Fine particle emissions from tropical peat fires decrease rapidly with time since ignition

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Key Points:

- In this study we show that emissions of $PM_{2.5}$ from Malaysian peat fires are likely three-times larger than previously assumed.
- We show that the emissions of fine particulate matter from peat fires in the field decrease rapidly with the age of the fire
- We show that the likely cause is the accumulation of an ash layer as the peat burns below the surface.

Abstract

Southeast Asia experiences frequent fires in fuel-rich tropical peatlands, leading to extreme episodes of regional haze with high concentrations of fine particulate matter ($PM_{2.5}$) impacting human health. In a study published recently, the first field measurements of $PM_{2.5}$ emission factors for tropical peat fires showed larger emissions than from other fuel types. Here we report even higher $PM_{2.5}$ emissions factors, measured at newly ignited peat fires in Malaysia, suggesting current estimates of fine particulate emissions from peat fires, may be underestimated by a factor of three or more. In addition, we use both field and laboratory measurements of burning peat to provide the first mechanistic explanation for the high variability in $PM_{2.5}$ emission factors, demonstrating that build-up of a surface ash layer causes the emissions of $PM_{2.5}$ to decrease as the peat fire progresses. This finding implies that peat fires are more hazardous (in terms of aerosol emissions) when first ignited than when still burning many days later. Varying emission factors for $PM_{2.5}$ also has implications for our ability to correctly model the climate and air quality impacts downwind of the peat fires. For modelers able to implement a

time varying emission factor, we recommend an emission factor for $PM_{2.5}$ from newly ignited tropical peat fires of 58 grams of $PM_{2.5}$ per kilogram of dry fuel consumed ($g.kg^{-1}$), reducing exponentially at a rate of 9% per day. If the age of the fire is unknown or only a single value may be used, we recommend an average value of 28 $g.kg^{-1}$.

1 Introduction

Tropical peatland fires in Southeast Asia release huge amounts of particulate and gaseous carbon to the atmosphere [*Page et al.*, 2002; *Turetsky et al.*, 2015], accounting on average for approximately 10-15% of the net estimated global total greenhouse gas emissions from deforestation and forest degradation, albeit with significant interannual variability [*Ballhorn et al.*, 2009; *van der Werf et al.*, 2009]. Fire emission inventories usually estimate emissions using the algorithm of *Seiler and Crutzen* [1980], which multiplies together the total area burned, the fuel loads, combustion efficiencies and the emission factors (the mass of each species emitted per unit of dry vegetation burned). Within the fire emissions inventory GFED4s (Global Fire Emissions Database 4s), from 1997 to 2016, on average peat fires in Indonesia account for 3% of total fire $PM_{2.5}$ emissions globally. This contribution increases during El Niño years when fires are elevated. The most extreme year on record was 1997 when this region accounted for 17% of total fire $PM_{2.5}$. During the more recent 2015 El Niño episode, the contribution was 8%, while the contribution is very small (less than a percent) during wet years [*van der Werf et al.*, 2017]. These seasonal peatland fires destroy unique ecosystems and release aerosols with significant impacts on air quality, agricultural productivity, human health and regional economies [*Gaveau et al.*, 2014]. Unlike forest fires on mineral soils, (which usually burn with great intensity, lofting emissions high into the atmosphere), peat fires typically smoulder for a long time producing enormous quantities of fine particulates, which become trapped in the planetary boundary layer [*G. Rein*, 2013]. For example, in 2015 smoke from peat fires in Indonesia blanketed much of Asia in a persistent plume of pollution throughout September and October [*Field et al.*, 2016; *Huijnen et al.*, 2016], contributing to an estimated 100,000 premature deaths due to exposure to fire-related air pollution [*Koplitz et al.*, 2016]. Due to long-range transport of these emissions, tropical peatland fires frequently affect large areas of the world [*Andreae*, 1983; *Edwards et al.*, 2006; *Viatte et al.*, 2015].

Emission factors are defined as the grams of a species emitted to the atmosphere per kilogram of dry fuel consumed and are used to calculate total emissions from fires [*Andreae and Merlet*, 2001]. Previous studies of fine particulate emission factors (EFs) from temperate peat/organic soils in North Carolina, USA found significant variability in EFs between sites and studies with *Geron and Hays* [2013] reporting $PM_{2.5}$ EFs ranging from 9 $g.kg^{-1}$ to 79 $g.kg^{-1}$. For tropical peat fires *Iinuma et al.* [2007] reported a PM_{10} EF of 33 $g.kg^{-1}$ and *May et al.* [2014] reported a PM_1 EF of 34.9 $g.kg^{-1}$ from laboratory burns of Indonesian peat. Emissions from temperate and tropical peat are likely to be different, since there are differences in composition (e.g. tropical peat has significantly higher carbon content) [*Hu et al.*, 2018]. Temperate and boreal peats are derived largely from sedges, shrubs and Sphagnum and other mosses, whereas the tropical peats of SE Asia are derived largely from the leaves, wood, and roots of trees because the peatlands are forested with diverse trees up to 70 m tall [*Yule*, 2010]. Consequently, tropical peats tend to be largely composed of lignin and the products of lignin degradation (e.g. tannins, humic acids and other phenolic compounds), whereas temperate and boreal peats have

much larger proportions of cellulose and hemicellulose, and less lignin and its derivatives [*Andriesse*, 1988].

Until recently there were no reported emission factors for $PM_{2.5}$ from tropical peat fires in the field, despite their extremely detrimental impact on regional air quality. Previous estimates of excess mortality in the region relied on predicted emissions extrapolated from measurements from other fuel types or regions [*Giglio et al.*, 2013; *Lelieveld et al.*, 2015; *van der Werf et al.*, 2010]: with GFED4 using an emission factor of 9.1 $g.kg^{-1}$ for $PM_{2.5}$ [*Van der Werf*, 2013]. The first EFs for $PM_{2.5}$ from tropical peat fires were reported by *Stockwell et al.* [2016] (whilst this study was in progress) and included EFs from five different smoke plumes at two different peat fires in Indonesia. Their observed $PM_{2.5}$ emission factors ranged from 15.7 $g.kg^{-1}$ to 29.6 $g.kg^{-1}$ [*Stockwell et al.*, 2016]. These EFs are significantly larger and more variable than emissions of $PM_{2.5}$ from other non-soil fuel types (with boreal forests showing the next largest and most variable emissions of $PM_{2.5}$ with values of 15 ± 7 $g.kg^{-1}$ dry fuel consumed) [*Akagi et al.*, 2011]. Neither studies of temperate peat EFs [*Geron and Hays*, 2013] nor that of *Stockwell et al.* [2016] explore the reasons behind the variability in $PM_{2.5}$ emissions. *Black et al.* [2016] measured $PM_{2.5}$ emissions from laboratory burns of peat cores from North Carolina, USA over 5 to 7 hours in duration, noting that emissions in the first 3 hours of the burn were 3 to 10 times larger than for the final few hours. This suggests that as the fire progresses, there are changes in the burning conditions that influence the emission of fine particulates. Given the evidence for premature mortality occurring as a result of $PM_{2.5}$ pollution [*Lelieveld et al.*, 2015], there is a need for an improved understanding of the magnitude and causes of variability of $PM_{2.5}$ emissions from tropical peat fires. In this paper we present $PM_{2.5}$ EFs from *in-situ* measurements of Malaysian peat fires that are considerably higher than the previous assumed value of 9.1 $g.kg^{-1}$, (as used in global fire emissions databases based on measurement in tropical forests because peat-specific measurements were lacking [*Giglio et al.*, 2013]). This finding means that recent estimates of deaths attributable to $PM_{2.5}$ for biomass burning in the region are likely to be underestimated. We also observed that the emission of fine particles decreased rapidly with the age of the peat fire (i.e. the time since ignition). We hypothesize that this phenomenon occurs because of an accumulation of peat ash over the surface of the burning peat, which impedes the fire's access to oxygen and acts as an aerodynamic filter, reducing particulate emissions to the atmosphere. Finally, we present strong evidence from a series of laboratory-based peat burn experiments to support this theory. This finding implies that newly ignited fires are particularly hazardous for human health due to their large emission of $PM_{2.5}$.

2 Methodology and field sites

In this study we present emission factors for $PM_{2.5}$ derived from measurements made *in situ* at peat fires burning in North Selangor, Malaysia. The experimental methodology involves coincident and collocated measurements of $PM_{2.5}$ and carbon monoxide (CO) in fresh smoke within a few meters of the burning peat in order to establish emission ratios (of $PM_{2.5}$ to CO). Emission factors of $PM_{2.5}$ can then be calculated by combining these emission ratios with emission factors of CO from the fires (see e.g. [*Paton-Walsh et al.*, 2014; *Smith et al.*, 2018; *Smith et al.*, 2014; *Stockwell et al.*, 2016]).. An aerosol monitor measured $PM_{2.5}$ concentrations (see Section 2.1), whilst mole fractions of CO were measured with a Thermo Scientific Model 48i CO analyser (see Section 2.2). The instruments' inlets were deployed in close proximity to one another and immediately downwind of peat fires burning in the vicinity of 3.68 ^{o}N, 101.05 ^{o}E. Fire plumes were sampled on six different days over one month, at four different locations,

with measurements made within 10 m of the burning peat. Most fires were the result of "slash and burn" practices, where palm oil fronds had been set alight above the peat, with the fire spreading into the peat and persisting for weeks after the surface slash fire had ceased. One fire had been ignited at numerous places on top of piles of peat, in preparation for planting a crop of okra. Photographs are provided in Figure 1, and further details of the fires are given in a supplementary information section.

2.1 Measurements of $PM_{2.5}$ and CO concentrations

We used a TSI DustTrak DRX 8533 aerosol monitor and a Thermo Scientific Model 48i CO analyser to estimate emission ratios of $PM_{2.5}$ to CO. An Ecotech Aurora 1000 integrating nephelometer was successfully deployed and sufficiently collocated on the 14th and 27th July 2016 to act as a separate measure of aerosol loading, and yielded agreement within 7%. The nephelometer and DustTrak instruments both measure light scattering at different wavelengths, from which total $PM_{2.5}$ concentrations may be inferred, by reference to an independent gravimetric measurement (the estimate of $PM_{2.5}$ concentrations from the measured light scattering assumes a size distribution of particles that matches that used to calibrate the instruments by reference to gravimetric samples). Prior to deployment at the Malaysia peat fires the DustTrak and nephelometer were calibrated against gravimetric standards in a smoke chamber experiment in Australia using wood-smoke and coal. The results agreed within the precision of determining the calibration factor (~ 5%) for wood-smoke and within 20% for coal (with the DustTrak reading lower than the gravimetric value). In addition, the DustTrak showed excellent agreement with continuous coincident $PM_{2.5}$ measurements made with a Met-One model BAM-1020 beta-attenuation monitor with the DustTrak reading 3% lower than the BAM-1020 for wood-smoke (with an R^2 of 0.94), and <2% lower for coal (R^2 of 0.97). We assume that the size distribution for peat smoke (and hence the DustTrak response) will lie between that of the wood-smoke and the coal (as most peat smoke particles are in the $PM_{2.5}$ size range [*Geron and Hays*, 2013; *Hu et al.*, 2018]), but we were unable to calibrate the DustTrak directly against gravimetric standards in peat smoke. We have estimated the uncertainty in the DustTrak measurements of $PM_{2.5}$ in peat smoke to be ±20%, which dominates the uncertainty in the emission ratio of $PM_{2.5}$ to CO. We have estimated a larger uncertainty in the final peat fire emission ratio (at site 4) due to possible interference from surface vegetation combustion.

The CO analyser was calibrated using a 5,092 ppm standard of CO, diluted using an Environics 6100 diluter to concentrations of 39.6 ppm, 29.7 ppm, 24.7 ppm, 19.8 ppm, 9.9 ppm, 4.9 ppm and 0.0ppm, with concentrations agreeing within 2%. Mole fractions of CO were converted to equivalent concentrations, assuming standard atmospheric pressure and an ambient temperature of 305 K, such that 1 ppm of CO is equivalent to 1.11 mg m^{-3} of CO.

Figure 1: (A) Site 1, 12 July 2016; (B) Site 2 (okra), 13 July 2016; (C) Site 3, 14 July 2016; (D) Site 4, 20 July 2016; (E) Site 4, 27 July 2016; (F) Site 4, 3 August 2016

2.2 Determining emission ratios of $PM_{2.5}$ to CO

The 5-second averages of $PM_{2.5}$ concentrations were shifted by approximately 90-seconds and averaged over one minute in order to yield the best correlation to one-minute averages of CO concentrations, by accounting for differences in integration/sampling time of the different measurements. Figure 2 shows scatter plots of $PM_{2.5}$ and CO concentrations at each of the fires.

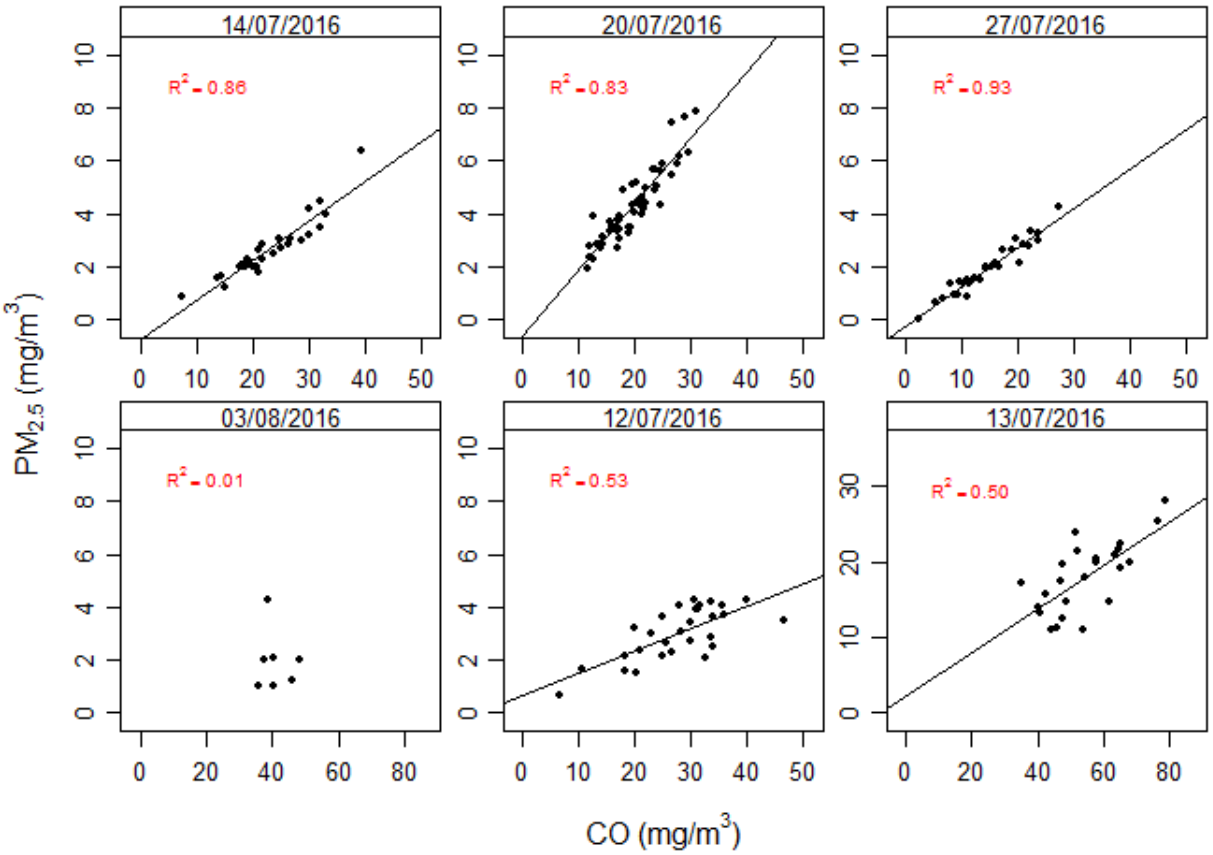

Figure 2: Correlation plots of $PM_{2.5}$ plotted against CO observed at the six peat fires sampled.

2.3 Measurements of Modified Combustion Efficiency (MCE)

Modified combustion efficiency (MCE) was measured using a handheld CO/CO_2 monitor (KANE 100-1). This uses a non-dispersive infrared sensor for measuring CO_2 and an electrochemical sensor for carbon monoxide. MCE is the ratio of excess CO_2 over background divided by the sum of excess CO and excess CO_2 over background [*Hao and Ward*, 1993] and is used to characterise the efficiency of burning within a fire. The instrument was calibrated prior to deployment and showed agreement within 1% of coincident measurements of MCE made at two fires with the FTIR described below.

2.4 Measurements of the Emission Factor for CO

The emission factor for CO (grams of CO emitted per kilogram of dry fuel burned) was calculated from *in situ* measurements of trace gas mole fractions using open-path Fourier transform infrared spectroscopy. A full description of the use of this method for determining emission factors from biomass burning can be found in *Smith et al.* [2014]. Here, we deployed a MIDAC M2000 series FTIR spectrometer to measure the spectra of an infrared lamp located 18–28 m from the spectrometer on two occasions (20 July 2016 and 27 July 2016) at Site 4. The aerosol sampling equipment was located approximately in the middle of the path. Carbon content of the peat is required for the calculation of emission factors. This was found to be 55.5%, as

determined from our peat carbon content measurements of four samples collected from Site 4 (see Section 2.5). Uncertainties in emission factors for CO (determined by calculating the combined uncertainties from: the spectral database; the impact of uncertainties in the temperature on the spectral line strengths; spectral fitting uncertainties and uncertainties in the gradient of best fit) are estimated at 16% [*Paton-Walsh et al.*, 2014]. When combined in quadrature with the 20% estimated uncertainties in the emission ratios of $PM_{2.5}$ to CO, this yields an uncertainty in emission factors of $PM_{2.5}$ of 25%.

2.5 Measurements of peat bulk density, moisture content, and carbon content

Fuel moisture content and bulk density measurements were determined after *in situ* sampling. Peat samples were taken from all four field sites from 2016 using a surface core sampler with a volume of 785 cm^3. Two samples were taken at Sites 2, 3 and 4, while only one sample was taken at Site 1. All samples were then separated into pre-weighed aluminium trays and their initial wet weight was measured on a Sartorius TR212 balance (S/N: 24003700 calibration date 17/08/2016). After which all samples were inserted into a Memmert UFB400 (S/N: EN60529) drying oven at 60°C for seven days. After drying was complete samples were removed from the oven and allowed to cool to room temperature over two hours. Samples were then weighed to determine their final dry weight for the calculation of moisture content. The bulk density of samples from sites 1, 3, 4 and the samples used for experimental burning (Section 5) were within 6% of each other, whilst Site 4 had significantly lower bulk density, having been ploughed in preparation for planting. Fuel moisture for all sites in July/August 2016 (Sites 1 and 4 determined as 53%, Site 3 as 53% and Site 2 at 62%), was lower than the samples collected experimental burns (in January 2017), which had a moisture content of 77%. For carbon content analysis, subsamples of peat and ash were oven dried at 105°C for 7 hours and then hand-milled for homogenization. Carbon content of the sub-samples was analysed using 20 mg of material enclosed in a tin capsule and measurements undertaken using a total element analyser (Thermo Flash EA 1112, CE Instruments).

3 Emission factors of $PM_{2.5}$

Average background amounts of 0.1 ppm of CO and 29 µg m^{-3} of $PM_{2.5}$ (measured upwind of the fires just before or after sampling the smoke plumes) were subtracted from the measurements within the smoke plumes and the emission ratio of $PM_{2.5}$ to CO at each of the fires (Table 1) was determined by calculating the total excess $PM_{2.5}$ divided by the total excess CO (described in *Paton-Walsh et al.* [2014]). Gaseous emission factors for two of our fires (20 July 2016 and 27 July 2016) are reported by *Smith et al.*, [2018]. They calculate emission factors of 200 and 201 grams of CO per kilogram of dry fuel consumed (EF CO, in $g.kg^{-1}$), assuming the fraction of carbon emitted as particulate matter (F_{PMC}) to be 0.0127 as reported by previous studies of PM EFs for tropical peatlands [*Jayarathne et al.*, 2017]. Our subsequent analysis of $PM_{2.5}$ emissions from these fires finds an F_{PMC} of 0.043, and so we recalculate the EF CO for these fires to be 194 and 195 $g.kg^{-1}$ (using Eqn. 3 in *Smith et al.*, [2018].). MCE showed little variability across the fires sampled and so the mean emission factor for CO of 194.5 $g.kg^{-1}$, was used to convert the emission ratios to emission factors of $PM_{2.5}$ in grams per kilogram of dry fuel burned (Table 1).

Very large variability in measured emission ratios of $PM_{2.5}$ to CO was found from the peat fires sampled, across six different days and at four different sites. There was more than a factor of six

between the largest and the smallest emission ratio of $PM_{2.5}$ to CO, despite relatively small variations in the bulk density and fuel moisture content at the sites. The fuel moisture content was determined from predominantly unburnt peat samples at the sites, but in reality will decrease as the fire progresses, drying the peat in its path. However, fuel moisture differences are unlikely to explain the variability in emissions of $PM_{2.5}$, because MCE was relatively constant at the fires sampled (at ~0.83 ± 0.02).

Emission factors for $PM_{2.5}$ measured in this study are surprisingly large, with only the lowest measured emission factor in agreement with the assumed value of 9.1 $g.kg^{-1}$ in the most widely used global biomass burning inventory (GFED4 [*van der Werf et al.*, 2017]). Note that GFED used $PM_{2.5}$ emission factors from tropical forest burning for peat fires, because no actual measurements for tropical peat were available previously [*Giglio et al.*, 2013]. The mean value measured of 28 $g.kg^{-1}$ is approximately three times the previous inventory value and similar to the largest value reported recently by *Stockwell et al.* [2016]. The largest emission factor was more than twice this value at 58 $g.kg^{-1}$ and was observed at an okra field when small, freshly ignited man-made piles of pure peat were burning on the surface of the okra field. As explained above, the variability could not readily be explained by differences in MCE or fuel moisture and since the humidity was consistently above 65% and temperatures were in the low 30s centigrade, meteorological differences are insufficient to explain the spread of emission factors measured.

Table 1: Dates, age of burn, modified combustion efficiency (MCE), fuel moisture content, bulk density, minutes of data recorded, mean and standard deviation of the concentration of CO and $PM_{2.5}$ measured, emission ratios (with 1σ uncertainty) and emission factors (with 1σ uncertainty) for $PM_{2.5}$ at each of the fires sampled.

Sampling Date and Location	Age of burn (days)	MCE	Fuel Moisture Content	Bulk Density	Minutes of Data	Mean conc CO mg/m3	Mean conc PM2.5 mg/m3	Emission Ratio $PM_{2.5}$/CO	Emission Factor $PM_{2.5}$ $g.kg^{-1}$
12/07/2016 Site 1	>10	0.84	54%	0.583 g cm^{-3}	60	31 ± 9	3.0 ± 1.0	0.10 ± 0.02	19 ± 5
13/07/2016 Site 2	0	0.81	62%	0.438 g cm^{-3}	48	61 ± 13	18 ± 5	0.30 ± 0.06	58 ± 15
14/07/2016 Site 3	12	0.85	53%	0.605 g cm^{-3}	60	25 ± 8	2.6 ± 1.2	0.10 ± 0.02	20 ± 5
20/07/2016 Site 4	6	0.84	54%	0.625 g cm^{-3}	100	22 ± 6	4.3 ± 1.4	0.20 ± 0.04	38 ± 10
27/07/2016 Site 4	13	0.85	54%	0.625 g cm^{-3}	60	16 ± 7	1.9 ± 0.9	0.12 ± 0.02	23 ± 6
03/08/2016 Site 4	20	0.8	54%	0.625 g cm^{-3}	14	46 ± 5	2.0 ± 1.1	0.04 ± 0.03	8 ± 6

4 Emission factors for $PM_{2.5}$ change with age of burn

The ignition dates for all but the first burn are known from regular reconnaissance in the area. The date of ignition of the first burn is unknown (due to cloud cover and the limited spatial extent of the fire, no satellite hotspots were detected), but was greater than ten days, having been observed burning 10 days prior during initial scouting of the area. We noticed that the emission ratio of $PM_{2.5}$ to CO showed a strong anti-correlation with the age of the peat fire (see Figure 3), with the emission ratio decreasing by approximately 9% per day. Both linear and exponential

regressions yield the same correlation coefficient (R^2 =0.97), however an exponential decay is more physically reasonable, since we do not expect the emissions to drop to zero after 3 weeks.

From this observation, and observations of ash layers (see Figure 1A, 1B and 1E), we theorized that the peat ash accumulating on the surface of the peat as it burned downwards , could be the cause of the drop in emissions. The accumulation of ash insulates the fire and reduces the availability of oxygen. In addition we hypothesized that the ash might act as an aerodynamic filter, thereby reducing the emissions of $PM_{2.5}$ from the surface of the burn. Given that we had only sampled five fires with known ignition dates (and that the rains had set in, preventing further field measurements), we could not be sure that variables other than time since ignition (e.g. moisture, rainfall, wind etc) were not driving the variability. For this reason we decided to test our theory via a series of experimental burns, using peat collected from one of the field sites (Site 3 on 1 January 2017). If our theory was correct, we would expect to see rapid decreasing $PM_{2.5}$ emission ratios from laboratory burns and increasing carbon content in the overlying ash.

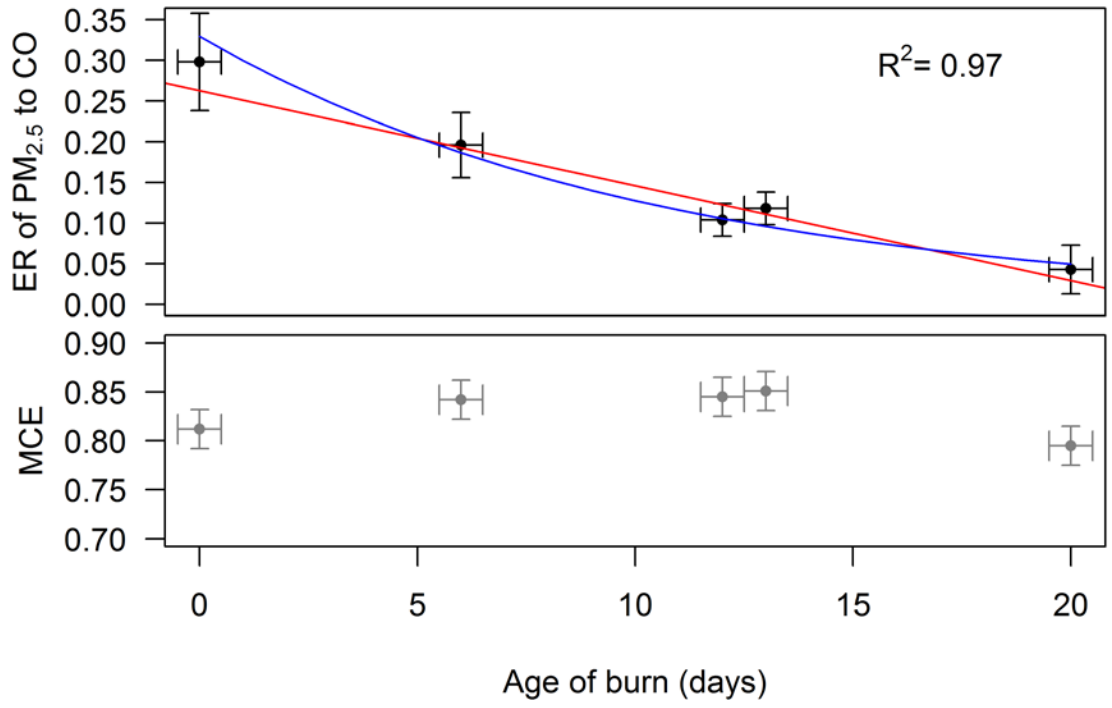

Figure 3: $PM_{2.5}$ to CO emission ratios (upper panel) and MCE (lower panel) as a function of age of burn in days. A linear fit to the data is given in red and an exponential fit in blue: both fits yield an R^2 value of 0.97. The error bars indicate the estimated uncertainties in the age of the burn (± 12 hours) ; in MCE (± 3%) and the 1σ uncertainties in the emission ratio (see Table 1).

5 Supporting evidence from controlled peat burn experiments

An insulating chamber was filled with approximately 4000 – 6000 $cm^3$of peat and ignited using a nichrome wire-bound ceramic ignition coil with 110 Watts of energy applied for 30 minutes (as described in *Wilson et al.* [2015] and [*Guillermo Rein et al.*, 2008]). Once burning independently, the chamber was placed under a custom-made fume hood, where the sample

heads for the DustTrak and CO analyser were situated. The peat was left to burn for 24 hours after which the accumulated ash was sampled and then removed using a spatula, leaving the actively burning peat as the surface layer once again. We undertook two experimental burns in this manner, yielding just less than four days of data. (Photographs of the experimental set-up are provided in an additional figure as supplementary information to this manuscript).

We found that the emission ratio of $PM_{2.5}$ to CO was initially high, with significant visible smoke emanating from the chamber. The $PM_{2.5}$ emissions decreased significantly with time after ignition, whilst the CO emissions remained relatively elevated, such that the emission ratio dropped steadily over time (at rates that varied between approximately 5% and 20% per hour). The time-series of emission ratios of $PM_{2.5}$ to CO during the first two experimental burns are shown in the top two panels of Figure 4. We observed a ~60-90% decline in the emission ratio of $PM_{2.5}$ to CO over the first 8 hours of our burns, which is similar to the 64% and 91% decline in measured $PM_{2.5}$ emission factors across 7 hours reported for peat fuels in *Black et al.* [2016]. Once the ash layer was removed, there was an obvious increase in visible smoke again and the emission ratio of $PM_{2.5}$ to CO increased to values similar to those recorded just after ignition, before decreasing again as fire progressed. This additional evidence from the small-scale laboratory burns, confirms the findings from the field campaign, that the emissions of $PM_{2.5}$ decrease with time since ignition as the peat fire progresses downwards and that the decrease is caused by the accumulation of the ash layer on the surface of the burning peat.

During the removal of the ash layer (*c.*3-4 cm in depth), samples were taken from the ash surface and ash base (at a minimum of 7 different sampling points) for later analysis of carbon content. In both experimental burns the ash from the surface layer had higher carbon content than that from the ash base (Burn 1: 5.3% for surface ash vs 2.3% for base ash and Burn 2: 40% for surface ash vs 15% for base ash). This supports our theory of the ash filtration effect, because the ash at the base is newly formed and has had less time than the surface ash to capture carbon-rich fine particulate matter emanating from the burning peat. Nevertheless, the carbon-content measurements are very variable and other mechanisms by which the ash layer reduces the emissions of $PM_{2.5}$ cannot be ruled out, (for instance by changing the burning conditions by reducing the supply of oxygen).

As a further test of the ash filtration theory, we ignited a third experimental burn and measured the emitted CO and $PM_{2.5}$ for approximately one hour. A layer of pre-incinerated ash was then applied to cover the surface of the burning peat and measurements continued. We noticed an immediate and substantial drop in the emitted $PM_{2.5}$, and the $PM_{2.5}$ to CO emission ratio. In the next few hours we observed visible smoke leaking out around the inside of the chamber walls and an accompanying increase in the $PM_{2.5}$ to CO emission ratio, until after 5 hours the emission ratio began to drop again as observed in the previous two experimental burns (see bottom panel of Figure 4). We took samples of the pre-incinerated ash (before addition to the surface of the experimental peat burn) and further samples of this added surface ash at regular intervals as the burn progressed, for subsequent analysis for carbon content. We found that the pre-incinerated ash (prior to its addition to the experimental burn surface) had a lower carbon content (0.6%) than all of the subsequent post burn re-retrieved surface ash samples, confirming that carbon rich smoke particles have been trapped in the pre-incinerated ash. The first sample, taken one hour after the ash addition, showed discolouration (black amongst the

original yellow) and had a carbon content of 7.4%. Subsequent samples showed very significant variability (as opposed to steadily increasing % carbon), with carbon content values varying from 1.1% to 4.7% in the centre of the chamber, to 38% at the edge of the chamber. This suggests that the added ash layer provided an inhomogeneous surface layer, with the smoke from the peat permeating through particular regions of the added ash. Despite the consistently higher carbon content of the ash that had been exposed to burning peat below it, the carbon content data showed sufficient variability that we cannot conclude with total certainty that the mechanism by which the particulate emissions decrease is through the $PM_{2.5}$ being captured by the accumulating ash layer.

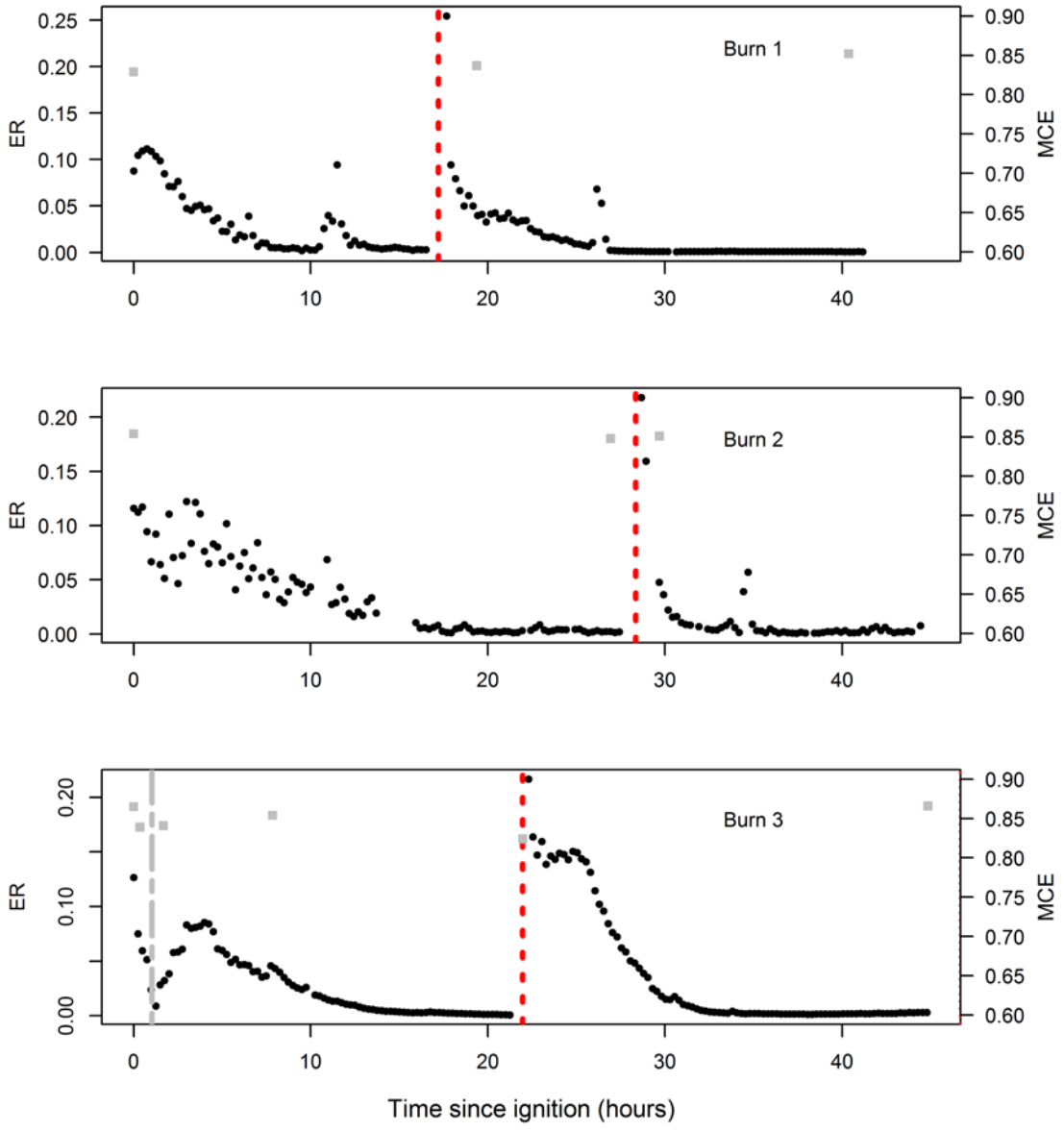

Figure 4. Time-series of 15-minute averages of emission ratios $PM_{2.5}$ to CO (black dots and left-hand axis) and MCE (grey dots and right-hand axis). The emission ratio is high after ignition, dropping steadily as the fire burns downwards. Note that Burn 2 was ignited in 3 sections (and the other burns in a single section) and shows considerably more variability in the emission ratio. During all 3 burns, the emission ratio increases immediately

when the ash layer is removed (red dotted vertical line) and then decreases steadily. The time-series also show occasional short-lived increases in the emission ratio above a very low baseline level, which we interpret as a collapse in the ash pile, causing a disturbance to the surface. MCE does not vary significantly as the fire progresses in any of the experimental burns. In Burn 3 the addition of the artificial ash layer is shown by the grey dotted vertical line.

6 Discussion & Conclusion

This study has identified unexpectedly large emission factors of $PM_{2.5}$ from newly-ignited fires on disturbed tropical peatlands. These new measurements imply that $PM_{2.5}$ emissions from these fires have been previously underestimated, possibly by a factor of three or more. These fires are known to cause widespread smoke (or haze) and to increase the incidence of respiratory illnesses and mortality [*Koplitz et al.*, 2016; *Sahani et al.*, 2014], such that accurate knowledge of these emissions is important for understanding the impacts of these fires on human health in the region. Southeast Asian peat fires account for about half of all biomass burning emissions in that region [van der Werf et al., 2017]. If our newly derived average emission factor were implemented in GFED, peat fires would contribute 10% of global total $PM_{2.5}$ emissions from biomass burning (not 3% as with current emission factors), with Indonesia by far the largest contributor to this. This is more than the total emissions from South America, despite the significantly smaller surface area of the peatlands, and implies that this region has the highest density of emissions anywhere in the world.

Several studies have aimed to estimate premature mortality from outdoor pollution. *Lelieveld et al.* [2015] estimated that 52,000 people died prematurely in 2010 in Indonesia from inhaling outdoor air pollution from various sources, with biomass burning being responsible for 27% of this number. Our results would boost this number through higher concentrations closer to fires and the resulting larger area exposed to lower-level concentrations. While uncertain, this boost may be largest during relatively low fire years, given that then the emission factors are highest and the relation between exposure and mortality is relatively linear [*Cohen et al.*]. In addition, *Lelieveld et al.* [2015] used emissions estimates from 2010, a year with substantially lower-than-average fire emissions in Southeast Asia. Our results do not directly impact mortality rates found by *Marlier et al.* [2013], because they boosted modelled $PM_{2.5}$ to better match satellite-derived aerosol optical depth (AOD). This has become common practice in aerosol studies as aerosol models underestimate AOD by roughly a factor three [*Kaiser et al.* [2012], although the degree to which scaling is necessary varies between studies. In fact, AOD can be a poor indicator of surface $PM_{2.5}$ [*Ford and Heald*, 2016] and the scaling will be influenced by the degree of vertical mixing, which is often not well reproduced by models [*Korhonen et al.*, 2014]. While our findings are not applicable globally, this mismatch, and thus the need for scaling, is one of the key open questions in biomass burning research. Use of higher emission factors for $PM_{2.5}$ as indicated by this study would lower the need for such scaling when modelling peat fires in this region.

The decrease of fine particulate matter emissions with the age of the peat fire, as described in this study, provides an explanation for the variability in emission factors observed in

this and other measurements reported in the literature recently [*Stockwell et al.*, 2016]. The observed decrease with age of the fire was replicated in our experimental burns, with the laboratory burns displaying more rapid decreases in emissions of $PM_{2.5}$. In a real peat fire (especially at a slash and burn site), there will usually be a much greater surface area, because of uneven ground. This will slow the accumulation of the ash layer barrier above the burning peat and hence reduce the rate of decrease of fine particulate emissions. A real peat fire will spread horizontally as well as downwards and so, in a larger scale fire, the peat is likely to be burning at different depths and hence may produce different emissions factors for $PM_{2.5}$ in different plumes from the same fire as has been reported in the literature [*Stockwell et al.*, 2016]. Other factors, such as burn temperature [*Kuwata et al.*, 2017], burn history [*Konecny et al.*, 2016; *Kuwata et al.*, 2017] and peat soil bulk density [*Wijedasa*, 2016] have been used to explain variability in aerosol properties and gas emissions and may also add to variability in $PM_{2.5}$ emissions.

Use of a single average emission factor for $PM_{2.5}$ from tropical peat fires, without accounting for the decreasing emissions as a fire ages, will underestimate emissions from newly-ignited fires and overestimate emissions from long-burning fires. In fact, fire emissions from the Southeast Asian peatlands have the highest interannual variability of any region [*van der Werf et al.*, 2010], due to the suppression of fires in wetter years when the forests flood more deeply and for longer than in drier years [*Fanin and Van Der Werf*, 2017]. However using the results of this study in models like GFED would dampen this interannual variability,. This is because low fire years usually coincide with a short less intense dry season, (when many fires are started but do not grow large because of the moist conditions) and our results show that these short-lived fires have the highest emission factors of $PM_{2.5}$.

Despite the drawbacks of using a single average emission factor for $PM_{2.5}$ from peat fires, it may not always be possible to implement a time varying emission factor. Knowledge of when the fire started in different areas of the peat may be missing, or the model may not allow for variable emission factors. In these instances we recommend the use of a $PM_{2.5}$ emission factor of 24 ± 6 $g.kg^{-1}$ which is the average value for all reported emission factors for $PM_{2.5}$ from tropical peat available in the literature (six from this study, averaging 26.4 $g.kg^{-1}$ and seven from Stockwell et al., 2016, averaging 21.5 $g.kg^{-1}$).

In this study we have shown that emissions of $PM_{2.5}$ from newly ignited peat fires are likely three-times larger than previously assumed. We have found that the emissions of fine particulate matter from peat fires decrease rapidly with the age of the fire, and shown that the likely cause is the accumulation of an ash layer on the surface as the peat burns from the surface downwards. This has important implications for understanding the impact of tropical peat fires on both air quality and climate. Further measurements of emissions from tropical peat are needed and future studies should ensure that the age of the fire is noted. In the meantime, for someone wishing to implement these findings, we recommend the use of an emission factor for $PM_{2.5}$ from newly ignited tropical peat fires of 55 $g.kg^{-1}$, reducing exponentially at a rate of 9% per day. Where implementation of a variable $PM_{2.5}$ emission factor is not feasible, we recommend use of an average $PM_{2.5}$ emission factor of 24 $g.kg^{-1}$.

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