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- 1 In situ tropical peatland fire emission factors and their variability, as
- 2 determined by field measurements in Peninsula Malaysia.
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18 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.
- 25

26 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.

32 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 33 environment. During August 2015 (under El Niño conditions) and July 2016, we embarked 34 on field campaigns to measure gaseous emissions at multiple peatland fires burning on 35 deforested land in Southeast Pahang (2015) and oil palm plantations in North Selangor 36 (2016), Peninsula Malaysia. Gaseous emissions were measured using open-path Fourier 37 38 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 39 these measurements used to calculate EFs. Peat samples were taken at each burn site for 40 physicochemical analysis and to explore possible relationships between specific 41 physicochemical properties and fire EFs. Here we present the first evidence to indicate that 42 substrate bulk density affects methane fire EFs reported here. This novel explanation of inter-43 plume, within-biome variability should be considered by those undertaking greenhouse gas 44 accounting and haze forecasting in this region, and is of importance to peatland management, 45 particularly with respect to artificial compaction. 46

47 **1 Introduction**

Fires in tropical peatlands account for 25% or more of the estimated total greenhouse gas 48 (GHG) emissions from global deforestation and forest degradation, amounting to 49 50 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-51 related fires in Indonesia were calculated to have released 13-40% of that year's total carbon 52 emissions from fossil fuel burning [Page et al., 2002]. Beyond the global climate forcing of 53 GHGs such as CO₂, methane (CH₄), and nitrous oxide (N₂O), peatland fire emissions of 54 carbon monoxide (CO), nitrogen oxides (NO_x) and volatile organic compounds (VOCs) react 55 to form ozone (O₃). This, combined with particulate matter (PM) emissions, leads to smog 56 (referred to as 'haze' in SE Asia). In the southern region of SE Asia, peatland fires are 57 responsible for 90% of trans-boundary haze events [Varkkey, 2013]. Air Pollution Index 58 readings in Palangkaraya in Borneo in October 2015 reached >2000, the highest values ever 59 recorded and far above the Emergency level of >500. Exposure to haze events has been 60 shown to cause both immediate and delayed effects, increasing respiratory related mortality 61 by 19-66% [Sahani et al., 2014] and resulting in over 100,000 excess deaths in Malaysia, 62 Indonesia and Singapore [Koplitz et al., 2016]. In the 2015 fires, an estimated >500,000 63 Indonesians were affected by haze-related respiratory infections [Jakarta Post, 2015]. 64 Economic losses related to haze are also significant, with estimates for the 2015 fires at \$16.1 65 billion for Indonesia [CIFOR, 2015]. Despite these significant global and regional impacts, 66 very little is known of the proportional contribution of the major chemical compounds being 67 emitted *in situ* from tropical peat fires. As a result, there are no published accounts addressing 68 how GHG and reactive (haze-forming) gaseous fire emissions vary among different types of 69 tropical peatlands, precluding both accurate greenhouse gas accounting and air quality (haze) 70

71 forecasting in this region.

The Indo-Malaysian region is the global centre of tropical peatswamp forests. Over the past 72 30 years regional peatswamp forests have been increasingly subject to degradation from 73 drainage, logging and agricultural conversion - particularly to oil palms (Elaeis guineensis). 74 Drainage reverses the environmental conditions that lead to peat accretion, resulting in 75 oxidation, compaction and drying of peat making it highly flammable [Langner and Siegert, 76 2009; Wösten et al., 2006]. Fire is often used to clear peatlands [Varkkey, 2013] and these 77 78 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 79 fire [Posa et al., 2011] because of the un-burnt biomass, removal of canopy shading and fire 80 prone secondary vegetation that will act to provide further fuel and ignitions [Siegert et al., 81 2001]. Due to the extensive degree of land degradation in the region, peatland fires now 82 occur twice a year in the Peninsula Malaysia and Sumatra regions, during each dry season 83 84 [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]), 85 depending on human and climate factors. 86

Understanding within-biome emissions variability is an important challenge for assessing the 87 atmospheric impacts of tropical peatland fires; in a broader context, van Leeuwen and van der 88 Werf [2011] suggest that explaining emission factor (EF, grams of a compound emitted per 89 kg of dry biomass burned) variability remains one of the biggest challenges for biomass 90 burning emissions science. Despite this, our understanding of specific gaseous emissions 91 from peat burning is limited to a handful of studies where small peat samples were burned in 92 laboratories [Christian et al., 2003; Stockwell et al., 2014] and two field studies [Huijnen et 93 al., 2016; Stockwell et al., 2016]. With the exception of Stockwell et al. [2014], these studies 94 95 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, 96 prohibiting any explanation of the substantial variability between samples and/or studies. For 97 98 example Huijnen et al. [2016] report that 7.8 g of methane is emitted per kilogram of dry peat 99 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 100 101 the literature exist for other important emission factors (e.g. ammonia, a reactive nitrogen species). 102

103

104 2 Tropical peatland fire emission factors

Unlike logging, the open burning of biomass directly consumes carbon stocks and rapidly 105 releases emissions (e.g. GHGs, haze-forming species, PM) to the atmosphere 'on site'. How 106 well peat fire emissions are accounted for depends upon uncertainties in the calculation of (i) 107 The amount of peat consumed in the fire; and (ii) the emission factors used to convert the 108 estimate of fuel consumption into an estimate of the mass of gas released per unit of dry fuel 109 burned (g kg⁻¹) [*Penman et al.*, 2003]. Emission factors for CH₄ and important haze-forming 110 compounds vary massively between the few studies of gaseous emission factors [Christian et 111 al., 2003; Stockwell et al., 2014; Stockwell et al., 2016]. The small number of samples in 112 these studies, combined with limited information on physicochemical peat properties beyond 113 elemental analysis, prohibits an explanation of the large differences in emissions factors 114 between studies. 115

Previous studies of peat fire gaseous emissions factors have predominantly undertaken Fourier transform infrared (FTIR) spectroscopic measurements of emissions from peat samples burning in a laboratory. These studies have investigated fire emissions from samples
of temperate peat [*Stockwell et al.*, 2014; *Wilson et al.*, 2015; *Yokelson et al.*, 1997]; boreal
peat [*Stockwell et al.*, 2014; *Yokelson et al.*, 1997]; and include studies of samples of tropical
peat from Sumatra [*Christian et al.*, 2003] and Kalimantan [*Stockwell et al.*, 2015; *Stockwell et al.*, 2014].

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

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

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

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

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.

169

170 **3 Methodology**

171 3.1 Site and fire descriptions

172 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 173 Pahang, Malaysia in August 2015, and at oil palm plantations located about 20 km north of 174 175 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 176 occasions in August 2016 at three different sites between Pekan and Lanjut, within a region 177 178 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. 179 According to weather observations at Kuantan, mean annual rainfall is 2,900 mm. Peaks in 180 rainfall occur in December (564 mm) and May (202 mm). August sees a mean maximum 181 temperature of 32.3°C and mean rainfall of 174 mm. The peat soils at the Pahang sites, which 182 were located on the coastal edge of the main southeast Pahang peat dome (Figure 1), were 183 very shallow (0.2–0.8 m), typical of degraded peatlands subject to drainage, land clearance, 184 and hence oxidative loss of the peat layer over some decades. Two further sets of 185 measurements were made in July 2016 at one site in north Selangor. This area has a similar 186 climate to that of southeast Pahang. Weather observations at the nearest weather station in 187 188 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 189 located towards the edge of the main north Selangor dome, were deeper than those in Pahang, 190 191 extending to 1.5 to 2 m depth.

192 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 193 and ferns as well as shrubs (particularly Melastoma sp. and Mimosa sp.) and trees such as 194 Acacia mangium and Macaranga pruinosa. A.mangium is a widespread introduced species 195 which can promote fire intensity and frequency due to flammable leaf litter and which 196 produces enormous numbers of seeds that remain viable for years and whose germination is 197 enhanced by fire. *M. pruinosa* is a native, secondary gap colonizer commonly found on the 198 fringes of regional peatswamp forests. Oil palm plantations are usually cleared manually and 199 treated with herbicides to remove other vegetation, leaving only a sparse ground cover of 200 grasses and ferns. We found evidence of ongoing chemical fertiliser use at the Selangor oil 201 palm plantation site (Site 4 in Figure 1), with open sacks of NPK fertiliser and pellets around 202 the base of oil palms. 203

204 Locations of actively burning peatland fires were identified by regular monitoring of the World Resources Institute Global Watch Fires website 205 Forest (http://fires.globalforestwatch.org/) and NASA's Worldview website 206 (https://worldview.earthdata.nasa.gov/). These websites provide the near-real-time location of 207 208 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 209

- 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
- 214 of the fire was established. Fewer fires were observed across Peninsula Malaysia in July
- 215 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
- 217 peatland fires were found on oil palm plantations.
- 218 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 m²). 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.





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).

246

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.

					Temp	Pressure	Bulk density	Soil moist			
Plume	Site	Lat (°)	Long (°)	Path (m)	(°C)	(hPa)	(g cm ⁻³)	(%)	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

253

255 3.2 Fire emissions measurements

Here we use an Open-Path Fourier Transform Infrared (OP-FTIR) approach to estimate path-256 averaged mole fractions of twelve trace gases. The OP-FTIR system consists of a MIDAC 257 Corporation M2000 Series FTIR spectrometer equipped with a Stirling-cooled mercury-258 259 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 260 a 12 V silicon carbide glowbar operating at 1500 K fitted in front of a 20 cm diameter gold-261 plated collimator. The MIDAC system and its use to characterise biomass burning emissions 262 via long OP-FTIR spectroscopy is detailed in Smith et al. [2014] and Wooster et al. [2011]. 263

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 271 Multiple Atmospheric Layer Transmission (MALT) program [Griffith, 1996], where selected 272 spectral regions of the measured OP-FTIR spectra are fitted with synthetic spectra. A more 273 comprehensive description of MALT can be found in Griffith [1996] and more details of the 274 specific spectral regions used for the retrieval can be found in Paton-Walsh et al. [2014] and 275 276 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 277 CO₂, CH₄, and CO are accurate to within 5% of true mole fractions. *Stockwell et al.* [2016] 278 279 also found retrieved mole fractions of CO₂, CH₄, and CO₂ to have an uncertainty of 3–5%.



280

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.

284 3.3 Calculation of emission ratios and emission factors

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

286
$$\operatorname{ER}_{i/y} = \frac{\Delta[i]}{\Delta[y]} = \frac{[i] - [i]_{background}}{[y] - [y]_{background}}$$
(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 CO₂ and CO) *i* are calculated via:

294
$$\mathrm{EF}_{i} = \mathrm{ER}_{i/\mathrm{CO}} \times \frac{\mathrm{MM}_{i}}{\mathrm{MM}_{\mathrm{CO}}} \times \mathrm{EF}_{\mathrm{CO}}$$
 (2)

where $\text{ER}_{i/\text{CO}}$ 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 CO₂ 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. $[i] - [i]_{\text{background}}$ in Eqn. 1); emission factors may then be calculated using the carbon mass balance method [*Ward and Radke*, 1993]:

302
$$\mathrm{EF_{CO}} = (F_{\mathrm{C}} - F_{\mathrm{PMC}}) \times 1000 \times \frac{\mathrm{MM_{CO}}}{\mathrm{AM_{C}}} \times \frac{\Delta_{\mathrm{CO}}}{\sum_{i=1}^{n} (\mathrm{NC}_{i} \times \Delta_{i})}$$
(3)

303
$$\mathrm{EF}_{\mathrm{CO2}} = (F_{\mathrm{C}} - F_{\mathrm{PMC}}) \times 1000 \times \frac{\mathrm{MM}_{\mathrm{CO2}}}{\mathrm{AM}_{\mathrm{C}}} \times \frac{\Delta_{\mathrm{CO2}}}{\sum_{i=1}^{n} (\mathrm{NC}_{i} \times \Delta_{i})}$$
(4)

where $F_{\rm C}$ is the measured carbon mass fraction of the fuel (see Sect. 2.4); $F_{\rm PMC}$ 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*; $\Delta_{\rm CO}$ and Δ_i are the summed excess mole fractions of CO and species *i* respectively. The emission factor for CO₂ is calculated using the molecular mass of CO₂ (MM_{CO2} = 44.01 g mol⁻¹) and $\Delta_{\rm CO2}$ excess mole fractions of CO₂.

The fraction of carbon emitted as particulate matter (F_{PMC}) is usually considered to be 310 negligible and ignored in the application of carbon mass balance approaches for the 311 calculation of gaseous fire emission factors (e.g. Wooster et al. [2011]). However, a recent in 312 situ study of PM EFs for tropical (Indonesian) peatlands [Javarathene et al., 2017] suggests 313 that PM emissions are not negligible for tropical peatland fuels and should therefore be 314 315 accounted for in calculations of gaseous emission factors. Jayarathene et al. [2017] report an EF of 17.3 g kg⁻¹ for PM_{2.5} with the PM_{2.5} consisting of 73% carbon. We therefore assume 316 $F_{\rm PMC}$ of 0.0127 for Eqns. 3 and 4. EFs are directly proportional to the combined carbon mass 317 fraction in Eqns. 3 and 4, enabling easy correction of determined average EFs should this be 318 warranted by additional future PM measurements. 319

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 CO_2) and is given by *Hao and Ward* [1993] as:

325
$$MCE = \frac{\Delta[CO_2]}{\Delta[CO_2] + \Delta[CO]}$$
(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.

329 3.4 Peat composition

Four substrate samples were taken from the uppermost section of the underlying unburnt 330 331 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 10 332 cm diameter and 10 cm deep brass corer. Wet and dry mass were measured to calculate bulk 333 density and moisture content on dry basis (Table 1). Total carbon (C) and nitrogen (N) 334 content were also determined (Table 1). For this, peat sub-samples were first oven dried at 335 105°C for 48 hours and then ball milled for homogenisation using a Planetary Ball Mill 336 (Retsch-PM400, Castleford, UK). Analysis of C and N used 20 mg of material enclosed in a 337 tin capsule with measurements undertaken using a total element analyzer (Thermo Flash EA 338 1112, CE Instruments, Wigan, UK). 339

340

341 **4 Results and Discussion**

342 4.1 Peat substrate composition

Given that the Pahang sites had been heavily degraded, with only a thin layer of peat 343 remaining, the majority of the Pahang substrate samples were not pure 'pristine' peat, with 344 our samples visibly containing sand or clay indicating that the peat fires had burned down to 345 the interface with the mineral soil substrate at the Pahang sites (carbon contents ranging from 346 15.2–43.3%), typical of degraded peats on the fringes of peat domes [Miettinen et al., 2017]. 347 The samples taken at the Selangor site were clearly peat, the mean carbon content of these 348 samples (55.6%) is close to that found for Sumatran peat (57.9%), as reported by *Stockwell et* 349 al. [2014]. The nitrogen content of our samples varied significantly between 0.39% and 350 1.76%. Nitrogen content of pristine peats is known to vary from 1.0% to 4.4% [Andriesse, 351 1988]. The lower nitrogen content measured here (<1%) is probably a further indicator of 352 degradation as the main source of nutrients to the soil (peatswamp forest leaf litter) had been 353 removed. The mean nitrogen content at the Selangor palm oil sites (1.34%) is higher than that 354 for the abandoned Pahang sites (0.96%) and we speculate that this is due to the use of 355 356 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, EFs are directly proportional to the 362 carbon mass fraction, enabling easy correction of EFs should this be warranted by additional363 future carbon mass fraction measurements.

All of our sites exhibited high bulk density for tropical peat, with bulk densities ranging from 364 0.28 to 0.63 g cm⁻³; this compares with 0.09–0.11 g cm⁻³ that might be expected for pristine 365 tropical peat [Page et al., 2011; Warren et al., 2012]. Drainage causes loss of water from peat 366 pores and shrinkage through drying, resulting in an increase in bulk density which is 367 exacerbated by the resulting aerobic conditions causing peat degradation due to oxidation, 368 increased microbial decomposition and compaction [Könönen et al., 2015]. Oil palm 369 plantation management with addition of lime and fertilizers further enhances microbial peat 370 breakdown, while artificial mechanical compaction (designed to increase root stability and 371 372 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 373 plantations (0.62 g cm⁻³) was significantly higher than the mean bulk density at our 374 deforested but abandoned Pahang sites (0.39 g cm⁻³), probably due to extensive use of heavy 375 machinery and purposeful artificial compaction of the peat soil at the Selangor sites. 376

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 383 384 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 385 summary of our study-averaged EFs and one standard deviation of the means for all species. 386 From measurements of ten tropical peat fire plumes, we find the major trace gas emissions by 387 mass (EF > 0.5 g kg⁻¹, as defined by *Stockwell et al.* [2016] to be: carbon dioxide (1579 \pm 388 58); carbon monoxide (251 ± 39); methane (11.00 ± 6.11); ammonia (7.82 ± 4.37): acetic 389 acid (5.02 \pm 1.64); hydrogen cyanide (3.79 \pm 1.97); methanol (2.83 \pm 0.84); ethylene (2.30 \pm 390 2.79); ethane (2.17 \pm 0.81); and formaldehyde (0.77 \pm 0.64). We report EFs for two further 391 species that may be categorised as minor trace gas emissions by mass: formic acid (0.25 \pm 392 0.04); and acetylene (0.06 \pm 0.01). The modified combustion efficiency (MCE) of the fire 393 burning to produce the plumes sampled here ranged from 0.774 to 0.839 (Table 2), with an 394 average of 0.800 ± 0.031 indicating pure smouldering combustion. 395

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

PLUME	P1	P2	P3	P4	P5	P6	P7	P8	S1	S2
SITE	1	1	2	2	3	2	3	2	4	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
СО	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_2H_2	0.06 ± 0.01	nr	0.05 ± 0.01	nr	nr	nr	nr	nr	nr	nr
C_2H_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_2H_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
нсоон	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	$6.24{\pm}1.00$	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



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411 **Table 3**. Emission factors (g kg⁻¹ of dry fuel burned) reported by this study of Malaysian 412 peatlands and those for the same trace gases reported by previous *in situ* Indonesian peatland 413 studies [*Huijnen et al.*, 2016; *Stockwell et al.*, 2016] and laboratory studies [*Christian et al.*, 414 2003; *Stockwell et al.*, 2014]. The mean and standard deviation (in parentheses) are calculated 415 from individual plumes (for *in situ* studies) or samples (for laboratory studies).

		Emissi	on Factor (g kg ⁻¹ dry fue	burned)	
Trace Gas	Malaysian peat (<i>in situ</i>) (this study)	Kalimantan peat (<i>in situ</i>)	Kalimantan peat (<i>in situ</i>) (Hujinen et al. 2016)	Sumatran peat (lab) (Christian et al. 2003)	Kalimantan peat (lab) (Stockwell et al. 2014)
MCE		0.772 (0.052)	(Hulfhen et al. 2010)	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_2H_2	0.06 (0.01)	0.121 (0.066)	-	0.06	0.18 (0.05)
C_2H_4	2.30 (2.79)	0.961 (0.528)	-	2.57	1.39 (0.62)
C_2H_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.



Figure 4. Example scatter plots of the measured trace gas pathlength-averaged mole fractions used to calculate emission ratios ($\text{ER}_{i/\text{CO}}$), 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 R^2 is given towards the top of each plot.

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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 434 2). In particular, we find significant variability (percentage difference between minimum and 435 maximum EF > 130%) for the EFs of methane (5.35–26.19 g kg⁻¹); ammonia (3.08–16.13 g 436 kg^{-1} ; and hydrogen cyanide (0.34–6.24 g kg^{-1}). The majority of the other trace gas species 437 exhibit substantial variability with the percentage difference between minimum and 438 maximum EFs close to 100%. Only CO₂ shows less variability (1535–1662 g kg⁻¹, a 439 percentage difference of 7.9%), while CO shows moderate variability (200–314 g kg⁻¹, a 440 percentage difference of 44.3%). Plume P1 is particularly anomalous, with significantly 441

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 444 EFs (e.g. Meyer et al. [2012]) [Stockwell et al., 2016]. In most biomass fires (e.g. savanna, 445 446 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 447 of smouldering combustion (e.g. CO) tending to anticorrelate with MCE [Burling et al., 448 2011]. The plumes measured here, however, originate from smouldering combustion only, 449 and we find no dependence of our EFs on MCE. This echoes the findings of Stockwell et al. 450 [2016] who also find no dependence of their Indonesian peat fire EFs on MCE. 451

The three plumes with the highest EFs for methane (P1, S1, S2) all originated from sites with 452 significantly higher bulk density of the burn substrate (a mean bulk density of 0.62 g cm⁻³, 453 compared with a mean of 0.35 g cm^{-3} for our other sites). A comparison of substrate bulk 454 densities with methane emission factors (Figure 5) shows a strong positive correlation 455 between these variables ($R^2 = 0.61$, p<0.01). Plume P1 is somewhat of an outlier with a 456 particularly high EF for CH₄. We decided to re-run the regression analysis without this 457 outlier; this results in a stronger relationship ($R^2 = 0.71$, p<0.01). This is a novel finding that 458 459 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 460 461 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 462 lack of oxygen (also as a result of high bulk density), will produce more emissions through 463 non-flaming pyrolysis and glowing combustion (gasification) [*Rein*, 2013]. Methane is 464 primarily a product of glowing combustion [Yokelson et al., 1997] and also a product of 465 pyrolysis [Lobert and Warnatz, 1993], and so we therefore propose a close link between peat 466 fire methane emission factors and site bulk density (a function of both drainage-related 467 degradation and artificial compaction). It is important to note that the bulk density at our 468 measurement sites was two-to-six times higher than that typical for pristine peats [Page et al., 469 2011] and we do not recommend extending the relationship we find here to emissions from 470 fires in pristine peatlands. Further controlled experimentation is required to investigate the 471 472 influence of lower bulk densities on fire emissions from peat fuels.

Both ammonia, an important reactive nitrogen species [Benedict et al., 2017]; and hydrogen 473 cyanide, a biomass burning tracer species [Duflot et al., 2012] exhibit substantial inter-plume 474 variability. It is known that smoke from wildfires contains enhanced concentrations of 475 reactive nitrogen species such as ammonia [Benedict et al., 2017], and the nitrogen content of 476 477 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 478 479 investigate whether fertilizer addition (i.e. as was evident at our Selangor site) alters peat chemical composition, and whether this in turn affects nitrogeneous compound emissions 480 during fires. 481



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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 CH_4 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.

496 As noted by two other in situ studies [Huijnen et al., 2016; Stockwell et al., 2016], we also 497 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). 498 499 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 500 just the smouldering peat remaining. We therefore consider the EFs presented here to be of 501 "pure" peat smoke, in line with the other studies presented in Table 3. The relative 502 contribution of emissions from combustion of the surface layer is small compared to the 503 emissions from burning peat [Page et al., 2002], and the complexities involved in the 504 505 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 510 notable decrease in CO_2 (-8%) and CH_4 (-55%) EFs, and an increase to the CO (+39%) EF. 511 Our findings also suggest that an adjustment is needed to the IPCC values. We also find a 512 decrease is needed for CO_2 (-5%) and CH_4 (-46%), and an increase needed for CO (+22%). 513 Both this study and Stockwell et al. [2016] find significantly smaller NH₃ EFs than Christian 514 et al. [2003], finding a decrease of 60% and 86%, respectively. The magnitude of these 515 516 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 517 field-measured values are more appropriate. We suggest that any future updates to the IPCC 518 guidelines and/or EF databases (e.g. Akagi et al. [2011]) use a combination of the improved 519 field-measured EFs presented here in Table 3 and those reported in other field studies, as well 520 as any future results from field campaigns. We find similar EFs to Stockwell et al. [2016] for 521 522 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 (C_2H_4) , which we find to have an EF closer to 523 that found by the previous laboratory studies. We present recommended inter-study averaged 524 EFs for tropical peatland fires in Table 4. 525

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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_2H_2	0.09 (0.06-0.12)
C_2H_4	1.63 (0.96-2.30)
C_2H_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)

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535 **5 Summary and conclusions**

We present results from open-path FTIR spectroscopy measurements of emission factors for 536 tropical peatland fires from ten fire plumes in Peninsular Malaysia. These represent the first 537 published emission factors for fires burning in Malaysian peatlands, and only the second 538 comprehensive set of field measurements for the tropical peatland ecosystem. We find EFs of 539 similar magnitude to those of Stockwell et al. [2016] (the only other comprehensive field 540 study of tropical peatland fire EFs) and we thus echo their suggestion that future total peat 541 fire emissions modelling uses field-measured EFs as a more reliable alternative to the earlier 542 laboratory studies. Mean EFs calculated from our findings, along with those of Stockwell et 543 al. [2016] and Huijnen et al. [2016] (CO₂, CO, and CH₄ only) are presented in Table 4. We 544 recommend these as the best available ecosystem-specific emission factors for tropical peat 545

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

Further to previous studies of tropical peatland fires, we present the first evidence that may be 549 550 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 551 [Hiraishi et al., 2014]. We find evidence, supported by a theoretical framework, which 552 suggests much of this variability may be determined by the bulk density of the burn substrate 553 (Figure 5). This relationship between burn substrate properties and fire emissions will need 554 further testing through both controlled laboratory experiments and through better 555 characterisation of fuels by in situ fire emissions field measurements. 556

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

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