ceiling (200 ppm), whereas 3 out of 20 measurements (15%) for lighting had peak concentrations that were higher than the ceiling value. A total of 9 out of 11 person-day samples (82%) that were assigned to “Other” had the concentrations that exceeded the ceiling value.

This result could be due to different combustion conditions that the firefighters had experienced during prescribed burns. Lighting involves the use of drip torches to ignite fires and generally exposure to WFS from flaming combustion. Contrarily, firefighters who performed holding were more exposed to smoke from smoldering combustion as they consistently worked at the boundary of prescribed burn areas to prevent the fires from spreading. Compared to flaming phase, higher emission factors for particulates and gases have been observed during smoldering phase of biomass burning in previous field and laboratory combustion studies (Balachandran et al., 2013; Kim Yong et al., 2018; Lee et al., 2005; Reisen et al., 2018).

Interestingly, out of 7 monitored prescribed burns, wildland firefighters who conducted tasks categorized as "Other" predominantly worked on the four prescribed burns with higher exposures of $PM_{2.5}$ and CO. Only one such firefighter worked any of the other prescribed burns (Table A.1).

Unlike the task-related difference in $PM_{2.5}$ and CO, the highest BC exposure concentrations were observed for firefighters who performed lighting and had mostly been exposed to WFS from flaming fires (Fig. 1). This result could be attributed to combustion efficiency in different fire phases. As demonstrated in previous experimental studies, flaming phase of biomass burning is capable of producing smoke particulates with higher level of soot (EC) compared to the smoldering phase due to better combustion efficiency (Aurell et al., 2015; McMeeking et al., 2009; Pósfai et al., 2003). Robinson et al. (2004) reported that EC level was about 5-fold higher during the flaming stage than during the smoldering stage in their study of $PM_{2.5}$ compositions during prescribed fires.

Another possible explanation for this result is the use of drip torches, which contains a mixture of three-part diesel to one-part gasoline, to ignite the fires during prescribed burns. Higher levels of BC in particulate matter emitted by diesel fuel combustion compared to biomass burning were observed in previous studies (Cheng et al., 2011; Du et al., 2014; Sandradewi et al., 2008). Also flaming combustion of wildland fuels emits particles with higher BC content but lower levels of reflective, albedo organic carbon (OC); on the contrary, smoldering combustion emits particles with lower BC content but higher OC content (Chakrabarty et al., 2006; Chen et al., 2006). It is noted that OC in PM from different sources including biomass combustion has been linked to elevated mutagenicity and increased cardiovascular morbidity and mortality (Kim Yong et al., 2018; Yang et al., 2019). Although we were not able to distinguish the carbonaceous aerosols contributed by different sources of combustion, the BC exposure of firefighters who lighted is likely more impacted by diesel and flaming biomass combustion as demonstrated by a higher mass percentage of BC to $PM_{2.5}$ (Fig. 1).

Although, the task-related difference in fractional abundance of trace metals was not significant, the exposure levels (μg/g) of the metals were generally higher for firefighters who performed lighting compared to those whose task was primarily holding or classified as others (Table A.5). Similar results were observed in another study in which higher concentrations of trace elements were measured in particles emitted during flaming compared to smoldering stage, presumably due to a drop in fire intensity, fire temperature, and fuel consumption rate (Robinson et al., 2004). A recent study also reported higher content of metals (μg/kg), such as chromium, copper, manganese, lead, and zinc, in $PM_{2.5}$ emissions from diesel fuel combustion compared to emissions from biodiesel combustion using the same vehicle engine (Coufalík et al., 2019). Firefighters that performed lighting were more likely exposed to smoke from diesel and flaming biomass combustion, so the metal content in the WFS to which they were exposed is expected to be higher.

However, a contrary result was observed when the levels of trace metals were presented by mass concentration (μg/m^3) (Table A.6). After the unit conversion (μg/g × particle mass concentration [μg/m^3]/ 1,000,000), firefighters that lighted tended to have lower exposure to metals in WFS, especially for manganese ($p = 0.02$), compared to those performing other tasks. This discrepancy might be due to the particle mass collected from WFS emissions during prescribed burns. The average particle mass obtained from firefighters that performed lighting was 163.47 μg, whereas averages of 215.19 and 269.53 μg were collected from holders and those with tasks classified as "Others", respectively.

4.5. Limitations

A couple of limitations should be considered when interpreting the findings of our study. First, personal exposure assessment for wildland firefighters was conducted at only 8 prescribed burns in Ohio. Therefore, there is need for exposure assessment in other locations in to confirm whether the relatively higher levels of exposure that were observed in this study are consistent across the region.

Secondly, work task classification was based on recalled information that was provided in the task questionnaire. Hence, there is potential for error in the classification. Nonetheless, the questionnaire was administered to each firefighter immediately after the completion of prescribed burn activity. The research personnel were also able to be at the fireline for most of the burns and observed that most of the firefighters consistently performed singular tasks during the burns. Therefore, we do not expect there was enough misallocation of task among the firefighters to change the results.

In addition, we were unable to collect more detailed information about the proportion of work time spent performing each task if the firefighters worked multiple tasks. Nonetheless, we were able to observe at the fireline that most of the firefighters that conducted lighting or holding worked a singular task at the fireline. Therefore, we do not expect that the addition of such detail in the analyses would have changed the results that are herein presented.

5. Conclusions

The current study assessed personal exposure to air pollutants in WFS emissions among the firefighters worked in the Midwestern United States. The results of this study showed that the average personal exposure concentration of wildland firefighters to $PM_{2.5}$ in WFS was about 2- to 5-fold higher than those reported in the southeastern and western regions. Similarly, the average CO exposure concentration was approximately 2- and 7-fold higher than those observed in the western and southeastern United States, respectively. No personal CO exposure concentration was above the OELs but 28 personal exposure concentrations had ever above the NIOSH REL Ceiling (200 ppm). The higher personal $PM_{2.5}$ and CO exposure concentrations that was observed in this study might be due to the different forest type groups and fuel loads.

Personal exposure concentrations of air pollutants in WFS emissions was associated with prescribed burn tasks. Although the difference is not significant, $PM_{2.5}$ exposure concentration was higher among the firefighters who performed holding than lighting. Personal CO exposure concentration was significantly higher among the firefighters who conducted holding compared to those who lighted. Contrarily, the average BC exposure concentration was highest for firefighters who lighted.

Our results showed wildland firefighters who worked at prescribed burns in Midwestern United States had higher WFS exposures compared to results observed in the southeastern and western United States. Personal exposure concentration of air pollutants in WFS emissions was associated with work tasks during prescribed burns. These results highlight the need for appropriate respiratory protection against WFS exposure for wildland firefighters.

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Ethics

This study was reviewed and approved by The Ohio State University Institutional Review Board (2017H0075).

CRediT authorship contribution statement

Chieh-Ming Wu: Conceptualization, Formal analysis, Investigation, Writing - original draft, preparation, Funding acquisition. **Chi (Chuck) Song:** Formal analysis, Writing - review & editing. **Ryan Chartier:** Methodology, Investigation, Resources, Writing - review & editing. **Jacob Kremer:** Methodology, Investigation, Resources, Writing - review & editing. **Luke Naeher:** Methodology, Resources, Writing - review & editing. **Olorunfemi Adetona:** Conceptualization, Validation, Investigation, Resources, Writing - review & editing, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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