Author/s:
Reisen, F;Meyer, CP;Weston, CJ;Volkova, L

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Reisen Fabienne (Orcid ID: 0000-0002-8500-6545)
Volkova Liubov (Orcid ID: 0000-0002-4556-3478)

Ground-based Field Measurements of $PM_{2.5}$ Emission Factors from Flaming and Smouldering Combustion in Eucalypt Forests

F. Reisen[1], C. P. Meyer[1], C. J. Weston[2], and L. Volkova[2]

[1]CSIRO Oceans and Atmosphere, Climate Science Centre, Aspendale, Victoria, Australia.

[2]School of Ecosystem and Forest Sciences, Faculty of Science, University of Melbourne, Creswick, Victoria, Australia

Corresponding author: Fabienne Reisen (Fabienne.reisen@csiro.au)

Key Points:

- Measured $PM_{2.5}$ emission factors (EFs) for flaming and smouldering combustion in eucalypt forests are consistent with EFs from other forest fuels.
- At low combustion efficiency, particle emission factors vary significantly due to the distinct combustion processes of glowing char combustion and pyrolysis.
- A single relationship between MCE and $PM_{2.5}$ emission factors does not explain the variability in EFs for a wide range of burning conditions.

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Abstract

In fire-prone areas such as southern Australia and parts of the United States, prescribed burning is a common fire management tool to reduce fuel load for wildfire suppression purposes. The burns are typically undertaken during calm and stable conditions when the burn extent and duration can be carefully controlled. This often coincides with poor atmospheric ventilation, leading to a build-up of smoke which can impact air quality and human health. The low intensity of these burns also means that the plume is less buoyant and the main impact on local populations is due to emissions during the slow and prolonged smouldering combustion of heavy fuels.

This study presents emissions measurements of $PM_{2.5}$ at prescribed burns in eucalypt forests of southern Australia using a smoke collection method suitable for both flaming combustion of fine fuels and residual smouldering combustion of heavy fuels and logs. The median $PM_{2.5}$ emission factors (EFs) measured were 16.9 g kg^{-1} fuel during flaming combustion and 38.8 g kg^{-1} fuel during smouldering combustion. The correlation between $PM_{2.5}$ EFs and modified combustion efficiency (MCE) highlights two distinct trends at low MCE, attributed to the distinct combustion processes of glowing char combustion and pyrolysis. Hence two distinct relationships were developed that best fitted the measurements and that can be used to extrapolate measured EFs to a wider range of fuel and burning conditions. The results from this study addressed a gap in our knowledge of particle emissions during burns in eucalypt forests under different burning conditions and help to better forecast and manage air quality impacts from prescribed burns on nearby communities.

1 Introduction

Biomass burning is a significant contributor to regional and global carbon emissions, with global emissions estimated at ~2.0 Pg C yr^{-1} (Kaiser et al., 2012; Li et al., 2014; Seiler & Crutzen, 1980; van der Werf et al., 2010). Global carbon emissions show substantial inter-annual variations; in particular higher carbon emissions were observed in the two El Nino years of 1997 and 1998 (van der Werf et al., 2010) and during the widespread forest and peatland fires in Southeast Asia in 2015 (Huijnen et al., 2016). In Australia, net carbon fluxes due to fire emissions are estimated at 26 Tg C yr^{-1} (Haverd et al., 2013). Approximately 95% of carbon emissions is in the form of carbon dioxide (CO_2), carbon monoxide (CO) and methane (CH_4) (Andreae & Merlet, 2001), with fine particles ($PM_{2.5}$) accounting for less than 5% of carbon emissions (Reid et al., 2005). Although $PM_{2.5}$ makes up only a small fraction of carbon emissions, it is a major component in bushfire smoke that consistently exceeds air quality guidelines (Keywood et al., 2015; Reisen & Brown, 2006; Reisen et al., 2015) and is responsible for the majority of adverse health impacts from biomass burning (Johnston et al., 2012). Additionally smoke particles lead to regional climate effects (Liu et al., 2014). Under a changing climate, large fire events are likely to occur more frequently in some parts of the world such as western US, Canada, Australia and Russia (Flannigan et al., 2009; Jolly et al., 2015; Keywood et

al., 2013; Spracklen et al., 2009; Turetsky et al., 2011), impacting on air quality and population health.

In Australia a program of prescribed burning is routinely carried out by fire and land management agencies to reduce fuel load (Fernandes & Botelho, 2003; McCaw, 2013), as well as for ecological purposes (Burrows, 2008). These burns are typically undertaken during calm and stable conditions when the burn extent and duration can be carefully controlled and risk minimised. Poor atmospheric ventilation in such conditions can lead to a build-up of smoke from the burn in the atmospheric boundary layer. As a result, these burns can have a significant impact on regional air quality and affect population health (Haikerwal et al., 2015). Prescribed burning practices may vary between countries; for example in the US conditions of low to moderate steady winds and some vertical convection are sought to increase smoke dispersion (https://www.bugwood.org/pfire/weather.html).

This study was prompted by the requirement for a PM emission sub-model for smoke dispersion and pollution impact modelling in forested landscapes dominated by the fire-adapted genus *Eucalyptus* (Wain et al., 2008). This is a core tool used by fire agencies in SE Australia to predict smoke dispersion from prescribed burning in order to minimise the risk of health impacts on regional populations. The sub-model was required to estimate both $PM_{2.5}$ emitted into the main smoke plume by flaming combustion at the fire front, and residual smouldering combustion (RSC) emissions in the hours following the fire. The latter form the weakly lofted plume that remains close to the ground and which is often the source of significant local pollution. Prescribed burns are usually of low to moderate intensity with fireline intensities of <500 - 2500 kW m^{-1} (Byram, 1959; McCaw, 2013) contrasting to severe wildfires with fireline intensities larger than 10,000 kW m^{-1} (Volkova et al 2014). For low-intensity prescribed burns buoyant plumes are often limited and RSC can be significant.

In order to manage smoke impacts from prescribed burns it is essential to have improved observational data on the characteristics of gaseous and particle pollutants released by fires for fuels and conditions typical of prescribed burns.

Emission factors (EFs) are a critical input into dispersion models to analyse or forecast smoke dispersion and estimate impacts on air quality and populations. EFs specify the mass of a gas or aerosol species emitted per unit mass of fuel burned. They are usually derived from emission ratios (ERs) and measured in controlled tests in the laboratory, or in smoke plumes from landscape fires which are sampled either from aircraft or at ground level. There are relatively few reliable measurements of $PM_{2.5}$ EFs from forest fires. Published $PM_{2.5}$ EFs in the literature data reviewed by Andreae and Merlet (2001), Akagi et al. (2011) and Urbanski (2014) provide a limited set of $PM_{2.5}$ EFs from wildfires or prescribed burns in temperate forests (either pine-oak forests or coniferous forests in the US). Data for $PM_{2.5}$ EFs from other globally important vegetation classes, such as the eucalypt forests of southern Australia, are rare (Schauer et al., 2001) .

Each of the three approaches to smoke sampling (laboratory experiments, plume sampling from aircraft, and ground-level field sampling) have their advantages and limitations. The advantage of laboratory studies lies in the capacity to control the combustion within well-defined limits and to deploy complex instrumentation to measure a wide range of smoke and fire properties (e.g. NOAA FIREX intensive laboratory experiments https://www.esrl.noaa.gov/csd/projects/firex/firelba/). Laboratory tests are often the only way of quantifying EFs for unstable, trace organic chemical species (Stockwell et al., 2015). The limitation of laboratory tests lies in accurately reproducing the properties of landscape sized fires.

Aircraft can sample smoke plumes produced as the fire front progresses across the landscape. These plumes consist mostly of the products of flaming combustion of the fine fuel, with smaller contribution from smouldering combustion of larger fuel elements in the wake of the fire front. Large wildfires can entrain and loft smoke from smouldering fuels, but for prescribed burns the long duration post-fire front of smouldering heavy fuels will often not be fully captured (Liu et al., 2017; Urbanski, 2013) These samples have very limited temporal coverage and therefore may underestimate the persistent smouldering emissions from burning logs. Smoke emissions from these large sized fuels can continue for many hours, and sometimes days after passage of the fire front (Ottmar, 2014).

The strength of ground-level sampling is the certainty with which the emission source, and combustion state are known and sampling can specifically focus on the smouldering component. The weaknesses of ground-based methods are the large variability in point source emission rates, and the very small fraction of total smoke emitted by the fire which is actually sampled. Therefore, the potential for sampling bias should be always in mind when interpreting point-source measurements.

The most appropriate method of sampling depends on the required application of the EF. Over large temporal and spatial scales, where emissions are estimated directly from fire area, averaged EFs measured on well mixed plumes are preferred. For emissions inventories that are stratified by fuel source (e.g. fine or coarse fuels) and used in model-based algorithms such as Consume (Prichard et al., 2006), FEPS (Anderson et al., 2004) and FINN (Wiedinmyer et al., 2011), EFs for each source and fire category (e.g. flaming, smouldering) are required.

Many studies have looked at particle emissions from the combustion of various fuels under controlled laboratory conditions (Aurell et al., 2015; Bertschi et al., 2003; Chen et al., 2007; Christian et al., 2003; Freeborn et al., 2008; Hays et al., 2002; Holder et al., 2016; Hosseini et al., 2013; McMeeking et al., 2009; Yokelson et al., 2008), however ground-based field measurements of PM are few (Aurell et al., 2015; Holder et al., 2016; Robertson et al., 2014; Urbanski et al., 2009). Very little is also known on particle emissions from RSC of coarse woody debris (CWD), e.g. logs larger than 50mm in diameter. Without specific emission inventories for RSC, emissions are likely to be underestimated in forecasting models and emission inventories.

The aims of this study are (1) to develop fuel specific $PM_{2.5}$ EFs measured during prescribed burns in eucalypt forests of southern Australia under different combustion phases and assess whether there is a consistency in $PM_{2.5}$ EFs across various forest fuels; (2) to find a relationship between $PM_{2.5}$ EFs and MCE that can explain much of the observed variation in $PM_{2.5}$ EFs and be used over a large range of combustion conditions; (3) to assess how PM emissions vary with combustion conditions in order to develop more process based emission algorithms.

2 Methods

Emission measurements were undertaken at four prescribed burns in dry shrubby eucalypt forests in Victoria, Australia (Figure 1): Greendale, 80 km northwest of Melbourne, on 13 April 2015 (293 ha, spring burn); Castlemaine, 130 km northwest of Melbourne, on 23 April 2015 (171 ha, spring burn); Bambra-Deans Marsh, 120km southwest of Melbourne, on 30 September 2015 (56 ha, autumn burn); and Campbells Creek, 130 km northwest of Melbourne, on 1 October 2015 (80 ha, autumn burn).
The burned fine surface fuels consisted of leaf litter, bark and twigs (diameter < 6 mm) and other loose organic materials. These were primarily consumed during flaming combustion. Sampling of smouldering combustion focused on heavy fuels (diameter > 50 mm), e.g. CWD and stumps on the forest ground. Meteorological and fuel data were collected at specific burn sites and are shown in Table 1. The CWD fuel load was quite variable between sites ranging from 1.9 t ha^{-1} to 32.5 t ha^{-1} (Table 1), yet within the range of the CWD fuel loads for the similar Victorian forests reported previously (34.6±2.95 t ha^{-1}, mean ± s.e.) (Volkova et al., 2018).

Fuel moisture was estimated using the fine fuel moisture meter (ME 2000, Wiltronics, Ballarat, Australia). Each measurement included two to three samples of suspended dead leaves collected within shaded locations prior to burning. Fuel moisture was estimated at 8-16%, which is based on the conditions under which fire agencies typically conduct prescribed burns in eucalypt forests in south eastern Australia.

Further information on the burn sites and fuel loads is provided in supplementary materials (Volkova & Weston, 2013).

Sampling was done using a portable backpack smoke collector suitable for sampling close to the emission sources of prescribed fires as described in Meyer et al. (2012). Air was sampled at approximately 1 L per minute via a 2.5 m × 12 mm diameter stainless steel sampling probe and in-line Teflon filters into a 10 L Tedlar bag mounted on the backpack. Sampling was done approximately 0.5 m above the flame where it is expected that the combustion has ceased due to cooling and dilution by entrained air. Three additional gas lines allowed for continuous measurement of $PM_{2.5}$, CO_2 and CO, and collection of total suspended particulate matter (TSP) on 47mm Fluoropore filters. Concentrations of CO_2 and CO were measured continuously with a Q-Trak (model 7565, TSI Inc., USA) and particulates were measured continuously with a

DustTrak (model 8530, TSI Inc., USA). Further details on the sampling and analysis methodology are provided in supplementary materials.

Smoke samples were collected (1) from flaming combustion by positioning the tip of the sampling probe approximately 0.5 m above the flame and (2) from smouldering-phase combustion behind the fire front and from burning of heavy fuels and logs. At each burn, 8-15 samples were collected during flaming combustion, 4-8 samples were collected during smouldering combustion, and two clean air samples were collected into Tedlar bags to provide background concentrations of gases. Background concentrations of CO and $PM_{2.5}$ were less than 1% of signal measured during the burns while background CO_2 concentrations were ~25% of measured concentrations during the burns.

All samples collected within the Tedlar bags were taken back to the CSIRO laboratory and analysed for CO, CO_2 and CH_4 within 12 hours of collection using a cavity ring-down spectrometer (Los Gatos Research Inc., CA) (Meyer et al., 2012). The pre-weighed 47mm Fluoropore membrane filters (1 μm pore size, Merck Millipore) were analysed for gravimetric mass using a Mettler UMT2 ultra-microbalance with a specialty filter pan in a temperature and humidity controlled environment. Filters collected for gravimetric mass concentrations were also used for analysis of anhydrosugars including levoglucosan and water-soluble ions as described in Meyer et al. (2008b).

EFs are usually derived from emission ratios (ERs) of combustion products. The ERs were calculated following Meyer et al. (2012):

$$ER_i = \frac{\Delta C_i}{\Delta CO_2 + \Delta CO + \Delta CH_4} \qquad (1)$$

where ΔCi is the difference between the molar concentration of trace species *i* in the smoke sample and its concentrations in ambient air up-wind of the combustion source.

EFs were expressed relative to combusted fuel mass (Andreae & Merlet, 2001) defined as:

$$EF_i = CC \times ER_i \quad (2)$$

where CC is the carbon content of the burned fuel, estimated at 50% (Meyer & Cook, 2015; Surawski et al., 2015). This approach is based on the simplified assumption that all fuel carbon is volatilized and emitted and does not take into account the changes between the pre- and post-fire carbon content (Surawski et al., 2016). This may lead to an overestimation in emissions, in particular for smouldering char combustion.

In order to characterize the relative amount of flaming and smouldering combustion, the modified combustion efficiency (MCE) is used defined as:

$$MCE = \frac{\Delta CO_2}{\Delta CO + \Delta CO_2} \qquad (3)$$

While pure flaming has an MCE near 0.99, smouldering combustion occurs at low MCE (~0.65-0.85) (Akagi et al., 2011).

3 Results

Table 2 shows the burn-average and study-average $PM_{2.5}$ EFs (in g kg^{-1} fuel) for the flaming (MCE of 0.94 ± 0.02) and smouldering combustion (MCE of 0.83 ± 0.04) phases. Statistical t-tests indicate that $PM_{2.5}$ EFs were significantly higher during the smouldering combustion ($p<0.01$). $PM_{2.5}$ EFs from flaming combustion (MCE of 0.91-0.97) were consistent among all burns and ranged from 3 to 34 g kg^{-1}. Higher averaged $PM_{2.5}$ EFs were measured at Bambra, when temperatures were lower and relative humidity higher. Median MCEs for the flaming samples were 0.92 at Bambra compared to 0.95 at the other burn sites. This is likely to explain the higher $PM_{2.5}$ EFs measured at Bambra. $PM_{2.5}$ EFs from smouldering combustion (MCE of 0.76-0.89) ranged from 10-133 g kg^{-1}; highest EFs were measured at Castlemaine and lowest EFs were measured at Campbells Creek, where temperatures were high and relative humidity low. There was no difference in EFs between autumn and spring burns (Figure 2).

Correlation between $PM_{2.5}$ and MCE shows an increase in $PM_{2.5}$ EFs with decreasing MCE (Figure 2), and an indication of two classes of $PM_{2.5}$ EFs at MCE < 0.9; visual assessment suggests that these corresponded to smoke samples from logs where there was either glowing char combustion or pyrolysis without a visible flame (Figure 3). Separating the smouldering combustion into the two different processes revealed that the pyrolytic process has the highest variability (standard deviation of 31 g kg^{-1}) and also the highest $PM_{2.5}$ EFs (Figure 4). No difference in EFs between the two smouldering combustion processes was observed for CO or CH_4 (Table S2).

The chemical speciation of $PM_{2.5}$ shows that higher emissions of potassium (K) were observed during flaming combustion whereas emissions of levoglucosan (a biomass burning marker) dominated the smouldering combustion (Table 3). The mass fraction of inorganic species was higher during flaming combustion than during smouldering combustion, while smouldering samples had higher concentrations of organic species such as levoglucosan, oxalate, acetic and formic acid (Figure 4). Among the inorganic species, chloride, potassium, sulfate and ammonium were the dominant soluble inorganic species accounting for 84-99% of the soluble inorganics and for 0.2-3.9% of the $PM_{2.5}$ mass concentrations.

Ion balance shows an excess of anions, which is more prevalent in the smouldering samples. Since the free acidity of the sample was not measured, no corrections were applied to distinguish between the dissociated (ionised) and undissociated species of the organic acids. This is likely to explain the excess of anions observed in the samples primarily for the smouldering samples that had a high proportion of organic acids. In fact higher free acidity has been observed

in wood smoke samples (Keywood et al., 2000) and this would lead to a larger proportion of undissociated species.

4 Discussion

4.1 Emission factors of $PM_{2.5}$ in eucalypt forests

This study reports ground-based measurements of $PM_{2.5}$ EFs for both flaming and smouldering combustion in the open eucalypt forests of south-eastern Australia, where prescribed burning is a valuable tool for managing fuels to mitigate the behaviour of high intensity fires (McCaw, 2013).

We observed a consistency in $PM_{2.5}$ EFs within forest fuels under similar combustion phases. The median value for flaming combustion of 16.9 g kg^{-1} fuel (Figure 4) is in the range of previously reported values for flaming combustion of forest fuels (11-29 g kg^{-1}) (Alves et al., 2011; Aurell & Gullett, 2013; Aurell et al., 2015; Balachandran et al., 2013; Burling et al., 2011; Holder et al., 2016; McMeeking et al., 2009; Robertson et al., 2014; Urbanski et al., 2009; Yokelson et al., 2011). Figure 6 further highlights the consistency in $PM_{2.5}$ EFs between measurements at prescribed burns in SE Australia and published data from fires or prescribed burns in temperate forests, including both field and laboratory measurements. Fires in grassland and shrubland were excluded from the comparison as the fuel type and burning conditions in those biomes differ from those in forest fuels.

Flaming combustion in this study produced slightly higher $PM_{2.5}$ EFs than those measured in smoke plumes measured by aircraft (Figure 6a). A similar trend was observed for tower-based field measurements (Figure 6b). This could be attributed to differences in sampling methods, e.g. near source versus plume measurements. The higher plume dilution in airborne measurements due to clean background air becoming entrained into the smoke plume would cause a shift in semi-volatile species into the gas-phase which may account for the difference between EFs (Holder et al., 2016; May et al., 2014). In low dilution smoke plumes with higher concentrations of organic aerosols, such as those measured on the ground, semi-volatile species would predominantly occur in the particle phase (May et al., 2013).

For laboratory and ground-based measurements, $PM_{2.5}$ EFs from this study were consistent with the literature (Figure 6c). For flaming combustion (MCE of 0.92-0.97), there was a good agreement between the field measurements from this study (average $PM_{2.5}$ EFs of 17.4 ± 7.2 g kg^{-1}) and laboratory measurements (average $PM_{2.5}$ EFs of 15.5 ± 11.1 g kg^{-1}). Similarly, there was good agreement between this data set (average $PM_{2.5}$ EFs of 17.4 ± 7.2 g kg^{-1}) and published data from ground-based field measurements (average $PM_{2.5}$ EFs of 20.8 ± 9.4 g kg^{-1}) (Figures 6b and d).

$PM_{2.5}$ EFs were determined over a wide range of MCE levels, thereby filling a significant gap in our understanding of $PM_{2.5}$ emissions from RSC. Previous studies using airborne and ground-based field measurements were dominated by flaming combustion with an MCE > 0.9.

Data on $PM_{2.5}$ EFs at low MCE are sparse, with a few laboratory-based experiments looking at the smouldering phase (Bertschi et al., 2003; Chen et al., 2007; May et al., 2014), one field-based study (Robertson et al., 2014), and two aircraft measurement studies (Hobbs et al., 1996; Liu et al., 2017). In addition, there are reported values of $PM_{2.5}$ EFs at low MCE from peat fires (Stockwell et al., 2016) and burns in organic soils (Geron & Hays, 2013). Due to differences in fuel composition and combustion for peat fires and fires in organic soils, those studies were not included in the comparison of $PM_{2.5}$ EFs.

The overall median $PM_{2.5}$ EF of 38.8 g kg^{-1} fuel for smouldering combustion is in agreement with a recent estimate of 33 g kg^{-1} from the combustion of smouldering stumps and logs (Urbanski, 2014) and with an estimate of ~34 g kg^{-1} for smouldering combustion measurements (Reid et al., 2005). The recent study by Robertson et al. (2014) showed higher particle EFs during the smouldering combustion (Figure 6d). Robertson et al. (2014) collected near-source smoke samples during prescribed burns in open-canopy pine-grasslands in Florida and Georgia and collected samples from both flaming combustion and smouldering combustion of fine fuels after the passage of the flaming front. The higher particle emissions observed by Robertson et al. (2014) were measured at lower MCE and are likely due to fuel composition and fuel structure (e.g. pine needles vs eucalypt leaf litter). Measured CWD fuel load was also higher in the Australian eucalypt forests (mean of 34.6 t ha^{-1}) compared to the measured woody fuel load in the pine-grasslands (mean of 1.4 t ha^{-1}) (Robertson et al., 2014). This may favour char combustion over pyrolysis and therefore result in lower $PM_{2.5}$ EFs.

4.2.Relationship between $PM_{2.5}$ EFs and MCE

Our results show that a single relationship between $PM_{2.5}$ EFs and MCE does not explain the variability in EFs for a wide range of MCE values. At an MCE value of approximately 0.87, the relationship between $PM_{2.5}$ EFs and MCE seems to split into two distinct trends (Figure 7). We attribute these distinct trends to the combustion processes as described in Yokelson et al. (1997) and Bertschi et al. (2003). During pyrolysis, carbon-enriched particles are emitted, and their emissions increase as combustion efficiency decreases. Once char is formed, gasification can occur in which solid char is converted to gas-phase products such as CO_2 and CO, also referred to as glowing combustion. This is characterised by an increase in CO emissions but no concurrent increase in PM emissions. A similar trend has been observed in domestic wood-heating combustion (Meyer et al., 2008a). Distinction between the two combustion processes at low MCE was based on a visual assessment from the data presented in Figure 7 as well as visual determination at the burn sites (Figure 3). Based on these visual assessments, we observed a 2.5-fold difference in median $PM_{2.5}$ EFs for smouldering combustion dominated by pyrolysis (no visible flame) and for smouldering combustion dominated by char combustion (Figure 4). The significantly higher $PM_{2.5}$ EFs observed during pyrolysis at low MCE have previously been observed in only one research study (Robertson et al., 2014).

Fitting a linear relationship between MCE and $PM_{2.5}$ EFs for all flaming and smouldering samples (eq.4) underestimates the higher $PM_{2.5}$ EFs observed at low MCE (dotted line in Figure

7). An empirical exponential function, the exponential Stirling curve (eq. 5), best explains the relationship between MCE and $PM_{2.5}$ EFs for flaming and smouldering combustion phases by pyrolysis. The same non-linear function was used to explain the relationship between CH_4 EFs and MCE for measurements conducted in eucalypt forests in southern Australia (Volkova et al., 2014) and in savanna grasslands in northern Australia (Meyer & Cook, 2015; Meyer et al., 2012) and can also be applied to the data set from this study (Figure S2) . Similarly the relationship between MCE and $PM_{2.5}$ EFs for glowing combustion can be explained with an exponential Stirling curve, but shows more variability (eq. 6).

$$EF_{PM} = (-313.4 \pm 30.5) \times MCE + (312.7 \pm 27.7) \quad (r^2 = 0.53) \qquad (4)$$

$$EF_{PM} = 26.3\left(e^{8.6(1-MCE)} - 1\right) \quad (r^2 = 0.92) \qquad (5)$$

$$EF_{PM} = 2.3E7\ (e^{9.8E-6(1-MCE)} - 1) \quad (r^2 = 0.57) \qquad (6)$$

Linear fits between $PM_{2.5}$ EFs and MCE, have previously been determined in laboratory studies of combustion of fine fuels and in airborne and ground-based measurements. Different gradients were observed, varying between -67 and -311 with a median of -204 ± 92 (Table 4). These results indicate that MCE alone does not explain all of the variance in EF for PM, but that other factors such as fuel loading, fuel characteristic (e.g. layering, structure), combustion processes, ambient air conditions (e.g. temperature and humidity) as well as differences in sampling, dilution and measurement methods may contribute to the variance in particle emissions. This variance in gradients was also determined by Meyer and Cook (2015) who showed that the relationship between methane EFs and MCE differed for fuel structure, e.g. having an open structure such as that observed for grass fires vs a more dense understorey layer as that observed in coniferous and deciduous forests for North America and Northern Europe. For example, the low $PM_{2.5}$ EFs at low CE observed by Ward & Hardy (1991) were observed during burning of piles residues. This may have favoured char combustion leading to low $PM_{2.5}$ EFs.

The observed variability in $PM_{2.5}$ EFs between burns may also be due to differences in fuel moisture. Research studies have shown that high fuel moisture increases emissions of organic aerosols (Chen et al., 2007; Hayashi et al., 2014), which in turn lead to partitioning of semi-volatiles into the particle phase in order to maintain phase equilibrium (Donahue et al., 2006; Robinson et al., 2010). Variation in fuel moisture for larger fuel elements is rarely done, but may be needed to improve PM emissions estimates for RSC.

As MCE alone is not sufficient to differentiate between the two combustion processes at low MCE, we assessed whether chemical speciation of filter samples may be able to provide patterns to differentiate the two combustion processes. Our measurements have shown that emissions of levoglucosan were dependent on both combustion state and fuel type. We observed higher levoglucosan EFs during smouldering combustion consistent with literature data (Schmidl et al., 2008). Measured levoglucosan EFs were also higher than those measured in chaparral fuel

(average of 95.9 ± 35.6 mg kg^{-1}) and oak savanna and woodlands (29.1-58.6 mg kg^{-1}) (Hosseini et al., 2013), but consistent with the EFs reported by Schauer et al. (2001) for wood burning of eucalypt (1.94 g kg^{-1}). Emissions of K were higher during flaming combustion as confirmed in previous studies (Amici et al., 2011; Balachandran et al., 2013; Vodacek et al., 2002). Separating smouldering combustion into char combustion and pyrolysis only provided a limited data set on chemical speciation. The data showed higher organic aerosol in the pyrolysis dominated combustion compared to glowing combustion which had higher concentrations of inorganics (e.g. Na, Cl and K) (Table S2). The levoglucosan to nssK ratio may be a possible marker to distinguish between the two combustion processes at low MCE, but further measurements are required to confirm this.

4.3. Relevance to emissions inventories

Due to the spatial and temporal complexities of fuel characteristics and combustion processes, it is difficult to derive one single EF that is applicable to emissions accounting and smoke forecasting on local to global scales. Biome-averaged $PM_{2.5}$ EFs presented in reviews such as Andreae and Merlet (2001) and Akagi et al. (2011) are relevant to determine fire emissions over large spatial and temporal scales. However to assess smoke impacts on local to regional scales, it is important to take into account the variability in EFs due to combustion chemistry of specific fuel types and the processes that control emissions. Therefore finding a relationship between EF and MCE that is primarily due to combustion state and is consistent across vegetation type and fire type could be very useful for emissions models (IPCC, 2000). Empirical relationships between MCE and EF($PM_{2.5}$) can then be used to extrapolate measured $PM_{2.5}$ EFs to a wider range of burning conditions and combustion processes. The derived explanatory relationships will help to better forecast and manage air quality impacts from prescribed burns on nearby communities. As an example, a linear relationship between EFs and MCE for major pollutants is used in the Fire Emission Production Simulator (FEPS) to calculate emissions from prescribed burns or wildfires (Anderson et al., 2004) and in CONSUME (Computer Models for Fire and Smoke) to predict hourly pollutant emissions (Prichard et al., 2006). These relationships were developed from experimental studies on common fuel types in the US and as shown in Table 4 and Figure S3 can be quite variable. None of the linear fits developed in previous studies are able to align with the high $PM_{2.5}$ EFs observed during the pyrolytic smouldering combustion observed in this study or the data set collected by Robertson et al (2014) at low MCE. Therefore, when developing emissions algorithms it is important to adopt a relationship that is appropriate for the fuel and combustion conditions of interest.

Having a better understanding of how $PM_{2.5}$ EFs vary as a function of combustion processes will improve smoke forecasting, in particular for RSC. While emissions during the rapid and intense flaming combustion of fine fuels are lofted by convection, particles emitted during the slow and prolonged smouldering combustion of heavy fuels remain closer to the ground and can have significant impacts on the local air quality. A research study done in the Huon Valley, Tasmania, Australia, has shown that surface impacts from prescribed burns were

not associated with the main smoke plume that was sufficiently buoyant to be mainly injected into the free troposphere, but due to RSC that occurred at the end of the burn in the evening when mixing depths and ventilation rates were smaller and dispersion was reduced (Meyer et al., 2011).

To assess the significance of residual smouldering combustion on total PM emissions, we used key parameters that underpin Australian national inventory calculations for fire emissions from a recent review by Roxburgh et al. (2015). Recommended fuel loads for Victorian forests were 12.2 t ha^{-1} for fine fuels (diameter< 6 mm) and 32.5t ha^{-1} for CWD, while recommended burning efficiencies during prescribed burns were 0.36 for fine fuels and 0.13 for CWD (Volkova & Weston, 2015). Burning efficiency is the product of fire patchiness and the proportion of available fuel that is burnt. Total $PM_{2.5}$ emissions per ha were calculated as the product of fuel load (t ha^{-1}), burning efficiency and $PM_{2.5}$ EF (g kg^{-1}). Using the measured EFs for $PM_{2.5}$ from this study (e.g. median values as shown in Figure 4), $PM_{2.5}$ emissions from RSC may contribute about 70% to total $PM_{2.5}$ emissions (Table 5). Less variability is observed for fine fuels that are mainly consumed by flaming combustion. However for large fuels consumed during the slow smouldering combustion, we observed a large variability and no single curve could be fitted to explain the relationship between MCE and $PM_{2.5}$ EFs. Currently it is not clear whether CWD is primarily consumed by smouldering pyrolysis or by char combustion and therefore the distinct relationships at low MCE are currently difficult to implement in emission inventories. Initial data indicates that chemistry may be able to distinguish between the two combustion processes. A focus on RSC of fuels is suggested as a prospective area for future studies.

5 Conclusions

The results of the study addressed a gap in our knowledge of particle emissions during burns in southern forests of Australia, both in terms of amount of particles emitted and chemical characterisation under different burning conditions. These are important inputs to both emission inventories and smoke forecasting models. Furthermore, studying $PM_{2.5}$ EFs over a large range of MCEs will help to improve accuracy of emissions estimates and to enhance our understanding on how burning conditions at low MCE influence particle emissions. Information on residual smouldering combustion is scarce, however its impact on the air quality of nearby communities can be significant. A lack of knowledge in emissions from RSC and fuel consumption of CWD by different combustion processes at low MCE can result in large uncertainties in local, regional and global estimates of PM emissions and its impact on air quality. The study clearly identified two distinct trends that are due to combustion processes that occur at low MCE. Due to the large variability in $PM_{2.5}$ EFs observed at low MCE, understanding what causes this variation and how we can distinguish between the various combustion processes can improve local to regional smoke forecasting of $PM_{2.5}$. While MCE distinguishes flaming from smouldering combustion, initial results indicate that PM chemistry (e.g. levoglucosan to K ratio) may be used to further distinguish between glowing char combustion and smouldering pyrolysis.

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The data used are listed in the tables and figures.

References

Akagi, S. K., Yokelson, R. J., Wiedinmyer, C., Alvarado, M. J., Reid, J. S., Karl, T., . . . Wennberg, P. O. (2011). Emission factors for open and domestic biomass burning for use in atmospheric models. *Atmospheric Chemistry and Physics, 11*(9), 4039-4072.

Alves, C., Vicente, A., Nunes, T., Goncalves, C., Fernandes, A. P., Mirante, F., . . . Pio, C. (2011). Summer 2009 wildfires in Portugal: Emission of trace gases and aerosol composition. *Atmospheric Environment, 45*(3), 641-649.

Amici, S., Wooster, M. J., & Piscini, A. (2011). Multi-resolution spectral analysis of wildfire potassium emission signatures using laboratory, airborne and spaceborne remote sensing. *Remote Sensing of Environment, 115*(8), 1811-1823.

Anderson, G. K., Sandberg, D. V., & Norheim, R. A. (2004). *Fire Emission Production Simulator (FEPS) User's Guide, Version 1.0*. Retrieved from http://www.fs.fed.us/pnw/fera/feps/FEPS_users_guide.pdf

Andreae, M. O., & Merlet, P. (2001). Emission of trace gases and aerosols from biomass burning. *Global Biogeochemical Cycles, 15*(4), 955-966.

Aurell, J., & Gullett, B. K. (2013). Emission Factors from Aerial and Ground Measurements of Field and Laboratory Forest Burns in the Southeastern US: PM2.5, Black and Brown Carbon, VOC, and PCDD/PCDF. *Environmental Science & Technology, 47*(15), 8443-8452.

Aurell, J., Gullett, B. K., & Tabor, D. (2015). Emissions from southeastern US Grasslands and pine savannas: Comparison of aerial and ground field measurements with laboratory burns. *Atmospheric Environment, 111*, 170-178. doi:10.1016/j.atmosenv.2015.03.001

Balachandran, S., Pachon, J. E., Lee, S., Oakes, M. M., Rastogi, N., Shi, W. Y., . . . Russell, A. G. (2013). Particulate and gas sampling of prescribed fires in South Georgia, USA. *Atmospheric Environment, 81*, 125-135.

Bertschi, I., Yokelson, R. J., Ward, D. E., Babbitt, R. E., Susott, R. A., Goode, J. G., & Hao, W. M. (2003). Trace gas and particle emissions from fires in large diameter and belowground biomass fuels. *Journal of Geophysical Research-Atmospheres, 108*(D13), -.

Burling, I. R., Yokelson, R. J., Akagi, S. K., Urbanski, S. P., Wold, C. E., Griffith, D. W. T., . . . Weise, D. R. (2011). Airborne and ground-based measurements of the trace gases and particles emitted by prescribed fires in the United States. *Atmospheric Chemistry and Physics, 11*(23), 12197-12216. doi:10.5194/acp-11-12197-2011

Burrows, N. D. (2008). Linking fire ecology and fire management in south-west Australian forest landscapes. *Forest Ecology and Management, 255*(7), 2394-2406.

Byram, G. M. (1959). Combustion of forest fuels. In K. P. Davis (Ed.), *Forest fire: control and use.* (pp. 61-89). New York, NY: McGraw Hill.

Chen, L. W., Moosmuller, H., Arnott, W. P., Chow, J. C., Watson, J. G., Susott, R. A., . . . Hao, W. M. (2007). Emissions from laboratory combustion of wildland fuels: emission factors and source profiles. *Environmental Science & Technology, 41*(12), 4317-4325.

Christian, T. J., Kleiss, B., Yokelson, R. J., Holzinger, R., Crutzen, P. J., Hao, W. M., . . . Ward, D. E. (2003). Comprehensive laboratory measurements of biomass-burning emissions: 1. Emissions from Indonesian, African, and other fuels. *Journal of Geophysical Research-Atmospheres, 108*(D23). doi:10.1029/2003jd003704

Donahue, N. M., Robinson, A. L., Stanier, C. O., & Pandis, S. N. (2006). Coupled partitioning, dilution, and chemical aging of semivolatile organics. *Environmental Science & Technology, 40*(8), 2635-2643.

Fernandes, P. M., & Botelho, H. S. (2003). A review of prescribed burning effectiveness in fire hazard reduction. *International Journal of Wildland Fire, 12*, 117-128.

Flannigan, M. D., Krawchuk, M. A., de Groot, W. J., Wotton, B. M., & Gowman, L. M. (2009). Implications of changing climate for global wildland fire. *International Journal of Wildland Fire, 18*(5), 483-507.

Freeborn, P. H., Wooster, M. J., Hao, W. M., Ryan, C. A., Nordgren, B. L., Baker, S. P., & Ichoku, C. (2008). Relationships between energy release, fuel mass loss, and trace gas and aerosol emissions during laboratory biomass fires. *Journal of Geophysical Research-Atmospheres, 113*(D1), 1301-1301.

Geron, C., & Hays, M. (2013). Air emissions from organic soil burning on the coastal plain of North Carolina. *Atmospheric Environment, 64*, 192-199.

Haikerwal, A., Reisen, F., Sim, M. R., Abramson, M. J., Meyer, C. P., Johnston, F. H., & Dennekamp, M. (2015). Impact of smoke from prescribed burning: Is it a public health concern? *Journal of the Air & Waste Management Association, 65*(5), 592-598.

Haverd, V., Raupach, M. R., Briggs, P. R., Canadell, J. G., Davis, S. J., Law, R. M., . . . Sherman, B. (2013). The Australian terrestrial carbon budget. *Biogeosciences, 10*(2), 851-869.

Hayashi, K., Ono, K., Kajiura, M., Sudo, S., Yonemura, S., Fushimi, A., . . . Tanabe, K. (2014). Trace gas and particle emissions from open burning of three cereal crop residues: Increase in residue moistness enhances emissions of carbon monoxide, methane, and particulate organic carbon. *Atmospheric Environment, 95*, 36-44.

Hays, M. D., Geron, C. D., Linna, K. J., Smith, N. D., & Schauer, J. J. (2002). Speciation of gas-phase and fine particle emissions from burning of foliar fuels. *Environmental Science & Technology, 36*(11), 2281-2295. doi:10.1021/es0111683

Hobbs, P. V., Reid, J. S., Herring, J. A., Nance, J. D., Weiss, R. E., Ross, J. L., . . . Liousse, C. (1996). Particle and trace-gas measurements in the smoke from prescribed burns of forest products in the Pacific Northwest. *Biomass Burning and Global Change* (pp. 697–715.). Cambridge, MA: MIT Press.

Holder, A. L., Hagler, G. S. W., Aurell, J., Hays, M. D., & Gullett, B. K. (2016). Particulate matter and black carbon optical properties and emission factors from prescribed fires in the southeastern United States. *Journal of Geophysical Research-Atmospheres, 121*(7), 3465-3483. doi:10.1002/2015jd024321

Hosseini, S., Urbanski, S. P., Dixit, P., Qi, L., Burling, I. R., Yokelson, R. J., . . . Cocker, D. R. (2013). Laboratory characterization of PM emissions from combustion of wildland biomass fuels. *Journal of Geophysical Research-Atmospheres, 118*(17), 9914-9929.

Huijnen, V., Wooster, M. J., Kaiser, J. W., Gaveau, D. L. A., Flemming, J., Parrington, M., . . . van Weele, M. (2016). Fire carbon emissions over maritime southeast Asia in 2015 largest since 1997. *Scientific Reports, 6*, 26886-26886.

IPCC. (2000). *Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories.* Retrieved from IPCC National Greenhouse Gas Inventories Programme, Technical Support Unit, Kanagawa, Japan: http://www.ipcc-nggip.iges.or.jp/public/gp/english/

Janhall, S., Andreae, M. O., & Poschl, U. (2010). Biomass burning aerosol emissions from vegetation fires: particle number and mass emission factors and size distributions. *Atmospheric Chemistry and Physics, 10*(3), 1427-1439.

Johnston, F. H., Henderson, S. B., Chen, Y., Randerson, J. T., Marlier, M., DeFries, R. S., . . . Brauer, M. (2012). Estimated Global Mortality Attributable to Smoke from Landscape Fires. *Environmental Health Perspectives, 120*(5), 695-701.

Jolly, W. M., Cochrane, M. A., Freeborn, P. H., Holden, Z. A., Brown, T. J., Williamson, G. J., & Bowman, D. (2015). Climate-induced variations in global wildfire danger from 1979 to 2013. *Nature Communications, 6*, 7537-7537.

Kaiser, J. W., Heil, A., Andreae, M. O., Benedetti, A., Chubarova, N., Jones, L., . . . van der Werf, G. R. (2012). Biomass burning emissions estimated with a global fire assimilation system based on observed fire radiative power. *Biogeosciences, 9*(1), 527-554.

Keywood, M., Cope, M., Meyer, C. P., Iinuma, Y., & Emmerson, K. (2015). When smoke comes to town: The impact of biomass burning smoke on air quality. *Atmospheric Environment*. doi:10.1016/j.atmosenv.2015.03.050

Keywood, M., Kanakidou, M., Stohl, A., Dentener, F., Grassi, G., Meyer, C. P., . . . Burrows, J. (2013). Fire in the Air: Biomass Burning Impacts in a Changing Climate. *Critical Reviews in Environmental Science and Technology, 43*(1), 40-83.

Keywood, M. D., Ayers, G. P., Gras, J. L., Gillett, R. W., & Cohen, D. D. (2000). Size distribution and sources of aerosol in Launceston, Australia, during winter 1997. *Journal of the Air & Waste Management Association, 50*(3), 418-427.

Li, F., Bond-Lamberty, B., & Levis, S. (2014). Quantifying the role of fire in the Earth system - Part 2: Impact on the net carbon balance of global terrestrial ecosystems for the 20th century. *Biogeosciences, 11*(5), 1345-1360.

Liu, X. X., Huey, L. G., Yokelson, R. J., Selimovic, V., Simpson, I. J., Muller, M., . . . Wolfe, G. M. (2017). Airborne measurements of western US wildfire emissions: Comparison with prescribed burning and air quality implications. *Journal of Geophysical Research-Atmospheres, 122*(11), 6108-6129.

Liu, Y. Q., Goodrick, S., & Heilman, W. (2014). Wildland fire emissions, carbon, and climate: Wildfire-climate interactions. *Forest Ecology and Management, 317*, 80-96.

May, A. A., Levin, E. J. T., Hennigan, C. J., Riipinen, I., Lee, T., Collett, J. L., . . . Robinson, A. L. (2013). Gas-particle partitioning of primary organic aerosol emissions: 3. Biomass burning. *Journal of Geophysical Research-Atmospheres, 118*(19), 11327-11338. doi:10.1002/jgrd.50828

May, A. A., McMeeking, G. R., Lee, T., Taylor, J. W., Craven, J. S., Burling, I., . . . Kreidenweis, S. M. (2014). Aerosol emissions from prescribed fires in the United States: A synthesis of laboratory and aircraft measurements. *Journal of Geophysical Research-Atmospheres, 119*(20), 11826-11849. doi:10.1002/2014jd021848

McCaw, W. L. (2013). Managing forest fuels using prescribed fire - A perspective from southern Australia. *Forest Ecology and Management, 294*, 217-224.

McMeeking, G. R., Kreidenweis, S. M., Baker, S., Carrico, C. M., Chow, J. C., Collett, J. L., . . . Wold, C. E. (2009). Emissions of trace gases and aerosols during the open combustion of biomass in the laboratory. *Journal of Geophysical Research-Atmospheres, 114*. doi:10.1029/2009jd011836

Meyer, C. P., & Cook, G. D. (2015). Biomass combustion and emission processes in the northern Australian savannas. In B. P. Murphy, A. C. Edwards, C. P. M. Meyer, & J. Russell-Smith (Eds.), *Carbon Accounting and Savanna Fire Management* (pp. 368). Clayton, VIC, Australia: CSIRO Publishing.

Meyer, C. P., Cook, G. D., Reisen, F., Smith, T. E. L., Tattaris, M., Russell-Smith, J., . . . Wooster, M. J. (2012). Direct measurements of the seasonality of emission factors from savanna fires in northern Australia. *Journal of Geophysical Research-Atmospheres, 117*, 20305-20305.

Meyer, C. P., Luhar, A. K., Gillett, R., & Keywood, M. D. (2008a). *Measurement of real-world PM10 emission factors and emission profiles from woodheaters by in situ source monitoring and atmospheric verification methods*. Retrieved from Final Report to the Department of Sustainability, Environment, Water, Population and Communities, Melbourne, Australia: http://www.environment.gov.au/atmosphere/airquality/publications/emission-factor.html

Meyer, C. P., Reisen, F., Keywood, M. D., & Crumeyrolle, S. (2011). *Impacts of smoke from regeneration burning on air quality in the Huon Valley, Tasmania. Final Report to Forestry Tasmania.* . Retrieved from FinalReport to Forestry Tasmania, CSIRO, Melbourne, Australia: https://cdn.forestrytasmania.com.au/uploads/File/pdf/pdf2011/huon_valley_csiro_report_2011.pdf

Meyer, C. P. M., Reisen, F., Luhar, A. K., Powell, J. C., Lee, S., Cope, M., . . . Tolhurst, K. (2008b). *Particles, Ozone and Air Toxic Levels in Rural Communities during Prescribed Burning Seasons*. Retrieved from Final Report to the Department of Sustainability, Environment, Water, Population and Communities, Melbourne, Australia: http://www.environment.gov.au/atmosphere/airquality/publications/particles-ozone-toxic.html

Ottmar, R. D. (2014). Wildland fire emissions, carbon, and climate: Modeling fuel consumption. *Forest Ecology and Management, 317*, 41-50. doi:10.1016/j.foreco.2013.06.010

Prichard, S. J., Ottmar, R. D., & Anderson, G. K. (2006). *Consume 3.0 User's Guide.* Retrieved from USDA Forest Service. Pacific Northwest Research Station, Seattle, WA.: https://www.fs.fed.us/pnw/fera/research/smoke/consume/consume30_users_guide.pdf

Reid, J. S., Koppmann, R., Eck, T. F., & Eleuterio, D. P. (2005). A review of biomass burning emissions part II: intensive physical properties of biomass burning particles. *Atmospheric Chemistry and Physics, 5*, 799-825.

Reisen, F., & Brown, S. K. (2006). Implications for community health from exposure to bushfire air toxics. *Environmental Chemistry, 3*(4), 235-243.

Reisen, F., Duran, S. M., Flannigan, M., Elliott, C., & Rideout, K. (2015). Wildfire smoke and public health risk. *International Journal of Wildland Fire, 24*(8), 1029-1044.

Robertson, K. M., Hsieh, Y. P., & Bugna, G. C. (2014). Fire environment effects on particulate matter emission factors in southeastern US pine-grasslands. *Atmospheric Environment, 99*, 104-111. doi:10.1016/j.atmosenv.2014.09.058

Robinson, A. L., Grieshop, A. P., Donahue, N. M., & Hunt, S. W. (2010). Updating the Conceptual Model for Fine Particle Mass Emissions from Combustion Systems. *Journal of the Air & Waste Management Association, 60*(10), 1204-1222.

Roxburgh, S. H., Volkova, L., Surawski, N., Meyer, C. P. M., & Weston, C. (2015). *Review of fuel loads, burn efficiencies, emissions factors and recovery functions used to estimate greenhouse gas emissions and removals associated with wildfire on temperate forested lands. Report prepared for the Department of the Environment, Australia.* Retrieved from Report for the Department of the Environment; CSIRO: Canberra, Australia:

Schauer, J. J., Kleeman, M. J., Cass, G. R., & Simoneit, B. R. T. (2001). Measurement of emissions from air pollution sources. 3. C-1-C-29 organic compounds from fireplace combustion of wood. *Environmental Science & Technology, 35*(9), 1716-1728.

Schmidl, C., Bauer, H., Dattler, A., Hitzenberger, R., Weissenboek, G., Marr, I. L., & Puxbaum, H. (2008). Chemical characterisation of particle emissions from burning leaves. *Atmospheric Environment, 42*, 9070–9079.

Seiler, W., & Crutzen, P. (1980). Estimates of gross and net fluxes of carbon between the biosphere and the atmosphere from biomass burning. *Climatic Change, 2*(3), 207-247.

Spracklen, D. V., Mickley, L. J., Logan, J. A., Hudman, R. C., Yevich, R., Flannigan, M. D., & Westerling, A. L. (2009). Impacts of climate change from 2000 to 2050 on wildfire activity and carbonaceous aerosol concentrations in the western United States. *Journal of Geophysical Research-Atmospheres, 114*, 20301-20301.

Stockwell, C. E., Jayarathne, T., Cochrane, M. A., Ryan, K. C., Putra, E. I., Saharjo, B. H., . . . Yokelson, R. J. (2016). Field measurements of trace gases and aerosols emitted by peat fires in Central Kalimantan, Indonesia, during the 2015 El Nino. *Atmospheric Chemistry and Physics, 16*(18), 11711-11732.

Stockwell, C. E., Veres, P. R., Williams, J., & Yokelson, R. J. (2015). Characterization of biomass burning emissions from cooking fires, peat, crop residue, and other fuels with high-resolution proton-transfer-reaction time-of-flight mass spectrometry. *Atmospheric Chemistry and Physics, 15*(2), 845-865.

Surawski, N. C., Sullivan, A. L., Meyer, C. P., Roxburgh, S. H., & Polglase, P. J. (2015). Greenhouse gas emissions from laboratory-scale fires in wildland fuels depend on fire spread mode and phase of combustion. *Atmospheric Chemistry and Physics, 15*(9), 5259-5273.

Surawski, N. C., Sullivan, A. L., Roxburgh, S. H., Meyer, C. P. M., & Polglase, P. J. (2016). Incorrect interpretation of carbon mass balance biases global vegetation fire emission estimates. *Nature Communications, 7*, 11536-11536.

Turetsky, M. R., Kane, E. S., Harden, J. W., Ottmar, R. D., Manies, K. L., Hoy, E., & Kasischke, E. S. (2011). Recent acceleration of biomass burning and carbon losses in Alaskan forests and peatlands. *Nature Geoscience, 4*(1), 27-31.

Urbanski, S. (2014). Wildland fire emissions, carbon, and climate: Emission factors. *Forest Ecology and Management, 317*, 51-60.

Urbanski, S. P. (2013). Combustion efficiency and emission factors for wildfire-season fires in mixed conifer forests of the northern Rocky Mountains, US. *Atmospheric Chemistry and Physics, 13*(14), 7241-7262. doi:10.5194/acp-13-7241-2013

Urbanski, S. P., Hao, W. M., & Baker, S. (2009). Chemical Composition of Wildland Fire Emissions. In A. Bytnerowicz, M. Arbaugh, A. Riebau, & C. Andersen (Eds.), *Developments in Environmental Science* (Vol. 8): Elsevier.

van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P. S., . . . van Leeuwen, T. T. (2010). Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997-2009). *Atmospheric Chemistry and Physics, 10*(23), 11707-11735.

Vodacek, A., Kremens, R. L., Fordham, A. J., Vangorden, S. C., Luisi, D., Schott, J. R., & Latham, D. J. (2002). Remote optical detection of biomass burning using a potassium emission signature. *International Journal of Remote Sensing, 23*(13), 2721-2726.

Volkova, L., Meyer, C. P., Haverd, V., & Weston, C. J. (2018). A data - model fusion methodology for mapping bushfire fuels for smoke emissions forecasting in forested landscapes of south-eastern Australia. *Journal of Envir Management*.

Volkova, L., Meyer, C. P., Murphy, S., Fairman, T., Reisen, F., & Weston, C. (2014). Fuel reduction burning mitigates wildfire effects on forest carbon and greenhouse gas emission. *International Journal of Wildland Fire, 23*(6), 771-780.

Volkova, L., & Weston, C. (2013). Redistribution and emission of forest carbon by planned burning in Eucalyptus obliqua (L. Herit.) forest of south-eastern Australia. *Forest Ecology and Management, 304*, 383-390.

Volkova, L., & Weston, C. J. (2015). Carbon loss from planned fires in southeastern Australian dry Eucalyptus forests. *Forest Ecology and Management, 336*, 91-98.

Wain, A., Mills, G., McCaw, L., & Brown, T. (2008). Managing Smoke from Wildfires and Prescribed Burning in Southern Australia. In A. Bytnerowicz, M. J. Arbaugh, A. R. Riebau, & C. Andersen (Eds.), *Developments in Environmental Science: Wildland Fires and Air Pollution* (Vol. 8, pp. 535-550): Elsevier Ltd.

Ward, D. E., & Hardy, C. C. (1991). Smoke emissions from wildland fires. *Environment International, 17*, 117-134.

Wiedinmyer, C., Akagi, S. K., Yokelson, R. J., Emmons, L. K., Al-Saadi, J. A., Orlando, J. J., & Soja, A. J. (2011). The Fire INventory from NCAR (FINN): a high resolution global model to estimate the emissions from open burning. *Geoscientific Model Development, 4*(3), 625-641.

Yokelson, R. J., Burling, I. R., Urbanski, S. P., Atlas, E. L., Adachi, K., Buseck, P. R., . . . Wold, C. E. (2011). Trace gas and particle emissions from open biomass burning in Mexico. *Atmospheric Chemistry and Physics, 11*(14), 6787-6808. doi:10.5194/acp-11-6787-2011

Yokelson, R. J., Christian, T. J., Karl, T. G., & Guenther, A. (2008). The tropical forest and fire emissions experiment: laboratory fire measurements and synthesis of campaign data. *Atmospheric Chemistry and Physics, 8*(13), 3509-3527.

Yokelson, R. J., Susott, R., Ward, D. E., Reardon, J., & Griffith, D. W. T. (1997). Emissions from smoldering combustion of biomass measured by open-path Fourier transform infrared spectroscopy. *Journal of Geophysical Research-Atmospheres, 102*(D15), 18,865-818,877.

Table 1 Weather and fuel data measured at burn sites

Burn site	Date of burn	Ambient temperature (°C)	Ambient RH (%)	Fuel moisture (%)	Fine fuel load (<6mm) (t ha^{-1})	CWD fuel load (>25mm) (t ha^{-1})
Greendale	13 Apr 2015	24	47	9-16	15.3±1.1	32.5
Castlemaine	23 Apr 2015	24	51	8-12	11.8±1.0	1.9
Bambra-Deans Marsh	30 Sep 2015	19	51	NM	NM	NM
Campbells Creek	1 Oct 2015	30	25	NM	NM	NM

NM – Not measured

Table 2 Emission factors (g kg^{-1} fuel) and MCE from ground measurements of flaming and smouldering combustion at the four prescribed burns (average ± stdev)

Samples collected at burn sites	MCE	EF (CO)	EF (CO_2)	EF (CH_4)	EF ($PM_{2.5}$)
Greendale (flaming, N=15)	0.94 ± 0.01	71.5 ± 10.6	1715 ± 18	2.3 ± 0.6	15.8 ± 5.2
Greendale (smouldering, N=8)	0.84 ± 0.04	188.0 ± 45.1	1512 ± 76	9.2 ± 3.4	50.2 ± 32.7
Castlemaine (flaming, N=19)	0.95 ± 0.01	54.0 ± 10.7	1743 ± 18	1.9 ± 0.4	14.4 ± 7.2
Castlemaine (smouldering, N=6)	0.81 ± 0.02	211.7 ± 23.4	1456 ± 32	16.1 ± 3.0	66.4 ± 33.6
Bambra (flaming, N=10)	0.93 ± 0.01	85.4 ± 14.3	1692 ± 24	2.7 ± 0.7	24.6 ± 7.3
Bambra (smouldering, N=10)	0.83 ± 0.04	196.1 ± 48.5	1488 ± 89	13.5 ± 6.1	51.7 ± 36.7
Campbells Creek (flaming, N=19)	0.95 ± 0.01	58.4 ± 14.8	1735 ± 25	2.2 ± 0.7	17.8 ± 6.3
Campbells Creek (smouldering, N=7)	0.86 ± 0.02	166.1 ± 24.3	1540 ± 45	11.7 ± 3.5	32.2 ± 16.9
Average all (flaming, N=63)	**0.94 ± 0.01**	**64.5 ± 16.7**	**1726 ± 28**	**2.2 ± 0.6**	**17.4 ± 7.2**
Average all (smouldering, N=31)	**0.83 ± 0.04**	**190.3 ± 40.2**	**1499 ± 72**	**12.5 ± 4.9**	**49.8 ± 32.1**
Average (smouldering pyrolysis, N=8)	**0.83 ± 0.03**	**194.3 ± 31.8**	**1491 ± 57**	**13.6 ± 4.8**	**87.4 ± 34.6**
Average (glowing, N=23)	**0.84 ± 0.04**	**188.0 ± 43.0**	**1505 ± 77**	**12.0 ± 5.0**	**34.4 ± 12.6**

Table 3 Emission factors (g kg^{-1} fuel) of levoglucosan and water-soluble ions from ground measurements of flaming and smouldering combustion at the four prescribed burns (average ± stdev) based on TSP filter samples

Compound	Flaming combustion	# samples	Smouldering combustion	# samples
Levoglucosan	1.54 ± 0.97	14	7.58 ± 3.11	8
Non sea-salt K (nssK)	0.092 ± 0.056	14	0.045 ± 0.054	6
K^+	0.093 ± 0.056	14	0.061 ± 0.056	6
Na^+	0.014 ± 0.010	12	0.039 ± 0.043	5
Cl^-	0.169 ±0.088	14	0.219 ± 0.167	7
Ca^{2+}	0.017 ± 0.013	12	0.024 ± 0.014	5
Mg^{2+}	0.003 ± 0.003	14	0.010 ± 0.012	6
NH_4^+	0.057 ± 0.018	14	0.113 ± 0.058	8
NO_3^-	0.016 ± 0.004	14	0.012 ± 0.007	7
SO_4^{2-}	0.082 ± 0.028	14	0.066 ± 0.029	7
$C_2O_4^{2-}$	0.010 ± 0.005	5	0.048 ± 0.028	5
Acetate	0.017 ± 0.006	9	0.104 ± 0.068	6
Formate	0.026 ± 0.012	11	0.094 ± 0.029	5
Cation/Anion	0.84 ± 0.08	14	0.65 ± 0.10	8

Table 4 Statistics for the linear regression relationships between $PM_{2.5}$ EFs and MCE

Study	Slope (g kg^{-1})	y-intercept (g kg^{-1})	R^2	MCE range
This study (n=94) (flaming + all smouldering)	-313.4 ± 30.5	312.7 ± 27.7	0.53	0.76-0.97
Burling et al. (2011) conifer forest	-231 ± 83	230 ±78	0.61	0.88-0.96
Burling et al. (2011) chaparral & oak savanna	-68 ± 29	71 ± 27	0.58	0.90-0.96
Hosseini et al. (2013)	-248.8	240.9	0.80	0.87-0.99
Janhall et al. (2010) forest	-89.8	93.2	0.6	0.80-0.98
McMeeking et al. (2009)	-311.1	310.4	0.39	0.80-0.99
Urbanski (2013) (n=50)	-203.6 ± 24.5	202.9 ± 22.5	0.59	0.80-0.95
Urbanski (2014) (n=54)	-212.3 ± 21.9	210.8 ± 20.2	0.64	
Ward and Hardy (1991)	-66.8	67.4	0.74	CE instead of MCE*

* Combustion efficiency (CE) was used instead of modified combustion efficiency (MCE)

Table 5 Key parameters for emissions estimates from prescribed burns in eucalypt forests of Victoria extracted from Roxburgh et al. (2015)

Parameter	Fine	CWD ave	CWD char	CWD pyr
Fuel load (t ha^{-1})	12.2	32.5	32.5	32.5
Burning efficiency	0.36	0.13	0.13	0.13
EF $PM_{2.5}$ (g kg^{-1})	16.9	38.8	33.9	84.2
Emissions $PM_{2.5}$ (kg ha^{-1})	73.7	163.9	185.7 (80% char, 20% pyr) 249.5 (50% char, 50% pyr)	

CWD_{ave} - the median EF $PM_{2.5}$ for smouldering (averaged for both combustion processes) was used
CWD_{char} - the median EF $PM_{2.5}$ for char combustion was used
CWD_{pyr} – the median EF $PM_{2.5}$ for smouldering pyrolytic combustion was used

Figure 1 Location of prescribed burn sites (blue dots represent autumn burns and red dots represent spring burns).

Figure 2 $PM_{2.5}$ emission factors (g kg^{-1} fuel) as a function of MCE.

Figure 3 Sampling during smouldering combustion, (left) pyrolysis dominated combustion; (right) glowing dominated combustion

Figure 4 Distribution of $PM_{2.5}$ emission factors by combustion phase, (a) separation into flaming and smouldering combustion phases and (b) separation into flaming, smouldering combustion dominated by pyrolysis or char combustion. Boxplots show median, 25-75 percentiles (box) and 10-90 percentiles (whiskers)

Figure 5 Chemical composition of $PM_{2.5}$ for flaming and smouldering combustion. For clarity, levoglucosan concentrations are multiplied by 0.1.

Figure 6 Comparison of $PM_{2.5}$ emission factors from this study to previous literature data

Figure 7 Relationship between $PM_{2.5}$ emission factors (g kg^{-1} fuel) and MCE. Fits between MCE and EF($PM_{2.5}$) are represented by (1) solid grey line for flaming and smouldering dominated by glowing char combustion (N=86); (2) solid black line for flaming and smouldering dominated by pyrolysis (N=71); (3) dotted line for all data (N=94).