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Combustion efficiency and emission factors for US wildfires

S. P. Urbanski

Missoula Fire Sciences Laboratory, Rocky Mountain Research Station, United States Forest Service, Missoula, Montana, USA

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Correspondence to: S. P. Urbanski (surbanski@fs.fed.us)

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Abstract

In the US wildfires and prescribed burning present significant challenges to air regulatory agencies attempting to achieve and maintain compliance with National Ambient Air Quality Standards (NAAQS) and Regional Haze Regulations. Wildland fire emission inventories (EI) provide critical inputs for atmospheric chemical transport models used by air regulatory agencies to understand and to predict the impact of fires on air quality. Fire emission factors (EF), which quantify the amount of pollutants released per mass of biomass burned, are essential input for the emission models used to develop EI. Over the past decade substantial progress has been realized in characterizing the composition of fresh biomass burning (BB) smoke and in quantifying BB EF. However, most BB studies of temperate ecosystems have focused on emissions from prescribed burning. Little information is available on EF for wildfires in the temperate forests of the conterminous US. Current emission estimates for US wildfires rely largely on EF measurements from prescribed burns and it is unknown if these fires are a reasonable proxy for wildfires.

Over 8 days in August of 2011 we deployed airborne chemistry instruments and sampled emissions from 3 wildfires and a prescribed fire that occurred in mixed conifer forests of the northern Rocky Mountains. We measured the combustion efficiency, quantified as the modified combustion efficiency (MCE), and EF for CO_2, CO, and CH_4. Our study average values for MCE, $EFCO_2$, EFCO, and $EFCH_4$ were 0.883, 1596 $g\,kg^{-1}$, 135 $g\,kg^{-1}$, 7.30 $g\,kg^{-1}$, respectively. Compared with previous field studies of prescribed fires in similar forest types, the fires sampled in our study had significantly lower MCE and $EFCO_2$ and significantly higher EFCO and $EFCH_4$. An examination of our study and 47 temperate forest prescribed fires from previously published studies shows a clear trend in MCE across US region/fire type: southeast (MCE = 0.933) > southwest (MCE = 0.922) > northwest (MCE = 0.900) > northwest wildfires (MCE = 0.883).

The fires sampled in this work burned in areas reported to have moderate to heavy components of standing dead trees and dead down wood due to insect activity and previous fire, but fuel consumption data was not available for any of the fires. However, fuel consumption data was available for 18 prescribed fires reported in the literature. For these 18 fires we found a significant negative correlation ($r = -0.83$, p-value = 1.7e-5) between MCE and the ratio of heavy fuel (large diameter dead wood and duff) consumption to total fuel consumption. This observation suggests the relatively low MCE measured for the fires in our study resulted from the availability of heavy fuels and conditions that facilitated combustion of these fuels. More generally, our measurements and the comparison with previous studies indicate that fuel composition is an important driver of variability in MCE and EF.

This study only measured EF for CO_2, CO, and CH_4; however, we used our study average MCE to estimate wildfire EF for $PM_{2.5}$ and 13 other species using EF–MCE linear relationships reported in the literature. The EF we derived for several non-methane organic compounds (NMOC) were substantially larger (by a factor of 1.5 to 4) than the published prescribed fire EF. Wildfire $EFPM_{2.5}$ estimated in our analysis is approximately twice that reported for temperate forests in a two widely used reviews of BB emission studies. Likewise, western US wildfire $PM_{2.5}$ emissions reported in a recent national emission inventory are based on an effective $EFPM_{2.5}$ that is only 40% of that estimated in our study. If the MCE of the fires sampled in this work are representative of the combustion characteristics of wildfires across western US forests then the use of EF based on prescribed fires may result in a significant underestimate of wildfire $PM_{2.5}$ and NMOC emissions. Given the magnitude of biomass consumed by western US wildfires, the failure to use wildfire appropriate $EFPM_{2.5}$ has significant implications for the forecasting and management of regional air quality. The contribution of wildfires to NAAQS $PM_{2.5}$ and Regional Haze may be underestimated by air regulatory agencies.

1 Introduction

Biomass burning (BB, defined here as the open burning of biomass which includes wildfires and prescribed fires in forests, savannas, grasslands, and shrublands, and agricultural fire such as the burning of crop residue) is a major source of global trace gases and particles (van der Werf et al., 2010; Wiedinmyer et al., 2011). In terms of total global source BB emissions are estimated to account for 40 % of carbon monoxide (CO), 35 % of carbonaceous particles, and 20 % of nitrogen oxides (NO_x) (Langmann et al., 2009). The contribution of BB in the conterminous US to global BB emissions is minor (van der Werf et al., 2010; Wiedinmyer et al., 2011). However; in the US wildland fires (defined here as BB excluding agricultural fires) have a significant impact on air quality and present major challenges to air regulatory agencies responsible for achieving and maintaining compliance with federal National Ambient Air Quality Standards (NAAQS; USEPA, 2012c) for ozone (O_3) and fine particulate matter ($PM_{2.5}$) and Regional Haze Regulations (USEPA, 1999). Because O_3 is a secondary pollutant resulting from complex chemistry, quantifying the contribution of wildfires to O_3 related air quality degradation is difficult. A thorough review of regulatory issues associated with wildfire O_3 production is provided by Jaffe and Wigder (2012). Acute impacts of wildfires and prescribed fires on $PM_{2.5}$ levels in urban areas have been reported in numerous studies (DeBell et al., 2004; Liu et al., 2009; Sapkota et al., 2005) and documented by air regulatory agencies (USEPA, 2012b). Wildland fires have also been identified as important contributors to visibility reduction in areas protected by the Regional Haze Rule (Brewer and Moore, 2009).

While prescribed burning (fires intentionally ignited for land management purposes) dominates fire activity in the southeastern US (~ 75 % of area burned between 2002–2010; (NIFC, 2012)), wildfires are dominant in the western US (defined here as: Arizona, California, Colorado, Idaho, Montana, Nevada, New Mexico, Oregon, Utah, Washington, Wyoming) (~ 85 % of area burned between 2002–2010; (NIFC, 2012)). Annually, wildland fires account for a sizeable fraction of total $PM_{2.5}$ and CO emissions

in the western US (as much as 39 % and 20 %, respectively) (Urbanski et al., 2011). Because wildfire emissions are episodic and highly concentrated both temporally and spatially (Urbanski et al., 2011), such annualized comparisons greatly understate the potential impact of the wildfires on the day time scale that is pertinent to air quality forecasting and management.

Wildland fire emission inventories (EI) provide critical input for atmospheric chemical transport models used by air regulatory agencies to understand and to predict the impact of fires on air quality. Fire emission factors (EF), which quantify the amount of pollutants released per mass of biomass burned, are essential input for the emission models used to develop EI. Over the past decade substantial progress has been realized in characterizing the composition of fresh BB smoke and in quantifying BB EF (see Akagi et al., 2011). Yet significant gaps in the current knowledge of EF remain in many areas. Little information is available on EF for wildfires in the temperate forests of the conterminous US. Emission estimates for US wildfires rely largely on EF measurements from prescribed fires. However, it is unknown if these prescribed fires are a reasonable proxy for wildfires. The combustion characteristics of a fire, in particular the relative amounts of flaming and smoldering combustion, have a significant influence on the chemical composition of the smoke. Smoldering combustion is less efficient than flaming combustion and per unit of fuel consumed produces more CO, CH_4, non-methane organic compounds (NMOC), and particulate matter and less CO_2 (Bertschi et al., 2003; Burling et al., 2010; Lobert, 1991; Yokelson et al., 1996, 2008). Smoldering combustion is prevalent in coarse woody debris (CWD, large diameter (> 7.62 cm) dead wood) and duff while fine fuels (grasses, shrubs, foliage, litter, and fine woody debris (FWD, small diameter (< 7.62 cm) dead wood)) tend to burn by mostly flaming combustion (Ottmar, 2001; Sandberg et al., 2002). Therefore, the characteristics of the fuels consumed in a wildland fire, which is determined by the fuels present and environmental conditions, should have an important influence on the composition of emissions.

Conditions during the western US wildfire season, low fuel moistures and high-intensity fire fronts, are favorable for the consumption of CWD and duff and these fuels may comprise a significant portion of total fuel consumed in a fire event (Campbell et al., 2007; Reinhardt et al., 1991). Conversely, prescribed burning is generally characterized by low-intensity fire when the moisture of CWD and duff are moderate to high (Finney et al., 2005; Hardy, 2002), conditions which minimize consumption of these fuels relative to fine fuels. Thus, wildfires might be expected to burn with more smoldering combustion than prescribed fires and have higher EF for species associated with smoldering combustion (and lower EF for species related to flaming combustion). This reasoning suggests EF based on prescribed fires may not be appropriate for modeling emissions from wildfires.

We present smoke emissions data from airborne field measurements of fires that occurred in conifer dominated montane forests of the western US during the 2011 wildfire season. We report our measurements of modified combustion efficiency (MCE) and EF for CO_2, CO, and CH_4 and compare these with previous field studies of temperate forest fires. The MCE measured in our field study are used to estimate wildfire EF for 14 additional species using previously published EF–MCE relationships. This new EF dataset for western US wildfires is compared with a recent review article and a national emissions inventory. We also examine MCE and fuel consumption data from previous studies of 18 prescribed fires to gain insight into regional MCE trends and to identify possible drivers of fire combustion characteristics.

2 Methodology

This study measured fresh smoke emissions from 3 large wildfires (Saddle Complex, Big Salmon Lake Fire, and Hammer Creek Fire) and a large prescribed fire (North Fork Prescribed Fire) in the northern Rocky Mountains in August 2011. Two of the wildfires were sampled on multiple days. We have treated these sampling days as separate fires, identifying each as a “fire-day” (see Sect. 3.1), resulting in a total of 9 fire-day

emission datasets. The fire activity and meteorological conditions associated with the fires on each day of sampling is provided in Table 1. All fires in this study were sampled using a US Forest Service Cessna 206 aircraft equipped with atmospheric chemistry instrumentation as described in Sect. 2.2.

2.1 Site descriptions

All 4 fires occurred in high elevation mixed conifer forests of Lodgepole Pine, Douglas-Fir, Englemann Spruce, and Subalpine Fir. The vegetation involved was determined from a combination of ICS-209 reports (NWCG, 2012) and geospatial overlays of the incident fire perimeters (MTBS, 2012) and a US Forest Service Forest Type map (Ruefenacht et al., 2008; USDA, 2012a). Fire elevation was obtained from geospatial overlays of the incident fire perimeters and a digital elevation map (LANDFIRE, 2012).

2.1.1 Hammer Creek Fire

The Hammer Creek Fire was ignited by lighting on 19 July 2011 in the Bob Marshall Wilderness in northwestern Montana (longitude = −113.281, latitude = 47.518) and burned an estimated 2555 ha before being declared under control on 7 October 2011 (Carbonari, 2011c). The fire occurred at an elevation of 1360 m to 2250 m a.m.s.l. (above mean sea level). The incident management team reported the fire was burning in "mature timber with moderate to heavy dead standing and dead down" trees and also in the area of a previous burn with "moderate to heavy component of dead/down fuel" (Carbonari, 2011b).

2.1.2 Big Salmon Lake Fire

The Big Salmon Lake Fire started, cause undetermined, 16 August 2011 in the Bob Marshall Wilderness in northwestern Montana (longitude = −113.411, latitude = 47.519), about 10 km northwest of the Hammer Creek Fire. The fire burned in steep terrain at elevation of 1320 m to 2400 m a.m.s.l. and its perimeter encompassed

~ 2200 ha when declared controlled on 7 October 2011 (Carbonari, 2011a). With the exception of a ~ 70 ha pocket, the area of the Big Salmon Lake Fire had not been significantly impacted by fire in over 25 yr (MTBS, 2012). An aerial forest health survey conducted in 2010 found ~ 10 % of the area burned by the Big Salmon Lake fire area was impacted by mortality due to beetles (USDA, 2012b).

2.1.3 Saddle Complex

The Saddle Complex was a fire complex along the Idaho–Montana border that formed when the Saddle Creek Fire and Stud Fire merged on 18 August 2012. The Saddle Creek Fire was ignited by lightning on 10 August in the Bitterroot National Forest in Montana. The Stud Fire which, was also caused by lightning, began on 14 August in the Salmon-Challis National Forest in Idaho. The fire complex was managed as two separate fire incidents (Salmon-Challis Branch and the Bitterroot Branch). The fire burned in complex terrain at an elevation of 1040 m to 2650 m a.m.s.l. with a final perimeter area of 13 770 ha. A substantial portion of the trees in the impacted forest were dead from insect kill (> 40 % Bitterroot Branch and 20–45 % Salmon-Challis Branch; Central Idaho Dispatch, 2011; McKee, K., 2011).

On the four days the Saddle Complex was sampled its perimeter grew 200 to 800 ha day^{-1} and fire activity included group torching of tree crowns as well as running crown fire (Table 1). MODIS active fire detections and daily burn scars (RSAC, 2012a,b) were consistent with our airborne observation that significant fire activity and smoke emissions occurred within the perimeter, especially on 24 and 25 August.

2.1.4 North Fork Prescribed Fire

The North Fork Prescribed Fire was actually two burns ignited on 12 August 2011 in the North Fork District of the Clearwater National Forest in northern Idaho. The prescribed burn targeted diseased and insect-infested areas (Chaney, 2011). The fires

were ignited using a combination of aerial and hand ignition and allowed to burn with the goal of 400 ha eventually burning. The fire was sampled on 13 August.

2.2 Cavity Ring-down Spectroscopy (CRDS) trace gas analyzer

Continuous measurements of CO_2, CO, CH_4, and H_2O were obtained using a flight ready CRDS trace gas analyzer (Picarro Inc., CA, USA, model G2401-m).[1] In the CRDS technique, the gas sample flows through an optical cavity with partially reflecting mirrors. Light of a specific wavelength from a continuous wave laser is injected into the optical cavity through one of the partially reflecting mirrors. While the laser is on, light builds up in the optical cavity. The laser is abruptly turned off and the decay of light intensity is monitored with a photodetector after the light exits the cavity through a second partially reflecting mirror. The measured light decay is used to determine the optical absorbance of the gas sample and provide a mixing ratio measurement of a particular gas species. A specific gas is measured by scanning a continuous wave laser over an individual spectral line of the targeted gas. The G2401-m analyzer used in this study scans lasers over the individual spectral lines of CO_2, CO, CH_4, and H_2O at wavelengths between 1560 nm and 1650 nm. The precise wavelengths used for monitoring are considered proprietary information and would not be released by the instrument's manufacturer. The analyzer tightly controls the gas sample pressure and temperature at ±0.005 °C and ±0.0002 atm to provide stable, well-resolved spectral features and ensure high precision measurements. The data acquisition rate was 2 s.

Frequent, in-flight, calibrations using 3 standard gases were used to maintain accuracy of the CRDS measurements and quantify the measurement precision. The in-flight standards were gas mixtures of CO_2, CO, and CH_4 in Ultrapure air and included or were cross-calibrated against two NIST-traceable gas mixtures (concentration in ppm ± reported analytical uncertainty: $CO_2 = 351 \pm 4$ and 510 ± 5;

[1] Tradenames are presented for informational purposes only and do not constitute endorsement by the US Department of Agriculture.

CO = 0.092 ± 0.0092 and 3.03 ± 0.06; $CH_4 = 1.493 \pm 0.015$ and 3.03 ± 0.03) (Scott-Marrin, Inc., Riverside, CA, USA). In the laboratory, a five point calibration, using an additional high span CO standard and Ultrapure air were used to ensure linearity of the CO calibration between the instrument limit of detection (~ 0.030 ppm CO, defined as the 15 s standard deviation while measuring a calibration standard) and 10 ppm.

In recent years, the CRDS technique has been successfully used for high accuracy/high precision measurements of CO_2, CH_4, and CO from airborne platforms (Beck et al., 2012; Chen et al., 2010a). To our knowledge, our study is the first to employ this technique for the in-situ measurement biomass burning emission factors.

2.3 Airborne sampling

Emissions were measured by sampling the smoke above the flame front and up to 40 km downwind at elevations between 300 m a.g.l. (above ground level) and plume top. It is not unusual for wildfires in complex forested terrain to spread unevenly across the landscape due to changing weather conditions and variability in fuels and terrain resulting in a burn with mixed severity (Arno, 1980; Hudec and Peterson, 2012; Schwind, 2008). The wildfires sampled in this study had active fire occurring, often discontinuously along a large portion of the fires' perimeters. Active fire was also typically scattered throughout the perimeter interior as areas unburned or lightly burned during progression of the initial fire front burned/re-burned. Pockets of vigorous fire activity within the perimeter appeared to entrain and loft smoke from the surrounding smoldering fuels. A typical sample run began a few km upwind of the fire in smoke free air, providing a background sample, and then penetrated the smoke plume immediately downwind of the fire front. Often, after passing over a segment of the fire front the sample run would continue to sample smoke in the plume for a several km downwind. Sample runs often encountered multiple smoke plumes as interior regions of the perimeter with active fire were transected. Smoke sampling also included level elevation transects perpendicular to the direction of the smoke transport at distances of 2–40 km downwind of the fire front. Sampling in this second mode typically crossed the entire width of the plume

and provided measurements of background air on one or both ends of the sample run. The extensive downwind transects of smoke emissions obtained in this study may be used for the validation of smoke dispersion models. However, the focus of this paper is limited to EF.

2.4 Emission Factor Calculations

Multiple smoke samples were collected on each day of fire sampling. For each smoke sample the excess mixing ratio (EMR) of compound X, ΔX, was calculated for each 2 s data point by subtracting the average background ($X_{background}$) for that sample run ($\Delta X = X_{smoke} - X_{background}$). Sample emission factors for the each compound X, EFX (grams of species X emitted per kilogram dry fuel burned), was calculated from the 2 s ΔX using the carbon mass balance method (Yokelson et al., 1999) following two approaches. Approach 1 (Eq. 1) used the integrated ΔX for each plume sample while, the second approach (Eq. 2) used emission ratios determined from linear regression fits, with the intercept forced to 0, of ΔX vs. ΔCO or ΔCO_2 using the 2 s data points. The emission ratio of CH_4 to CO_2, $\Delta CH_4/\Delta CO_2$, was calculated as the product of $\Delta CH_4/\Delta CO \times \Delta CO/\Delta CO_2$. In Eq. (1) ΔC_i are the excess mass mixing ratios of carbon (C) in each species. In Eqs. (1) and (2) MM_X is the molar mass of X ($g\,mole^{-1}$), 12 the molar mass of carbon ($g\,mole^{-1}$), and F_C is the mass fraction of carbon in the dry biomass, assumed to be 0.50. We assumed $F_C = 0.50$ based on studies which found that F_C ranged between 0.45 and 0.55 for the vegetation types involved in this study (Burling et al., 2010). The majority of carbon mass (> 95 %) in biomass smoke is contained in CO_2, CO, and CH_4, therefore our neglect other carbon-containing species in the carbon mass balance method over estimates the EF by ∼ 5 % (Yokelson et al.,

2007b).

$$EFX = F_C \times 1000\,(g\,kg^{-1}) \times \frac{MM_X}{12} \times \frac{\Delta X}{\Delta C_{CO_2} + \Delta C_{CO} + \Delta C_{CH_4}} \quad (1)$$

$$EFX = F_C \times 1000\,(g\,kg^{-1}) \times \frac{MM_X}{12} \times \frac{\frac{\Delta X}{\Delta CO_2}}{\frac{\Delta CO_2}{\Delta CO_2} + \frac{\Delta CO}{\Delta CO_2} + \frac{\Delta CH_4}{\Delta CO_2}} \quad (2)$$

The chemical composition of emissions from biomass fires are related to the combustion characteristics of the fire, in particular the relative amounts of flaming and smoldering combustion. Some species are emitted almost exclusively by flaming or smoldering, while the emissions of others are significant from both processes. Flaming combustion produces the gases CO_2, NO, NO_2, HCl, SO_2, HONO and N_2O (Burling et al., 2010; Lobert, 1991) and black carbon particles (Chen et al., 2007; McMeeking et al., 2009). The species CO, CH_4, NH_3, many NMOC, and primary organic aerosol (OA) are associated with smoldering combustion (Burling et al., 2010; McMeeking et al., 2009). Several NMOC have been linked with both flaming and smoldering combustion (Burling et al., 2010; Lobert, 1991; Yokelson et al., 1996).

Modified combustion efficiency (MCE; Eq. 3) is used to characterize the relative amount of flaming and smoldering combustion (Akagi et al., 2011; Ward and Radke, 1993). Laboratory studies have shown MCE is ∼ 0.99 for pure flaming combustion (e.g. fine fuels completely engulfed in flame, (Chen et al., 2007; Yokelson et al., 1996)), while the MCE for smoldering combustion varies over ∼ 0.65–0.85, with 0.80 being a typical value (Akagi et al., 2011). Since many species are predominantly emitted during either flaming or smoldering combustion, the EF of many compounds correlate with MCE. Laboratory studies of the combustion of fine fuels (Burling et al., 2010; Christian et al., 2003; McMeeking et al., 2009; Yokelson et al., 1997) and recent field measurements of emissions from prescribed fires (Burling et al., 2011) have found a strong correlation between EF and MCE for many species. Given the utility of MCE for characterizing combustion characteristics and its potential for estimation of EF for a range

of compounds, we have calculated MCE for all fresh smoke samples. But, we note that two laboratory studies of pure smoldering combustion of duff, organic soils, and CWD found poor correlation between MCE and EF (Bertschi et al., 2003; Yokelson et al., 2007a). However, since the combustion of CWD and duff in the natural environment is dependent on fuel bed characteristics such as the loading and arrangement of CWD and the presence of fine dead wood and litter (Albini et al., 1995; Ottmar et al., 1989), laboratory studies may not be a good proxy for these fuels. Nonetheless, it is possible that wildland fires involving a large component of CWD and duff may not show a strong MCE–EF relationship.

$$\mathrm{MCE} = \frac{\Delta CO_2}{\Delta CO_2 + \Delta CO} = \frac{1}{1 + \frac{\Delta CO}{\Delta CO_2}} \tag{3}$$

3 Results and discussion

3.1 Emission measurements

Fire perimeters, area of active burning, and region of smoke sampling from a representative fire-day, the Saddle Complex on 24 August, are shown in Fig. 1. The perimeters, as observed via airborne IR sensor on the evenings (23 and 24 August), indicate that on 24 August the fire growth occurred mostly on the west and east ends, with some minor growth along the northern and southern edges. In addition to the active fire fronts on the perimeter we also observed many pockets of burning scattered within the perimeter while sampling on the afternoon of 24 August. The MODIS burn scar data (RSAC, 2012b) and active fire detections (RSAC, 2012a) for that day captured some of this activity (Fig. 1). On this day, fresh smoke samples were obtained along the northern edge of the fire perimeter. Winds were from the WSW and the initial portion of our sampling runs captured emissions emanating from the within the western area of the perimeter just downwind as they reached neutral buoyancy. The runs proceeded to

the ENE sampling smoke above the fire front on the northern perimeter and then continued downwind with the plume that entrained smoke from across the fire complex. CRDS measurements for a fresh smoke sample run on 24 August (Table 2, sample SC2402) are shown in Fig. 2. The dashed line in each plot marks the background mixing ratios measured upwind of the fire. The background mixing ratios for this sample (CO_2 = 382.56 ppm, CH_4 = 1.856 ppm, and CO = 0.110 ppm) were typical of the background for all fire-days.

EF, MCE, and average ΔX for all 63 fresh smoke samples are given in Table 2. The EF in Table 2 were calculated from integrated ΔX using Eq. (1) (Sect. 2.4). The fire-day average EF calculated using Eqs. (1) and (2) agreed within 10 %. Some plume samples were taken significant distances downwind of the source. In particular, on 17 August, samples were taken 40 km downwind of the Big Salmon Lake Fire. The afternoon atmospheric sounding at Great Falls, Montana (NOAA, 2012) on this day indicated the transport winds were $\sim 11\,\mathrm{m\,s^{-1}}$ implying a smoke age of ~ 60 min for these samples. However, since CO_2, CO, and CH_4 are fairly non-reactive in the atmosphere (CO, the most chemically reactive of the 3 gases, has a lifetime > 30 days with respect to chemical reaction; Seinfeld and Pandis, 2006) the age will not impact the measured EF for these gases.

The Big Salmon Lake Fire and the Saddle Complex were sampled on multiple days and as mentioned previously we have treated these sampling days as separate fires, identifying each as a "fire-day". We believe this treatment is justified given the complex terrain, heterogeneous fuels, and the inter-day variability in metrological conditions and observed fire behavior (see Table 1). Furthermore, one day is an appropriate temporal scale for atmospheric chemical modeling applications since most biomass burning emission inventories provide estimates on a daily basis, from which models then create an hourly profile based on assumptions about diurnal fire behavior cycles. Our study average values (average of the 9 fire-day values) for MCE, $EFCO_2$, EFCO, and $EFCH_4$ are 0.883, 1596 $\mathrm{g\,kg^{-1}}$, 135 $\mathrm{g\,kg^{-1}}$, 7.30 $\mathrm{g\,kg^{-1}}$, respectively. Figure 3a–c shows the average, range, and $\pm 1\sigma$ of MCE, EFCO, and $EFCH_4$ for each fire-day. The

fire-day average values of EFCO and $EFCH_4$ are confined to a fairly narrow span of 26 % and 21 % of the study average, respectively, and the standard deviations are only ~ 10 % of the study average (Table 2). This muted inter-fire-day variability supports the idea that the dataset average values are more broadly representative of wildfire season forest fires in the western US. We note that despite the limited span of MCE and $EFCH_4$ observed in our study, our measurements are sufficiently precise to reveal an MCE–$EFCH_4$ relationship. CH_4 is produced by smoldering combustion processes, and as expected, $EFCH_4$ has a strong inverse correlation with MCE (Fig. 3d; $r = -0.87$, p-value = 0.002).

3.2 Comparison with other studies

We compare our results with previous field studies of emissions from fires in temperate conifer dominated forests in the US and southern British Columbia, Canada: the airborne studies of Burling et al. (2011, hereafter B11), Hobbs et al. (1996, hereafter H96) and Radke et al. (1991, here after R91) (Burling et al., 2011; Hobbs et al., 1996; Radke, 1991), and the tower based study of Urbanski et al. (2009, hereafter U09) (Urbanski et al., 2009). B11 studied understory prescribed fires in conifer dominated forests of North Carolina and the Sierra Nevada Mountains of California. H96 studied 3 prescribed burns of clear cut logging slash on the Washington and Oregon coasts and the Corral-Blackwell Complex wildfire which occurred in northern Idaho. From R91 we consider their results for 2 wildfires in western Oregon and a prescribed burn of hemlock, deciduous, Douglas-fir logging debris in British Columbia. U09 reported on understory prescribed fires in mixed conifer forests of the interior mountain west and in conifer and conifer/hardwood forests of the southeast US.

For comparison we grouped the prescribed fires of the 4 previous studies into 3 regions: southeast (SE), southwest (SW), and northwest (NW). The U09 Arizona fires were assigned to SW and the Montana, Oregon, and British Columbia fires were assigned to NW. The southeastern conifer and conifer/hardwood forest fires in U09 were assigned to SE. We included 6 fires listed in the grasslands and shrublands section

of Table A1 in U09 (EB1, EB2, FL5, SC9, FS1, ICl3) in SE since these fires were, in fact conifer/hardwood understory burns. They were listed as grassland/shrub in U09 since the fuel consumed was overwhelmingly grass and shrubs in the understory. In this sense these fires were very similar to the southeast burns studied in B11. We assigned the 6 North Carolina fires of B11 to SE and the 2 Sierra Nevada fires to NW. The H96 and R91 prescribed fires were included in the NW set.

First we compare our wildfire results (WF) with the prescribed fire data. We include the North Fork Prescribed Fire in our WF results since it burned during the wildfire season. Fire average MCE, EFCO, and $EFCH_4$ from this study, B11, U09, H96, and R91 are shown in Fig. 4a–c. Figure 4a–c includes the wildfire measurements of H96 and R91, however in the ensuing discussion WF refers strictly to the results of our study (Table 2). On average the fires sampled in our study burned with a lower combustion efficiency compared to the prescribed fires. The data show a clear trend in average MCE across categories: SE (0.933) > SW (0.922) > NW (0.900) > WF (0.883). There is no overlap of the WF MCE with those of the SE and SW fires. The MCE of B11's Shaver fire (0.885) and the average MCE of H96 (0.877) are both close to the WF average MCE. These four prescribed fires involved heavy loads of down dead wood due to logging in the case of H96 and pine beetle activity in the Shaver fire (see Sect. 3.3). There is also a pronounced trend across categories for EFCO (WF (135) > NW (111) > SW (88) > SE (76)) and $EFCH_4$ (WF (7.30) > NW (6.29) > SW (3.32) > SE (2.13)). This work does not report $EFPM_{2.5}$; however, we note that the $EFPM_{2.5}$ of B11 and U09 exhibit a similar trend (NW (18.0) > SW (14.5) > SE (12.6)).

There is limited temperate forest wildfire data with which we can compare our measurements. Figure 4a shows the R91 wildfires had MCE (0.921, 0.907) above the range measured in our study while the Corral-Blackwell Complex MCE (0.810) was significantly lower. The Corral-Blackwell Complex burned in forest and terrain very similar to that of the fires studied in this work. The fire, which occurred 120 km west of the Saddle Complex, burned in mixed stands of Lodgepole pine, Engelmann spruce, and subalpine fir at an elevation of 600 m to 2735 m (Farris et al., 2008). H96 provide little

information on the Corral-Blackwell Complex, but do note that it was sampled "during smoldering combustion". The MCE of smoldering combustion has been found to range from ~0.65–0.85, but typically being near 0.80 (Akagi et al., 2011). Using a ground based FTIR, B11 measured post-flame front emissions from nearly pure smoldering combustion of dead tree stumps for a prescribed at Camp Lejeune, NC (The ground-based measurements of pure smoldering are not included in the B11 results discussed thus far and reproduced in Fig. 4). Since CWD can smolder for an extended period of time and can comprise a large share of fuel consumed in western forest fires (Brown et al., 1991; Reinhardt et al., 1991), the smoldering stump measurements of B11 serve as a reasonable analog for assessing the Corral-Blackwell Complex data reported by H96. The four dead stumps measured by B11 yielded averages of MCE = 0.795 and $EFCH_4 = 17.4\,g\,kg^{-1}$, similar to the Corral-Blackwell Complex values reported by H96 (MCE = 0.81, EFCH4 = $18.0\,g\,kg^{-1}$). If H96 sampled almost exclusively smoldering combustion this may explain the very low value of their MCE compared to that measured in this work for wildfires in similar terrain and forest. During our sampling we observed active flaming combustion on all days for all fires, usually included torching of tree crowns, and the emissions we measured originated from both active flaming and post-flame front smoldering combustion (see Fig. 1 and related discussion).

The Silver and Myrtle/Fall Creek wildfires sampled by R91 occurred in southwestern Oregon, which has a Mediterranean climate, and they burned in different vegetation and elevation than the wildfires sampled in our work. The Silver Fire burned in Douglas-Fir/Tanoak/Pacific Madrone forest between elevations of 75 m and 1500 m with an average of 800 m. R91 described the vegetation involved in the Myrtle/Fall Creek wildfires as "standing pine, brush, and Douglas-fir" and they burned at elevation of 300 m and 900 m (average = 615 m). The different vegetation involved in the R91 fires may explain the relatively high MCE they measured compared to the wildfires sampled in this work. The vegetation involved in the Silver Fire was determined from a combination of geospatial overlays of the fire perimeters (MTBS, 2012) and an existing vegetation map (LANDFIRE, 2012) and literature (Thompson and Spies, 2010). Fire elevation for both

R91 wildfires was obtained from geospatial overlays of the fire perimeters and a Digital Elevation Map (LANDFIRE, 2012).

3.3 Estimated wildfire emission factors and comparison with reference data

This work only measured EF for CO_2, CO, and CH_4. However, our study average MCE can be used to estimate EF for other species using the EF–MCE linear relationships reported by B11. We believe comparison of our $EFCH_4$ data with that of B11 justifies this approach. Figure 4d plots fire average $EFCH_4$ vs. MCE for this work and B11 and region average prescribed fire $EFCH_4$ based on data of U09, H96, and R91. Included in Fig. 4d is the $EFCH_4$ vs. MCE regression line of B11 with 95 % confidence intervals (calculated for this work using data from B11). We cite the following observations in support our MCE based EF estimates:

- Our study average MCE (0.883) falls just outside the range of the B11 MCE (0.885 to 0.957) and therefore is not a significant extrapolation of the B11 relationships
- Our study average $EFCH_4$ sits along the B11 $EFCH_4$ vs. MCE regression line and all of our fire average $EFCH_4$ fall within the 95 % confidence intervals (Fig. 4d)
- The coefficients obtained from a linear regression of combined fire average $EFCH_4$ from this work and B11 are not significantly different from the coefficients for the B11 data alone (based on Chow's test for heterogeneity in two regressions, F-value = 1.56, $df_1 = 2$, $df_2 = 21$, p-value = 0.23).

We used EF–MCE linear relationships of B11 and our study average MCE to estimate wildfire EF for 14 species which are given in Table 3 along with the EF measured in this work. For comparison we have included the temperate forest EF from the recent review of Akagi et al. (2011) (hereafter, A11) and effective EF from the US EPA 2008 National Emission Inventory version 2 (USEPA, 2012a) (hereafter, NEI). The NEI documentation reports that wildland fire EF were estimated "using the Fire Emissions Prediction Simulator which relies on EFs from the literature apportioned by flaming and

smoldering combustion", but does not provide the complete set of EF used ((USEPA, 2012a), documentation, Chapter 5, page 125). For comparison with EF from this work and A11 we derived effective forest fire EF from the NEI Supporting Data and Summaries, Sect. 5 ("Fires") supplemental data (USEPA, 2012a). The effective EF were derived as follows: from the wildland fire location file (WF_locations_ALL.xlsx) we extracted all wildfires in forest ecosystems (fires type = "WF" and "canopy" > 0) in the western US and then from the extracted fires calculated effective EF for species X as the sum of emissions of X for all fires divided by the sum of fuel consumed for all fires. The NEI effective MCE was based on $\Delta CO/\Delta CO_2$ calculated from the effective EF and molecular masses of CO and CO_2 ($\Delta CO/\Delta CO_2 = MM_{CO_2}/MM_{CO} \times EFCO/EFCO_2$).

For most species our measured and estimated wildfire EF are substantially larger than those in A11. This is expected given our wildfire MCE is much lower than their temperate forest MCE. The ratio of wildfire EF to the recommendations of A11, $R_{WF/A}$, were 1.9 for CH_4, ~ 3 for phenol and furan, and 4.1 in the case of glycolaldehyde. Wildfire EF for $PM_{2.5}$, methanol and acetic acid were also markedly higher, with $R_{WF/A} = 2.0$, 1.6 and 1.9, respectively. For a few compounds the EF were little changed, and EFHCOOH is actually lower than A11. For NO_x ($NO + NO_2$), a product of flaming combustion, $R_{WF/A}$ was 0.77. We note that the nitrogen content of fuel also plays an important role in the emissions of both NO_x and NH_3 (Burling et al., 2010).

The NEI effective EF (hereafter referred to as simply NEI EF) are only partially consistent with those measured in this study and that predicted by the EF–MCE regression equations of B11. The NEI MCE of 0.847 is significantly lower than our wildfire MCE and the $EFNH_3$ and EFNOx are commensurately higher and lower, respectively, each being within 10 % of that predicted based on the MCE equations of B11. However, for other species the NEI EF are significantly lower than expected. Despite the large difference in MCE, the NEI $EFCH_4$ is only 10 % larger than our measured wildfire $EFCH_4$. The NEI EF for CH_4, HCHO, and $PM_{2.5}$ are substantially lower than the B11 EF–MCE regression equations predict at MCE = 0.847. The ratios of NEI EF to predicted EF are 0.70, 0.38, and 0.40 for CH_4, HCHO, and $PM_{2.5}$, respectively. The NEI includes VOC

(the fire emissions documentation does not provide a speciation of this category) and their effective EFVOC (39.6 $g\,kg^{-1}$) is comparable to the estimated wildfire EF for the sum of identified and unidentified NMOC (Table 3). Extrapolating the B11 EF–MCE regression equations to the NEI MCE for the 11 NMOC predicts EFNMOC = 22.0 $g\,kg^{-1}$. Adding 1.46 $g\,kg^{-1}$ to this value to account for ethane, propane, and butane (see Table 3) and then doubling this sum to account for unidentified NMOC (see A11) leads to EFNMOC = 47.0 $g\,kg^{-1}$, 19 % larger than the NEI EFVOC. This indicates that despite the apparent large underestimate in $EFPM_{2.5}$, the NEI EFVOC is roughly in-line with expectations base on previous studies (B11; A11).

Our field measurements suggest that western US wildfires burn with an MCE significantly lower than most of the temperate forest prescribed fires reported in the literature (U09, B11, R91) and used to develop EF recommendations for atmospheric modeling (A11; Andreae and Merlet, 2001). The lower MCE of western US wildfires indicates these fires have larger EF for species associated with smoldering combustion processes ($PM_{2.5}$ and NMOC) than are reported in previous studies and reviews (U09; B11; A11; Andreae and Merlet, 2001). If the wildfires sampled in our study are representative more generally of wildfires in western US forests, then use of EF based on temperate forest prescribed fires will significantly underestimate $PM_{2.5}$ and NMOC emissions. Because large wildfires dominate fire burned area, fuel consumption, and emissions in the western US (Urbanski et al., 2011), this has important implications for the forecasting and management of regional air quality. The western US wildfire $PM_{2.5}$ emissions reported in the most recent national emission inventory is based on an effective $EFPM_{2.5}$ that is approximately a factor of 2 lower than that expected based on our wildfire field measurements and published $EFPM_{2.5}$–MCE relationships. Given the magnitude of biomass consumed by western US wildfires, the failure to use wildfire appropriate $EFPM_{2.5}$ has significant implications for the forecasting and management of regional air quality. The contribution of wildfires to NAAQS $PM_{2.5}$ and Regional Haze may be underestimated by air regulatory agencies. This is especially true considering

that compared with anthropogenic and biogenic emission sources, wildfire emissions are highly concentrated both temporally and spatially (Urbanski et al., 2011).

3.4 MCE, EF, and fire characteristics

The MCE we measured for wildfires are significantly lower than those reported in the literature for prescribed fires in temperate conifer forests. There are also distinct regional differences in the published prescribed fire MCE (Fig. 4a). Factors that affect the combustion process, in particular environmental conditions (e.g. wind speed, topography) and fuel characteristics (e.g. moisture, chemistry, soundness of dead wood, geometry and arrangement of fuel particles) (Ottmar, 2001; Sandberg et al., 2002) will also influence MCE. Fine fuels, those with high surface to volume ratios, such as grasses, conifer needles, and fine woody debris (diameter < 7.6 cm) have a tendency to burn by flaming combustion with a high MCE (Chen et al., 2007; Ottmar, 2001; Sandberg et al., 2002; Yokelson et al., 1996). Smoldering combustion, which has a lower MCE, is more prevalent in CWD, duff, and organic soils (Bertschi et al., 2003; Burling et al., 2011; Ottmar, 2001; Sandberg et al., 2002; Yokelson et al., 1997). Reviews of field studies show that fires in ecosystems dominated by fine fuels such as grasslands and savannas burn with a higher MCE than forest fires (Akagi et al., 2011; Andreae and Merlet, 2001; Urbanski et al., 2009). In addition to fuel geometry and arrangement, recent laboratory studies suggest a linkage between fuel moisture and MCE, with MCE tending to increase with decreasing fuel moisture for a constant fuel type and fuel mass (Chen et al., 2010b; McMeeking et al., 2009). An analysis of emission field measurements for multiple biomes found evidence that the spatio-temporal variability in MCE could be partially attributed to fraction of tree cover and monthly precipitation (van Leeuwen and van der Werf, 2011), the later which is presumably a surrogate for fuel moisture.

Considering the influence of fuel moisture and the tendency of certain fuel types to favor flaming or smoldering combustion, one might expect higher fuel moisture and/or the involvement of heavy fuels (CWD and duff) to result in fires with lower MCE. However, the combustion completeness of CWD and duff increases with decreasing fuel

moisture (Albini and Reinhardt, 1997; Brown et al., 1991; Ottmar et al., 2006; Ottmar, 2001), while that of fine woody debris, grasses, and litter is relatively insensitive to moisture once ignition is achieved (Ottmar et al., 2006; Ottmar, 2001). Because the moisture contents of different types of fuel particles respond to environmental conditions with different time-lags, there can be a large difference in the moisture content of fuel bed components. The moisture content of fine fuels like cured grasses, litter, and small twigs (< 0.64 cm diameter) adjusts to environmental conditions with a time-lag on the order of 1 h (these are often referred to as 1-h fuels; (Bradshaw et al., 1984)). In contrast, CWD and duff respond with a time-lag of around 1000 h (1000-h fuels; Bradshaw et al., 1984; Brown et al., 1985; Harrington, 1982). Therefore, at a given forest stand, under conditions typical of a springtime prescribed burn, consumption of heavy fuels may be minimal due to the high fuel moisture content of these components. However, at the same site under wildfire conditions, when the moisture content of heavy fuels is low, these components may comprise the majority of fuel consumed. Thus, despite the lower fuel moisture during the wildfire season, one might expect a fire with lower MCE compared with a springtime prescribed fire in the same forest stand due to the greater involvement of heavy fuels which favor smoldering combustion processes.

We believe the relatively low MCE of the wildfires and the general trend in MCE across regions is partially attributable to the differential involvement of heavy fuels. The Big Salmon Lake, Hammer Creek, Saddle Complex wildfires and the North Fork Prescribed fire involved significant areas of dead standing and dead down trees (Sect. 2.1). The 6 SE understory conifer fires reported in B11 occurred under conditions of high duff moisture and the fuels burned were predominantly shrubs, litter, grass, and fine woody debris (B11; Reardon, 2012). Pre and post fuel loading measurements taken at two of the B11 NC sites (the two Camp Lejeune burns) indicate CWD and duff were < 15 % of the fuel mass consumed (Reardon, 2012). While the SE burns of B11 involved predominantly fine fuels, their Sierra Nevada burns (Turtle burn and Shaver burn) involved moderate to heavy loadings of dead wood. At the Turtle burn site litter and 1-h dead wood comprised only ∼ 1/3 of the surface dead fuel loading (Gonzalez, 2009). The

primary fire carrier was expected to be dead woody fuels and pockets of chaparral were not expected to burn except where covered with pine needle drape (Gonzalez, 2009). The site of the Shaver burn had dead woody fuel loadings of up to 28 $kg\,m^{-2}$ due to mountain pine beetle activity and the lack of previous fire (B11). Perhaps coincidently, the MCE measured for the Shaver burn (0.885) was roughly equal to the average MCE (0.883) of the wildfires studied in this work which also burned in forests with areas of standing dead trees and heavy down dead wood.

In contrast to the B11 SE burns, which were characterized by high fuel moistures, the region of the Shaver and Turtle burns experienced only ~ 0.50 cm of precipitation in the 27 days preceding the burns, and none in the two weeks prior (WFAS, 2012). Consequently, at the time of the Shaver and Turtle burns, the heavy fuels had fairly low moisture content (1000-h = 18 %, (WFAS, 2012)) and likely comprised a significant portion of the fuel mass consumed. This comparison of the B11 prescribed fires and the wildfires suggests the presence of heavy fuels (CWD and duff) and conditions favorable for their burning results in fires with a greater fraction of smoldering combustion, a lower MCE, and higher emissions of species associated with smoldering.

Given the lack of fuel consumption data for the wildfires and all but 2 of the B11 prescribed fires our argument is highly speculative. However, fuel consumption data is available for 13 prescribed fires from U09 and for the 3 prescribed fires of H96. To test our argument that the consumption of heavy fuels favors lower MCE we compared the ratio of heavy fuel consumption to total fuel consumption (HFF) versus MCE for the 18 prescribed fires with fuel consumption data (see Appendix A for details). The results, plotted in Fig. 5, show a strong negative correlation between HFF and MCE ($r = -0.83$, p-value = 1.7e-5), as heavy fuels comprise a larger fraction of the total fuel consumed the fire average MCE decreases.

The analysis presented in Fig. 5 indicates the consumption of heavy fuels favors smoldering combustion, a finding consistent with previous ground based studies of prescribed burns in logging slash and guidelines for smoke management (Ottmar, 2001; Sandberg et al., 2002). However, we emphasize that our conclusion is based on a small

sample size and involves significant uncertainty regarding the representativeness of emission sampling. The fuel consumption measurements quantify the fuel consumed over the entire life of the burn. Since smoldering combustion may continue for many hours after the active flame front has passed (Ottmar, 2001; Sandberg et al., 2002) it is unlikely the emissions sampling is properly weighted for smoldering emissions. Due to this temporal mismatch between emissions and fuel sampling it is possible the contribution of smoldering emissions may be underrepresented in the MCE and EF measurements. Further, given the variability in fuel loading and fire characteristics (spread rate, ignition method) the degree of sampling bias with respect to smoldering emissions may vary among burns. For these reasons the data may not be suitable for predicting MCE. Nonetheless, the analysis identifies relative heavy fuel consumption as a driver of fire average MCE and provides an explanation for the differences in MCE measured for temperate forest fires.

van Leeuwen and van der Werf (2011) developed a global, biome-independent MCE model. This continuous MCE model, a multivariate regression of field measured MCE versus coarse-scale (monthly, 0.5° × 0.5°) environmental parameters, was driven primarily by monthly precipitation and fraction tree cover (FTC), and explained about 34 % of the variability in the field measured MCE. They were unable to account for fuel composition due to lack of consistent data, but suggested it may be a crucial factor driving the MCE variability not captured by their analysis. The authors also explored biome stratified emissions data and highlighted a strong negative correlation between MCE and FTC for fires in Australian Savannas and deforestation fires in Brazil. If the loading of CWD is proportional to FTC, then the heavy fuel combustion–MCE dependence we have identified may help explain their observed FTC-MCE relationship. Since their model is biome-independent and aggregates across grasslands, savannas, and forests it is possible the global correlation they have observed between FTC–MCE reflects fuel composition, with FTC serving as a proxy for CWD loading.

4 Conclusions

Over 8 days in August of 2011 we sampled emissions from 3 wildfires and a prescribed fire that occurred in mixed conifer forests of the northern Rocky Mountains. We measured MCE and EF for CO_2, CO, and CH_4 using a CRDS gas analyzer. We believe this study may be the first to apply in-flight CRDS technology to characterize the emissions from open biomass burning in the natural environment. The combustion efficiency, quantified by MCE, of the fires sampled in this work was substantially lower than the average MCE measured in previous field studies of prescribed fires in similar forest types (conifer dominated temperate forests) and that reported in recent review articles of biomass burning emissions. In comparison to previous field studies of prescribed fires and review articles, the fires studied in this work measured lower MCE and $EFCO_2$ and higher EFCO and $EFCH_4$. An examination of results from our study and 47 temperate forest fires from previously published studies show a clear trend in MCE across region/fire type: southeast (MCE = 0.933) > southwest (MCE = 0.922) > northwest (MCE = 0.900) > wildfires (MCE = 0.883). The fires sampled in this work burned in areas reported to have moderate to heavy components of standing dead trees and dead down wood due to insect activity and in the case of one fire, a previous burn. Of previously published field measurements of prescribed fires the few with MCE similar to that measured in our study also burned in forests with heavy loadings of large dead wood and/or duff.

Fuel consumption data was not available for any of the fires sampled in this study; however, it was available for 18 prescribed fires reported in the literature. For these 18 fires we found a significant negative correlation between MCE and the ratio of heavy fuel (CWD and duff) consumption to total fuel consumption. This observation suggests the comparatively low MCE measured for the fires in our study results from the availability of heavy fuels and conditions that facilitate combustion of these fuels (e.g. low moisture content). More generally, our measurements and the comparison with previous studies indicate that fuel composition is an important driver of EF variability.

Considering the accumulation of heavy fuels in western US forests due to factors such as fire exclusion and insect induced mortality (see for example Klutsch et al., 2009), the MCE and EF measured in this study and those we have estimated based on EF–MCE relationships, may be representative of wildfires in forests across the western US.

The temperate forest EF reported in the literature are based on fires which burned with higher combustion efficiency (i.e. a lower relative fraction of smoldering combustion) than the wildfires sampled in our study. Because the EF of many smoldering combustion species have a strong negative correlation with MCE, the EF found in the literature may significantly underestimate the true EF for smoldering species for fires with combustion characteristics similar to the wildfires measured in this work. EF–MCE linear relationships from the literature and our study average MCE were used to estimate wildfire EF for 14 species. If the MCE of the fires sampled in this work are representative of the combustion characteristics of wildfires in western US forests, this analysis indicates that the use of literature EF will result in a significant underestimate of wildfire $PM_{2.5}$ and NMOC emissions. The most recent national emission inventory reports western wildfire emissions of $PM_{2.5}$ based on an effective $EFPM_{2.5}$ that is less than half that estimated in this study. Given the magnitude of biomass consumed by western wildfires, the failure to use wildfire appropriate $EFPM_{2.5}$ has significant implications for the forecasting and management of regional air quality. The contribution of wildfires to NAAQS $PM_{2.5}$ and Regional Haze may be underestimated by air regulatory agencies.

Our study sampled 4 fires over 8 days for a total of 9 fire-day observations. The fires burned in similar environments: high elevation, mixed conifer forest of Lodgepole Pine, Douglas-Fir, Engelmann Spruce, and Subalpine Fir with significant insect induced tree mortality and moderate to heavy loadings of standing dead and down dead wood. Our measured MCE and EF and the EF estimated from EF–MCE relationships may not be applicable to all wildfires in western US forests. The presence of heavy loadings of standing dead trees and dead down wood may have been the main factor driving the MCE and EF of these fires. Additionally, our wildfire measurements did not include

fires in ponderosa pine dominated forests which are characterized by lower loadings of CWD (Graham et al., 1994). Other forests types or forests with a different disturbance history may not have similar loadings of heavy fuels and the therefore the MCE and EF (measured and estimated) reported here may not be applicable. Future emission studies focusing on other regions (e.g. Southern Rocky Mountains), forest types (e.g. ponderosa pine dominated), and forests with different disturbance histories are needed to better quantify $PM_{2.5}$ and NMOC emissions from wildfires in the western US.

Appendix A

Qualitative reports indicate the low MCE fires sampled in our study involved significant consumption of CWD and standing dead trees. In contrast, previous studies of prescribed burns in the southeastern US (B11, U09) measured relatively high MCE and mostly anecdotal observations suggested these fires consumed mostly fine fuels with the consumption of CWD and duff being minimal. This pattern is not unexpected since fine fuels have a tendency to burn by flaming combustion, while CWD and duff favor smoldering combustion processes (Sandberg et al., 2002). Using previous studies of 18 prescribed burns for which detailed fuel consumption data was available, we tested for a relationship between fire average MCE and the composition of fuel consumed. Specifically we tested for a significant correlation between the relative amount of flaming and smoldering combustion, quantified by MCE, and the relative amount of heavy fuel and fine fuel consumption. The later fire characteristic was quantified with the heavy fuel fraction (HFF), defined as the sum of CWD and duff fuel loading consumed divided by the sum of total fuel loading consumed. HFF is given by equation A1 where C_i is consumption ($\mathrm{kg\,m^{-2}}$) of fuel component i and fine fuels includes grasses, shrubs, foliage, litter, and fine woody debris (small diameter (< 7.62 cm) dead wood):

$$\mathrm{HFF} = \frac{C_{\mathrm{CWD}} + C_{\mathrm{duff}}}{C_{\mathrm{CWD}} + C_{\mathrm{duff}} + C_{\mathrm{fine\,fuels}}} \tag{A1}$$

Name and location of the prescribed burns, MCE, fuel consumption by class, and references for the 18 fires used in this analysis are provided in Table A1.

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Table 1. Fire activity and meteorological conditions for fires on the days they were sampled.

Date	Fire	Growth (ha)	T^1_{MAX} (K)	RH^1_{MIN} (%)	Fuel moisture[2] (%) 10-h	1000-h	Fire activity[3]
13 Aug 2011	North Fork Prescribed	N/A					Prescribed fire with initial ignition on 12 August.
17 Aug 2011	Big Salmon Lake	N/A	300.4	17	5	14	Significant spotting up to 0.4 km and sustained crown runs with wind-driven and terrain induced spread.
22 Aug 2011	Big Salmon Lake	20[5]	302.0	24	5	14	Fire spread was low to moderate with creeping and smoldering combined with some single tree torching
22 Aug 2011	Hammer Creek	0[5]			5	14	Fire activity was mostly creeping and smoldering with some isolated single tree and group torching observed along the perimeter.
24 Aug 2011	Saddle Complex	767[4]	300.4	22	4	10	Bitterroot branch (BR): Upslope runs Salmon-Challis branch (SC): group torching and short runs
25 Aug 2011	Saddle Complex	352[4]	298.2	25	5	10	BR: backing, isolated torching and moderate fire behavior SC: Group torching and short runs
			297.6	17			
26 Aug 2011	Saddle Complex	235[4]	302.0	14	4	10	BR: Fire backing down slope, upslope crown runs from rolling material, small columns developed in isolated areas of the fire SC: Group torching and short runs
			297.6	18			
27 Aug 2011	Saddle Complex	471[4]	301.5	14	5	9	BR: moderate fire behavior, isolated and group torching, spotting, and crown runs SC: NA
28 Aug 2011	Big Salmon Lake	405[5]	304.8[2]	12[2]	4	13	Moderate to high with active ground fire with group torching.

[1] Weather observations are from the Incident Command System 209 Reports (ICS-209; (NWCG, 2012)) unless otherwise noted.
[2] Fuel moisture data and some T and RH data from fire weather stations archived by the USFS–Wildland Fire Assessment System (WFAS, 2012). Observations are from the following weather stations: Big Salmon Lake and Hammer Creek – WIMS# 241596; Saddle Complex – WIMS# 101019.
[3] Fire activity from ICS-209 (NWCG, 2012).
[4] Daily burned area growth estimated from fire perimeters (Geospatial Multi-Agency Coordinating Group (GeoMAC) online database: http://www.geomac.gov/index.shtml; last accessed: 30 November 2011.
[5] Daily burned area growth estimated from ICS-209.

Table 2. Smoke sample EMR, MCE, and EF and fire–day average (±1 standard deviation) MCE and EF. EMR are sample average and are in units of ppmv. EF are in units of $g\,kg^{-1}$.

Sample	Time	Location	n^1	ΔCO_2	ΔCO	ΔCH_4	MCE	$EFCO_2$	EFCO	$EFCH_4$
North Fork Prescribed Fire – 13 August 2011										
NF1301	17:03	at source	13	8.19	1.12	0.11	0.879	1594	139.1	7.48
NF1302	17:04	at source	14	18.15	2.75	0.28	0.868	1571	151.7	8.66
NF1303	17:07	at source	27	8.06	1.17	0.11	0.873	1582	146.5	7.74
NF1304	17:18	4–8 km	85	5.09	0.90	0.08	0.850	1537	172.2	9.18
NF1305	17:47	at source	36	8.46	1.27	0.11	0.869	1575	150.8	7.71
NF1306	18:06	at source	25	16.07	2.50	0.20	0.865	1569	155.5	7.26
NF1307	18:14	at source	49	6.26	0.95	0.08	0.868	1574	152.4	7.32
NF1308	18:18	at source	11	14.00	2.27	0.19	0.861	1559	160.7	7.81
		Average					0.867 ± 0.009	1570 ± 17	153.6 ± 9.8	7.89 ± 0.68
Big Salmon Lake Fire – 17 August 2011										
BSL1701	15:55	17–20 km	61	9.96	1.07	0.09	0.903	1641	112.6	5.51
BSL1702	16:04	30 km	62	7.92	0.88	0.08	0.900	1637	115.3	5.65
BSL1703	16:51	40 km	79	5.52	0.78	0.07	0.876	1588	143.2	7.34
BSL1704	17:03	23–29 km	78	14.29	1.58	0.14	0.901	1637	114.9	5.87
BSL1705	17:18	at source	47	13.60	1.54	0.15	0.898	1631	117.4	6.55
BSL1706	17:24	at source	38	8.03	0.99	0.09	0.890	1615	126.9	6.74
BSL1707	17:30	at source	55	13.84	1.71	0.16	0.890	1615	126.8	6.80
BSL1708	17:42	16–35 km	132	7.35	0.89	0.08	0.892	1619	124.9	6.62
BSL1709	17:49	40 km	81	6.42	0.80	0.07	0.889	1613	128.4	6.77
		Average					0.893 ± 0.008	1622 ± 17	123.4 ± 9.6	6.43 ± 0.61
Big Salmon Lake Fire – 22 August 2011										
BSL2201	15:25	source to 10 km	80	6.25	0.82	0.07	0.884	1605	133.5	6.65
BSL2202	15:52	at source	32	5.56	0.72	0.06	0.886	1608	131.8	6.80
BSL2203	16:01	at source	30	8.96	1.18	0.12	0.884	1601	134.0	7.83
BSL2204	16:03	at source	16	9.74	1.45	0.15	0.871	1576	149.0	8.53
BSL2205	16:05	at source	31	3.02	0.56	0.05	0.845	1527	178.7	9.46
		Average					0.874 ± 0.017	1583 ± 34	145.4 ± 19.9	7.85 ± 1.18
Hammer Creek Fire – 22 August 2011										
HC2201	16:10	at source	53	13.30	1.44	0.12	0.903	1642	112.8	5.27
HC2202	16:14	source to 10 km	38	11.68	1.27	0.10	0.902	1641	113.6	5.15
HC2203	16:21	at source	20	13.91	2.15	0.21	0.866	1567	154.2	8.57
HC2204	16:36	at source	27	11.00	1.29	0.12	0.895	1624	121.7	6.53
HC2205	16:39	at source	15	21.37	2.35	0.23	0.901	1636	114.5	6.31
HC2206	16:41	at source	14	10.47	1.18	0.11	0.899	1632	117.2	6.28
HC2207	16:50	at source	40	16.05	1.99	0.21	0.890	1612	127.4	7.57
HC2208	16:53	4–8 km	79	8.76	1.04	0.10	0.894	1621	122.9	7.05
HC2209	17:08	at source	41	9.53	1.01	0.10	0.904	1642	110.9	6.32
HC2210	17:09	at source	14	47.39	4.35	0.35	0.916	1668	97.4	4.50
		Average					0.897 ± 0.013	1628 ± 26	119.3 ± 14.7	6.36 ± 1.20
Saddle Complex Fire – 24 August 2011										
SC2401	15:32	at source	52	9.22	1.47	0.13	0.863	1562	158.4	8.17
SC2402	15:37	at source	55	20.00	2.86	0.28	0.875	1585	144.0	8.09
SC2403	15:42	at source	41	5.02	0.78	0.07	0.866	1568	154.6	8.13
SC2404	15:46	at source	65	3.07	0.45	0.04	0.873	1582	147.1	7.46
SC2405	15:51	at source	49	4.95	0.57	0.05	0.897	1629	119.2	6.20
SC2406	16:05	8 km	125	4.74	0.66	0.06	0.878	1592	140.7	7.36
SC2407	17:02	5 km	126	1.57	0.22	0.02	0.876	1590	142.5	7.20
SC2408	17:19	at source	62	4.09	0.56	0.05	0.879	1596	139.5	6.41
SC2409	17:23	at source	62	3.88	0.62	0.06	0.863	1562	158.3	8.14
SC2410	17:29	at source	69	6.71	1.06	0.10	0.864	1563	157.0	8.52
		Average					0.873 ± 0.011	1583 ± 21	146.1 ± 12.0	7.57 ± 0.79

Table 2. Continued.

Sample	Time	Location	n[1]	ΔCO_2	ΔCO	ΔCH_4	MCE	$EFCO_2$	EFCO	$EFCH_4$
Saddle Complex – 25 August 2011										
SC2501	14:55	at source	83	1.58	0.21	0.02	0.885	1603	132.9	7.74
SC2502	14:60	at source	23	11.30	1.40	0.15	0.889	1612	127.5	7.76
SC2503	15:06	at source	68	1.78	0.28	0.03	0.865	1566	155.1	8.65
SC2504	15:09	at source	57	5.97	0.65	0.06	0.902	1639	113.7	5.68
		Average					0.885 ± 0.015	1605 ± 30	132.3 ± 17.2	7.46 ± 1.26
Saddle Complex Fire – 26 August 2011										
SC2601	15:06	6 km	44	3.14	0.37	0.03	0.895	1625	121.4	6.39
SC2602	15:58	at source	33	4.65	0.66	0.08	0.876	1582	143.1	9.62
SC2603	16:04	at source	85	3.56	0.53	0.05	0.871	1578	149.0	7.70
SC2604	16:15	source to 8 km	115	5.11	0.66	0.06	0.886	1607	131.3	7.15
		Average					0.882 ± 0.011	1598 ± 22	136.2 ± 12.3	7.71 ± 1.38
Saddle Complex Fire – 27 August 2011										
SC2701	14:47	22–24 km	143	3.05	0.32	0.03	0.904	1643	110.9	5.66
SC2702	15:08	28–34 km	156	4.58	0.41	0.04	0.918	1671	94.9	4.95
SC2703	15:39	at source	95	3.86	0.55	0.05	0.876	1588	142.7	7.69
SC2704	15:50	at source	174	3.03	0.43	0.04	0.876	1587	143.1	7.78
SC2705	16:01	at source	109	3.74	0.56	0.06	0.869	1572	150.8	8.71
		Average					0.889 ± 0.021	1612 ± 42	128.5 ± 24.4	6.96 ± 1.58
Big Salmon Lake Fire – 28 August 2011										
BSL2801	14:57	9–13 km	43	14.65	1.22	0.11	0.923	1681	89.3	4.40
BSL2802	15:14	at source	20	21.65	2.63	0.23	0.892	1620	125.1	6.23
BSL2803	15:18	at source	48	2.59	0.37	0.03	0.875	1586	144.8	7.07
BSL2804	15:22	at source	22	10.63	1.60	0.17	0.869	1571	150.9	9.00
BSL2805	15:26	at source	27	10.50	1.50	0.17	0.875	1581	143.8	9.50
BSL2806	15:27	at source	33	6.43	0.71	0.07	0.901	1637	114.4	6.03
BSL2807	15:32	at source	14	10.91	1.94	0.24	0.849	1528	173.0	12.09
BSL2808	15:54	12–16 km	15	8.35	1.08	0.10	0.885	1605	132.5	7.18
		Average					0.884 ± 0.022	1601 ± 46	134.2 ± 25.3	7.69 ± 2.41
Study Average							0.883 ± 0.010	1596 ± 23	135 ± 11	7.30 ± 0.58

[1] n = number of 2-s data points.

Table 3. MCE and EF, this study (measured and estimated) A11, and NEI and the ratio of EF for this study to A11 and NEI.

Species	This study	A11	NEI	This study / A11	This study / NEI
MCE	0.883 ± 0.018	0.921	0.847	Ratio	
	EF ($g\,kg^{-1}$)				
Carbon Dioxide (CO_2)[1]	1600 ± 35	1637	1466	0.98	1.09
Carbon Monoxide (CO)[1]	135 ± 20	89	169	1.52	0.80
Methane (CH_4)[1]	7.32 ± 1.10	3.92	8.06	1.87	0.91
	Estimated from B11 EF–MCE Relationships				
Acetylene (C_2H_2)[2]	0.21 ± 0.20	0.29		0.72	
Ethylene (C_2H_4)[2]	1.71 ± 0.23	1.12		1.53	
Propylene (C_3H_6)[2]	0.96 ± 0.13	0.95		1.01	
Formaldehyde (HCHO)[2]	2.6 ± 0.25	2.27	1.29	1.15	2.02
Methanol (CH_3OH)[2]	3.14 ± 0.26	1.93		1.63	
Formic Acid (HCOOH)[2]	0.26 ± 0.03	0.35		0.74	
Acetic Acid (CH_3COOH)[2]	3.72 ± 0.38	1.97		1.89	
Phenol (C_6H_5OH)[2]	1.02 ± 0.25	0.33		3.09	
Furan (C_4H_4O)[2]	0.6 ± 0.06	0.20		3.00	
Glycolaldehyde ($C_2H_4O_2$)[2]	1.03 ± 0.46	0.25		4.12	
Hydrogen Cyanide (HCN)[2]	0.83 ± 0.09	0.73		1.14	
Ammonia (NH_3)[2]	1.9 ± 0.3	0.78	2.75	2.44	0.69
Nitrogen Oxides (NO_x)[2]	1.93 ± 0.55	2.51	1.35	0.77	1.43
$PM_{2.5}$[2]	25.8 ± 9.27	12.70	13.84	2.03	1.86
Σ (ethane, propane, butane)[3]	1.46	1.46			
NMOC (identified)[4]	17.54	11.85		1.51	
NMOC (identified + unidentified)[5]	35.08	23.7	39.57[6]	1.51	0.85

[1] EF value for this study is the fire-day average from Table 3 with uncertainty of 1.833σ which approximates 90 % confidence interval for df = 9.
[2] EF value for this study is based on EFX vs. MCE regression equations of Burling et al. (2011) at average MCE of this study (0.833). Uncertainty is 90 % confidence interval of Burling et al. (2011) linear regression fit at MCE = 0.833.
[3] Value is the sum of EF for ethane, propane, and butane from Akagi et al. (2011) which were not reported by Burling et al. (2011).
[4] Sum of EF for NMOC in table. [5] Unidentified NMOC is estimated as 50 % of identified NMOC after Akagi et al. (2011).
[6] Effective EFVOC (see text).

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Table A1. Fuel consumption and MCE for 18 prescribed fires.

Fire name	Location	MCE	Fuel consumption ($kg\,m^{-2}$)			Reference
			Fine fuels[1]	CWD[1]	Duff	
Quinalt	NW	0.850	2940	8050	11 000	Hobbs et al. (1996)
Creamery	NW	0.905	2270	1320	2500	Hobbs et al. (1996)
Raymond	NW	0.877	1980	1320	2500	Hobbs et al. (1996)
Camp Lejeune IA	North Carolina	0.943	1541	0	0	Burling et al. (2011); Reardon (2012)
Camp Lejeune ME	SE	0.945	1127	204	0	Burling et al. (2011); Reardon (2012)
MT1	western Montana	0.891	556	462	222	Urbanski et al. (2009); Harrington (2012)
MT2	western Montana	0.908	952	632	1076	Urbanski et al. (2009); Harrington (2012)
MT3	western Montana	0.913	649	439	918	Urbanski et al. (2009); Harrington (2012)
MT4	western Montana	0.910	536	632	545	Urbanski et al. (2009); Harrington (2012)
OR1	Oregon	0.906	513	268	848	Urbanski et al. (2009); Ottmar and Ward (1996)
OR2	Oregon	0.900	1339	424	1406	Urbanski et al. (2009); Ottmar and Ward (1996)
OR3	Oregon	0.916	1004	223	1429	Urbanski et al. (2009); Ottmar and Ward (1996)
FL5	Florida	0.933	923	0	383	Urbanski et al. (2009); Ottmar and Vihnanek (1995)
SC1	SE	0.921	423	0	169	Urbanski et al. (2009); Ottmar and Vihnanek (1995)
SC12A	SE	0.942	374	0	300	Urbanski et al. (2009); Ottmar and Vihnanek (1995)
SC12B	SE	0.923	414	0	284	Urbanski et al. (2009); Ottmar and Vihnanek (1995)
MN4	MW	0.953	4444	0	112	Urbanski et al. (2009); Ottmar and Vihnanek (1995)
MN5	MW	0.936	8836	0	1377	Urbanski et al. (2009); Ottmar and Vihnanek (1995)

[1] Fine fuels include litter, grasses, shrubs, and dead wood with diameter < 7.62 cm; CWD (coarse woody debris) is dead wood with diameter > 7.62 cm.

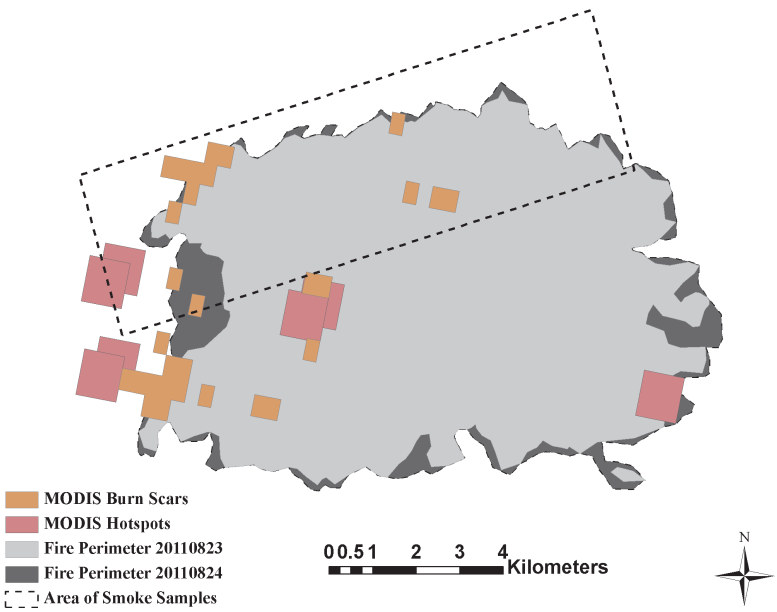

Fig. 1. Region of smoke sampling, fire perimeters, and area of active burning for Saddle Complex on 24 August 2011.

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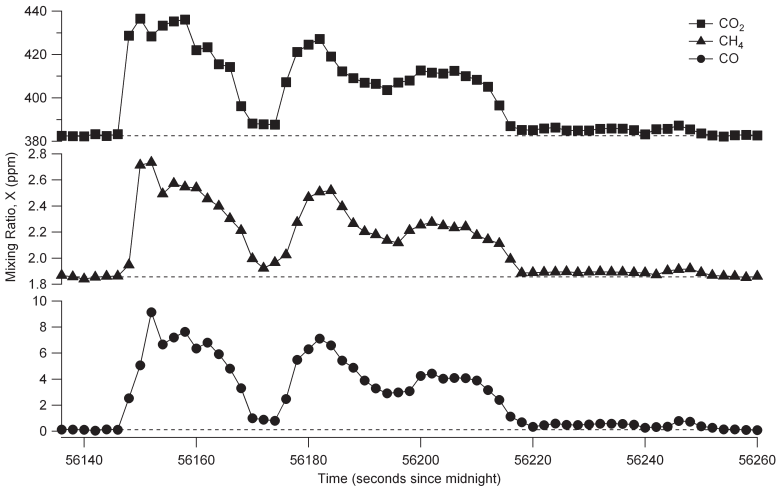

Fig. 2. CRDS measurements of CO_2, CH_4, and CO for a smoke sample run on the Saddle Complex on 24 August 2011. The dashed line in each panel shows the background mixing ratios measured upwind of the fire on approach for the smoke sample.

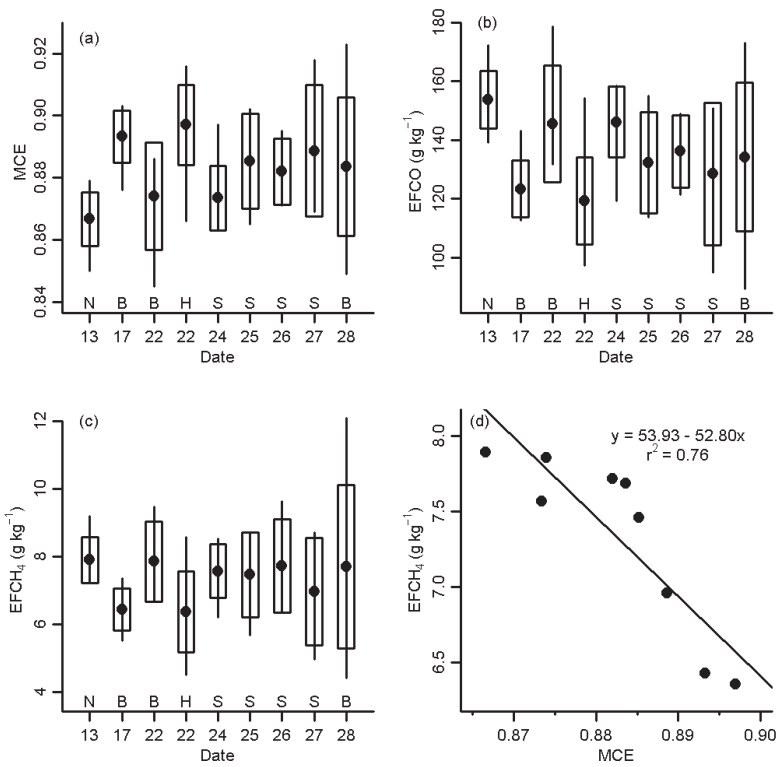

Fig. 3. (a) MCE, **(b)** EFCO, and **(c)** EFCH4 by fire-day. Date is day of August 2011, N = North Fork Prescribed Fire, B = Big Salmon Lake Fire, H = Hammer Creek Fire, S = Saddle Complex. Solid circles are the fire-day average, boxes are $\pm 1\sigma$, vertical bars mark the minimum and maximum. **(d)** Fire-day average EFCH4 vs. MCE. Solid line is linear least square fit.

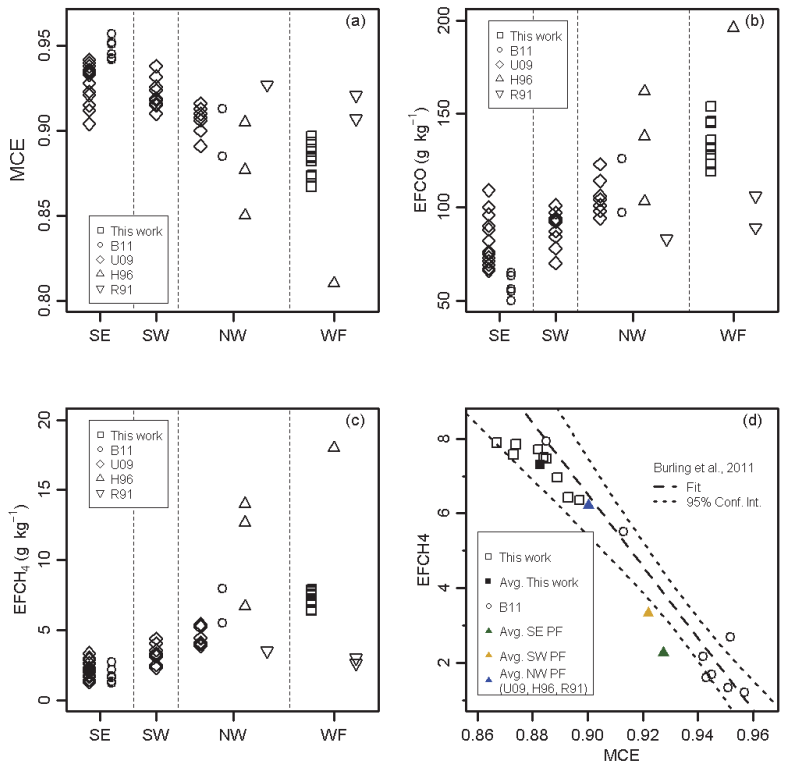

Fig. 4. (a) MCE, **(b)** EFCO, and **(c)** EFCH4 for this work and from several other studies plotted by region for prescribed fires (SE = southeast, SW = southwest, NW = northwest). WF = wildfires. **(d)** EFCH4 vs. MCE with best fit line from B11.

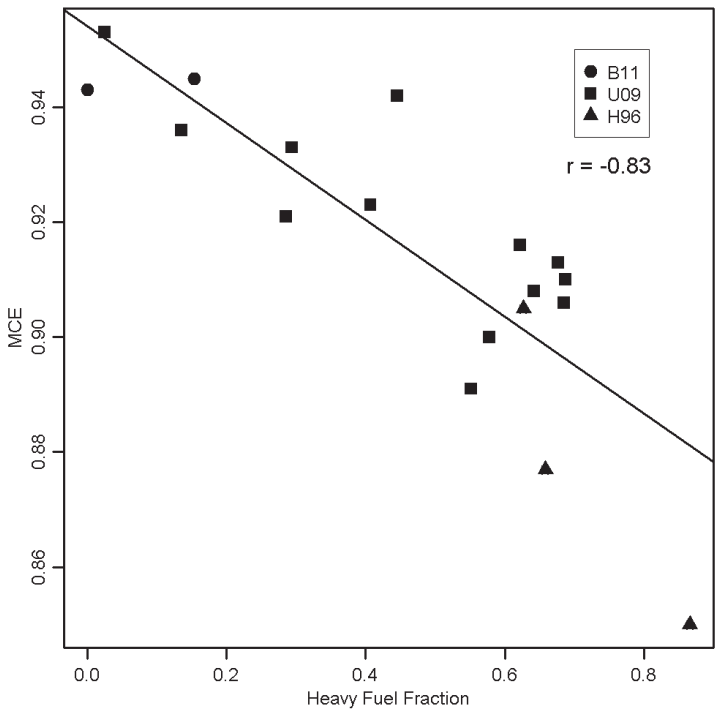

Fig. 5. Plot of MCE vs. heavy fuel fraction (the ratio of heavy fuel consumption to total fuel consumption) for 18 prescribed fires from previous studies (B11; U09; H96; Ottmar and Ward, 1996; Ottmar and Vihnanek, 1995; Reardon, 2012; Harrington, 2012). Heavy fuel is CWD and duff. See Sect. 3.3 and Appendix A for details.

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