Atmospheric Environment 99 (2014) 104–111

Contents lists available at ScienceDirect

Atmospheric Environment

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

Fire environment effects on particulate matter emission factors in southeastern U.S. pine-grasslands

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HIGHLIGHTS

- We tested fire environment effects on particulate matter emission factors ($EF_{PM2.5}$).
- 41 prescribed burns were measured in pine-grasslands of Florida and Georgia, USA.
- $EF_{PM2.5}$ increased from winter to summer and with pine needle content.
- $EF_{PM2.5}$ decreased with grass content and frequency of burning.
- Timber thinning and frequent prescribed burning should reduce $EF_{PM2.5}$.

ARTICLE INFO

Article history:
Received 24 June 2014
Received in revised form
18 September 2014
Accepted 22 September 2014
Available online 29 September 2014

Keywords:
Prescribed fire
Biomass burning
Fire behavior
Combustion efficiency
Structural equation model

ABSTRACT

Particulate matter (PM) emission factors (EF_{PM}), which predict particulate emissions per biomass consumed, have a strong influence on event-based and regional PM emission estimates and inventories. PM < 2.5 μm aerodynamic diameter ($PM_{2.5}$), regulated for its impacts to human health and visibility, is of special concern. Although wildland fires vary widely in their fuel conditions, meteorology, and fire behavior which might influence combustion reactions, the $EF_{PM2.5}$ component of emission estimates is typically a constant for the region or general fuel type being assessed. The goal of this study was to use structural equation modeling (SEM) to identify and measure effects of fire environment variables on $EF_{PM2.5}$ in U.S. pine-grasslands, which contribute disproportionately to total U.S. $PM_{2.5}$ emissions. A hypothetical model was developed from past literature and tested using 41 prescribed burns in northern Florida and southern Georgia, USA with varying years since previous fire, season of burn, and fire direction of spread. Measurements focused on $EF_{PM2.5}$ from flaming combustion, although a subset of data considered MCE and smoldering combustion. The final SEM after adjustment showed $EF_{PM2.5}$ to be higher in burns conducted at higher ambient temperatures, corresponding to later dates during the period from winter to summer and increases in live herbaceous vegetation and ambient humidity, but not total fine fuel moisture content. Percentage of fine fuel composed of pine needles had the strongest positive effect on $EF_{PM2.5}$, suggesting that pine timber stand volume may significantly influence $PM_{2.5}$ emissions. Also, percentage of fine fuel composed of grass showed a negative effect on $EF_{PM2.5}$, consistent with past studies. Results of the study suggest that timber thinning and frequent prescribed fire minimize $EF_{PM2.5}$ and total $PM_{2.5}$ emissions on a per burn basis, and that further development of PM emission models should consider adjusting $EF_{PM2.5}$ as a function of common land use variables, including pine timber stocking, surface vegetation composition, fire frequency, and season of burn.

1. Introduction

Particulate matter (PM) emission factors (EF_{PM}), typically expressed as the mass of PM emitted per mass of fuel consumed (g kg^{-1}), are essential for estimating regional and event-based atmospheric emissions from wildland fires. Emission of PM < 2.5 μm

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http://dx.doi.org/10.1016/j.atmosenv.2014.09.058

aerodynamic diameter ($PM_{2.5}$) is of particular concern because of its effects on human health (Naeher et al., 2007), reduction of visibility, radiative forcing (Reid et al., 2005a), formation of secondary pollutants (Koppmann et al., 2005), and role as condensation nuclei (Reid et al., 2005b). For these reasons it is regulated by the U.S. Environmental Protection Agency (EPA). Calculation of regional $PM_{2.5}$ emissions from wildland fires typically involves multiplication of the estimated burned area, estimated fuel consumption per unit area, and $EF_{PM2.5}$, followed by model-based predictions of $PM_{2.5}$ dispersion, longevity, and deposition (Battye and Battye, 2002). Although wildland fires vary widely in their fuel conditions, meteorology, and fire behavior, the $EF_{PM2.5}$ component of this equation is typically a constant for the region being assessed (Andreae and Merlet, 2001) or the general fuel or vegetation type burned (van der Werf et al., 2010; Akagi et al., 2011; Urbanski et al., 2011), although it additionally may be weighted by estimated contributions from flaming versus smoldering phases of combustion (Prichard et al., 2007; Hardy et al., 2010; Lutes, 2013). These approaches depend on the assumption that the applied $EF_{PM2.5}$ is acceptably robust over a wide range of geographic, climatic, and local environmental conditions.

Evidence suggests that certain local fuel and environmental conditions affect $EF_{PM2.5}$ through their influence on combustion processes. Such processes are often described in terms of combustion efficiency (CE), the proportion of carbon (C) released as CO_2 relative to C in all other emissions, which is inversely related to $EF_{PM2.5}$ (Janhäll et al., 2010) and often used to calculate $EF_{PM2.5}$ indirectly. Fuel moisture tends to decrease CE and increase $EF_{PM2.5}$ because it absorbs energy that would otherwise be available for combustion, and emitted water vapor dilutes volatized gases and reduces the rate of oxidation reactions (Ward et al., 1989). Increasing fuel moisture tends to shift the emission source from flaming to smoldering combustion, the later having a much lower CE and higher $EF_{PM2.5}$ (Hardy et al., 2010). Variation in fuel moisture, reflecting proportion of live fuel and response of dead fuel to ambient conditions, has been attributed to seasonal differences in $EF_{PM2.5}$ in tropical savanna fires (Hao et al., 1996; Scholes et al., 1996; Ward et al., 1996; Hoffa et al., 1999; Korontzi et al., 2003). Research on gasoline combustion engines has shown higher ambient temperature of intake air to decrease PM emissions relative to energy released (Nam et al., 2008) and higher humidity to increase emissions (McCormick et al., 1997; Rahai et al., 2011), although such direct effects of ambient air conditions on wildland fire $EF_{PM2.5}$ has not been studied. $EF_{PM2.5}$ also responds to oxygen availability (Hegg et al., 1990), which is influenced by fuel particle size and bulk density (packing ratio) (Ward et al., 1980, 1983).

Fire behavior, reflecting fuel, weather, and topography as well as direction of fire spread relative to the wind, might also influence $EF_{PM2.5}$. Field experiments have suggested that $EF_{PM2.5}$ decreases with increasing reaction intensity (RI, rate of heat released per unit area) in prescribed burns because of stronger heat feedback and convection resulting in higher CE (Sandberg, 1974; Ward and Hardy, 1984). Results for fireline intensity (FI, rate of heat release per length of fire line) suggest that $EF_{PM2.5}$ initially decreases with increasing FI but above some level begins to increase due to oxygen deficiency as the depth of the flaming zone increases (Ward et al., 1980, 1983; Ward and Hardy, 1991). FI is typically an order of magnitude higher for fires running with the wind (head fire) than those spreading against the wind (backing fire) (Hmielowski, 2013), such that location on the fire perimeter or prescribed fire ignition pattern might influence $EF_{PM2.5}$.

Wildland fire $EF_{PM2.5}$ might also be influenced by ecological characteristics of the area burned, including plant community type and changes in fuel characteristics during post-fire succession. $EF_{PM2.5}$ has been shown to vary among general plant community types, such as forest, savannas, grasslands, and brushlands (Urbanski et al., 2009; Janhäll et al., 2010), attributable to variation in physical and chemical characteristics of the fuel matrix reflecting the proportions of shrub, grass, and litter fuels (Ward et al., 1996). Grass dominance is generally associated with low $EF_{PM2.5}$ because it tends to burn readily through flaming combustion (Ward et al., 1996; Urbanski et al., 2009; Janhäll et al., 2010). Pine needle litter has been found to have a disproportionately high $EF_{PM2.5}$ (Sandberg, 1974) despite its high flammability and energy content (Reid and Robertson, 2012). In most community types, time since previous fire corresponds to an increase in total fine fuel, woody plant dominance, leaf litter, and duff and a decrease in grass, forbs, and percentage of live fuel (Binkley et al., 1992; Peterson et al., 2007; Reid et al., 2012). These changes correspond to an overall decrease in fuel energy content (Hough, 1969; Reid and Robertson, 2012) and increase in fuel bulk density, which might promote higher $EF_{PM2.5}$ (Ward and Hardy, 1991). However, these changes also correspond to a reduction in the percentage of live fuel, making it difficult to predict the net effect of time since fire on $EF_{PM2.5}$.

To the degree that such factors predict $EF_{PM2.5}$, there is an opportunity to improve $PM_{2.5}$ emission models by considering their effects. The goal of this study was to identify which if any commonly measured fuel, fire, and weather variables during prescribed fires in southeastern U.S. pine-grasslands influences $EF_{PM2.5}$ to provide a theoretical foundation for further empirical model development. Estimates of $PM_{2.5}$ emissions are especially important in this region because of its frequent prescribed burning and wildfire and resulting disproportionate contribution to the nation's annual $PM_{2.5}$ emissions (Aurell and Gullett, 2013) and non-attainment of EPA standards for $PM_{2.5}$ in certain urban areas within the region (EPA, 2014). The study was designed to incorporate the range of variables most commonly considered by prescribed fire managers in planning burns: time since last fire, season of burn, ignition pattern (head versus backing fire), ambient air conditions, and fuel composition. Our approach was to measure these and associated environmental variables and $EF_{PM2.5}$ during burns under a wide range of fire conditions, then assess the relative effects of these variables on $EF_{PM2.5}$ using Structural Equation Modeling (SEM). The SEM analyses focused on fire behavior dominated by the flaming phase of combustion with an emphasis on comparing effects of environmental variables rather than estimating total emissions or event-based (all phases combined) emission factors, although smoldering-dominated combustion and MCE were measured for a subset of burns and reported for purposes of discussion.

2. Materials and methods

2.1. Fire environment measurements

Field work was conducted on the 1619-ha Tall Timbers Research Station and Land Conservancy (30°40′N, 8°14′W) and the 1222-ha Pebble Hill Plantation (PHP) (30°46′N, 84°3′W) between Tallahassee, Florida, and Thomasville, Georgia, USA. The communities studied were open-canopy pine-grasslands with either native (never plowed) or old-field (post-agriculture) surface vegetation (Ostertag and Robertson, 2007). They have been managed with single tree selection forestry and prescribed fire applied at mostly 1–2 year intervals since European settlement or abandonment of agriculture in the early 20th century (Reid et al., 2012), although certain burn units were recently fire-excluded up to four years for purposes of this and other studies.

Prescribed burns were applied in 2010, 2011, and 2012 on dates ranging from January to August to include the period when burns are typically applied in the region and include fires in the dormant and growing seasons (beginning in late March) (Table 1). Burns were approximately stratified according to occurrence 1, 1.5, 2, 3, and 4 years since last fire. For most burns, a portion of the burn unit was lit with a backing fire, extinguished, then lit with a head fire, and a separate set of measurements were taken for each for a total of 41 sets of measurements (hereafter "burns").

For each burn, fuels were measured in the vicinity of fire behavior and emissions measurements (ca. 400 m^2 area) within two weeks prior to the burn. The study was limited to fine fuels (<0.6 cm thickness) as these compose more than 90% of the total available fuel load in this community type (Robertson and Ostertag, 2007) and virtually all of the fuel consumed in the flaming front where emissions measurements were taken. Stems and leaves of live woody fuels <0.6 cm thickness were measured in four randomly placed 1 m^2 frames. Other fine fuels (<0.6 cm thickness) were collected in 4–6 0.25 m^2 frames and separated into the following categories: 1) live herbs, 2) dead herbs 2) pine needle litter, and 3) other litter (including broad leaf litter, twigs, and finely broken fuel on the soil surface). Also, the percentage of live and dead herbs composed of grass was visually estimated. Using these data, we calculated the percentage of non-woody fine fuels composed of dead fuel versus live fuel (total = 100%) and of live forbs, grass (live and dead), pine needle litter, and other litter (total = 100%) (Table 1). Fuels were dried to constant weight then measured for dry mass. Duff was not present owing to a history of frequent fire. Larger dead fuels (branches, pine cones) were present but sparsely distributed and not considered to contribute significantly to smoke in the flaming front measured in this study. Fine fuel bed height was estimated at each location and used to calculate fine fuel bed bulk density (kg m^{-3}, Table 1).

Within one hour prior to burning, a grab sample from multiple locations throughout the study area was taken for each fuel category, sealed in a plastic bag, weighed, dried to constant weight, then weighed dry to calculate fuel moisture content ([wet mass − dry mass]/dry mass × 100) for each fuel category, and total fine fuel moisture content was calculated as a weighted average. Post-burn fuels were measured and weighed in the same manner as pre-burn fuels to estimate fuel consumption and fuel percent consumed on a per area basis.

Table 1
Abbreviations, mean, and standard deviations for environmental and emissions variables measured for each burn ($n = 41$) unless indicated otherwise.

Variable	Abbrev.	x	SD
$EF_{PM2.5}$ (g kg^{-1})	EF	21.0	8.9
Modified Combustion Efficiency[a]	MCE	0.942	0.020
Date of burn	JULIAN	4 MAY	59.7
Temp (C)	TP	27.6	6.0
Relative humidity	RH	43.4	14.2
Specific humidity	SH	0.869	0.374
Keetch-Byrum drought index	KBDI	314	178
Wind speed (kph)	WS	4.3	2.3
Years since fire	YSF	2.4	1.1
Live woody fuel (Mg ha^{-1})	WOODY	1.4	1.1
Fine fuel (Mg ha^{-1})	FUEL	8.2	2.7
Dead fine fuel (Mg ha^{-1})	D FUEL	7.7	2.9
Fine fuel % dead	% DEAD	91.8	7.4
Fine fuel % live	% LIVE	8.2	7.3
Fine fuel % live forbs	% L FORBS	3.8	4.0
Fine fuel % grass	% GRASS	14.9	10.9
Fine fuel % needles	% NEEDLES	23.0	9.2
Fine fuel % other litter	% LITTER	58.3	15.9
Fine fuel moisture content (%)	FUEL MC	28.6	11.0
Fine fuel bulk density (kg m^{-3})	DENSITY	12.2	7.6
Fine fuel consumed (Mg ha^{-2})	FUEL C	4.7	2.0
Fine fuel % consumed	FUEL % C	57.3	14.8
Heat per unit area (kJ m^{-2})	HUA	8583	3895
Reaction intensity (kJ m^{-2} s^{-1})	RI	154	98
Fireline intensity (kJ m^{-1} s^{-1})	FI	335	606
Rate of spread (m min^{-1})	ROS	2.24	3.50

[a] Available for a subset of burn units.

Fire behavior was measured with the aid of high temperature type-K thermocouple wires (0.125 mm, Omega Engineering, Inc.) attached to dataloggers (HOBO U12-014, Onset Corporation) buried shallowly in the ground in plastic bags to measure the wire junction temperature approximately 3 cm above the soil surface at 2 s intervals during fires. Their purpose was to measure flaming combustion residence time, which was approximated as time >260C based on experimentation, and fire rate of spread between thermocouples placed at fixed intervals parallel to the direction of fire spread (5–9 locations per burn). Assuming fuel energy content averaged 20,000 J kg^{-1} (Reid and Robertson, 2012), we calculated heat released per unit area (HUA, kJ m^{-2}), reaction intensity (RI, kJ m^{-2} s^{-1}), and fireline intensity (FI, kJ m^{-1}). Direction of spread was expressed as a dummy variable. All measurements were averaged for each burn, which served as the unit of replication in analyses ($n = 41$).

During burns, ambient temperature, relative humidity, wind speed, and wind direction were measured per minute using a portable weather station (NovaLynx Corporation). Specific humidity (g kg^{-1}) was calculated from measurements of relative humidity and ambient temperature (Wagner and Pruss, 1993) because of its potentially more direct effects on dead fuel moisture and combustion processes (Choi et al., 2000). Keetch–Byrum Drought Index (KBDI) was calculated from precipitation and temperature measurements (Keetch and Byrum, 1968) taken at the Tall Timbers Research Station weather station within 10 km of all study sites.

2.2. Emission factor and combustion efficiency measurements

Emission factors were determined using the excess concentration (excess mixing ratio) method described in other studies (Agaki et al., 2011). Gases and $PM_{2.5}$ were simultaneously measured from the ground, first under pre-burn ambient conditions and then at the tip of the longest flames within the convection column during burns. Sampling was through a hand-held galvanized aluminum tube (10 cm diameter, 3 m long) from which a flexible aluminum foil conduit (maximum 30 m) lead to the sampling station outside of the burn unit. Intake was driven by an in-series vent fan (2200 L min^{-1} flow rate). Earlier experiments confirmed that length of conduit did not cause differential transport of gases and PM to the sampling station.

At the sampling station, the gas and $PM_{2.5}$ mixture was released from the conduit vertically into the air approximately 10 cm from the intake vent of a PQ 200 $PM_{2.5}$ air sampler (BGI Inc., Waltham, MA) within a 50 cm diameter cylindrical baffle (lamp shade) oriented vertically to protect the gas-PM mixture from wind dispersal prior to sampling. When CO_2 concentration reached 2000 ppm (as monitored by a Qubit S151 CO_2 analyzer), the air sampler collected for 10 min at a flow rate of 16.7 L min^{-1} and collected $PM_{2.5}$ on a 47-mm prebaked and pre-weighed quartz filter (SKC Inc., Eighty Four, PA). Gas was sampled from within the cylindrical baffle within approximately 10 cm of the air sampler intake vent and collected in 10 L Tedlar bags. For a subset of burns (31 out of 41), CO concentration was measured in the same manner. For each burn, three gas-PM samples were taken for backing fires and averaged per burn and one measurement was taken for head fires because of the rapid rate of spread and limited time and space for sampling. For a subset of burns ($n = 22$), smoldering combustion from fine fuels was also sampled after the passage of the flaming front in the same manner as for flaming combustion, with air sampled a few centimeters away from the emission source. After sample collection, the filter was removed and placed in a desiccator before reweighing using an analytical balance (Mettler Toledo, Columbus, OH) with resolution of 0.01 mg, and $PM_{2.5}$ mass was calculated as the difference between the pre- and post-sampling filter weight. Sample CO concentrations were measured in the laboratory using a calibrated analyzer (Environmental Sensors Co., Boca Raton, Florida). The concentration of C in sampled $PM_{2.5}$ was obtained Multi-elemental scanning thermal analysis (MESTA) as described in Hsieh (2007).

$PM_{2.5}$ emission factors (g kg^{-1}) were calculated as:

$$EF_{PM2.5} = \{(PM_{2.5burn} - PM_{2.5ambient})/[(C_{CO2burn} - C_{CO_2ambient}) + C_{COburn} + (C_{PM2.5burn} + C_{PM2.5ambient})]\}\cdot w \tag{1}$$

where $EF_{PM2.5}$ is in g kg^{-1}, $PM_{2.5burn}$ and $PM_{2.5abmient}$ are in μg m^{-3}, and $C_{CO2burn}$, $C_{CO2ambient}$, C_{COburn}, $C_{PM2.5burn}$, and $C_{PM2.5ambient}$ are in mg C m^{-3}, and w is the correction factor to convert unit mass of C emitted to unit mass of fuel consumed, approximately 0.44 for the studied fuel type (Matamala et al., 2008). For samples without CO measurements, CO concentration was estimated at 6% of excess CO_2 concentration based on the mean of samples with CO (SD = 2.1%). It was assumed that all measured CO in the convection column was released through combustion and that the sum of net C mass released represented the total carbonaceous emission from the fuel (i.e., additional source contributions to total C emitted were negligible; Reid et al., 2005b).

For the subset of burns where CO was measured, MCE was calculated as $(\Delta C_{CO2})/(\Delta C_{CO2} + \Delta C_{CO})$, where ΔC represents the proportional moles of carbon released as CO_2 or CO (Janhäll et al., 2010). MCE was used to test the assumption that it is inversely related to $EF_{PM2.5}$ (Kaufman et al., 1992; Janhäll et al., 2010) and to provide a basis for comparison to other studies reporting MCE. Data for fuel, fire behavior, and $EF_{PM2.5}$ and MCE measurements averaged per burn and used in analyses are provided in Supplementary Material 1 for flaming combustion and in Supplementary Material 2 for smoldering combustion.

2.3. Data analysis using structural equation modeling

Structural equation modeling (SEM) was used to analyze potential influences of fire environmental variables on $EF_{PM2.5}$. Our approach was to present a reasonable *a priori* model based on past research and then simplify the model to contain only relationships among variables that were statistically significant or which otherwise contributed to the overall model fit (Grace, 2006). The model used only manifest (observed) variables (Grace, 2006) and was analyzed using SPSS Amos 22.0 (IBM Corporation).

Before constructing the *a priori* model, we used a principal components analysis (PCA) using PC-ORD (MjM Software Design) to assess which of the variables measured or calculated (Table 1) had high multivariate correlations to identify potentially redundant variables, which reduce SEM model fit and degrees of freedom. Each variable was tested for normality using the Shapiro–Wilk test and skewness and kurtosis using Systat 13.0 (Systat Software Inc.), resulting in RI and % live fuel being natural log transformed to improve normality, while all variables had acceptably low skewness and kurtosis levels (<1.5). The first three principal components of the PCA accounted for 32.1, 17.5, and 11.1 percent of total variance (total explained = 60.7 percent). Variables closely clustered (Fig. 1) with a logical reason for being correlated were considered redundant and were represented by a single variable for inclusion in the SEM. Thus, total fine fuel represented variables associated with years since previous fire (Fig. 1), fire direction of spread (0 = backing, 1 = heading) represented associated fire behavior variables, temperature represented Julian date of burn (Jan–Aug) and associated weather variables (Schroeder and Buck, 1970), and fine fuel moisture content represented variables relating to live fuel content. Fine fuel percent needles, RI, and fine fuel % grass were considered stand-alone variables (Fig. 1).

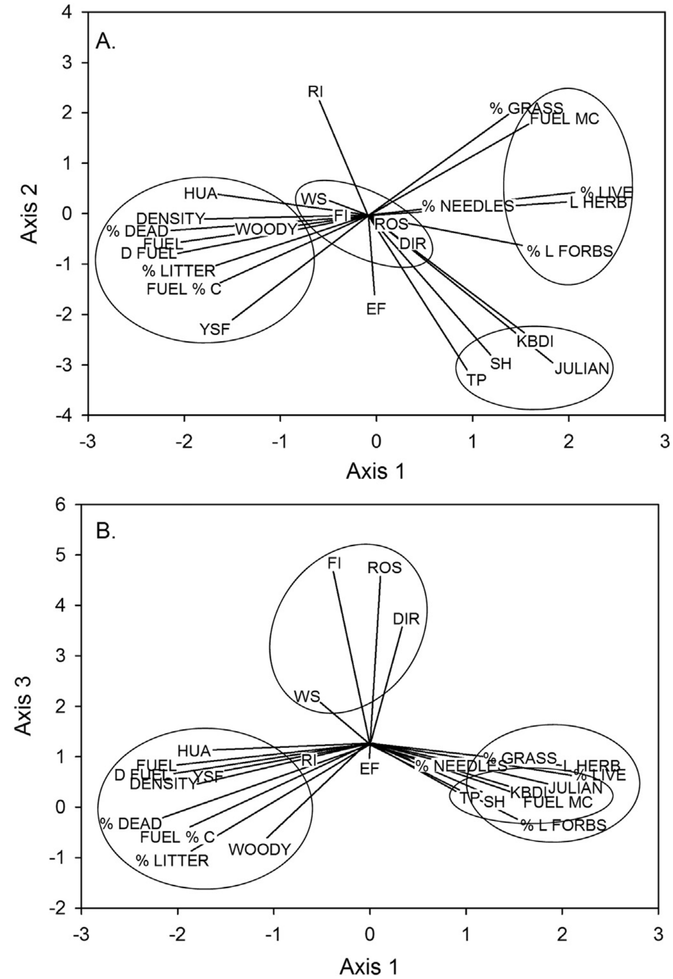

Fig. 1. Results of principal components analysis showing loadings of variables measured during prescribed on Axis 1, 2, and 3. Variables grouped within ellipses were reduced to a single proxy variable for use in Structural Equation Modeling. Variable abbreviations are explained in.Table 1.

The *a priori* model in the SEM analysis considered potential direct effects of variables on $EF_{PM2.5}$ as well potential indirect effects through RI (Fig. 2). It predicted direction of effects among most variables based on past research, although direction of some

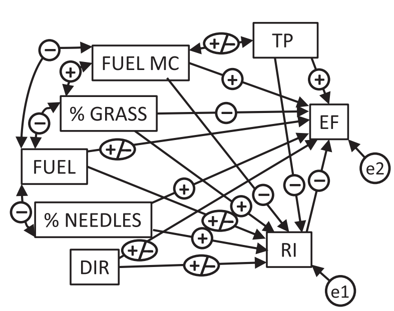

Fig. 2. Initial (*a priori*) model showing expected directions of correlations (+, ,+/) among the reduced set of variables measured during prescribed burns. Variable abbreviations are provided in.Table 1.

effects was not predicted (±) because of competing potential influences, as cited above.

After testing the initial model using the collected data, certain non-significant relationships and associated variables were removed to simplify the model, although other non-significant relationships were retained for their indirect effects and contribution to model fit as assessed using the χ^2 goodness-of-fit test in AMOS (Grace, 2006). Models with a non-significant ($P > 0.05$) χ^2 results were considered to be acceptably well fit (Grace, 2006).

In addition to the SEM analysis, an ANOVA using Systat 13.0 was used to test the effects of time since fire (1–2 years versus 3–4 years) and season of burn (dormant versus growing) as categorical independent variables on $EF_{PM2.5}$ and MCE in separate analyses for flaming-dominated and smoldering-dominated combustion. A regression of $EF_{PM2.5}$ on MCE was also run to confirm the previously reported inverse relationship between these variables (Ward and Hardy, 1991; Janhäll et al., 2010).

3. Results

Following analysis of the initial SEM model, most relationships among variables showed the predicted directional effects (Fig. 3), including the negative effect of temperature on RI, positive effect of temperature on $EF_{PM2.5}$, positive effect of fine fuel % grass on RI, negative effect of fine fuel % grass on $EF_{PM2.5}$, and positive effect of fine fuel % needles on $EF_{PM2.5}$, as well as correlations among variables (Fig. 3). Additionally, total fine fuel showed a significant positive effect on RI, which was not specifically predicted. All other paths were non-significant (Fig. 3).

Model simplification involved removal of RI which, although fairly well explained by the environmental variables ($r^2 = 0.49$), did not have a significant effect on $EF_{PM2.5}$ (Fig. 3). Also removed was direction of fire spread and fine fuel moisture content, which were neither significantly correlated with RI nor $EF_{PM2.5}$ (Fig. 3). Total fine fuel, although showing a non-significant direct effect on $EF_{PM2.5}$, was retained because it had significant correlations with fine fuel % grass and % needles, which did have significant effects on $EF_{PM2.5}$ (Fig. 3). The reduced model passed the χ^2 goodness-of-fit test ($P = 0.651$, 3 df) and had an acceptably low multivariate kurtosis of −3.195 (Gao et al., 2008), and Pearson's r^2 for $EF_{PM2.5}$ remained the same at 0.40 (Fig. 4).

ANOVA results showed that $EF_{PM2.5}$ for flaming combustion was higher at 3–4 years since fire than at 1–2 years since fire ($P = 0.052$, $F = 4.01$, 1 df) and higher in the growing season than in the dormant season ($P = 0.032$, $F = 4.94$, 1 df) (Table 2), with a non-significant interaction between the two factors. Years since fire had a non-significant effect on MCE ($P = 0.486$, $F = 0.50$, 1 df), although the

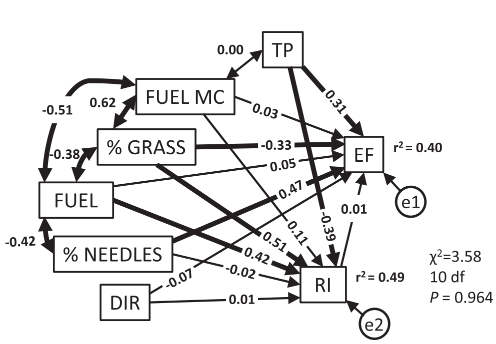

Fig. 3. Results of Structural Equation Model analysis testing the initial model. Bold arrows indicated significant correlations ($P < 0.05$). Arrows are labeled with the standardized path coefficients. Endogenous variables are labeled with the Pearson's r^2 describing the degree to which they are explained by exogenous variables. The results of the overall χ^2 goodness-of-fit test are provided.

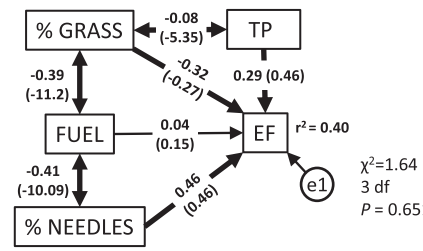

Fig. 4. Results of Structural Equation Model analysis testing the final reduced model, presented as in Fig. 3, except that arrows are also labeled with unstandardized coefficients in parentheses.

pattern was opposite that of $EF_{PM2.5}$ as expected, while the effect of season of burn on MCE was significant ($P = 0.014$, $F = 6.89$, 1 df) and also opposite of $EF_{PM2.5}$ (Table 2) with no significant interaction between factors. The ANOVA results for smoldering combustion were not significant but showed the same pattern as for flaming combustion (Table 2). Regression results of $EF_{PM2.5}$ for flaming combustion on MCE ($N = 31$) showed the expected negative relationship between the two variables, producing the equation $EF_{PM2.5} = -259.4(MCE) + 266.5$ ($P = 0.004$, $r^2 = 0.250$).

4. Discussion

The results of the SEM and ANOVAs suggest that variation in the fuel and fire environment significantly influence $EF_{PM2.5}$ within the relative narrow range of conditions represented by the open-canopy, frequently burned pine-grasslands studied. Overall, the variables showing the greatest effect were those associated with fuel and ambient air conditions rather than fire behavior. The overall pattern of higher $EF_{PM2.5}$ later in the season was reflected in the ANOVA results, which additionally predicted high $EF_{PM2.5}$ with longer times since fire. The consistency in effects between the initial and final SEM models suggest that the initial model was at least partially confirmed, although to the degree that it was altered it should be considered hypothesis-generating (Grace, 2006).

Although the predicted positive correlation between temperature and $EF_{PM2.5}$ was observed, it did not appear to be related to fuel moisture as a function of increasing live fuel and humidity during the transition from winter to summer. Apparently any effects of increasing live fuel and humidity on total fine fuel moisture was offset by increases in incident solar radiation, day length, and KBDI, which promote lower dead fine fuel moisture (Schroeder and Buck, 1970). It is also possible that fine fuel moisture content is equalized by desiccation prior to combustion as the flaming front approaches. These results contrast past studies identifying a negative effect of proportion of live fuels and associated moisture content on MCE in tropical grassland ecosystems (Ward et al., 1992, 1996; Hoffa et al., 1999), perhaps because seasonal variation in dead fuel moisture is not as strong in those environments.

It is possible that the increase in $EF_{PM2.5}$ with time from winter to summer was in response to ambient air conditions. Although higher temperature has been predicted to decrease $EF_{PM2.5}$ in other combustion environments (Nam et al., 2008), specific humidity, which increased in the warmer months, has been found to increase $EF_{PM2.5}$ in studies of combustion engines through the absorption of energy and displacement of oxygen by water in the intake air (McCormick et al., 1997; Choi et al., 2000; Rahai et al., 2011). Specific humidity may have a similar effect in natural wildland fires, but such an effect has not yet been studied.

The negative effect of % grass on $EF_{PM2.5}$ is consistent with studies in tropical savannas (Ward et al., 1992, 1996; Hoffa et al.,

Table 2
Mean and standard error of $EF_{PM2.5}$ and MCE for flaming combustion (EF-F, MCE-F) and smoldering combustion (EF-S, MCE-S) measurements (n) for prescribed burns conducted in the dormant season (Jan–Mar) and growing season (Apr–Jul) and at 1–2 or 3–4 years since fire (YSF).

Season	YSF	EF-F	n	MCE-F	n	EF-S	n	MCE-S	n
Dormant	1–2	14.7 ± 2.5	11	0.957 ± 0.005	6	66.2 ± 52.8	5	0.913 ± 0.031	4
Growing	1–2	22.9 ± 2.6	16	0.938 ± 0.004	15	96.9 ± 89.7	9	0.871 ± 0.073	9
Dormant	3–4	23.6 ± 4.1	6	0.953 ± 0.011	4	85.5 ± 104	3	0.817 ± 0.141	2
Growing	3–4	27.9 ± 3.0	9	0.931 ± 0.012	6	191 ± 108	5	0.782 ± 0.079	5

1999). This pattern is attributable to most grass species having a highly aerated structure and thin membranes which promote rapid drying and well-oxygenated combustion dominated by the flaming phase, as well as relatively high energy content compared to other fuel components (Hough, 1969).

According to the SEM, percent pine needles was the strongest single predictor of $EF_{PM2.5}$, consistent with a previous study showing logging slash with ponderosa pine and Douglas fir needles to have an EF_{PM} up to seven times that of slash without needles (Sandberg, 1974). This mechanism is not known, but pine needles are thicker than most grasses and have relatively high concentrations of various terpenes (White, 1994; Zhao et al., 2011), which during increases in sub-flaming temperatures are readily volatized, degrade through exothermic oxidation reactions, and rapidly condense into PM (McGraw et al., 1999). The effect of pine needle content on $EF_{PM2.5}$ may be considerable in southeastern U.S. pine communities where percentage of fine fuel composed of pine needles varies from nearly zero to nearly 100 in correspondence with pine timber volume and its competitive effects on herbaceous fuels (Wolters, 1981; Harrington and Edwards, 1999; Pecot et al., 2007; Robertson and Ostertag, 2007).

According to the ANOVA analysis, prescribed fire frequency may have a measurable negative effect on $EF_{PM2.5}$ in pine-grasslands. This effect most likely reflects the influence of fine fuel accumulation on fine fuel bulk density, which increased from an average of 6.1 kg m^3 one year following fire to 19.0 kg m^3 four years following fire. In the SEM, total fine fuel load, serving as a proxy for years since previous fire and fine fuel bulk density, had a positive effect on $EF_{PM2.5}$ primarily through its negative influence on fine fuel % grass. The dependence of grass dominance on high fire frequency within the range of 1–4 years since fire is well known (Waldrop et al., 1992; Glitzenstein et al., 2003), attributable to reduction of woody plant dominance and release of herbs from competition (Blair, 1971; Harrington and Edwards, 1999) as well as promotion of sexual reproduction (Platt et al., 1991). However, the SEM also suggests that effects of time since fire on $EF_{PM2.5}$ can be canceled out by the decrease in fine fuel % needles, attributable to increases in the other fine fuel litter category.

Results of the study have implications for management of pine communities with the goal of reducing $PM_{2.5}$ emissions from prescribed burning. Timber thinning is predicted to reduce $EF_{PM2.5}$ during prescribed burns or wildfires by reducing pine needle litter loads. Both timber thinning and frequent burning release grasses and forbs from competition (Harrington and Edwards, 1999; Pecot et al., 2007), which is predicted to decrease $EF_{PM2.5}$. Thus, the recommendation to thin pine timber to less than 15 m^2 ha^{-1} and apply frequent (1–2 year interval) fire for native wildlife management and plant biodiversity (Masters et al., 2003) is predicted to minimize $EF_{PM2.5}$ as well as total $PM_{2.5}$ emissions on an area per burn basis. Such frequent burning would likely increase the total $PM_{2.5}$ emissions when integrated over multiple years, but on average it would decrease the likelihood of exceeding daily and annual standards as monitored by EPA and reduce emissions in the case of wildfire. Dormant season burns are predicted to have lower $EF_{PM2.5}$ and are often easier to accomplish because of the higher proportion of dead fuel and predictable weather patterns within the region. However, growing season burns are advantageous for reducing woody shrub vegetation (Robertson and Hmielowski, 2014), releasing grasses and forbs (Glitzenstein et al., 2003), and mimicking the historic pattern of lightning ignitions and associated native grass reproduction (Platt et al., 1991), which could possibly reduce $PM_{2.5}$ emissions over time.

Although it is beyond the scope of this study to present a useable model for predicting $EF_{PM2.5}$ over a wide range of wildland fire applications, the results from this and past studies suggest that models predicting $EF_{PM2.5}$ based on fire environment inputs can and should be further developed using additional empirical data. For example, our results predict $EF_{PM2.5}$ to nearly double between burning every 1–2 years in the dormant season and burning every 3–4 years in the growing season, which has significant implications for emissions estimates. A productive approach toward further development of PM emission models would be to accommodate the input of common land use variables, including pine timber stocking, frequency of fire, surface vegetation composition (proportion of grass, forbs, shrubs), and season of burn, for predicting $EF_{PM2.5}$.

Acknowledgments

We thank the Pebble Hill Foundation for providing field sites and land management support, Djanan Nemours, Kristina Hatakka, Timothy Malo, Tracy Hmielowski, George Bruner, Bailey Spitzner, Meredith Liedy, Devin Joseph, Rachel Burnette, and Christine Sloan for assistance with field and laboratory work, and Kaye Gainey for editing. We thank two anonymous reviewers for their helpful comments which greatly improved the article. The project was funded by National Science Foundation Atmospheric Chemistry Program Award (Grant no. #AGS-0962970), a grant from Gene Phipps, and Tall Timbers Research Station and Land Conservancy.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2014.09.058.

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