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**Article (Accepted version)
(Refereed)**

Original citation:

Smith, T. E. L. and Evers, S. and Yule, C. M. and Gan, J. Y. (2018) *In situ tropical peatland ire emission factors and their variability, as determined by field measurements in peninsula Malaysia*. [Global Biogeochemical Cycles](#). ISSN 0886-6236

DOI: [10.1002/2017GB005709](https://doi.org/10.1002/2017GB005709)

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Available in LSE Research Online: February 2018

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***In situ* tropical peatland fire emission factors and their variability, as determined by field measurements in Peninsula Malaysia.**

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Key Points:

- From measurements at 10 peat fires in SE Pahang and N Selangor, we present the first fire emission factors for Malaysian peatlands.
- We find substantial inter-plume fire emission factor variability for key greenhouse gases (e.g. methane) and reactive gases (e.g. ammonia).
- We present the first discussion of relationships between peat bulk density and methane fire emission factors.

Abstract

Fires in tropical peatlands account for >25% of estimated total greenhouse gas emissions from deforestation and degradation. Despite significant global and regional impacts, our understanding of specific gaseous fire emission factors (EFs) from tropical peat burning is limited to a handful of studies. Furthermore, there is substantial variability in EFs between sampled fires and/or studies. For example, methane EFs vary by 91% between studies.

Here we present new fire EFs for the tropical peatland ecosystem; the first EFs measured for Malaysian peatlands, and only the second comprehensive study of EFs in this crucial environment. During August 2015 (under El Niño conditions) and July 2016, we embarked on field campaigns to measure gaseous emissions at multiple peatland fires burning on deforested land in Southeast Pahang (2015) and oil palm plantations in North Selangor (2016), Peninsula Malaysia. Gaseous emissions were measured using open-path Fourier transform infrared spectroscopy. The IR spectra were used to retrieve mole fractions of twelve different gases present within the smoke (including carbon dioxide and methane), and these measurements used to calculate EFs. Peat samples were taken at each burn site for physicochemical analysis and to explore possible relationships between specific physicochemical properties and fire EFs. Here we present the first evidence to indicate that substrate bulk density affects methane fire EFs reported here. This novel explanation of inter-plume, within-biome variability should be considered by those undertaking greenhouse gas accounting and haze forecasting in this region, and is of importance to peatland management, particularly with respect to artificial compaction.

1 Introduction

Fires in tropical peatlands account for 25% or more of the estimated total greenhouse gas (GHG) emissions from global deforestation and forest degradation, amounting to approximately 3% of total global anthropogenic GHG emissions [Ballhorn *et al.*, 2009; IPCC, 2013; van der Werf *et al.*, 2009; van der Werf *et al.*, 2017]. In 1997 alone, El Niño-related fires in Indonesia were calculated to have released 13–40% of that year's total carbon emissions from fossil fuel burning [Page *et al.*, 2002]. Beyond the global climate forcing of GHGs such as CO₂, methane (CH₄), and nitrous oxide (N₂O), peatland fire emissions of carbon monoxide (CO), nitrogen oxides (NO_x) and volatile organic compounds (VOCs) react to form ozone (O₃). This, combined with particulate matter (PM) emissions, leads to smog (referred to as 'haze' in SE Asia). In the southern region of SE Asia, peatland fires are responsible for 90% of trans-boundary haze events [Varkkey, 2013]. Air Pollution Index readings in Palangkaraya in Borneo in October 2015 reached >2000, the highest values ever recorded and far above the Emergency level of >500. Exposure to haze events has been shown to cause both immediate and delayed effects, increasing respiratory related mortality by 19–66% [Sahani *et al.*, 2014] and resulting in over 100,000 excess deaths in Malaysia, Indonesia and Singapore [Kopplitz *et al.*, 2016]. In the 2015 fires, an estimated >500,000 Indonesians were affected by haze-related respiratory infections [Jakarta Post, 2015]. Economic losses related to haze are also significant, with estimates for the 2015 fires at \$16.1 billion for Indonesia [CIFOR, 2015]. Despite these significant global and regional impacts, very little is known of the proportional contribution of the major chemical compounds being emitted *in situ* from tropical peat fires. As a result, there are no published accounts addressing how GHG and reactive (haze-forming) gaseous fire emissions vary among different types of tropical peatlands, precluding both accurate greenhouse gas accounting and air quality (haze) forecasting in this region.

The Indo-Malaysian region is the global centre of tropical peat swamp forests. Over the past 30 years regional peat swamp forests have been increasingly subject to degradation from drainage, logging and agricultural conversion – particularly to oil palms (*Elaeis guineensis*). Drainage reverses the environmental conditions that lead to peat accretion, resulting in oxidation, compaction and drying of peat making it highly flammable [Langner and Siegert, 2009; Wösten *et al.*, 2006]. Fire is often used to clear peatlands [Varkkey, 2013] and these fires can continue smouldering for months both at the surface and reaching deeper underground. Peatlands subjected to fire incidents have a high probability of recurrence of fire [Posa *et al.*, 2011] because of the un-burnt biomass, removal of canopy shading and fire prone secondary vegetation that will act to provide further fuel and ignitions [Siegert *et al.*, 2001]. Due to the extensive degree of land degradation in the region, peatland fires now occur twice a year in the Peninsula Malaysia and Sumatra regions, during each dry season [Wooster *et al.*, 2012], occurring in February–May and August–September, but may occur outside of these periods (e.g. the fires in Riau, Sumatra, in June 2013; [Gaveau *et al.*, 2014]), depending on human and climate factors.

Understanding within-biome emissions variability is an important challenge for assessing the atmospheric impacts of tropical peatland fires; in a broader context, van Leeuwen and van der Werf [2011] suggest that explaining emission factor (EF, grams of a compound emitted per kg of dry biomass burned) variability remains one of the biggest challenges for biomass burning emissions science. Despite this, our understanding of specific gaseous emissions from peat burning is limited to a handful of studies where small peat samples were burned in laboratories [Christian *et al.*, 2003; Stockwell *et al.*, 2014] and two field studies [Huijnen *et al.*, 2016; Stockwell *et al.*, 2016]. With the exception of Stockwell *et al.* [2014], these studies do not characterise the physicochemical properties of samples, and with the exception of Stockwell *et al.* [2016], nor were specific location, fire history or sample depth stated, prohibiting any explanation of the substantial variability between samples and/or studies. For example Huijnen *et al.* [2016] report that 7.8 g of methane is emitted per kilogram of dry peat that burns, whereas Christian *et al.* [2003] report 20.8 g, a difference of 91%, leading to major implications when scaled-up to regional GHG accounting. Similar large differences in the literature exist for other important emission factors (e.g. ammonia, a reactive nitrogen species).

2 Tropical peatland fire emission factors

Unlike logging, the open burning of biomass directly consumes carbon stocks and rapidly releases emissions (e.g. GHGs, haze-forming species, PM) to the atmosphere ‘on site’. How well peat fire emissions are accounted for depends upon uncertainties in the calculation of (i) The amount of peat consumed in the fire; and (ii) the emission factors used to convert the estimate of fuel consumption into an estimate of the mass of gas released per unit of dry fuel burned (g kg^{-1}) [Penman *et al.*, 2003]. Emission factors for CH_4 and important haze-forming compounds vary massively between the few studies of gaseous emission factors [Christian *et al.*, 2003; Stockwell *et al.*, 2014; Stockwell *et al.*, 2016]. The small number of samples in these studies, combined with limited information on physicochemical peat properties beyond elemental analysis, prohibits an explanation of the large differences in emissions factors between studies.

Previous studies of peat fire gaseous emissions factors have predominantly undertaken Fourier transform infrared (FTIR) spectroscopic measurements of emissions from peat

118 samples burning in a laboratory. These studies have investigated fire emissions from samples
 119 of temperate peat [Stockwell *et al.*, 2014; Wilson *et al.*, 2015; Yokelson *et al.*, 1997]; boreal
 120 peat [Stockwell *et al.*, 2014; Yokelson *et al.*, 1997]; and include studies of samples of tropical
 121 peat from Sumatra [Christian *et al.*, 2003] and Kalimantan [Stockwell *et al.*, 2015; Stockwell
 122 *et al.*, 2014].

123 Christian *et al.* [2003] report emission factors from just a single small sample of Sumatran
 124 peat, while Stockwell *et al.* [2014] report emission factors from two further samples from
 125 Kalimantan. Neither study reports any biological or chemical properties of the peat that might
 126 help to explain the large differences in emissions factors between the two studies. Both the
 127 2013 IPCC ‘Supplement to the 2006 Guidelines for National Greenhouse Gas Inventories:
 128 Wetlands’ [Hiraishi *et al.*, 2014] and the emission factors database of Akagi *et al.* [2011]
 129 summarise emission factors from tropical peatland fires using only the laboratory findings of
 130 Christian *et al.* [2003], with the IPCC stating that these were “derived from a very limited
 131 number of studies”. Stockwell *et al.* [2014] and Stockwell *et al.* [2016] offer an update to the
 132 emission factors summarised by the IPCC and Akagi *et al.* [2011] by taking an average of
 133 their findings and those of Christian *et al.* [2003].

134 Despite these advances, it is difficult to assess the representativeness of emissions from
 135 laboratory-scale burns [Stockwell *et al.*, 2016], particularly for peat fuels, that under
 136 landscape-scale burning conditions would smoulder at the surface or underground, with less
 137 access to oxygen than might be available under laboratory conditions [Rein *et al.*, 2009]. As
 138 far as we are aware, the first *in situ* field measurements of gaseous fire emissions from
 139 tropical peatland fires are those by Hamada *et al.* [2013], whose study of landscape-scale
 140 peat fires near Palangka Raya (Kalimantan, Indonesia) in August 2009 reports emission ratios
 141 of CH₄, N₂O, and CO, to CO₂. Huijnen *et al.* [2016] report emission factors for CO₂, CO, and
 142 CH₄ from peatland fires measured *in situ* using a portable cavity enhanced laser absorption
 143 spectrometer, also near Palangka Raya, in October 2015. The most comprehensive field
 144 study to date is that of Stockwell *et al.* [2016] who reported gaseous and particulate emission
 145 factors for ~90 species from measurements at 35 smoke plumes during the 2015 El Niño in
 146 Central Kalimantan, Indonesia. There have been no published laboratory or *in situ* derived
 147 emission factors for Malaysian peat fires.

148 van Leeuwen and van der Werf [2011] argue that future fire emissions measurement
 149 campaigns would be far more beneficial to the global modelling community if measurements
 150 were distributed spatially across biomes and paid more attention to fuel properties. The
 151 physical properties of peat fuels (e.g. bulk density, fuel moisture) are important because they
 152 will determine the combustion dynamics [Rein *et al.*, 2009]. Therefore, degradation impacts
 153 (such as the lowering of the water table or artificial peat compaction) on the physical
 154 properties of tropical peats (e.g. moisture content, bulk density) may influence the relative
 155 abundances of CO₂ and haze-forming compounds during fires [Moreno *et al.*, 2011; Rein *et al.*,
 156 *et al.*, 2009].

157 The chemical properties of fuels are important because they will determine the chemical
 158 composition of fire emissions. Whilst the carbon content of vegetation and tropical peat does
 159 not vary much beyond 50–56% [Christian *et al.*, 2003; Stockwell *et al.*, 2014], nitrogen
 160 content of tropical peats is known to vary from 1.0–4.4% and may be heavily influenced by
 161 land conversion [Andriess, 1988] and management. This will influence nitrogenous fire EFs
 162 (e.g. NO_x, N₂O, NH₃, HCN) which are important for O₃ and haze formation. Direct impacts
 163 of degradation include the use of fertilizers on converted peatlands (increasing peat nitrogen),
 164 leading to the potential for increasing fire EFs for these nitrogen compounds.

Here we report new emission factors for twelve gas species measured using open-path Fourier transform infrared (OP-FTIR) spectroscopy *in situ* at ten plumes from actively burning tropical peatland fires in Southeast Pahang and North Selangor, Malaysia. These represent the first emission factors reported for Malaysian peatlands.

3 Methodology

3.1 Site and fire descriptions

Tropical peatland fire emissions were measured at wildfires on peatlands along a stretch of road (Federal Route 3) connecting Pekan and Lanjut in the Pekan District of the State of Pahang, Malaysia in August 2015, and at oil palm plantations located about 20 km north of Sekinchan, off Federal Route 5, in the Sabak Bernam District of the State of Selangor, Malaysia in July 2016. Measurements at the Pahang sites were made on eight separate occasions in August 2016 at three different sites between Pekan and Lanjut, within a region 80 km south of Kuantan. This area has a tropical wet climate (Koppen-Geiger classification: Af), with no well-defined dry or wet seasons, being hot and humid throughout the year. According to weather observations at Kuantan, mean annual rainfall is 2,900 mm. Peaks in rainfall occur in December (564 mm) and May (202 mm). August sees a mean maximum temperature of 32.3°C and mean rainfall of 174 mm. The peat soils at the Pahang sites, which were located on the coastal edge of the main southeast Pahang peat dome (Figure 1), were very shallow (0.2–0.8 m), typical of degraded peatlands subject to drainage, land clearance, and hence oxidative loss of the peat layer over some decades. Two further sets of measurements were made in July 2016 at one site in north Selangor. This area has a similar climate to that of southeast Pahang. Weather observations at the nearest weather station in Kuala Lumpur show a mean annual rainfall of 2,600 mm. July sees a mean maximum temperature of 32°C and mean rainfall of 150 mm. The peat soils at the Selangor sites, also located towards the edge of the main north Selangor dome, were deeper than those in Pahang, extending to 1.5 to 2 m depth.

The flora of these areas is typical of deforested and degraded tropical peatlands across the Malay Peninsula with many introduced, weedy species. There is a ground cover of grasses and ferns as well as shrubs (particularly *Melastoma* sp. and *Mimosa* sp.) and trees such as *Acacia mangium* and *Macaranga pruinosa*. *A.mangium* is a widespread introduced species which can promote fire intensity and frequency due to flammable leaf litter and which produces enormous numbers of seeds that remain viable for years and whose germination is enhanced by fire. *M.pruinosa* is a native, secondary gap colonizer commonly found on the fringes of regional peat swamp forests. Oil palm plantations are usually cleared manually and treated with herbicides to remove other vegetation, leaving only a sparse ground cover of grasses and ferns. We found evidence of ongoing chemical fertiliser use at the Selangor oil palm plantation site (Site 4 in Figure 1), with open sacks of NPK fertiliser and pellets around the base of oil palms.

Locations of actively burning peatland fires were identified by regular monitoring of the World Resources Institute Global Forest Watch Fires website (<http://fires.globalforestwatch.org/>) and NASA's Worldview website (<https://worldview.earthdata.nasa.gov/>). These websites provide the near-real-time location of satellite-detected fires (thermal hotspots); true-colour and false-colour imagery that can be used to identify the presence of smoke plumes; as well as information on the location of

peatlands. Many active fires were observed in SE Pahang (Sites 1–3 in Figure 1) in July 2015 shortly before the ground measurement campaign; this led the research team to be based at Pekan, just north of these July fire clusters. Once a possible peatland fire had been identified, the research team drove in the direction of the fires until a visible confirmation of the location of the fire was established. Fewer fires were observed across Peninsula Malaysia in July 2016, however a cluster of hotspots were detected just north of Sekinchan in N Selangor (Site 4 in Figure 1) in early July which led the research team to this area where a number of small peatland fires were found on oil palm plantations.

The opportunistic nature of this campaign allowed for emissions to be measured at a number of sites (Figure 1) that had different degradation characteristics, and at sites that had been burning for different lengths of time. Table 1 summarises the main characteristics of the fire sites and conditions studied for this paper.

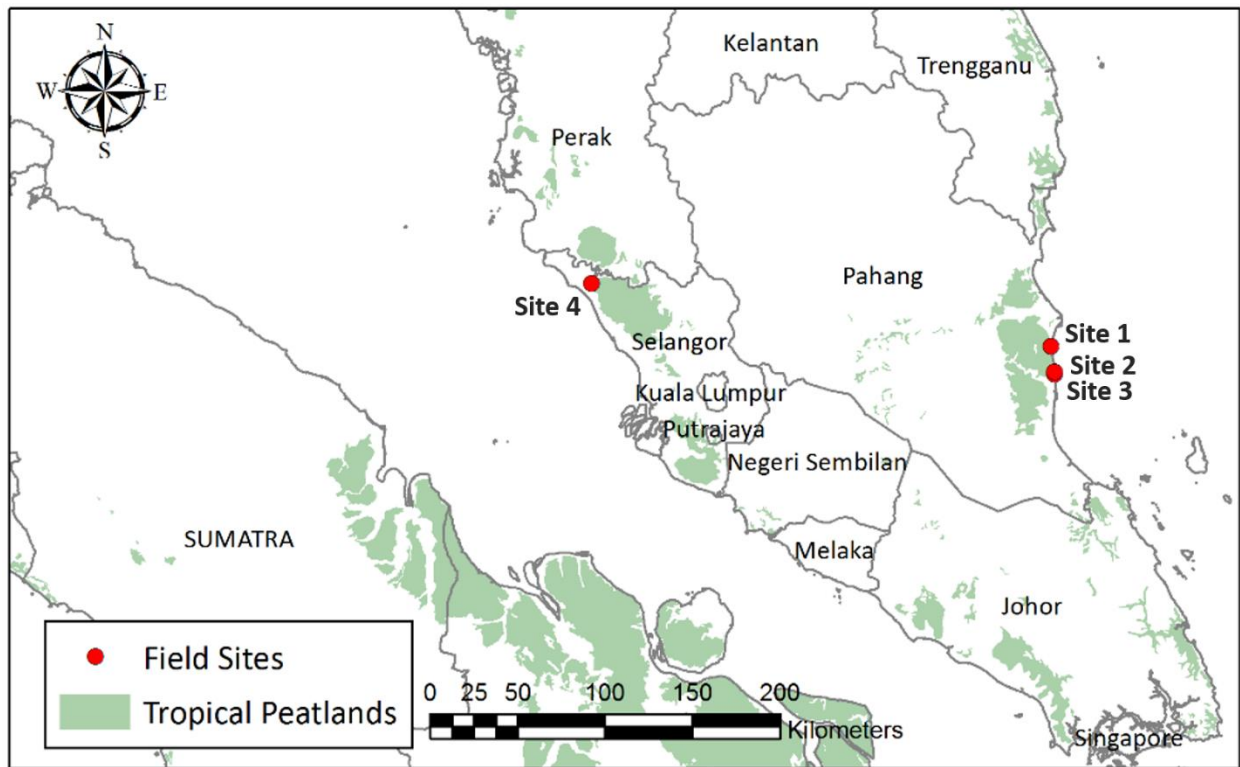
Site 1 (SE Pahang) was visited twice, on 6 August 2015, following rain, when small areas (patches of a few square metres) of the landscape were still smouldering following the large fires in mid-July; and on 12 August 2015, when relatively larger areas were burning (patches of $>10\text{ m}^2$). Site 1 was largely devoid of any tree vegetation and surface vegetation was dominated by shrubs and leaf litter.

Site 2 (SE Pahang) was visited four times. On 17, 18, and 20 August 2015, when a large landscape-scale peat fire was burning across many hectares, and on 24 August, following rain, when smaller patches of the landscape fire were persisting. Site 2 had also been burning since the large fires in the region in mid-July and is dominated by shrub vegetation with some secondary tree growth.

Site 3 (SE Pahang) was visited twice, on 20 August 2015, when a large landscape-scale fire was burning through the area, and on 24 August, following rain, when smaller areas of the landscape ($\sim 10\text{ m}^2$) were burning. Site 3 had more secondary growth tree cover than the other sites.

Measurements were made at Site 4 (N Selangor) on two occasions at a palm slash fire that was burning into the peat on 20 July 2016, and at the same fire seven days later on 27 July 2016.

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Figure 1. Map of southern Peninsula Malaysia and Sumatra, showing the location of sites measured in our two field campaigns; Sites 1–3 (Plumes P1–8) in August 2015, and Site 4 (Plumes S1–2) in July 2016. The map also shows the location of Malaysian peatlands (Wetlands International, 2017) and Indonesian peatlands (Ministry of Agriculture, 2017).

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Table 1. List of the ten plumes studied using OP-FTIR during the two Malaysian measurement campaigns in SE Pahang (P1–8) and N Selangor (S1–2), with information about the site number (Figure 1), plot location, OP-FTIR pathlength, meteorological variables, as well as peat physicochemical characteristics, including bulk density, soil moisture, nitrogen-content, and carbon-content. MCE calculated using Eqn. 5 is also presented. Standard deviations are presented in parentheses.

Plume	Site	Lat (°)	Long (°)	Path (m)	Temp (°C)	Pressure (hPa)	Bulk density (g cm ⁻³)	Soil moist (%)	N (%)	C (%)	MCE
P1	1	3.361	103.420	17	34	1008	0.63 (0.03)	24 ± 0.0	-	-	0.82
P2	1	3.361	103.420	12	36	1011	0.46 (0.20)	20 ± 1.3	0.39 (0.46)	15.2 (7.0)	0.77
P3	2	3.221	103.440	35; 29	40	1008	0.28 (0.09)	33 ± 0.1	0.69 (0.57)	20.9 (11.9)	0.83
P4	2	3.221	103.440	23; 13	38	1009	0.31 (0.10)	31 ± 0.1	0.58 (0.51)	18.3 (13.4)	0.79
P5	3	3.231	103.437	20; 12	40	1010	0.25 (0.05)	41 ± 0.6	1.75 (0.27)	43.3 (7.8)	0.79
P6	2	3.221	103.440	17	40	1010	0.38 (0.04)	29 ± 1.2	0.51 (0.21)	23.2 (15.7)	0.75
P7	3	3.231	103.437	12; 11; 10	34	1011	0.41 (0.11)	16 ± 0.3	1.76 (0.27)	41.1 (2.2)	0.78
P8	2	3.221	103.440	11; 11	34	1011	0.37 (0.10)	28 ± 0.9	1.02 (0.58)	24.3 (13.5)	0.79
S1	4	3.686	101.054	28	32	1008	0.62 (0.04)	53 ± 1.1	1.34 (0.22)	55.6 (2.6)	0.84
S2	4	3.686	101.054	18	33	1006	0.61 (0.04)	52 ± 1.2	1.34 (0.22)	55.6 (2.6)	0.84

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3.2 Fire emissions measurements

Here we use an Open-Path Fourier Transform Infrared (OP-FTIR) approach to estimate path-averaged mole fractions of twelve trace gases. The OP-FTIR system consists of a MIDAC Corporation M2000 Series FTIR spectrometer equipped with a Stirling-cooled mercury-cadmium-telluride (MCT) detector, and fitted with a MIDAC custom-built 76 mm Newtonian telescope. The spectrometer is used to view a remotely located infrared source, consisting of a 12 V silicon carbide glowbar operating at 1500 K fitted in front of a 20 cm diameter gold-plated collimator. The MIDAC system and its use to characterise biomass burning emissions via long OP-FTIR spectroscopy is detailed in *Smith et al.* [2014] and *Wooster et al.* [2011].

At all sites, the OP-FTIR path was positioned directly above the actively burning peat (see Figure 2). There was no evidence of flaming combustion at any of the burns, with all smoke being produced by pyrolysis and smouldering combustion. Given that there were no quick changes to burning conditions during any one deployment, spectra were collected using sixteen co-added scans to increase signal-to-noise (generating approximately one measured spectrum every 10 s). Observations of atmospheric temperature and pressure (Table 1) were made using a co-located Kestrel 5500 Weather Meter (Nielsen-Kellerman Co.).

Path-averaged trace gas mole fractions were retrieved from the OP-FTIR spectra using the Multiple Atmospheric Layer Transmission (MALT) program [*Griffith*, 1996], where selected spectral regions of the measured OP-FTIR spectra are fitted with synthetic spectra. A more comprehensive description of MALT can be found in *Griffith* [1996] and more details of the specific spectral regions used for the retrieval can be found in *Paton-Walsh et al.* [2014] and *Smith et al.* [2014]. An accuracy assessment of greenhouse gas mole fraction retrievals using this approach can be found in *Smith et al.* [2011], whose findings suggest that retrievals of CO₂, CH₄, and CO are accurate to within 5% of true mole fractions. *Stockwell et al.* [2016] also found retrieved mole fractions of CO₂, CH₄, and CO₂ to have an uncertainty of 3–5%.

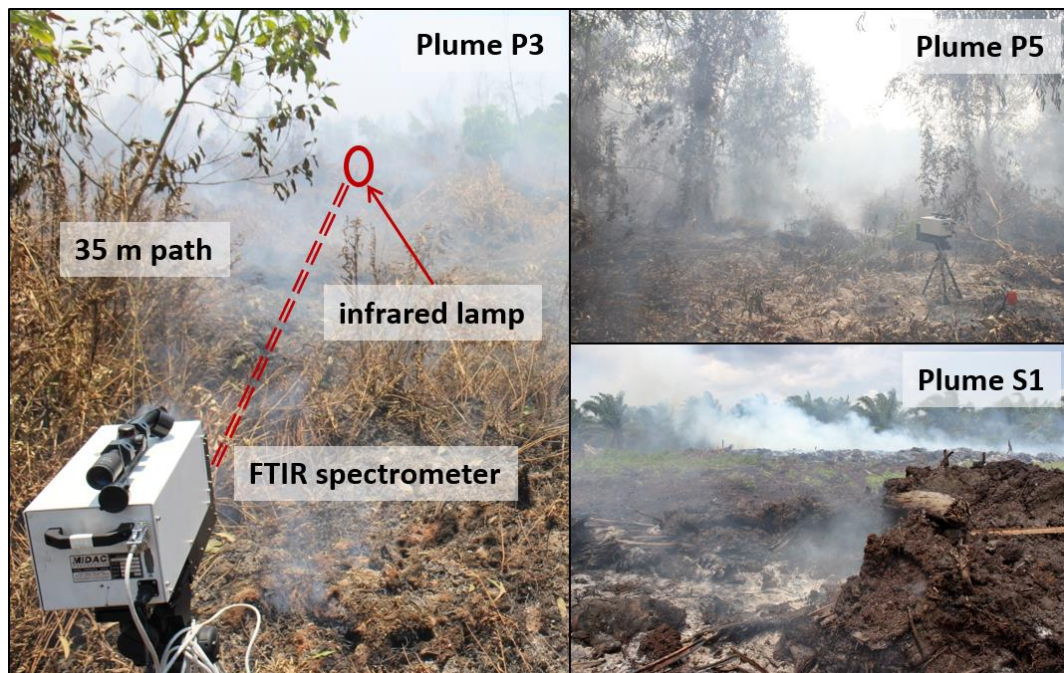


Figure 2. (left) Photograph of a typical OP-FTIR setup measuring Plume P3 using a 35 m path between the FTIR spectrometer and infrared lamp (see Table 1); (right) Photographs from the sites of Plume P5 and Plume S1.

3.3 Calculation of emission ratios and emission factors

The emission ratio of species i to a reference species y is given by:

$$ER_{i/y} = \frac{\Delta[i]}{\Delta[y]} = \frac{[i] - [i]_{background}}{[y] - [y]_{background}} \quad (1)$$

where $\Delta[i]$ is the excess mole fraction of species i . Following the same method as *Paton-Walsh et al.* [2014], here we derive emission ratios via the gradient of the linear best fit of all measurements of the abundance of species i against the abundance of reference species y for each fire.

The emission factor (EF_i) refers to the mass of species i emitted per unit of dry fuel consumed, expressed in units of g kg^{-1} . Here, the emission factors for all species (except CO_2 and CO) i are calculated via:

$$EF_i = ER_{i/\text{CO}} \times \frac{MM_i}{MM_{\text{CO}}} \times EF_{\text{CO}} \quad (2)$$

where $ER_{i/\text{CO}}$ is the emission ratio of species i to CO ; MM_i is the molecular mass of species i ; MM_{CO} is the molecular mass of CO (28.01 g mol^{-1}); and EF_{CO} is the emission factor for CO . The emission factor for CO_2 and CO is determined using the “summation method” [*Paton-Walsh et al.*, 2014] where it is necessary to calculate the total excess amounts of each gas species by summing the excess amounts retrieved for each spectrum (i.e. $[i] - [i]_{background}$ in Eqn. 1); emission factors may then be calculated using the carbon mass balance method [*Ward and Radke*, 1993]:

$$EF_{\text{CO}} = (F_C - F_{\text{PMC}}) \times 1000 \times \frac{MM_{\text{CO}}}{AM_C} \times \frac{\Delta_{\text{CO}}}{\sum_{i=1}^n (NC_i \times \Delta_i)} \quad (3)$$

$$EF_{\text{CO}_2} = (F_C - F_{\text{PMC}}) \times 1000 \times \frac{MM_{\text{CO}_2}}{AM_C} \times \frac{\Delta_{\text{CO}_2}}{\sum_{i=1}^n (NC_i \times \Delta_i)} \quad (4)$$

where F_C is the measured carbon mass fraction of the fuel (see Sect. 2.4); F_{PMC} is the carbon mass fraction of the fuel that is emitted as particulate matter (see below); AM_C is the atomic mass of carbon (12 g mol^{-1}); NC_i is the number of carbon atoms in species i ; Δ_{CO} and Δ_i are the summed excess mole fractions of CO and species i respectively. The emission factor for CO_2 is calculated using the molecular mass of CO_2 ($MM_{\text{CO}_2} = 44.01 \text{ g mol}^{-1}$) and Δ_{CO_2} excess mole fractions of CO_2 .

The fraction of carbon emitted as particulate matter (F_{PMC}) is usually considered to be negligible and ignored in the application of carbon mass balance approaches for the calculation of gaseous fire emission factors (e.g. *Wooster et al.* [2011]). However, a recent *in situ* study of PM EFs for tropical (Indonesian) peatlands [*Jayarathene et al.*, 2017] suggests that PM emissions are not negligible for tropical peatland fuels and should therefore be accounted for in calculations of gaseous emission factors. *Jayarathene et al.* [2017] report an EF of 17.3 g kg^{-1} for $\text{PM}_{2.5}$ with the $\text{PM}_{2.5}$ consisting of 73% carbon. We therefore assume F_{PMC} of 0.0127 for Eqns. 3 and 4. EFs are directly proportional to the combined carbon mass fraction in Eqns. 3 and 4, enabling easy correction of determined average EFs should this be warranted by additional future PM measurements.

To enable comparison with other studies and understanding of variability between sites, we also report the modified combustion efficiency (MCE). MCE is an approximation of the combustion efficiency (the proportion of total carbon emitted as CO₂) and is given by *Hao and Ward* [1993] as:

$$\text{MCE} = \frac{\Delta[\text{CO}_2]}{\Delta[\text{CO}_2] + \Delta[\text{CO}]} \quad (5)$$

The calculation of emission factors and MCE require knowledge of the background mole fractions of all species. Background OP-FTIR spectra were collected upwind of each of our fire sites prior to measuring the fire emissions.

3.4 Peat composition

Four substrate samples were taken from the uppermost section of the underlying unburnt substrate/peat layer, or from areas immediately adjacent to burning areas at each burn site. Samples were taken along a transect between the spectrometer and infrared lamp using a 10 cm diameter and 10 cm deep brass corer. Wet and dry mass were measured to calculate bulk density and moisture content on dry basis (Table 1). Total carbon (C) and nitrogen (N) content were also determined (Table 1). For this, peat sub-samples were first oven dried at 105°C for 48 hours and then ball milled for homogenisation using a Planetary Ball Mill (Retsch-PM400, Castleford, UK). Analysis of C and N used 20 mg of material enclosed in a tin capsule with measurements undertaken using a total element analyzer (Thermo Flash EA 1112, CE Instruments, Wigan, UK).

4 Results and Discussion

4.1 Peat substrate composition

Given that the Pahang sites had been heavily degraded, with only a thin layer of peat remaining, the majority of the Pahang substrate samples were not pure ‘pristine’ peat, with our samples visibly containing sand or clay indicating that the peat fires had burned down to the interface with the mineral soil substrate at the Pahang sites (carbon contents ranging from 15.2–43.3%), typical of degraded peats on the fringes of peat domes [*Miettinen et al.*, 2017]. The samples taken at the Selangor site were clearly peat, the mean carbon content of these samples (55.6%) is close to that found for Sumatran peat (57.9%), as reported by *Stockwell et al.* [2014]. The nitrogen content of our samples varied significantly between 0.39% and 1.76%. Nitrogen content of pristine peats is known to vary from 1.0% to 4.4% [*Andriessse*, 1988]. The lower nitrogen content measured here (<1%) is probably a further indicator of degradation as the main source of nutrients to the soil (peatswamp forest leaf litter) had been removed. The mean nitrogen content at the Selangor palm oil sites (1.34%) is higher than that for the abandoned Pahang sites (0.96%) and we speculate that this is due to the use of artificial fertiliser on the palm oil plantations.

We use a carbon fraction of 0.556 for the calculation of our peat fire emission factors (in Eqn. 3), in line with samples that we determine to have been closer in composition to the overlying peat that was consumed by the fire, and also similar to the carbon fraction of 0.5793 ± 0.0252 used by *Stockwell et al.* [2016], as found in the samples analysed *Stockwell et al.* [2014]. As is the case for the fraction of carbon emitted as PM_{2.5}, EFs are directly proportional to the

carbon mass fraction, enabling easy correction of EFs should this be warranted by additional future carbon mass fraction measurements.

All of our sites exhibited high bulk density for tropical peat, with bulk densities ranging from 0.28 to 0.63 g cm⁻³; this compares with 0.09–0.11 g cm⁻³ that might be expected for pristine tropical peat [Page *et al.*, 2011; Warren *et al.*, 2012]. Drainage causes loss of water from peat pores and shrinkage through drying, resulting in an increase in bulk density which is exacerbated by the resulting aerobic conditions causing peat degradation due to oxidation, increased microbial decomposition and compaction [Könönen *et al.*, 2015]. Oil palm plantation management with addition of lime and fertilizers further enhances microbial peat breakdown, while artificial mechanical compaction (designed to increase root stability and per volume nutrient status [Melling *et al.*, 2009], also results in further increases to bulk [Evers *et al.*, 2016]. The mean bulk density at our two Selangor sites located on palm oil plantations (0.62 g cm⁻³) was significantly higher than the mean bulk density at our deforested but abandoned Pahang sites (0.39 g cm⁻³), probably due to extensive use of heavy machinery and purposeful artificial compaction of the peat soil at the Selangor sites.

4.2 Trace gas emission factors and modified combustion efficiency

Figures 3 and 4 show example time series for each of the investigated species and their corresponding emission ratio plots, respectively, for Plume P5. Our emission ratio plots show a good correlation between species and mole fractions elevated far above background, indicating that the plumes sampled at fires in this study were well-mixed [Stockwell *et al.*, 2016], and giving confidence to the individual EFs calculated for each fire (Table 2).

Table 2 presents the individual trace gas EFs and their associated uncertainties (in accordance with Paton-Walsh *et al.* [2014]) for each of the ten plumes sampled. An EF is calculated only when a trace gas species has a strong emission ratio correlation (>0.4). Table 3 presents a summary of our study-averaged EFs and one standard deviation of the means for all species. From measurements of ten tropical peat fire plumes, we find the major trace gas emissions by mass (EF > 0.5 g kg⁻¹, as defined by Stockwell *et al.* [2016] to be: carbon dioxide (1579 ± 58); carbon monoxide (251 ± 39); methane (11.00 ± 6.11); ammonia (7.82 ± 4.37); acetic acid (5.02 ± 1.64); hydrogen cyanide (3.79 ± 1.97); methanol (2.83 ± 0.84); ethylene (2.30 ± 2.79); ethane (2.17 ± 0.81); and formaldehyde (0.77 ± 0.64). We report EFs for two further species that may be categorised as minor trace gas emissions by mass: formic acid (0.25 ± 0.04); and acetylene (0.06 ± 0.01). The modified combustion efficiency (MCE) of the fire burning to produce the plumes sampled here ranged from 0.774 to 0.839 (Table 2), with an average of 0.800 ± 0.031 indicating pure smouldering combustion.

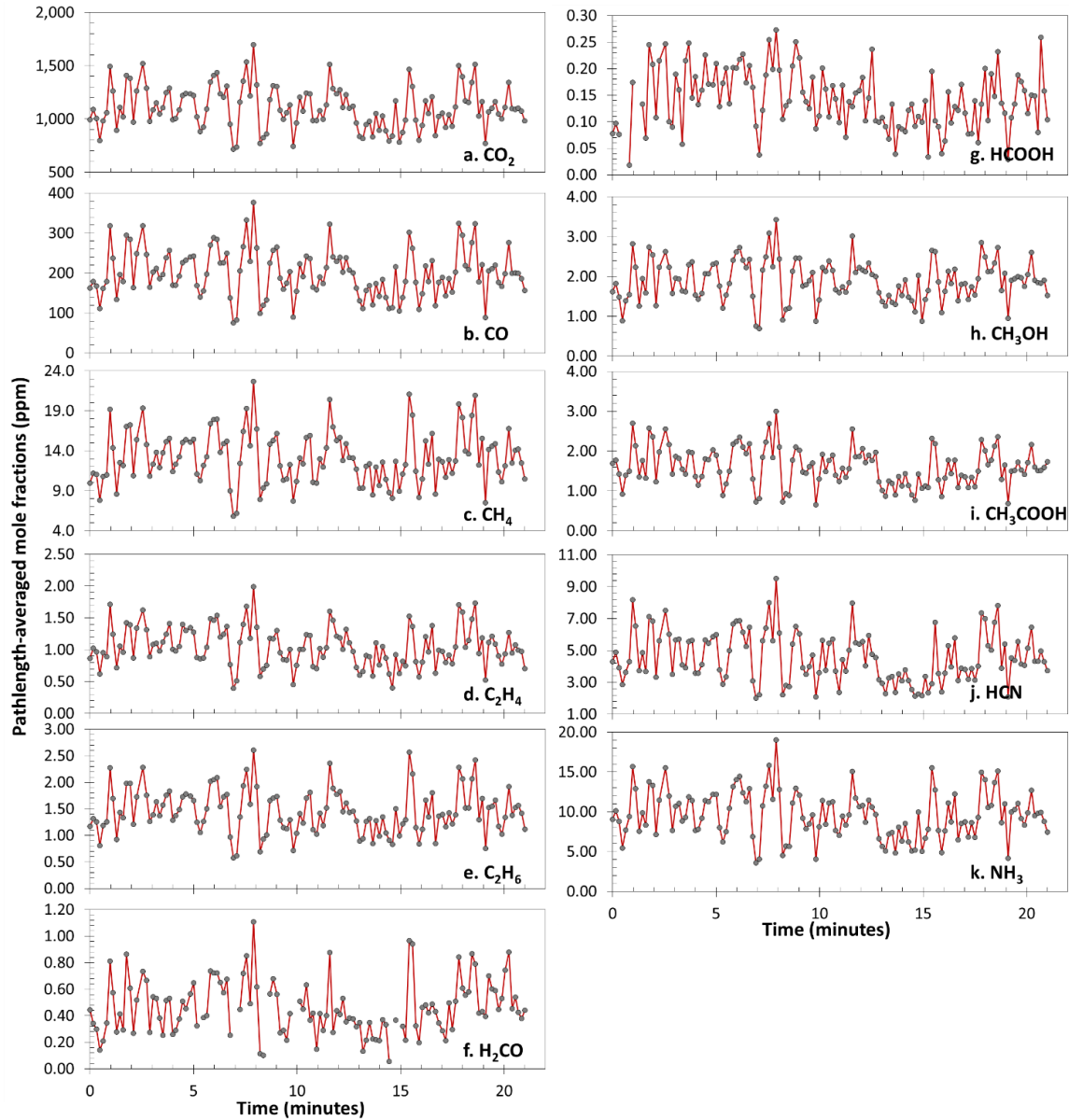
Table 2. Emission factors (g kg^{-1} of dry fuel burned) for each individual plume, calculated using the standardised method outlined in *Paton-Walsh et al.* [2014]. Uncertainties were calculated in quadrature from those associated with the trace gas emission ratios and a $\pm 10\%$ uncertainty in the assumed fuel carbon.

PLUME SITE	P1 1	P2 1	P3 2	P4 2	P5 3	P6 2	P7 3	P8 2	S1 4	S2 4
MCE	0.82	0.77	0.83	0.79	0.79	0.75	0.78	0.79	0.84	0.84
CO ₂	1545±154	1541±154	1662±166	1579±158	1575±157	1488±149	1535±153	1563±156	1653±165	1648±165
CO	216±34	286±46	218±35	261±42	261±42	314±50	283±45	266±43	201±32	200±32
CH ₄	26.19±4.20	6.67±1.08	5.35±0.88	7.68±1.27	8.38±1.37	9.32±1.47	8.83±1.37	8.83±1.37	14.76±2.35	14.07±2.25
C ₂ H ₂	0.06±0.01	nr	0.05±0.01	nr	nr	nr	nr	nr	nr	nr
C ₂ H ₄	10.16±1.02	1.57±0.16	1.23±0.13	1.40±0.14	1.71±0.17	1.25±0.13	1.65±0.17	1.46±0.15	0.80±0.08	1.68±0.17
C ₂ H ₆	3.88±0.62	0.90±0.15	1.66±0.26	2.24±0.36	2.02±0.32	2.83±0.46	2.07±0.32	2.05±0.32	2.56±0.41	1.44±0.22
H ₂ CO	nr	1.83±0.19	0.66±0.07	0.46±0.05	0.76±0.08	0.16±0.02	nr	nr	nr	nr
HCOOH	nr	nr	0.28±0.05	nr	0.23±0.04	nr	nr	nr	nr	nr
CH ₃ OH	2.79±0.45	3.82±0.62	1.62±0.26	1.94±0.31	2.50±0.40	2.37±0.38	2.91±0.47	3.28±0.53	2.68±0.43	4.46±0.71
CH ₃ COOH	8.91±1.43	3.66±0.59	3.65±0.59	4.14±0.66	3.78±0.61	4.46±0.71	4.14±0.66	5.65±0.90	6.20±1.00	5.61±0.90
HCN	3.43±0.55	3.26±0.52	0.34±0.06	1.96±0.31	5.99±0.96	3.26±0.52	6.24±1.00	5.92±0.95	3.71±0.22	nr
NH ₃	14.46±2.34	3.08±0.49	3.74±0.60	5.24±0.84	8.32±1.33	5.75±0.92	9.06±1.46	5.34±0.85	7.07±1.13	16.13±2.60

Table 3. Emission factors (g kg^{-1} of dry fuel burned) reported by this study of Malaysian peatlands and those for the same trace gases reported by previous *in situ* Indonesian peatland studies [*Huijnen et al.*, 2016; *Stockwell et al.*, 2016] and laboratory studies [*Christian et al.*, 2003; *Stockwell et al.*, 2014]. The mean and standard deviation (in parentheses) are calculated from individual plumes (for *in situ* studies) or samples (for laboratory studies).

Trace Gas	Emission Factor (g kg^{-1} dry fuel burned)				
	Malaysian peat (<i>in situ</i>) (this study)	Kalimantan peat (<i>in situ</i>) (Stockwell et al. 2016)	Kalimantan peat (<i>in situ</i>) (Huijnen et al. 2016)	Sumatran peat (lab) (Christian et al. 2003)	Kalimantan peat (lab) (Stockwell et al. 2014)
MCE	0.800 (0.030)	0.772 (0.053)	-	0.838	0.816 (0.065)
CO ₂	1579 (58)	1564 (77)	1625 (170)	1703	1637 (204)
CO	251 (39)	291 (49)	234 (47)	210.3	233 (72)
CH ₄	11.00 (6.11)	9.51 (4.74)	7.8 (2.5)	20.8	12.8 (6.6)
C ₂ H ₂	0.06 (0.01)	0.121 (0.066)	-	0.06	0.18 (0.05)
C ₂ H ₄	2.30 (2.79)	0.961 (0.528)	-	2.57	1.39 (0.62)
C ₂ H ₆	2.17 (0.81)	1.52 (0.66)	-	-	-
H ₂ CO	0.77 (0.64)	0.867 (0.479)	-	-	1.25 (0.79)
HCOOH	0.25 (0.04)	0.180 (0.085)	-	0.79	0.55 (0.05)
CH ₃ OH	2.83 (0.84)	2.14 (1.22)	-	8.69	3.24 (1.39)
CH ₃ COOH	5.02 (1.64)	3.89 (1.65)	-	8.97	7.65 (3.65)
HCN	3.79 (1.97)	5.75 (1.60)	-	8.11	-
NH ₃	7.82 (4.37)	2.86 (1.00)	-	19.92	1.39 (0.97)

^a Extratropical peat emission factors are taken from *Stockwell et al.* [2014], which are a combination of their findings with those from *Yokelson et al.* [1997]



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Figure 3. Time series of path-averaged trace gas mole fractions (in ppm) for Plume P5 as measured using a 12 m path (20 August 2015) during the strong El Niño dry period of 2015. Any gaps in the time series of specific trace gases are due to periods of low signal-to-noise within the spectral window used for the retrieval of that particular species. A photograph taken during the measurements of P5 is presented in Fig. 2. Emission ratio plots for this plume are presented in Figure 4.

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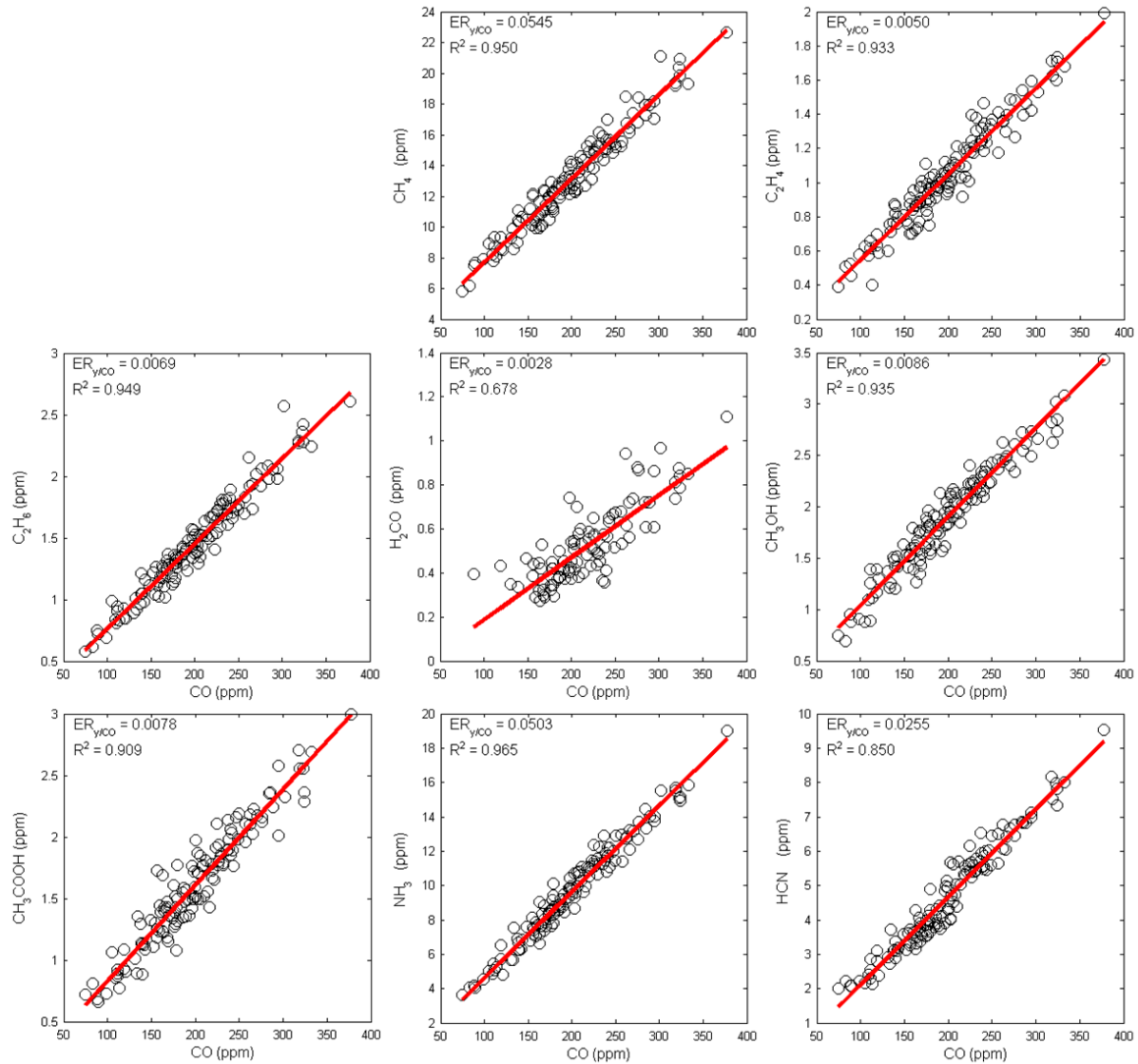


Figure 4. Example scatter plots of the measured trace gas pathlength-averaged mole fractions used to calculate emission ratios (ER_{i/CO}), which are in turn used to calculate emission factors through Eqn. 2. The data presented here are from Plume P5 (Figs. 2 and 3). For each of the eight trace gases shown above, the emission ratio and the R² is given towards the top of each plot.

4.3 Emission factor inter-plume variability and comparison with bulk density

We found substantial inter-plume variability for EFs of a number of trace gas species (Table 2). In particular, we find significant variability (percentage difference between minimum and maximum EF > 130%) for the EFs of methane (5.35–26.19 g kg⁻¹); ammonia (3.08–16.13 g kg⁻¹); and hydrogen cyanide (0.34–6.24 g kg⁻¹). The majority of the other trace gas species exhibit substantial variability with the percentage difference between minimum and maximum EFs close to 100%. Only CO₂ shows less variability (1535–1662 g kg⁻¹, a percentage difference of 7.9%), while CO shows moderate variability (200–314 g kg⁻¹, a percentage difference of 44.3%). Plume P1 is particularly anomalous, with significantly

elevated EFs for methane (26.19 g kg⁻¹); ammonia (14.46 g kg⁻¹); ethylene (10.16 g kg⁻¹); and acetic acid (8.91 g kg⁻¹).

Many biomass burning studies use MCE to explain variability of non-CO₂ trace gas species EFs (e.g. *Meyer et al.* [2012]) [*Stockwell et al.*, 2016]. In most biomass fires (e.g. savanna, boreal forest) there is a mix of flaming and smouldering combustion, with the EFs of products of flaming combustion (e.g. CO₂) tending to correlate with MCE, and the products of smouldering combustion (e.g. CO) tending to anticorrelate with MCE [*Burling et al.*, 2011]. The plumes measured here, however, originate from smouldering combustion only, and we find no dependence of our EFs on MCE. This echoes the findings of *Stockwell et al.* [2016] who also find no dependence of their Indonesian peat fire EFs on MCE.

The three plumes with the highest EFs for methane (P1, S1, S2) all originated from sites with significantly higher bulk density of the burn substrate (a mean bulk density of 0.62 g cm⁻³, compared with a mean of 0.35 g cm⁻³ for our other sites). A comparison of substrate bulk densities with methane emission factors (Figure 5) shows a strong positive correlation between these variables ($R^2 = 0.61$, $p < 0.01$). Plume P1 is somewhat of an outlier with a particularly high EF for CH₄. We decided to re-run the regression analysis without this outlier; this results in a stronger relationship ($R^2 = 0.71$, $p < 0.01$). This is a novel finding that suggests an important influence of the physical properties of the burn site on the resultant emissions. We suggest that our results provide evidence to support the influence of bulk density on burn dynamics [*Rein*, 2013; *Wijedasa et al.*, 2016], whereby a higher bulk density maintains both a higher fire temperature and slower spread rate which when combined with a lack of oxygen (also as a result of high bulk density), will produce more emissions through non-flaming pyrolysis and glowing combustion (gasification) [*Rein*, 2013]. Methane is primarily a product of glowing combustion [*Yokelson et al.*, 1997] and also a product of pyrolysis [*Lobert and Warnatz*, 1993], and so we therefore propose a close link between peat fire methane emission factors and site bulk density (a function of both drainage-related degradation and artificial compaction). It is important to note that the bulk density at our measurement sites was two-to-six times higher than that typical for pristine peats [*Page et al.*, 2011] and we do not recommend extending the relationship we find here to emissions from fires in pristine peatlands. Further controlled experimentation is required to investigate the influence of lower bulk densities on fire emissions from peat fuels.

Both ammonia, an important reactive nitrogen species [*Benedict et al.*, 2017]; and hydrogen cyanide, a biomass burning tracer species [*Duflot et al.*, 2012] exhibit substantial inter-plume variability. It is known that smoke from wildfires contains enhanced concentrations of reactive nitrogen species such as ammonia [*Benedict et al.*, 2017], and the nitrogen content of fuels is known to influence nitrogen species emissions during a fire [*Burling et al.*, 2010; *Coggon et al.*, 2016; *Yokelson et al.*, 1996]. Further controlled experimentation is required to investigate whether fertilizer addition (i.e. as was evident at our Selangor site) alters peat chemical composition, and whether this in turn affects nitrogenous compound emissions during fires.

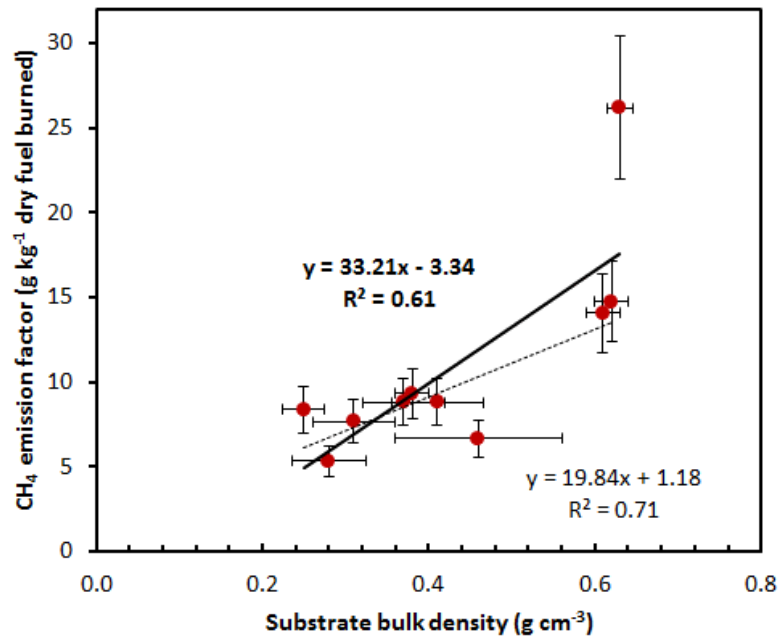


Figure 5. Scatter plot showing the relationship between peat substrate bulk density and the methane emission factor for each of our plumes. A linear regression line of best-fit is presented using all of the data points (solid black line) as well as a best-fit line that excludes the outlier with the highest CH₄ EF (Plume P1). Both lines of best-fit have a statistically significant non-zero slope ($p < 0.01$).

4.4 Representativeness and comparison of emission factors to previous studies

Here we present the first tropical peat fire emission factors for Peninsula Malaysia. All previous studies of tropical peat emission factors have been laboratory studies of Sumatran [Christian *et al.*, 2003] or Kalimantan [Stockwell *et al.*, 2014] peat, and only one other comprehensive *in situ* study of peat fires in Kalimantan [Stockwell *et al.*, 2016]. Hamada *et al.* [2013] and Huijnen *et al.* [2016] both provide information for a limited range of species (CO₂, CO, CH₄) as measured *in situ* at peat fires in Kalimantan. These emission factors are summarised in Table 3 for comparison.

As noted by two other *in situ* studies [Huijnen *et al.*, 2016; Stockwell *et al.*, 2016], we also found little evidence of surface fuel combustion at our Pahang sites; the surface grasses and shrubs remained unburned although heavily dried by the heat from the fire (see Figure 2). The peat fires at our Selangor sites had clearly been ignited by a surface palm slash fire, although the fire in the palm slash had long been extinguished upon our arrival on site, with just the smouldering peat remaining. We therefore consider the EFs presented here to be of “pure” peat smoke, in line with the other studies presented in Table 3. The relative contribution of emissions from combustion of the surface layer is small compared to the emissions from burning peat [Page *et al.*, 2002], and the complexities involved in the calculation of combined surface-peat EFs is discussed at length in Stockwell *et al.* [2016].

An important finding from the comparison of our EFs with those from previous studies (Table 3) is the difference between our EFs and those measured in the laboratory study of Christian *et al.* [2003]. The EFs for CO₂, CH₄, and CO from Christian *et al.* [2003] were subsequently adopted by IPCC greenhouse gas guidelines [Hiraishi *et al.*, 2014]. Stockwell *et*

al. [2016] suggest some significant adjustments to the values used by the IPCC, with a notable decrease in CO₂ (−8%) and CH₄ (−55%) EFs, and an increase to the CO (+39%) EF. Our findings also suggest that an adjustment is needed to the IPCC values. We also find a decrease is needed for CO₂ (−5%) and CH₄ (−46%), and an increase needed for CO (+22%). Both this study and *Stockwell et al.* [2016] find significantly smaller NH₃ EFs than *Christian et al.* [2003], finding a decrease of 60% and 86%, respectively. The magnitude of these adjustments are similar to those suggested by the only other comprehensive *in situ* study of tropical peatland emissions [*Stockwell et al.*, 2016], lending confidence to our assertion that field-measured values are more appropriate. We suggest that any future updates to the IPCC guidelines and/or EF databases (e.g. *Akagi et al.* [2011]) use a combination of the improved field-measured EFs presented here in Table 3 and those reported in other field studies, as well as any future results from field campaigns. We find similar EFs to *Stockwell et al.* [2016] for the majority of the other gas species in Table 3 (and thus similar differences from those used by the IPCC), with the exception of ethylene (C₂H₄), which we find to have an EF closer to that found by the previous laboratory studies. We present recommended inter-study averaged EFs for tropical peatland fires in Table 4.

Table 4. Recommended tropical peatland EFs for any future updates to *Akagi et al.* [2011] and/or the IPCC Greenhouse Gas Guidelines [*Hiraishi et al.*, 2014] (an asterisk indicates those species that are provided by the IPCC). The recommended EF is calculated from the mean of EFs from this study and the two other *in situ* studies [*Huijnen et al.*, 2016; *Stockwell et al.*, 2016]. A standard deviation is presented in parentheses where three studies are available (for CO₂, CO, CH₄), otherwise we report the range of values from the two available studies.

Trace Gas	EF (g kg ^{−1})
CO ₂ *	1589 (32)
CO*	259 (29)
CH ₄ *	9.44 (1.6)
C ₂ H ₂	0.09 (0.06–0.12)
C ₂ H ₄	1.63 (0.96–2.30)
C ₂ H ₆	1.84 (1.52–2.17)
H ₂ CO	0.82 (0.77–0.87)
HCOOH	0.22 (0.18–0.25)
CH ₃ OH	2.49 (2.14–2.83)
CH ₃ COOH	4.46 (3.89–5.02)
HCN	4.77 (3.79–5.75)
NH ₃	5.34 (2.86–7.82)

5 Summary and conclusions

We present results from open-path FTIR spectroscopy measurements of emission factors for tropical peatland fires from ten fire plumes in Peninsular Malaysia. These represent the first published emission factors for fires burning in Malaysian peatlands, and only the second comprehensive set of field measurements for the tropical peatland ecosystem. We find EFs of similar magnitude to those of *Stockwell et al.* [2016] (the only other comprehensive field study of tropical peatland fire EFs) and we thus echo their suggestion that future total peat fire emissions modelling uses field-measured EFs as a more reliable alternative to the earlier laboratory studies. Mean EFs calculated from our findings, along with those of *Stockwell et al.* [2016] and *Huijnen et al.* [2016] (CO₂, CO, and CH₄ only) are presented in Table 4. We recommend these as the best available ecosystem-specific emission factors for tropical peat

fires, as determined by field measurements. Field-derived emission factors for a further ~80 gas species and aerosol optical properties from Indonesian peatland fires can be found in *Stockwell et al.* [2016]

Further to previous studies of tropical peatland fires, we present the first evidence that may be used to explain the large inter-plume variability found by our study and others. We find substantial inter-plume variability in emission factors for methane, a potent greenhouse gas [Hiraishi *et al.*, 2014]. We find evidence, supported by a theoretical framework, which suggests much of this variability may be determined by the bulk density of the burn substrate (Figure 5). This relationship between burn substrate properties and fire emissions will need further testing through both controlled laboratory experiments and through better characterisation of fuels by *in situ* fire emissions field measurements.

Acknowledgments, Samples, and Data

Both the 2015 and 2016 campaigns were supported by a Royal Geographical Society Small Grant (£3,000) and by a King's College London Department of Geography Research Innovation Fund Grant (£2,000). The 2016 campaign was in-part supported by a Monash Large Strategic Grant. We would like to thank Christopher MacLellan of NERC FSF for his wide-ranging support, including the loan of the IR lamp used here. We are grateful for the much-needed field support provided by Dr Tajang Jingtut, Vicky Ting, and Christopher Roulston. We also extend thanks to Dr Bruce Main for technical and logistical support, to Dr James Millington for informing our statistical analysis; and to Dr Clare Paton-Walsh for her FTIR expertise and field support at the Selangor field sites.

The authors declare no financial interests. Raw mole fraction retrievals and emission ratios used to calculate emission factors may be found here: <https://doi.org/10.5281/zenodo.910770>

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