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# Article (Published version) (Refereed)

**Original citation:** 

Hu, Yuqi and Fernandez-Anez, Nieves and Smith, T. E. L. and Rein, Guillermo (2018) *Review of emissions from smouldering peat fires and their contribution to regional haze episodes*. International Journal of Wildland Fire, 27. pp. 293-312. ISSN 1049-8001 DOI: <u>10.1071/WF17084</u>

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International Journal of Wildland Fire **2018**, 27, 293–312 https://doi.org/10.1071/WF17084

### Review of emissions from smouldering peat fires and their contribution to regional haze episodes

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**Abstract.** Smouldering peat fires, the largest fires on Earth in terms of fuel consumption, are reported in six continents and are responsible for regional haze episodes. Haze is the large-scale accumulation of smoke at low altitudes in the atmosphere. It decreases air quality, disrupts transportation and causes health emergencies. Research on peat emissions and haze is modest at best and many key aspects remain poorly understood. Here, we compile an up-to-date inter-study of peat fire emission factors (EFs) found in the literature both from laboratory and from field studies. Tropical peat fires yield larger EFs for the prominent organic compounds than boreal and temperate peat fires, possibly due to the higher fuel carbon content (56.0 vs 44.2%). In contrast, tropical peat fires present slightly lower EFs for particulate matter with diameter  $\leq 2.5 \ \mu m (PM_{2.5})$  for unknown reasons but are probably related to combustion dynamics. An analysis of the modified combustion efficiency, a parameter widely used for determining the combustion regime of wildfires, shows it is partially misunderstood and highly sensitive to unknown field variables. This is the first review of the literature on smouldering peat emissions. Our integration of the existing literature allows the identification of existing gaps in knowledge and is expected to accelerate progress towards mitigation strategies.

Additional keywords: emission factor, modified combustion efficiency, smoke, wildfires.

Received 20 May 2017, accepted 20 March 2018, published online 22 May 2018

#### Introduction

Smouldering peat fires propagate horizontally and vertically through deep layers, leading to the largest fires on Earth (in terms of mass of fuel consumed per unit surface) (Rein 2013; Huang and Rein 2017). They are frequently reported in tropical, temperate and boreal regions including North America, Siberia, the British Isles, the sub-Arctic, south-east Asia and southern Africa (for example, Botswana in 2000, Scotland in 2006, the USA in 2004, central Russia in 2010 and Indonesia in 2015) (Rein 2013). Peatlands are an important terrestrial carbon pool, storing one-third of the world's soil carbon (500–600 Gt C (1 Gt =  $1 \times 10^9$  tonnes)), as much carbon as surface vegetation globally (IPCC 2006; Turetsky *et al.* 2014). Carbon emissions derived from peat fires are equivalent to 15% of anthropogenic emissions, creating large perturbations to the global carbon cycle (Poulter *et al.* 2006).

Peat is a carbon-rich organic soil that contains at least 12% organic carbon ( $\sim 20\%$  organic material) with less than 20–35% mineral content (Couwenberg 2009; Turetsky *et al.* 2014). It is composed of partially decomposed organic material (mostly from plants) accumulated in waterlogged,

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anaerobic, acidic and nutrient-deficient conditions, over centuries to millennia (Turetsky et al. 2014; IPCC 2006). Peatlands are areas with a naturally accumulated peat layer at the surface (peat thickness at least 20 cm if drained, 30 cm if undrained), and are the most widespread of all wetland types in the world, covering over  $4\,000\,000 \text{ km}^2$  (~3% of the Earth's land surface) (World Energy Council 2013; Turetsky et al. 2014). Peatlands are mainly distributed at northern high latitudes, while tropical peatlands comprise  $\sim 11\%$  of global peatland areas (mainly in south-east Asia) (Page et al. 2011). A total of 85% of the global peatland areas are in four countries only: Russia, Canada, the USA and Indonesia (World Energy Council 2013). A full description of the global and regional distribution of peatlands can be found in World Energy Council (2013). In addition to peatland area, Page et al. (2011) provided the best estimates of mean peat thickness in tropical countries (varying from 1.3 to 11 m) and adopted a default best estimate of 0.5 m peat thickness for most other countries in calculating global peat volumes. It is estimated that global peat volume is 7093  $\text{Gm}^3$  (1 $\text{Gm}^3 = 1 \times 10^9 \text{ m}^3$ ) (Page et al. 2011).

Review



Fig. 1. Schematics of a 3-day-long peat forest flaming fire (left), and the subsequent 3-month-long smouldering peatland fire (right).

Because peat accumulation only takes place in watersaturated environments, the bulk of peat in pristine peatlands usually has a very high moisture content and is thus naturally protected from burning. However, natural droughts (e.g. El Niño) or human activities (anthropogenic drainage, peat harvesting) lower the watertable in peatlands (Turetsky *et al.* 2014) and reduce the moisture content of the peat profile (Kettridge *et al.* 2015), rendering them susceptible to smouldering fires – the slow, low-temperature, flameless burning of porous fuels and the most persistent type of combustion phenomena (Rein 2015). Smouldering peat fires emit substantial quantities of aerosols that lead to severe regional haze episodes. However, information regarding emissions and health effects of regional haze episodes from smouldering peat fires is limited to a handful of studies.

The present paper is a review of the emissions from smouldering peat fires and their contribution to regional haze episodes for the first time. There are three main objectives: (1) to provide an overview of past haze episodes evolved from peat fires and to summarise the potential health effects of haze; (2) to conduct an up-to-date and comprehensive compilation of pollutant emission factors produced from peat fires; (3) to review the origins and to verify the applicability of modified combustion efficiency, a parameter widely used to evaluate the combustion regime of peat fire. This analysis will allow identification of knowledge gaps and move us forward in developing mitigation strategies.

#### Overview of past haze episodes

This section reviews regional haze episodes that have resulted from smouldering peat fires. Notorious haze episodes are frequently reported, especially when extended droughts intensify the vulnerability of peatlands to fire (Page *et al.* 2002; Heil 2007; Rein 2013; Turetsky *et al.* 2014). These fires generate smoke plumes that remain close to the ground owing to their weak buoyancy. The whitish colour of the haze indicates a predominant content of organic carbon particles, which is characteristic for haze aerosols (Heil 2007; Black *et al.* 2016). Dispersal of haze is slow, with the smoke plume tending to accumulate near the ground, driven by wind. It can migrate long distances into populated areas, causing regional haze for long periods of time (days or weeks) (Rein 2013). In contrast, flaming wildfires generate a strong fire plume of black or dark grey colour with a lower concentration of organic carbon particles but a higher concentration of soot than smouldering peat fires (Akagi *et al.* 2011; Drysdale 2011). A comparison between the plumes of the two types of fire is presented in Fig. 1.

Since 1991, the haze that results from smouldering peat fires has been a particularly acute and recurrent problem in south-east Asia, especially in 1997, 2006, 2009, 2013 and 2015 (Rein 2013; Koplitz et al. 2016). Because the main source of the south-east Asia haze is ascribed to peat fires occurring on the islands of Sumatra and Borneo, Indonesia has been the target of criticism from affected countries in the Association of South-East Asian Nations (ASEAN) region (Heil and Goldammer 2001; Glover and Jessup 2006; Heil 2007). In addition to south-east Asia, the USA and Russia are among other areas suffering from haze evolving from peat fires. For example, in June 2008, large-scale burning of peat deposits occurred in the eastern plains of North Carolina, and the emitted air pollutants caused a haze episode in this region (Black et al. 2016). During the summer of 2010, large urban regions in Russia were blanketed in smoky haze from severe peat fires raging outside Moscow, threatening a total population exceeding 15 million (Konovalov et al. 2011).

The effects of haze are diverse. Local regional air quality can seriously deteriorate. In addition, haze leads to strong and persistent impairment in visibility, which can severely affect local transportation, construction, tourism and agriculture-based industries (Heil and Goldammer 2001). An example of wide-spread disruption due to peatland fires is the unprecedented peatland mega-fire that occurred on the islands of Indonesia during the 1997 El Niño event, resulting in the transboundary 1997–98 south-east Asia haze episode (Fig. 2) (Page *et al.* 2002;



**Fig. 2.** Aerosol imaging by NASA satellite in October 1997 shows the vast smoke haze released by smouldering peat fires in Indonesia.

Rein 2013). This event led to severe air pollution with more than  $2000 \,\mu g \, m^{-3}$  total suspended particulates (TSP, particles with an aerodynamic diameter ≤100 µm) detected in Kalimantan and Sumatra (Heil 2007). An airplane crash with 234 deaths in northern Sumatra and ship collisions in the Strait of Malacca were partly attributed to the impaired visibility at that time. This haze affected 100 million people across five south-east Asian countries, with total damages estimated to be US\$4.5 billion (Heil and Goldammer 2001). During the 2010 Russian haze event, the Moscow region suffered from extremely high levels of daily mean carbon monoxide (CO) and coarse particle ( $PM_{10}$ , particles with an aerodynamic diameter  $\leq 10 \ \mu m$ ) concentrations (estimated up to  $10 \text{ mg m}^{-3}$  and  $700 \text{ }\mu\text{g m}^{-3}$  respectively) (Konovalov et al. 2011). The hourly Singapore fine particle (PM<sub>2.5</sub>, particle with an aerodynamic diameter  $\leq$ 2.5 µm) concentration during the acute 2013 south-east Asian haze conditions soared  $\sim 22$  times above background levels, degrading local air quality from a 'Good' to 'Hazardous' level. In the fall (autumn) of 2015, widespread peatland fires burned once again over large parts of equatorial Asia and induced the worst haze episode since 1997 (Huijnen et al. 2016; Koplitz et al. 2016).

More importantly, haze exposes populations to high concentrations of various pollutants. However, little is known about the health effects associated with haze emanating from smouldering peat fire emissions (Hinwood and Rodriguez 2005; Rein 2015). Generally, exposure to pollutants during haze events results in various deleterious physiological responses, predominantly to the respiratory and cardiovascular systems (World Health Organisation 2006). Specifically, haze-related diseases include conjunctivitis, acute upper respiratory tract infection, allergic rhinitis, asthma, bronchitis, eczema, pneumonia, emphysema, acute myocardial infarct and other ischaemic heart diseases (Kunii et al. 2002; Sastry 2002; Glover and Jessup 2006; Rein 2013; Durán et al. 2014; Ho et al. 2014). Individual adverse health outcomes are influenced by the composition and the amount of pollutants, the exposure duration, and personal health status (age, physique) (Hinwood and Rodriguez 2005). The health effects of some individual components found in haze are briefly discussed below.

CO exposure leads to health effects that range from tensiontype headaches and dizziness symptomatic of mild poisoning to unconsciousness or death after prolonged exposure at high concentrations. Also, trace gas species such as sulfur dioxide (SO<sub>2</sub>) emitted from peat fires are associated with pulmonary dysfunction and respiratory symptoms (Hinwood and Rodriguez 2005; World Health Organisation 2006).

Particulate matter (PM) exposure can cause cardiovascular disease and may aggravate arrhythmias; PM can affect the central nervous system and the reproductive system, and can induce cancer (World Health Organisation 2006; Durán et al. 2014).  $PM_{10}$  can be inhaled into the human respiratory tract, and the deposition of these particles in the lungs can induce a systemic inflammatory response (Hinwood and Rodriguez 2005; Glover and Jessup 2006; Heil 2007). PM<sub>2.5</sub> from peat fires can penetrate into lower respiratory tracts and exacerbate respiratory diseases including chronic bronchitis, emphysema and asthma (World Health Organisation 2006; Durán et al. 2014). Epidemiologic studies have shown a correlation between PM2.5 uptake and an increase of morbidity, as well as an increase in death rates related to respiratory complications, heart problems and lung cancer (Dockery et al. 1993; Lighty et al. 2000; Kunii et al. 2002; Sastry 2002; Hinwood and Rodriguez 2005; Heil 2007; Jayachandran 2009; Durán et al. 2014). Ultrafine particles (UFP, particles with an aerodynamic diameter  $\leq 0.1 \,\mu\text{m}$ ) can deposit deep in the lungs by diffusion and can enter the blood through the alveoli of the lung (Lighty et al. 2000). Recently, a cardiopulmonary toxicity study showed that PM10 from smouldering peat fires induced larger lung inflammatory responses, whereas UFP caused significant adverse cardiac effects (Kim et al. 2014).

Metals (Fe, Cu, Ni) that are enriched in PM can catalyse the formation of reactive oxygen species, which are implicated via cellular activity to a variety of inflammatory responses including cardiovascular diseases (Lighty *et al.* 2000). Additionally, peat fires release mercury (Hg) into the atmosphere at a rate 15 times greater than upland forest fires, creating serious human health concerns (Durán *et al.* 2014; Turetsky *et al.* 2014). The inhalation of mercury can induce harmful effects on the nervous, digestive and immune systems, lungs and kidneys, with increased risks of cardiovascular disease and severe neurological damage to humans (Durán *et al.* 2014).

Volatile organic compounds (VOCs) are by-products of the incomplete combustion of smouldering peat that can pose great health threats to people suffering from exposure (Heil 2007; Durán et al. 2014). VOCs include a wide range of chemical substances such as hydrocarbons (e.g. methane, benzene and toluene), halocarbons and oxygenates. The prolonged exposure to typical hydrocarbons like benzene may increase susceptibility to leukaemia. Among the VOCs, one of the few classes established for epidemiological purposes are the polycyclic aromatic hydrocarbons (PAHs) (e.g. fluorene, phenanthrene, pyrene). Substances identified in haze like xylene and PAHs are known or suspected carcinogens, mutagens and teratogens, with the potential to cause serious long-term effects (Muraleedharan et al. 2000a). Exposure to PAHs has also been linked with cardiovascular disease and poor foetal development (Kunii et al. 2002).

#### Epidemiological evidence of haze effects

There are a few epidemiological studies of haze from peat fire events in Indonesia, Malaysia, Singapore and the USA (Kunii et al. 2002; Sastry 2002; Glover and Jessup 2006; Jayachandran 2009; Rappold et al. 2011; Ho et al. 2014). The 1997 south-east Asian transboundary haze event caused acute health risk to 105 million people, mainly in Indonesia, Singapore, Malaysia, Brunei and Thailand. The number of acute respiratory infection cases increased by 3.8 times in south Sumatra from 1996 to 1997 (Hinwood and Rodriguez 2005). A total of 16 400 Indonesian infant and foetal deaths were attributable to this haze pollution (Jayachandran 2009). Kunii et al. (2002) interviewed 543 persons in terms of their response to haze and conducted lungfunction tests. More than 90% of the respondents had respiratory symptoms, and elderly individuals suffered a serious deterioration of their overall health. According to their statistics, there were 527 haze-related deaths, 298 125 cases of asthma, 58 095 cases of bronchitis, and 1446120 cases of acute respiratory infection reported in the country of Indonesia for this haze episode alone (Kunii et al. 2002). Compared with 1995, the attendance at hospital accident and emergency (A and E) in Singapore in the 1997 haze period increased by 5588 cases for asthma, bronchitis, emphysema and pneumonia (Glover and Jessup 2006). In Malaysia, the haze of 1997 increased total allcause mortality by 22%, and non-traumatic mortality among the population aged 65-74 increased by 72% (Sastry 2002). Overall, the number of additional premature deaths in adults related to this haze exposure was estimated to be between 22200 and 53 700 (total mortality among the adult population increased by ~3%) (Heil 2007).

A study of the 2013 south-east Asia haze showed that the most common physical symptoms during the crisis were mouth, throat and nose discomfort, eye discomfort, headache and breathing difficulty (Ho *et al.* 2014). More than 28 000 000 people in Indonesia were affected by the 2015 south-east Asian haze, of which more than 140 000 residents reported respiratory illness. It is estimated that this recent haze episode resulted in 91 600 excess deaths across Indonesia, 6500 in Malaysia and 2200 in Singapore (Koplitz *et al.* 2016).

Russia and the USA are also among countries suffering from haze. There were close to 11000 excess deaths from nonaccidental causes during the 2010 Russian peat fire event; the most pronounced effects were cardiovascular, respiratory, genitourinary and nervous system diseases or deaths (Shaposhnikov *et al.* 2014). During the 2008 North Carolina severe peat fire event (Black *et al.* 2016), asthma-related visits accounted for 44% of all respiratory events (2081 cases) considered, and heart failure accounted for 33% of all cardiac events (1817 cases) (Rappold *et al.* 2011).

#### **Smouldering peat fires**

This section summarises the characteristics of smouldering peat fires and their feedback on local ecosystems and climate systems. In physical terms, peat is a porous and charring natural fuel that consists of a permeable medium. When above the watertable and available to burn, this aggregate nature provides a large surface area per unit volume, which facilitates the heterogeneous reaction with oxygen, and permits in-depth transport of oxygen through the fuel bed, which is thus prone to smouldering (Rein 2013, 2015; Rein et al. 2008). Smouldering combustion is sustained by the heat released when oxygen directly attacks the surface of a solid fuel, whereas flaming combustion dominates when the oxidation takes place in the gas phase (Rein 2015). Owing to the difference in combustion regimes, the temperature and spread rate characteristics in smouldering combustion are low compared with flaming combustion (Rein 2013, 2015). Typical peak temperatures for smouldering are in the range from 450 to 700°C, whereas the typical temperature for flaming is ~1500°C (Rein 2015; Huang et al. 2016). Additionally, smouldering spreads in a creeping fashion, typically  $\sim 1 \text{ mm min}^{-1}$ , which is two orders of magnitude slower than flame spread (Rein 2015). Despite the large differences between smouldering and flaming combustion, they are closely related, and one can lead to the other (Rein 2015). Transition from flaming to smouldering can occur as residual smouldering combustion, i.e. persistent smouldering of thick fuels and biomass accumulated below ground that can be observed for days after a flaming wildfire has passed by (Bertschi et al. 2003; Urbanski 2014). The reverse, the transition from smouldering to flaming, can happen under enhanced oxygen supply (e.g. strong winds) (Rein 2013).

In the presence of a large quantity of fuel, a smouldering fire can be initiated with a weak ignition source and sustained for very long periods (months, years or even decades) despite firefighting attempts, extensive rains or weather changes. For these two reasons - easy ignition and difficult suppression smouldering fires present the most persistent combustion phenomena on Earth (Rein 2013). Moisture content is the most important property governing the ignition and the spread of smouldering peat fires (Rein 2015; Archibald et al. 2017), while oxygen supply and heat transfer are the two mechanisms controlling its spread rate (Huang and Rein 2015; Rein 2015). Real smouldering fires are a multidimensional phenomenon, which includes two main components: in-depth vertical and surface lateral spread (Rein 2013; Huang and Rein 2017). Complex behaviours like the 'overhang' phenomenon, where smouldering spreads fastest beneath the free surface, is formed by the spread rate difference between the top and lower peat layers as well as the competition between oxygen supply and heat losses (Fig. 3) (Huang et al. 2016).

Smouldering peat fires result in the widespread destruction of ecosystems, bringing fatal damage to the forest soil, its microflora and micro-fauna (Rein et al. 2008). The long residence time of smouldering means that heat can penetrate deep into the soil layers and largely consume the fuel bed (>90% mass loss) (Rein 2013). In contrast, flaming combustion is a surface phenomenon that produces high temperatures above the ground for short periods of time (in the order of 15 min), thus leading to perhaps less severe impacts on the soil and its micro-fauna and flora (Doerr and Cerdà 2005; Rein 2013). Thermal conditions in smouldering peat are more severe than medical sterilisation treatments, and mean that the soil is exposed to conditions that are lethal to biological agents. Furthermore, peat fires can burn down to the watertable, exposing tree roots and leading to forest vegetation instability. This long-term impact of peat fires on local vegetation (e.g. changes in flora species, incomplete vegetation recovery) has been observed in boreal, temperate



**Fig. 3.** Schematic diagram of smouldering spread laterally along the surface and vertically in depth with an overhang and depth of burn (Huang *et al.* 2016).

and tropical peatland settings (Maltby *et al.* 1990; Hoscilo *et al.* 2011; Kettridge *et al.* 2015).

Smouldering fires in peatlands involve the burning of ancient (up to >10 000 years) carbon (Rein 2013; Archibald et al. 2017). The sheer volume of carbon being emitted from peat fires contributes greatly to greenhouse gas (GHG) emissions to the atmosphere, accelerating climate change. Warmer climates at a global scale will have the most pronounced effect on peatlands that have been impacted by anthropogenic activities (i.e. where hydrology, vegetation cover, etc., have been modified). The warmer climate will also lead to more frequent and more extensive smouldering fires from areas where warmer and drier peatlands are induced (Rein 2013). This is because smouldering ignition and self-heating are known to have a strong dependence on peat moisture and temperature (Rein et al. 2008; Drysdale 2011; Huang and Rein 2015, 2017; Huang et al. 2016). Drying as a result of climate change and human activities has raised questions regarding the vulnerability of peatlands to fire (Rein 2013; Turetsky et al. 2014). In addition, more frequent flaming wildfires under warmer climates will lead to more frequent residual smouldering peat events (Flannigan et al. 2009). As a result, peat fires form a positive feedback within the climate system, a self-accelerating process (Rein 2013).

#### **Pollutant emissions**

Peat fire emissions are a rich and complex mixture of gases (more than 100 detectable gas species) and aerosols (Stockwell et al. 2014, 2015, 2016; Hatch et al. 2015). The trace gas species detected during peat fire events mainly comprise CO<sub>2</sub>, CO, CH<sub>4</sub>, HCN, NH<sub>3</sub> and a series of alkanes (ethane, propane and butane), alkenes (e.g. ethylene, propylene), aldehydes (formaldehyde, acetaldehyde, glycolaldehyde) and other organic compounds (methanol, acetic acid, furan, etc.) (Yokelson et al. 1997; Christian et al. 2003; Wilson et al. 2015; Stockwell et al. 2014). The sum of CO<sub>2</sub> and CO comprises the largest mass or mole fraction of emissions from peat fires (alongside water vapour). A detailed characterisation of the emission species can be found in Hatch et al. (2015) and Stockwell et al. (2015). Among all the gaseous species, CO<sub>2</sub> and CH<sub>4</sub> are significant sources of GHGs, while trace gases including CO, NH3 and the other non-methane organic compounds (NMOCs) play a main role in radiative forcing through their photochemical processing. These trace gases can impact atmospheric levels of CO<sub>2</sub>, CH<sub>4</sub>, tropospheric O<sub>3</sub>, and PM (Urbanski 2014). PM derived from peat fires varies greatly in size, ranging from nanoscale ultrafine fraction UFP to microscale PM<sub>1</sub> (particles with an aerodynamic diameter  $\leq 1.0 \ \mu$ m), PM<sub>2.5</sub>, PM<sub>10</sub>, and TSP.

Although peat fires contribute a large fraction of atmospheric pollutant emissions, they have not been explicitly considered in most of the previous studies of biomass combustion emissions (IPCC 2006; Yokelson *et al.* 2008; McMeeking *et al.* 2009; Wilson *et al.* 2015). Specifically, this section comprises a review of the emissions from peat fires; an up-to-date and comprehensive compilation of gas and PM emission factors (EFs) from both laboratory experiments and field measurements available in peat fire emission studies (all EFs are categorised into and compared between boreal and temperate peat, and tropical peat); and a discussion of the uncertainties in peat fire emission estimates.

#### Emissions and emission factors

Inventories of biomass fire emissions are an essential input for atmospheric chemical transport models, used to understand the influence of fire emissions on the atmosphere and climate change (Urbanski 2014). Overall, there are two different groups of methodologies for calculating total emissions from biomass fires: top–down and bottom–up approaches. Top–down approaches generally utilise satellite atmospheric emission observations in combination with modelling techniques to derive total emission estimates (Page *et al.* 2002; Huijnen *et al.* 2016). For bottom–up methods, information regarding the individual components that contribute to the total amount of fuel burned, and the amount of gas emitted per unit of burned fuel are used to approximate the total emissions (IPCC 2006) (Eqn 1).

$$E_T = A \times B \times C \times EF \tag{1}$$

where, for any given emission species,  $E_T$  is the total emission from biomass fires, usually expressed in the form of emission mass flux; A is the annual burnt area (m<sup>2</sup> year<sup>-1</sup>), B is the fuel load (g dry matter m<sup>-2</sup>), C is the burning efficiency (sometimes called the combustion completeness), and EF is the emission factor, defined as the mass of the species emitted per mass of dry fuel consumed, usually expressed in units of grams per kilogram (Urbanski 2014).

Most total emissions studies focus on carbon emissions flux ( $m_c$ , equivalent carbon mass per unit area), as substantial carbon losses from the terrestrial carbon reservoir are of great importance in terms of global carbon budgets and climate change (Rein 2013; Turetsky *et al.* 2014). Established approaches for estimating total carbon emissions from peat fires relied on direct field-based measurement campaigns, where information including peat bulk density, moisture content and depth of burn were used to estimate the carbon emission fluxes during a specific peat fire episode. Benscoter and Wieder (2003) found that the boreal western Canadian peatlands have an average carbon emission of  $2.1 \pm 0.4$  kg carbon m<sup>-2</sup>. They also provided a summary of carbon emissions from early boreal region peat combustion studies using both field measurements and remote sensing data.

Davies *et al.* (2013) surveyed a temperate peatland fire episode in the Scottish Highlands during the summer of 2006 and estimated that average carbon emissions reached  $9.6 \pm 1.5$  kg carbon m<sup>-2</sup>. Davies *et al.* (2013) summarised peat fire carbon emission fluxes obtained from different field-based estimates, finding that drained boreal peat fire carbon emissions can reach 16.8 kg carbon m<sup>-2</sup> (Turetsky *et al.* 2011), while maximum tropical peat fire carbon emissions have been reported up to 31.5 kg carbon m<sup>-2</sup> (Page *et al.* 2002).

The development of satellite remote sensing algorithms and products has enabled the provision of estimates of global or regional peat fire emissions on an annual basis. Page et al. (2002) estimated that the 1997 Indonesian forested peatland fires emitted between 0.81 and 2.57 Gt carbon. This was equivalent to 13-40% of mean annual global carbon emissions from fossil fuels. Poulter at al. (2006) proposed that temperate peatland fires may emit up to 0.32 Gt of carbon during drought years. van der Werf et al. (2008) looked into fire emissions from Indonesia, Malaysia and Papua New Guinea during 2000-06 and found that average fire emissions (peat fires, forest fires, etc.) from the region reached  $0.128 \pm 0.051$  Gt carbon year van der Werf et al. (2010) revealed that during 1997-2009, carbon emissions from tropical deforestation, degradation and peatland fires were on average 0.5 Gt carbon year<sup>-1</sup>, while tropical peat fire contributed to 3% of global fire emissions from 2001 to 2009. Huijnen et al. (2016) estimated that the total carbon released by the 2015 south-east Asia peatland fires was  $0.227\pm0.067$  Gt; the daily 0.0113 Gt CO\_2 emissions during the fire episode exceeded the fossil fuel CO2 release rate of the European Union (0.0089 Gt  $CO_2$  day<sup>-1</sup>). In recent work by van der Werf et al. (2017), it is estimated that the carbon emissions from peat fire contribute 3.7% of the global fire emissions estimates during 1997–2016.

In addition to the emission fluxes approaches discussed above, a different approach for quantifying peat fire emissions is to measure the mass concentration (usually expressed in the units of milligram per cubic metre or microgram per cubic metre) of different emission species during a specific peat fire episode. For example, Muraleedharan *et al.* (2000*a*) reported the concentration of both PM<sub>10</sub> and a series of VOCs during the 1998 Brunei haze episodes evolved from local peat fires. Fujii *et al.* (2014) found that PM<sub>2.5</sub> aerosols emitted from the 2012 Sumatra peatland fire were at high concentrations of 7120  $\pm$  3620 µg m<sup>-3</sup>. These peat fire emission concentrations data are widely used in public haze health impact studies (Koplitz *et al.* 2016).

Characterisation of EFs is critical for the calculation of total peat fire emissions, and to determine their impacts in the context of global climate change. EF for species i can be calculated with Eqn 2:

$$EF_i = \frac{\dot{m}''_i}{\dot{m}''} \tag{2}$$

where  $\dot{m}_i^{"}$  is the mass flux of the released species *i* and  $\dot{m}^{"}$  is the total mass loss rate (fuel consumption rate) of the dry fuel.

Among the studies to reveal the importance of biomass fire emissions, Andreae and Merlet (2001) and Akagi *et al.* (2011) Y. Hu et al.

compiled the biomass burning EFs (which include peat fires) from previous studies. Their synthesised EFs are widely used as the recommended EFs in the atmospheric modelling community to calculate total fire emissions such as Global Fire Emissions Database (GFED), a global fire emissions database that provides the estimates of fire emissions from different areas and time periods (van der Werf et al. 2010, 2017). With more attention paid to peat fire emissions, more gas and particle EFs have been reported from both laboratory and field measurements in recent years. In the next two subsections, we provide an up-to-date and comprehensive compilation of inter-study peat fire gas and particle species EFs. According to the geographical origins of the peat used in fire emission studies, we classified the samples into two categories: boreal and temperate peat (we merge these two climate zones into one category owing to the limited sampling location information reported in the literature), and tropical peat. It is worth noting that the separation of boreal and temperate peats from tropical peats is based only on data reported in studies of peat combustion or peat fire emissions. By doing this, in the following sections, the best estimate peat fire EFs were calculated and compared between the two peat categories for the first time.

Owing to the different forms of the EFs reported in the peat fire emission studies (for example, Christian et al. (2003) reported study-averaged EF means, Geron and Hays (2013) reported the EFs as a range, whereas other studies reported both study-averaged EF means and their variability - usually expressed in standard deviation), a reasonably simple approach was adopted in the present paper to conservatively estimate the overall EFs for each species in a peat category (boreal and temperate-, or tropical). When the variability is not given in the original data, a general range of 20-50% of the EF mean is assumed. For EFs reported in ranges, the mean and variability are calculated from the data provided by the authors (e.g. Geron and Hays 2013). We assumed different studies were independent random variables that were normally distributed. Based on this assumption, the overall EFs of different peat categories shown in the present paper are reported in the form of inter-study EF mean  $\pm$  variability (variability is defined as the inter-study standard deviation). Inter-study EF means are calculated by averaging the mean EFs for each study; for variability, we first summed the EF variances (square of standard deviation) of each study, and then divide them by the number of studies. Next, we took the square root of the averaged variance to derive the inter-study standard deviation for a peat category. By doing this, the EF variability within each peat fire emission study was included in calculating the overall inter-study EF variability.

#### Gas species EFs

In this subsection, EFs of peat fire gas species that are commonly reported in the literature (19 species) are compiled in Table 1 and compared with savanna-burning EFs from Akagi *et al.* (2011) in Fig. 4 (left). Compared with smouldering-dominated peat fires, the majority of biomass in a savanna fire is consumed by flaming combustion. Generally, peat fires have higher EFs of incomplete combustion products like CO and  $CH_4$  than flaming savanna fires owing to departure from stoichiometric Review of haze emissions from peat fires

Pyres         Intermediate	erence	Yokelson et al. (1997)	Stockwell et al. (2014)	Black et al. (2016)	Black <i>et al.</i> (2016)	Chakrabarty et al. (2016)	Stockwell et al. (2014)	Geron and Hays (2013) <sup>C</sup>	sWilson <i>et al.</i> (2015)	Rein <i>et al.</i> (2009)	Chakrabarty <i>et al.</i> (2016)	Boreal and Temperate neat <sup>D</sup>	Tropical peat <sup>D</sup>	Christian <i>et al.</i> (2003) <sup>B</sup>	Stockwell et al. (2014)	Stockwell et al.	Smith et al.	Hujnen <i>et al.</i>
Matrix         Matrix<	y type	Laboratory	Laboratory	Laboratory	Laboratory	Laboratory	Laboratory	Laboratory	Laboratory	Laboratory	Laboratory	beau		Laboratory	Laboratory	Field	Field	Field
manonenticiale 20 (65) 26(1) (67) (67) (51) (51) (52) (52) (51) (51) (51) (51) (51) (51) (51) (51	location <sup>A</sup> oon dioxide	NA #1 1395 (52)	NA #2 1066 (287)	NA #3 861 (112)	NA #4 922 (47)	NA #5 1418 (180)	NA #6 1274 (19)	NA #7 1071 (79)	NE #1 1346 (31)	NE #2 420 (134)	EE #1 1565 (133)	1133.8 (139)	1615.4	SA #1 1703 (340)	SA #2 1637 (204)	SA #3 1564 (77)	SA #4 1579 (58)	SA #5 1594 (61)
Munc(Li) $(8 (56))$ $(10 (5))$ $(11)$	02) bon monoxide	209 (68)	276 (139)	108 (20)	122 (14)	105.5 (22.5)	197 (9)	262 (18)	218 (22)	170 (33)	126.5 (77.5)	179.4 (60.5)	(184) 248.06 (40.8)	210.3 (42)	233 (72)	291 (49)	251 (39)	255 (39)
opene (CH)         0.1 (0.0)         0.18 (0.0)         0.18 (0.0)         0.18 (0.0)         0.18 (0.0)         0.18 (0.0)         0.13 (0.	D) hane (CH4)	6.85 (5.66)	10.9 (5.3)	I	I	I	6.25 (2.17)	I	8.35 (1.3)	I	I	8.1 (4.07)	(49.0) 12.30	20.8 (4.2)	12.8 (6.6)	9.51	11.00	7.4 (2.3)
	tylene (C <sub>2</sub> H <sub>2</sub> )	0.1 (0.05) <sup>b</sup>	0.16 (0.08)	I	I	I	$0.1 \ (0.05)^{\rm B}$	I	I	I	I	0.12 (0.06)	(5.0.5) 0.11	0.06 (0.03)	0.18 (0.05)	(4./4) 0.121	0.06	I
	ylene (C <sub>2</sub> H <sub>4</sub> )	1.37 (0.51)	1.27 (0.77)	I	I	I	0.81 (0.29)	I	1.74 (0.23)	I	I	1.3 (0.50)	(0.04) 1.81	2.57 (0.51)	1.39 (0.62)	(0.066) 0.961	(0.01) 2.30	I
	ane (C <sub>2</sub> H <sub>6</sub> )	I	I	I	I	I	I	I	1.53 (0.17)	I	I	1.53 (0.17)	(1.48) 2.17	Í	I	- (826.0)	(2.79) 2.17	I
thand         404 (3.4)         2.8 (2.8)         -         -         0.5 (0.87)         -         -         0.5 (0.87)         0.21         0.23         0.87         0.77         0.84         0.87         0.77	pylene (C <sub>3</sub> H <sub>6</sub> )	2.79 (0.44)	1.17 (0.63)	I	I	I	$0.5 (0.2)^{\rm B}$	I	I	I	I	1.49 (0.46)	(0.50) 1.98 (1.30)	3.38 (0.66)	1.49 (0.63)	1.07	(0.81) -	I
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	thanol	4.04 (3.43)	2.83 (2.87)	I	I	I	0.75 (0.35)	I	0.6 (0.87)	I	I	2.06 (2.28)	(10.01) 4.17	8.46 (1.68)	3.24 (1.39)	(cc.0) 2.14	2.83	I
	(H <sub>3</sub> OH) maldehyde	1.99 (2.67)	1.41 (1.16)	I	I	I	1.43 (0.37)	I	I	I	I	1.61 (1.69)	(1.32)	1.4 (0.07)	1.25 (0.79)	(1.22) 0.867	(0.84) 0.77	I
	HCHO) staldehyde	I	I	I	I	I	I	I	I	I	I	I	(0.56) 1.98	3.27 (0.64)	I	(0.479) 0.697	(0.64)	I
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	CH <sub>3</sub> CHO) /colaldehyde	1.66 (2.64)	I	I	I	I	I	I	I	I	I	1.66 (2.64)	(0.56) 0.85	1.59 (0.32)	Ι	(0.46) 0.108	I	I
COH         Control         Control <thcontrol< th=""> <thcontrol< th=""> <thcont< td=""><td><math>^{2}_{2}H_{4}O_{2})</math></td><td>0.80.01.51</td><td>0 44 (0 34)</td><td> </td><td></td><td></td><td>0.4.00.060</td><td></td><td>I</td><td></td><td>I</td><td>0 58 (0 80)</td><td>(0.24) 0.38</td><td>0.54.00.11)</td><td>0 55 (0.05)</td><td>(0.089) 0.18</td><td>560</td><td>I</td></thcont<></thcontrol<></thcontrol<>	$^{2}_{2}H_{4}O_{2})$	0.80.01.51	0 44 (0 34)				0.4.00.060		I		I	0 58 (0 80)	(0.24) 0.38	0.54.00.11)	0 55 (0.05)	(0.089) 0.18	560	I
tic acid 7.29 (4.89) $8.46$ (4.6) $   1.86$ (1.35) $        5.87$ (5.70) (5.3) (5.3) (3.65) (3.75) (3	HCOOH)	((-1) (0.0	(10.0) 11.0	I	I	I	(00.0) 1.0	I	I	I	I	(10.0) 00.0	(0.08)	(11:0) +0:0	(00.0) 00.0	(0.085)	(0.04)	I
Targeound an (C4H4O)-1.78 (1.84)0.88 (0.04)0.88 (0.04)1.33 (1.3)1.181.91 (0.38)0.89 (0.27)0.736	etic acid	7.29 (4.89)	8.46 (8.46)	I	I	I	1.86 (1.35)	I	I	I	I	5.87 (5.70)	6.38	8.97 (1.8)	7.65 (3.65)	3.89	5.02	I
drogen cyanide $5.09(5.64)$ $4.45(3.02)$ $   -$ <td>an (C4H4O)</td> <td>I</td> <td>1.78 (1.84)</td> <td>I</td> <td>I</td> <td>I</td> <td>0.88 (0.04)</td> <td>I</td> <td>I</td> <td>I</td> <td>I</td> <td>1.33 (1.3)</td> <td>(+C-2) 1.18</td> <td>1.91 (0.38)</td> <td>0.89 (0.27)</td> <td>0.736</td> <td>(1.04)</td> <td>I</td>	an (C4H4O)	I	1.78 (1.84)	I	I	I	0.88 (0.04)	I	I	I	I	1.33 (1.3)	(+C-2) 1.18	1.91 (0.38)	0.89 (0.27)	0.736	(1.04)	I
$ \begin{array}{rcccccccccccccccccccccccccccccccccccc$	drogen cyanide	5.09 (5.64)	4.45 (3.02)	I	I	I	1.77 (0.55)	I	2.21 (0.35)	I	I	3.38 (3.21)	(0.35) 5.24 2.7	8.11 (1.6)	3.3 (0.79)	(0.392) 5.75 (1.6)	3.79	I
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	ric oxides (NC	- -×	0.51 (0.12)	I	I	I	I	I	I	I	I	0.51 (0.12)	(cc.1) 1.05	1 (0.2)	1.85 (0.56)	0.307	(1.9.1)	I
$ \begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$	rogen dioxide	I	2.31 (1.46)	I	I	I	I	I	I	I	I	2.31 (1.46)	(0.40) 2.36	I	2.36 (0.03)	(0.36) -	I	I
40NO) (0.05) (0.05) (0.05) (0.05) (0.05)	VO <sub>2</sub> ) rous acid	I	0.48 (0.5)	I	I	I	$0.18\ (0.1)^{\rm B}$	I	I	I	I	0.33 (0.36)	(0.03) 0.15	I	$0.1 (0.05)^{B}$	0.208	I	I
	10N0) monia (NH <sub>3</sub> )	8.76 (13.76)	1.87 (0.37)	I	I	I	2.21 (0.24)	I	0.73 (0.5)	I	I	3.39 (6.89)	(0.05) 8.00 (3.04)	19.92 (3.98)	1.39 (0.97)	(0.059) 2.86 (1)	7.82 (4.37)	I

sampling locations (detailed sampling location information is omitted here). <sup>B</sup>Uncertainty was not given in original data; here, a general range of 20–50% is assumed.

<sup>CEF</sup> values for wildfire were reported from multi-fire emission study. Here, we only report the EFs obtained from the sampling of Pocosin Lakes National Wildlife Refuge ground fire (strict organic peat soil). Mean and

variability values are calculated from the data provided by the author. DEF values for overall boreal and temperate peats, and tropical peats are an inter-study average (the final EF average values are determined by averaging the EF mean of each study; the average variabilities are the square root of the averaged variances in each study).



Fig. 4. Gas-species emission factors (EFs) from peat and savanna burning (left); main gas species (CO<sub>2</sub>, CO, CH<sub>4</sub>, HCN, NH<sub>3</sub>) EFs from peat burning in boreal and temperate, and tropical regions (right).

combustion (Rein 2015). Peat fires emit significant amounts of  $CO_2$  and  $CH_4$ , with the potential to shift peatlands from being a major atmospheric and terrestrial carbon sink to becoming a major GHG emission source (Turetsky *et al.* 2014). Smouldering peat fire EFs of HCN and NH<sub>3</sub> are approximately 10 times those for flaming savanna fires; these two species are thought to be primarily emitted by smouldering and not flaming combustion, and were therefore proposed as atmospheric tracers for peat fires (McMeeking *et al.* 2009; Akagi *et al.* 2011). As a result,  $CO_2$ , CO, CH<sub>4</sub>, HCN and NH<sub>3</sub> are selected as the main characteristic peat fire gas species analysed in this review.

 $CO_2$  is the dominant gas species released during a peat fire event, with EFs reported in the order of  $10^3$  g kg<sup>-1</sup>. Fig. 5 shows  $CO_2$  EFs from peat burning available in the literature. The largest  $CO_2$  EF value (1703 g kg<sup>-1</sup>) was reported from the burning of south Sumatra tropical peat (Christian *et al.* 2003), whereas the smallest value of  $CO_2$  EFs in the literature (149– 185 g kg<sup>-1</sup>) was reported from the combustion of Brunei peat (Muraleedharan *et al.* 2000*b*). Given that this unusually low EF range was derived from peat thermodecomposition rather than open combustion, this EF is not included in our compilation. Comparatively, tropical peat fires emit higher amounts of  $CO_2$ with an average EF of 1615 g kg<sup>-1</sup> (n = 5, where *n* is the number of studies), whereas boreal and temperate peat fires have an average  $CO_2$  EF of 1134 g kg<sup>-1</sup> (n = 10).

CO is the second most predominant gaseous species emitted during peat fire events. Compared with flaming biomass, smouldering peat has a much higher CO EF; the highest value of CO EF (291 g kg<sup>-1</sup>) reported in the literature comes from a peat fire field measurement (Stockwell *et al.* 2016). This may be explained by the predominance of incomplete smouldering combustion in peat fires, from which CO emissions are more predominant (Rein 2015). Similarly to CO<sub>2</sub>, tropical peat fires emit higher amounts of CO, with an average EF of 248 g kg<sup>-1</sup> (n = 5), whereas boreal and temperate peat fires have an average CO EF of 179 g kg<sup>-1</sup> (n = 10). Peat fires EFs of CO from different studies are summarised in Fig. S1 available as Supplementary Material to this paper.



Fig. 5. Peat fire  $CO_2$  emission factors (EFs) from different emission studies. Peat samples used in the literature are classified into two categories: boreal and temperate peat, and tropical peat. According to the United Nations geo-scheme, 'NA' refers to northern America; 'NE' refers to northern Europe; 'EE' refers to eastern Europe; 'SA' refers to south-east Asia; '#1–7' represents different sampling locations (detailed peat sampling location information is omitted here).

The third most abundant compound in peat fire emissions is methane (CH<sub>4</sub>). CH<sub>4</sub> EFs from peat fires are approximately two times higher than those for tropical forest fires, and are almost six times higher than savanna fire EFs (Akagi *et al.* 2011). Consistent with CO<sub>2</sub> and CO, tropical peat fires are found to emit more CH<sub>4</sub> per unit mass of dry fuel burned (12.3 g kg<sup>-1</sup>, n = 5) than boreal and temperate peat fires (8.1 g kg<sup>-1</sup>, n = 4).

The EFs for CH<sub>4</sub>, HCN and NH<sub>3</sub> from peat fires are summarised in Figs S2–S4. To summarise, by categorising the 60

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different studies.

Peat carbon content (C%)

Boreal and temperate peat

Heres of the state A alal in the second A COLORADA AND A COLO a. culorfielde peak Crattabarty Fig. 6. Peat carbon contents used in each peat fire emission study. Variability of the peat carbon content within one study is shown as the error bar where applicable. Peat samples used in the literature are classified into two categories: boreal and temperate peat, and tropical peat. According to the United Nations geo-scheme, 'NA' refers to northern America; 'NE' refers to northern Europe; 'EE' refers to eastern Europe; 'SA' refers to southeastern Asia; '#1-6' represents different sampling locations (detailed peat sampling location information is omitted here). The mean and variability for the boreal and temperate peat, and for tropical peat were calculated from the average and the standard deviation of the peat carbon content(s) from

Tropical peat

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peat into tropical peat and boreal and temperate peat, it is found that tropical peat fires have overall larger EFs of main gas species (CO<sub>2</sub>, CO, CH<sub>4</sub>, HCN and NH<sub>3</sub>) than boreal and temperate peat fires (Fig. 4).

The carbon content of peat, which is of critical importance in terms of the carbon emissions, varies among different locations and climate zones. Several non-fire studies provide comprehensive and systematic databases of soil carbon (which includes peat soil). Page et al. (2011) briefly reviewed peat soil carbon and found that the carbon content of south-east Asian peat is in the range of 41.6 to 62%. Liu et al. (2013) provided a unified North American soil map, where they assumed that soil organic carbon accounts for 58% of soil organic matter. Consistent with peat classification ruled for EF compilation, for this review, we rely mostly on peat carbon content measurements associated with peat combustion or peat fire emissions to classify and compare the peat carbon content. Our comparison shows that by contrast, tropical peat has an average carbon content of 56.0% (n = 5), whereas the boreal and temperate peat samples have a lower average carbon content of 44.2% (n = 10) (Fig. 6). To summarise the peat carbon content associated with fire emission studies, we first investigate the correlation of peat carbon content with the corresponding inter-study EFs of CO2 and CO, the most abundant carbon-containing gas species from peat fires.

Fig. 7 reveals a positive correlation between the carbon content of the peat and the  $CO_2$  EFs, with higher peat carbon



Fig. 7. Peat fire CO<sub>2</sub> emission factors (EFs) as a function of peat carbon content. With the increase of peat carbon content, the CO2 EFs increase.



Fig. 8. Peat fire CO<sub>2</sub> emission factors (EFs) as a function of CO EFs. The relationship between CO2 EF and CO EF for all peat samples is weakly correlated ( $R^2 = 0.116$ ).

contents associated with higher CO<sub>2</sub> EFs. CO<sub>2</sub> is the main product of complete combustion and, taking into account that it is formed by one atom of carbon and two of oxygen, its formation directly depends on the availability of both elements. Compared with boreal and temperate peat, tropical peat samples have higher carbon content, and as such will produce higher CO<sub>2</sub> emissions per unit of dry fuel mass. As observed for CO<sub>2</sub> emissions, the carbon content is positively correlated with CO EFs (Fig. S5). Tropical peats have higher carbon content, emitting more CO on average, possibly owing to the same reasons as explained above for  $CO_2$ .

Regarding the relationships between different gas species EFs, CO EFs from tropical peat fires showed a negative correlation with  $CO_2$  EFs, whereas the relationship in boreal and temperate peat remains inconclusive (Fig. 8). The relationships between CO<sub>2</sub> EFs and the EFs of CH<sub>4</sub>, HCN and NH<sub>3</sub> are depicted in Figs S6-S8. The relationships remain inconclusive for most of the gas species, possibly owing to the limited amount of available data reported in the literature. For example, there are only four studies that have quantified  $CH_4$ , HCN and  $NH_3$  EFs from boreal and temperate peat fires, and only four HCN and  $NH_3$  EF values are reported for tropical peat fires.

Generally, the EFs of all other trace gases (gas species listed in Table 1, exclusive of CO<sub>2</sub>, CO, CH<sub>4</sub>, HCN and NH<sub>3</sub>) are below 10 g kg<sup>-1</sup> and most of them are NMOCs, which can influence secondary organic aerosol and ozone formation (Akagi *et al.* 2011). However, most NMOCs are reactive, and it is still challenging to identify and quantify all trace gas species properly. Additional studies measuring peat carbon content and emissions and advanced gas species detection technologies could improve their incorporation into EF quantification.

#### Particle EFs

Research focusing on PM emissions from wildland fires, especially smouldering peat fires, is limited. Tissari *et al.* (2008) found that for smouldering combustion, PM<sub>1</sub> yields were six times those from flaming combustion. Iinuma *et al.* (2007) found that peat burning yields significantly higher PM<sub>2.5</sub> emissions in comparison with other biofuels (softwood, hardwood and grass). Tropical peat fires can emit 3–6 times as much PM as grassland, forest or plantation fires per unit carbon combusted (Turetsky *et al.* 2014).

In this subsection, we have compiled the EFs for PM ( $PM_{10}$ ,  $PM_{2.5}$  and  $PM_1$ ) reported in peat fire emission studies (Table 2). Geron and Hays (2013) found that most peat smoke particles are in the PM<sub>2.5</sub> size range. Reported peat fire PM<sub>2.5</sub> EFs in the literature vary from 5.9 g kg<sup>-1</sup> (Black *et al.* 2016) to 44.5 g kg<sup>-1</sup> (Geron and Hays 2013). Fig. 9 provides a comparison of PM EFs for both boreal and temperate peat and tropical peat fires, with flaming savanna EFs from Akagi et al. (2011) for contrast. Generally, peat fires emit larger quantities of PM<sub>2.5</sub> than flaming savanna (averaged PM2.5 EFs for all types of peat fires and savanna burning are 18.7 and 7.17 g  $kg^{-1}$  respectively). In comparison, boreal and temperate peat fires emit slightly more  $PM_{2.5}$  (EF = 19.17 g kg<sup>-1</sup>, n = 3) than tropical peat fires (EF = 17.3 g kg<sup>-1</sup>, n = 1), although the number of studies is low. This variation may partly be caused by differences in the carbon content of the peat samples and the PM EF determination method used in each study. It is clear that there is a serious shortage of PM EFs, especially for tropical peat fires; additional thorough peat fire PM studies are required to fill this knowledge gap.

Organic carbon (OC) constitutes the main component of  $PM_{2.5}$ , accounting for 71% of  $PM_{2.5}$  aerosols in a study of a Sumatran peat fire (Fujii et al. 2014). Black carbon (BC), a major component of soot and an important compound for atmospheric warming owing to its low albedo, is present only in trace amounts ( $\sim 0.1 \text{ g kg}^{-1}$ ) in boreal and temperate peat fires. There is no available BC EF data for tropical peat fires. A recent study showed that aerosols emitted from the burning of Alaskan and Siberian peatlands are predominantly brown carbon (BrC), a class of visible-light-absorbing OC, with a negligible amount of BC (Chakrabarty et al. 2016). Although BrC is very unsteady and can be lost quickly through chemical loss and evaporation, it can amplify the warming effects of BC emitted from these fires (Forrister et al. 2015). However, it is noticeable that OC is light-scattering in nature; it can offset the warming effects of GHGs and BC. Therefore, the co-existence of OC, BC

and BrC complicates assessing radiative impacts and adds large uncertainties to the atmospheric feedback from peat fire PM emissions (Chakrabarty *et al.* 2016).

A range of VOCs including BTEX (benzene, toluene, ethylbenzene and xylene) and PAHs can also be enriched in the PM (Durán *et al.* 2014). Specifically, PAH emissions from boreal and temperate peat fires were dominated by naphthalene, fluorene and phenanthrene (Black *et al.* 2016). Black *et al.* (2016) found that dioxins, a group of toxic compounds, can also be produced from peat fires. The polychlorinated dibenzodioxins and dibenzofurans (PCDD-F) EFs (8.1 ng kg<sup>-1</sup>) from boreal and temperate peat are noticeably higher than the EFs from other various biomass types. No dioxin EFs from tropical peat fires are reported in the literature.

### Uncertainties in peat fire EF determination and total emission estimates

Theoretically, EFs can be calculated by using species mass flux and dry fuel mass loss rate (Eqn 2); however, the mass loss rate is difficult to obtain under most measurement conditions, especially for field measurements (Huijnen *et al.* 2016; Stockwell *et al.* 2016; Smith *et al.* 2018). As a result, a carbon mass balance approach is widely used to calculate the EFs from peat fires. This approach utilises the fuel carbon content and the amount of carbon species released to approximate the fuel consumption rate (Eqn 3) (Yokelson *et al.* 1997; Christian *et al.* 2003; Stockwell *et al.* 2014; Wilson *et al.* 2015):

$$EF_i = F_c \times 1000(g \, kg^{-1}) \times \frac{MM_i}{12} \times \frac{C_i}{C_T}$$
(3)

where  $F_c$  is the measured carbon content of the peat,  $MM_i$  is the molar mass of species *i*,  $C_i$  is the number of moles of species *i*;  $C_T$  is the total number of moles of carbon emitted.

Detailed different forms of calculating gas species EFs by using the carbon balance approach (e.g. calculating directly from species excess mole fractions or emission ratios) are summarised in Paton-Walsh et al. (2014). Particle EFs are generally determined by representative PM sampling and gravimetric analysis (Black et al. 2016). There are inherent advantages and uncertainties rooted in both laboratory experiments and field measurements for calculating EFs for peat fires. The availability of advanced gas identification techniques and instrumentation (e.g. high-resolution proton-transfer-reaction time-of-flight mass spectrometry and two-dimensional gas chromatography-time-of-flight mass spectrometry) in laboratory studies enables best practice for the identification of the gas species present in peat smoke (Hatch et al. 2015; Stockwell et al. 2015). However, any peat samples used for laboratory tests or peat in situ in field measurements have an inherent inhomogeneity in composition (e.g. carbon content, moisture content), cracks and gas permeability. Owing to the complexity of the peat burning environment, challenges remain in terms of how to scale up the EFs derived from laboratory fires to a natural peat fire.

Stockwell *et al.* (2016) provide the best available EF for peat fires from field measurements. However, the transient emission composition and concentration from field measurements are highly dependent on the combustion efficiency and sampling environment (e.g. wind and smoke aging) (Akagi *et al.* 2011).

Reference	Black <i>et al.</i> (2016)	Black <i>et al.</i> (2016)	Chakrabarty et al. (2016) <sup>D</sup>	Geron and Hays (2013) <sup>F</sup>	Iinuma et al. (2007)B,E	Chakrabarty et al. (2016) <sup>D</sup>	Boreal and temperate	Tropical peat <sup>C</sup>	Christian et al.	Iinuma et al. (2007) <sup>B,E</sup>	May <i>et al.</i> (2014)	Jayarathne et al.
Study type	Laboratory	Laboratory	Laboratory	Laboratory	Laboratory	Laboratory	pcar		Laboratory	Laboratory	Laboratory	Field
Peat location <sup>A</sup>	NA #1'	NA #2'	NA #3'	NA #4'	WE #1'	EE #1'			SA #1'	SA #2'	SA #3'	SA #4'
PM10	I	I	I	I	44 (22)	I	44 (22)	33 (16.5)	I	33 (16.5)	I	I
PM2.5	7.1 (5.6)	5.9 (6.7)	Ι	44.5 (8.01)		I	19.17 (6.84)	17.3 (6.0)	I	, I	I	17.3 (6.0)
$PM_1$	Ι	I	Ι	Ι	Ι	Ι	I	34.9	Ι	I	34.9 (17) <sup>B</sup>	I
								(c./1)				
0C	6.3(4.1)	4.3(4.1)	13 (3)	I	12.8 (6.4)	5.5 (1.5)	8.38 (4.14)	8.8 (4.24)	6.02 (3)	8 (4)	I	12.4 (5.4)
EC	0.05(0.06)	0.08(0.09)	I	I	0.96 (0.48)	I	0.36 (0.28)	0.28	0.04~(0.02)	0.57 (0.29)	I	0.24(0.10)
		0.07.70.017	0 15 (0 05)			015 (0.05)	011 (0.04)	(01.0)				
BC	0.07 (0.04)	0.06 (0.01)	(c0.0) c1.0	I	I	(c0.0) c1.0	0.11 (0.04)	I	I	I	I	I
<i>n</i> -Alkanes (mg kg <sup>-1</sup> )	I	I	I	I	910 (455)	I	910 (455)	470 (235)	I	470 (235)	I	I
<i>n</i> -Alkenes (mg kg <sup><math>-1</math></sup> )	I	I	I	I	180(90)	I	180 (90)	110 (55)	I	110 (55)	I	I
Fluorene ( $\operatorname{mg} \operatorname{kg}^{-1}$ )	13.12	9 (6.56)	I	I	18 (9)	I	13.4 (7.52)	11 (6)	I	11 (6)	I	I
	(6.74)											
Phenanthrene (mg kg <sup><math>-1</math></sup> )	14.99	8.6 (6.35)	Ι	I	3 (1.3)	Ι	8.86 (4.88)	2.7 (1.4)	I	2.7 (1.4)	Ι	Ι
-	(5.42)											
Pyrene (mg kg <sup><math>-1</math></sup> )	2.84 (1.4)	1.55(1.08)	Ι	I	1.5(0.8)	I	1.96 (1.12)	1.5 (0.8)	Ι	1.5(0.8)	Ι	Ι
Total PAHs (mg kg <sup><math>-1</math></sup> )	16(8.9)	14 (9)	I	I	28 (14)	I	19.3 (10.9)	20 (10)	I	20(10)	I	I
Dioxins (PCDD-F), (ng kg <sup>-1</sup> )	8.7 (4.35)	7.4 (3.7) <sup>b</sup>	I	I	I	I	8.1 (4.05)	I	I	I	I	I
AL ocation classified accordi	no to United N	ations geo-sche	me: 'NA' refers	s to northern A	merica: 'WE' 1	refers to western	n Eurone: 'EE' 1	efers to easter	rn Eurone: 'SA	refers to sout	n-eastern Asia:	# represents
different sampling locations	(detailed samp	oling location in	nformation is or	mitted here).	-							
<sup>B</sup> Uncertainty was not given	in original data	a; here, a genera	al range of 20-:	50% is assume	d.							
<sup>C</sup> EF values for overall borea	l and temperate	peats, and tropi	ical peats are an	inter-study ave	prage (the final	EF average valu	ues are determin	ed by averagi	ng the EF mean	of each study;	the average var	iabilities are
the square root of the average	ged variances in	n each study).										
<sup>D</sup> EF values were calculated	from the avera	ged values fron	n different exne	rimental settin	as reported in	the naner						

<sup>E</sup>PM samples were collected in five different size stages: 0.05–0.14, 0.14–0.42, 0.42–1.2, 1.2–3.5 and 3.5–10 µm. It is noticeable that the authors reported the particle EFs by summing all the five stages; thus,

<sup>F</sup>EFs values for wildfire were reported from multi-fire emission study. Here we only reported the EFs obtained from the sampling of Pocosin Lakes National Wildlife Refuge ground fire (strict organic peat soil). Mean and variability values are calculated from the data provided by the author. we report their result here as PM<sub>10</sub>.

Review of haze emissions from peat fires

Table 2. Emission factors (EFs) of particle species and their main components commonly reported from peat fire emission studies (g kg<sup>-1</sup>, unless otherwise specified)



Fig. 9. Particulate matter (PM:  $PM_{10}$ ,  $PM_{2.5}$  and  $PM_1$ ) and its main component emission factors (EFs) from different peat burning emission studies. In Akagi *et al.* (2011),  $PM_1$ – $PM_5$  were categorised as  $PM_{2.5}$ .

Many species emitted from peat burning are unidentified and semivolatile, rendering them very difficult to quantify by fieldbased techniques. Furthermore, when using the carbon balance approach in calculating the EFs from field measurements, the large natural peat carbon variations that cannot be measured in field studies can become an issue affecting the emission estimation results. The variability of peat carbon content from smallscale laboratory studies (for example, 56.0% for tropical peats vs 44.2% for boreal and temperate peat, discussed above) needs to be taken into consideration when applying the EFs to total peat fire emissions estimates. Careful comparison and synthesis of controlled laboratory results with field measurements have a key role in improving the accuracy of EF determinations into the future (Akagi *et al.* 2011).

Importantly for total fire emission estimates for use in atmospheric modelling, peat must not be considered as a onedimensional surface ground fuel owing to its uneven depth distribution in natural peatlands (Page et al. 2011). Smouldering peat fires are a multidimensional phenomenon. They can burn deep into underground peat layers, resulting in variable burn depth within a single fire episode or among the recurrent fires at the same location (Lukenbach et al. 2015; Konecny et al. 2016). Given that peat fires tend to keep smouldering underground without becoming outwardly apparent on the surface, fire 'hotspot' and burnt area remote sensing detection technologies developed from studies of flaming combustion become ineffective and produce large uncertainties when applied to smouldering peat fires (Rein 2013). To summarise, unavoidable uncertainties exist in this application; significant advances could be achieved by improved detection technologies and more extensive research in this area.

### Combustion regimes, 'combustion efficiency' and 'modified combustion efficiency'

Emissions from peat fires are highly dependent on the combustion regime. There are four different sub-fronts in a



**Fig. 10.** The composition of peat and possible decomposition paths and products (Huang and Rein 2014).

smouldering fire: preheating, drying, pyrolysis and oxidation (Rein et al. 2009). The preheating front, occurring in a range of temperatures up to 100°C, emits water vapour in large quantities and is not involved in chemical reactions (some volatiles may also be released at this stage but are in trace amounts) (Usup et al. 2004; Rein et al. 2009; Yang et al. 2016). At temperatures above 200°C, pyrolysis dominates. Subsequent heating above this temperature increases the pyrolysis rate, forming carbonaceous char as a product. In this front, peat emits VOCs, PAHs, trace levels of CO, CO<sub>2</sub> and water vapour (Usup et al. 2004; Yang et al. 2016). Peat oxidation starts at temperatures of  $\sim$ 350°C and is the main source of CO and CO<sub>2</sub> (Rein *et al.* 2009). It produces a layer of ash as solid residue. This oxidation front could overlap with the pyrolysis front, depending on oxygen availability (Fig. 10). Rein et al. (2009) verified that by changing the combustion dynamics (heat losses and oxygen supply), the emissions from peat burning vary significantly. This section provides a critical review of the validity of the indices available to quantify the combustion regime that are widely used in wildfire emission studies.

Combustion efficiency (CE) is defined as the fraction of fuel carbon converted to  $CO_2$  and it indicates the completeness of a combustion process (Ward and Hardy 1991). Compared with flaming, smouldering combustion has lower combustion efficiency due to the predominant incomplete combustion (Rein 2015). Based on their seven fuel types test results, Ward and Hardy (1991) suggested that smouldering and flaming fires can be easily differentiated by the CE: values of CE from 50 to 80% are characteristic of smouldering fires, whereas values from 80 to 95% represent flaming fires.

The main disadvantage of determining CE is that it requires the measurement of all the carbon released, including CH<sub>4</sub>, NMOC and PM, which is impractical under most field and laboratory conditions. As a solution, Ward and Hao (1991) defined a parameter named modified combustion efficiency (MCE) as a substitute for CE (Eqn 4):

$$MCE = \frac{CO_{2-c}}{CO_{-c} + CO_{2-c}}$$
(4)

where  $CO_{2-c}$  is the mass of carbon emitted as  $CO_2$ , and  $CO_{-c}$  is the mass of carbon emitted as CO. However, the most cited



**Fig. 11.** Modified combustion efficiency reported from literature ( $MCE_{\Delta}$ ) as a function of  $CO_2(a)$ ; CO (*b*); and  $CH_4(c)$  emission factors (EFs) in peat fires. Generally,  $MCE_{\Delta}$  shows no correlation with  $CO_2$ , CO and  $CH_4$  EFs in peat fires (linear correlation  $R^2 < 0.1$ ). Boreal and temperate peat fire MCE data source: Yokelson *et al.* (1997); Stockwell *et al.* (2014); Black *et al.* (2016); Wilson *et al.* (2015); Chakrabarty *et al.* (2016). Tropical peat fire MCE data source: Christian *et al.* (2003); Stockwell *et al.* (2014); Stockwell *et al.* (2016); Smith *et al.* (2018). It is noticeable that Geron and Hays (2013) reported MCE in the smouldering ground fire field measurement at Green Swamp (MCE ranged from 65 to 80%). However, as we only included EF results from Pocosin Lakes National Wildlife Refuge (strict peat soil fuel, no corresponding MCE value is reported) (see Table 1), the MCE reported in their paper is not included in the discussion here. No MCE is reported in Rein *et al.* (2009) and Huijnen *et al.* (2016).

paper in terms of the historical origin of MCE is the one written by Ward and Radke (1993), which recommended the use of MCE in correlating carbon emissions to combustion regimes. Using previous field and laboratory measurement results dating back to the 1980s (for example, Ward and Hardy (1984) found that  $CO_2$  and CO comprise ~95% of the total carbon emissions for most fire types) (Ward and Hardy 1991), MCE was found to be linearly correlated with CE (Eqn 5) and was used as an independent variable to model the release of carbon-containing combustion products (Ward and Hao 1991):

$$MCE = 0.15 + 0.86 \cdot CE, R^2 = 0.96.$$
 (5)

For practical measurement purposes, MCE was used in correlating species EFs instead of CE during biomass fire studies, from which Eqn 6 is used to calculate MCE (Yokelson *et al.* 1996):

$$MCE_{\Delta} = \frac{\Delta CO_2}{\Delta CO + \Delta CO_2} \tag{6}$$

where the term MCE<sub> $\Delta$ </sub> represents the MCE reported from literature (calculated with Eqn 6); excess mixing ratio (EMR) is defined as the mole ratio of species *X* in smoke minus its mole ratio in background air ( $X_{\text{smoke}} - X_{\text{background}}$ ). The EMR of *X* is often denoted by ' $\Delta X$ ' (Akagi *et al.* 2011):  $\Delta CO_2$  is the EMR of CO<sub>2</sub>;  $\Delta CO$  is the EMR of CO.

Yokelson *et al.* (1997) built on MCE<sub> $\Delta$ </sub> as an index of the relative amount of flaming and smouldering in biomass fires. Since then, MCE<sub> $\Delta$ </sub> has been widely used with this objective and

(b)

CO emission factor (g kg<sup>-1</sup>)

150

100

50 0

0.6



8

4

0

0.6

Fig. 12. Modified combustion efficiency calculated by using emission factors (MCE<sub>EF</sub>) as a function of CO<sub>2</sub> (*a*); CO (*b*); and CH<sub>4</sub> (*c*) emission factors (EFs) in peat fires. Boreal and temperate peat fire MCE data source: Yokelson et al. (1997); Stockwell et al. (2014); Black et al. (2016); Wilson et al. (2015); Chakrabarty et al. (2016); Geron and Hays (2013); Rein et al. (2009). Tropical peat fire MCE data source: Christian et al. (2003); Stockwell et al. (2014); Stockwell et al. (2016); Smith et al. (2018); Huijnen et al. (2016).

MCEEF

0.9

it has been regarded as an authoritative index to determine the importance of flaming or smouldering in a fire (Christian et al. 2003; Stockwell et al. 2014; Urbanski 2014; Wilson et al. 2015). It is believed that pure flaming combustion presents an MCE<sub> $\Delta$ </sub> of 0.99 (Akagi et al. 2011; Stockwell et al. 2014); in contrast, the  $MCE_{\Delta}$  of smouldering widely differs in the literature (Urbanski 2014). For example, Stockwell et al. (2016) proposed that an  $MCE_{\Delta} \sim 0.75$  to 0.84 designates pure biomass smouldering. Akagi *et al.* (2011) summarised that smouldering MCE<sub> $\Lambda$ </sub> ranges from  $\sim 0.65$  to 0.85 but most often is near 0.8, whereas an overall  $MCE_{\Delta}$  near 0.9 suggests approximately equal amounts of flaming and smouldering.

 $-397.8x + 519, R^2 = 0.201$ 

0.7

0.8

The relationships between real-time MCE<sub> $\Delta$ </sub> and the real-time EFs of the biomass fire emission have received some attention in the literature. MCE<sub> $\Delta$ </sub> is found to be linearly correlated with the EFs for many compounds (for example, CH<sub>4</sub>, CH<sub>3</sub>OH and CH<sub>3</sub>COOH) during the combustion of fine fuels (savanna fires) with a high MCE<sub> $\Delta$ </sub> value (>~0.9) (Christian *et al.* 2003; Stockwell et al. 2014; Urbanski 2014). This dependence allows the calculation of the EFs of a specific species under a specific MCE<sub> $\Delta$ </sub>. Meyer *et al.* (2012) found that for tropical savanna fires

with a larger range of MCE ( $\sim 0.7$  to 0.99), an exponential relationship provides a better empirical description ( $R^2 = 0.74$ ) between MCE and CH<sub>4</sub> EFs. However, during the measurements of smouldering fires, little or no dependence of EFs on MCE was observed (Akagi et al. 2011; Burling et al. 2011; Stockwell et al. 2016; Smith et al. 2018).

0.08

0.7

0.8

0.9

In practice, fire-averaged  $MCE_{\Delta}$  are reported in the literature to represent the overall combustion regime of a peat fire. Fig. 11 shows the relationship between the fire-averaged MCE $_{\Lambda}$  and the corresponding reported EF values of CO2, CO and CH4 listed in Table 1. There is no significant difference in MCE<sub> $\Delta$ </sub> between boreal and temperate peat, and tropical peat. A larger MCE<sub> $\Delta$ </sub> implies more complete combustion, which means CO<sub>2</sub> EFs are expected to be higher; however, almost no correlation exists between MCE<sub> $\Delta$ </sub> and CO<sub>2</sub> EFs ( $R^2 = 0.045$ ) in Fig. 11*a*. MCE<sub> $\Delta$ </sub> and the EFs of CO and CH<sub>4</sub> correlate poorly ( $R^2$  for MCE<sub> $\Delta$ </sub> and CO EFs, and CH<sub>4</sub> EFs are 0.003 and 0.06 respectively) (Fig. 11b, c). Owing to the limited amount of studies reporting EFs of HCN, NH<sub>3</sub> and other trace gases in the literature, the relationships between MCE<sub> $\Delta$ </sub> and the EFs of those species are omitted here. This is the first time that relationships between inter-study MCE<sub> $\Delta$ </sub> and the EFs of CO<sub>2</sub>, CO and CH<sub>4</sub> have been investigated. Based on the result, we confirm that EFs for smouldering peat fires are poorly correlated with MCE<sub> $\Delta$ </sub>, and that a new proxy is needed to describe the relationship between combustion regimes and emissions.

According to Eqn 2, EF is derived from the mass of species emitted per mass of dry fuel consumed. Factoring the original definition of MCE (Ward and Hao 1991), it can be alternatively calculated with the EFs of CO<sub>2</sub>, CO and their molar mass (Eqn 7):

$$MCE_{EF} = \frac{EF_{CO_2}}{EF_{CO_2} + \frac{MM_{CO_2}}{MM_{CO_2}}EF_{CO}}$$
(7)

where  $\text{EF}_{CO2}$  and  $\text{EF}_{CO}$  are the EF of CO<sub>2</sub> and CO,  $MM_{CO}$  and  $MM_{CO2}$  are the molar mass of CO<sub>2</sub> and CO.

Here we introduce the term 'MCE<sub>EF</sub>', which stands for the MCE calculated with EFs. As can be seen in Fig. 12, compared with MCE<sub> $\Delta$ </sub>, MCE<sub>EF</sub> correlates better with EFs of CO<sub>2</sub> and CO from peat fires listed in Table 1. MCE<sub>EF</sub> and CO<sub>2</sub> EFs show a positive correlation ( $R^2 = 0.40$ ). In contrast, MCE<sub>EF</sub> and CO EFs show a negative correlation ( $R^2 = 0.2$ ). These correlation results correspond to the rationale that more complete combustion emits more CO<sub>2</sub> but less CO. There is no correlation between MCE<sub>EF</sub> and the EFs of CH<sub>4</sub> ( $R^2 = 0.08$ ).

Ideally, in spite of the different expressions,  $MCE_{EF}$  and  $MCE_{\Delta}$  should have a strong correlation with each other, as both variables are representative of CE (Ward and Hao 1991; Ward and Radke 1993). The relationship between MCE<sub>EF</sub> and MCE<sub> $\Delta$ </sub> is shown in Fig. 13. Given the experimental methods reported in the literature, we have arranged the data according to three different types of peat fire EF measurement conditions: controlled laboratory conditions; uncontrolled laboratory conditions and field measurements. Controlled laboratory conditions use preconditioned moisture content peat that is ignited in a controlled manner to ensure a smouldering combustion process (Rein et al. 2009; Stockwell et al. 2014; Wilson et al. 2015), whereas uncontrolled laboratory conditions utilise strong flames or a pilot igniter to initiate the burning of peat (Yokelson et al. 1997; Christian et al. 2003; Black et al. 2016; Chakrabarty et al. 2016). Field measurements unavoidably entail a mixture of peat fire and the burning of aboveground vegetation (Geron and Hays 2013; Huijnen et al. 2016; Stockwell et al. 2016; Smith et al. 2018). As can be seen from Fig. 13, peat fire MCE<sub> $\Delta$ </sub> are generally lower than 0.85, suggesting a smouldering-dominant combustion regime. However, variations of MCE<sub> $\Delta$ </sub> are substantial, especially under the uncontrolled laboratory environment.  $\text{MCE}_{\Delta}$  and  $\text{MCE}_{\text{EF}}$  showed almost no correlation ( $R^2 = 0.018$ ), possibly due to two significantly deflected data points reported by Chakrabarty et al. (2016) as a MCE of less than 0.7, without reporting any exact value. Stockwell et al. (2014) presented a significant uncertainty of MCE (the longest error bars shown in Fig. 12) in the measurement reported by Yokelson et al. (1997). When excluding the deflected data from Chakrabarty et al. (2016), MCE<sub> $\Delta$ </sub> and  $MCE_{EF}$  show good correlation ( $R^2 = 0.65$ ). As pointed out in Ward and Hao (1991), although the MCE measurement provides insight regarding the completeness of the combustion process, it does not explain the variances.  $MCE_{\Delta}$  is highly sensitive to



**Fig. 13.** Modified combustion efficiency (MCE) reported in peat fire emission studies (calculated by using excess mixing ratio, EMR; error bars are standard deviations reported in the studies) as a function of MCE calculated by using emission factors (EFs). 'Controlled lab' data source: Wilson *et al.* (2015); Stockwell *et al.* (2014). 'Uncontrolled lab' data source: Yokelson *et al.* (1997); Black *et al.* (2016); Christian *et al.* (2003); Chakrabarty *et al.* (2016). 'Field measurement' data source: Stockwell *et al.* (2016); Smith *et al.* (2018). The rest of the literature that reported EFs (listed in Table 1) did not provide an MCE<sub>Δ</sub> result, thus this is not included in this figure. Akagi *et al.* (2011) proposed that pure flaming has an MCE near 0.99; smouldering MCE ranges from ~0.65 to 0.85 but is most often near 0.8; an overall MCE near 0.9 suggests approximately equal amounts of flaming and smouldering. Chakrabarty *et al.* (2016) reported an overall MCE ≤0.7. Stockwell *et al.* (2014) used the raw data published in Yokelson *et al.* (1997) and calculated an MCE with 0.809 ± 0.327 (s.d.).

uncontrolled variables, and more well controlled measurements are needed to verify the validity of its use as a universal criterion to determine the combustion regime of a peat fire.

Factoring all trace gas species, a parameter named ' $CE_{EF}$ ' is defined as the mass of carbon emitted in the form of CO<sub>2</sub>, divided by the mass of carbon emitted as the carbon-containing species reported in peat fire emission studies. Similarly to Eqn 7, the  $CE_{EF}$  can be calculated from the EFs of all the carboncontaining species reported in the literature and the molar mass of each species (Eqn 8):

$$CE_{EF} = \frac{EF_{CO_2}}{EF_{CO_2} + \sum_{i=1}^{n} \frac{MM_{CO_2}}{MM_i} EF_i}$$
(8)

where  $\text{EF}_i$  is the EF of carbon-containing species *i*;  $MM_i$  is the molar mass of carbon-containing species *i*. For practical purposes, here we employ the carbon-containing species listed in Table 1 (CO<sub>2</sub>, CO, CH<sub>4</sub>, C<sub>2</sub>H<sub>2</sub>, C<sub>2</sub>H<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, C<sub>3</sub>H<sub>6</sub>, CH<sub>3</sub>OH, HCHO, CH<sub>3</sub>CHO, C<sub>2</sub>H<sub>4</sub>O<sub>2</sub>, HCOOH, CH<sub>3</sub>COOH, C<sub>4</sub>H<sub>4</sub>O and HCN) to calculate the 'best-approximate CE' (CE<sub>EF</sub>) in peat fire emission measurements.

Although the  $CE_{EF}$  is not identical to CE (the accurate calculation of CE requires the EFs of all species including NMOC and PM, which has never been done with current analysis methods), in theory it is closer to CE than MCE, owing to its consideration of a larger number of carbon-containing





Fig. 14. Combustion efficiency calculated by using emission factors and species' molar mass ( $CE_{EF}$ ) as a function of  $CO_2(a)$ ; CO (*b*); and CH<sub>4</sub> (*c*) emission factors (EFs) in peat fires.

species (MCE only considers CO<sub>2</sub> and CO). In the present work,  $CE_{EF}$  is also used to correlate with the EFs of CO<sub>2</sub>, CO and CH<sub>4</sub>. Surprisingly, the *R* coefficient between  $CE_{EF}$  and CO<sub>2</sub> EFs decreases to 0.274 (Fig. 14*a*), meaning that it is a poorer correlation than MCE<sub>EF</sub>. This result is unexpected as the  $CE_{EF}$  is thought to indicate the completeness of a combustion process better than MCE (Ward and Hao 1991). However, when using  $CE_{EF}$  to correlate with CO EFs, it reveals a better *R* coefficient than MCE<sub>EF</sub> ( $R^2 = 0.298$ ) (Fig. 14*b*). CE<sub>EF</sub> and peat fire CH<sub>4</sub> EFs are found to be uncorrelated ( $R^2 = 0.003$ ) (Fig. 14*c*).

To summarise, the correlation existing between CO<sub>2</sub> EFs and MCE<sub>EF</sub> ( $R^2 = 0.397$ ), CE<sub>EF</sub> ( $R^2 = 0.274$ ) and MCE<sub>Δ</sub> ( $R^2 = 0.045$ ) successively decreases; as for CO, the highest correlation observed is between CO EFs and CE<sub>EF</sub> ( $R^2 = 0.298$ ), followed by the one observed with MCE<sub>EF</sub> ( $R^2 = 0.201$ ) and MCE<sub>Δ</sub> ( $R^2 = 0.003$ ). Regarding CH<sub>4</sub> EFs, the highest correlation is the one observed with MCE<sub>EF</sub> ( $R^2 = 0.08$ ), the second highest is the one with MCE<sub>Δ</sub> ( $R^2 = 0.003$ ), indicating that the peat fire CH<sub>4</sub> EFs are correlated with neither MCE nor CE. The variation of these correlations shows that the validity of CE<sub>EF</sub> and MCE<sub>EF</sub> is inconclusive, and not as good as it is assumed in the literature. As a result, we believe that currently no single

parameter is capable of describing accurately the relationship between the combustion regime and comprehensive emissions from smouldering peat fires. Therefore, more studies investigating the effect of combustion dynamics are needed.

#### Conclusions

This paper provides the first review of fire emissions for smouldering peat, and their contribution to haze events. Smouldering peat fire is an incomplete combustion process and generates weak buoyant smoke plumes that accumulate near the ground. Substantial quantities of pollutants emitted from these fires lead to acute and recurrent regional haze episodes, especially in southeast Asia, Russia and the USA. Haze induces air quality deterioration, transportation disruption, impaired visibility and vast economic losses. It also carries broad negative health effects, predominantly to the respiratory and cardiovascular systems.

Overall, research investigating emissions from peat fire is limited to a handful of laboratory and field studies. The relationships between peat fire emissions and the combustion dynamics associated with fire evolution remain poorly understood. Our up-to-date compilation of inter-study gas species and PM EFs reveals that there is large variability of EFs among the literature: tropical peat fires exhibit larger EFs for the prominent gas species (CO<sub>2</sub>, CO, CH<sub>4</sub>, HCN and NH<sub>3</sub>) than those from boreal and temperate peat fires, whereas boreal and temperate peat fires have slightly higher PM<sub>2.5</sub> EFs (19.2 g kg<sup>-1</sup>) than tropical peat fires (17.3 g kg<sup>-1</sup>). We conclude that this EF variability is mainly attributable to the variations of carbon content between tropical peats (56.0%) and boreal and temperate peats (44.2%), together with the complex combustion dynamics.

The discussion of the uncertainties of peat fire emission indicates that laboratory-based techniques enable the thorough detection of species that are present in the peat fire emissions (more than 100 gas species), while uncertainties remain in terms of scaling up the EFs derived from laboratory experiments and applying to total fire emission estimation. In contrast, given the complexity of natural fires, field measurements provide the best available EFs for peat fires. However, high uncertainties are introduced in these EFs owing to the limitation of field-based techniques. The variability in the fuel carbon content needs to be taken into consideration when applying the carbon balance approach to calculate field-based EFs; additional field studies measuring the carbon content across the regions of interest are required. This allows the conclusion that the combination of controlled laboratory experiments with field measurements helps minimise the large uncertainties that exist in peat fire EF quantification and total emission estimates.

The review of MCE, a parameter widely used in biomass fire combustion regime determination, shows that it is often misunderstood and highly sensitive to several uncontrollable variables. The validity of MCE,  $MCE_{EF}$  (calculating from the EFs of CO<sub>2</sub>, CO and their molar mass) and  $CE_{EF}$  (calculating from the EFs of all the carbon-containing species reported in the literature and the molar mass of each species) in correlating EFs for smouldering peat fires is inconclusive. There is a lack of a clear smouldering combustion signature in the literature, emphasising the importance of more studies investigating peat combustion dynamics.

Although international efforts such as haze control agreements have been made, regional haze episodes as a result of peat fires still remain an unresolved environmental and health crisis. Given the massive impact of haze episodes, more experimental and theoretical studies and a multidisciplinary research approach to smouldering peat fires are needed to develop both fundamental understanding and provide technological solutions to these overlooked fires and their associated haze phenomena.

#### **Conflicts of interest**

The authors declare they have no conflicts of interest.

#### Acknowledgements

The research was funded by the China Scholarship Council (CSC) and the European Research Council (ERC) Consolidator Grant HAZE (682587). The authors thank Eirik Christensen and Dr Francesco Restuccia from Imperial College London for valuable discussions. Data supporting this publication can be obtained from Hu Y and Rein G (2018) Review of emissions from smouldering peat fires [Data set]. Zenodo. http://doi.org/ 10.5281/zenodo.1216074 (Verified 10 April 2018).

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