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Development of gas signatures of smouldering peat wildfire from emission factors

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ABSTRACT

Smouldering peat fires are responsible for regional haze episodes and cause environmental, social and health crises. Owing to the unique burning characteristics of smouldering peat, identifying and detecting this kind of fire remains a challenge. This work explores smouldering peat gas signatures using emission factor (EF) data from literature. Systematic comparisons and statistical analyses were carried out to investigate 28 forms of EF combinations created from the four most abundant gas species: carbon dioxide (CO_2), methane (CH_4), carbon monoxide (CO) and ammonia, from smouldering peat, flaming savanna and grassland, agricultural residue and forest fires. Among the candidate gas signatures, the ratio of EF(CO_2) to EF(CH_4) for smouldering peat showed a significant improvement with statistically different ranges of values (134.6) compared to those from flaming savanna and grassland fire (940.2), agricultural residue fire (434.4), forest fire (368.8) and mixed burning peat fires (207.7). Additionally, we found that EF(CO_2)/EF(CH_4) is independent from fuel composition and could differentiate early ignition from the subsequent spread, making it the best gas signature among those analysed, including CO/CO_2 ratio and the Modified Combustion Efficiency. This work presents the first scientific endeavour developing smouldering gas signatures, contributing to the scientific understanding and remote sensing and early detection of smouldering peat wildfires.

Keywords: detection, emissions, fire, haze, mitigation, peatland, smouldering, wildfire.

Introduction

Peatlands[1] are the most widespread of all wetland types in the world, covering ~3% of the Earth's land surface (Yu 2012). They are formed in water-saturated environments and are usually of high moisture contents[2] (up to 300%) which serve as natural fire barriers (Turetsky *et al.* 2015). Natural droughts (e.g. El Niño) and human activities (e.g. agricultural drainage and peat harvesting) lower the water table in peatlands (Turetsky *et al.* 2015) and reduce the moisture content in peat (Kettridge *et al.* 2015), rendering them more vulnerable to fire. Uniquely, peat fire is dominated by smouldering combustion, a slow, low-temperature and flameless burning mode that differs from flaming combustion in terms of chemistry and heat transfer (Rein 2016). 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 2016). As a result, smouldering peat spreads and produces fundamentally different emissions from flaming wildfires (for example, grass and savanna fires) observed on the Earth. Flaming forest fires occurring across the continents (for example, in California (Fig. 1)) typically show strongly buoyant smoke plumes of dark colour, and flames spreading above the ground with high temperatures (~1500°C) for short periods of time (in the order of minutes) (Rein 2013).

[1]Peat is a carbon-rich organic soil that contains at least 12% organic carbon (20% organic material) with less than 20–35% mineral content (Belyea and Clymo 2001; Turetsky *et al.* 2015).
[2]Moisture content of peat is defined as the mass of water divided by the mass of a dried sample (%).

Fig. 1. Caldoro flaming forest fire occurred in California, on 29 August 2021. Intense flame and dark smoke plume was observed. Photograph: Xinhua/Reuters.

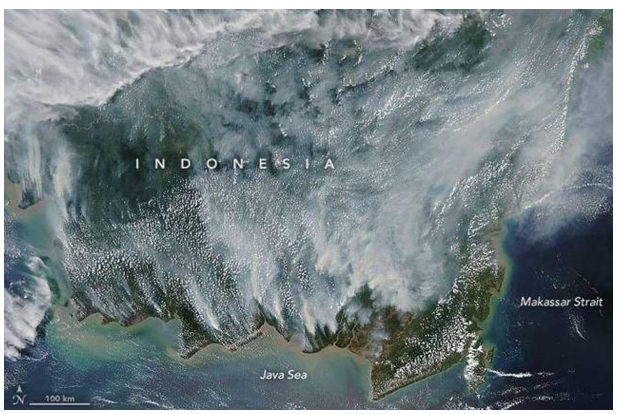

Fig. 2. NASA satellite image showing the extent of the haze on 14 September 2019. Thick grey smoke generated from fires in Borneo blanketed both islands and neighbouring regions.

In contrast, smouldering peat fires are frequently observed in tropical, temperate and boreal regions across the globe, and are characterised by weakly buoyant smoke plumes with a light colour that accumulate near the ground (Wiggins *et al.* 2018). These fires have a lower peak temperature (in the range of 450–700°C) and propagate both horizontally and vertically, spreading in a creeping fashion (typically two orders of magnitude slower than flaming fire spread) that can last weeks or even months (Rein 2013; Wiggins *et al.* 2018). In the presence of a large quantity of peat, a smouldering fire can be initiated with a weak ignition source and sustained for very long periods of time despite fire-fighting attempts, extensive rains or weather changes (Rein 2013). For example, in Indonesia, where slash and burn, a traditional farming method in which natural vegetation is cut down and burned, is widely used for clearing the land for palm plantations or agriculture (Cochrane 2003). Heat generated from these flames ignites the peat underneath, initiating long-lasting and persistent smouldering fires (Rein 2013; Kettridge *et al.* 2015).

Emissions from smouldering peat fires, the largest fires on Earth in terms of fuel consumption, are an important contributor of atmospheric carbon that drives climate change (Rein 2013). For example, it is estimated that the 1997 Indonesian peatland fires emitted 0.81–2.57 Gt carbon, equivalent to 13–40% of mean annual global carbon emissions from fossil fuels (Page *et al.* 2002). Peat fires are also the dominant source of aerosols, causing regional haze episodes especially under dry weather (Wiggins *et al.* 2018). Haze is the large-scale atmospheric accumulation of peat fire smoke at low altitudes (Fig. 2) (Hu *et al.* 2018*a*). Driven by wind, these emissions can migrate long distances and evolve into transboundary haze episodes. Haze is notorious for inducing surges of health emergencies among local populations, disrupting shipping and aviation routes for weeks or even months, and posing a major threat to public health and property. Among the regions suffering from haze, Southeast Asian countries frequently experience exceptional haze in response to fire season fuelled by El Niño-induced drought (Smith *et al.* 2018; Wiggins *et al.* 2018). For example, more than 28 000 000 people in Indonesia were affected by the Southeast Asian Haze event in 2015, from which more than 140 000 people reported respiratory illness (Hu *et al.* 2018*a*). It is estimated that this haze event led to economic losses of up to US$35 billion. Despite the severe effects of haze on people's health and property, it remains an unresolved environmental and health crisis and could evolve into regional disputes (Forsyth 2014).

Understanding the emission characteristics of peat burning can contribute to developing successful regional peat fire technologies (Rein 2016). Emission factor (EF), defined as the mass of species emitted per mass of dry fuel consumed (expressed as $g\,kg^{-1}$), is widely used in atmospheric chemistry modelling to quantify fire emissions and understand their influence on the atmosphere and climate change (Hu *et al.* 2018*a*). Several EFs derived from both laboratory peat fire experiments, for example, in Christian *et al.* (2003), Stockwell *et al.* (2016) and Hu *et al.* (2018*b*, 2019) and field measurements from Huijnen *et al.* (2016), Stockwell *et al.* (2016), Smith *et al.* (2018) and Wooster *et al.* (2018) with diverse peat origins and burning conditions were reported in recent decades. Compilations of the EFs of the various gas species from Andreae and Merlet (2001) and Akagi *et al.* (2011) to the most recent of Hu *et al.* (2018*a*) and Andreae (2019) have greatly expanded the amount of available data for emission inventory and furthered the scientific understanding of pollution from biomass fires.

Peat fires are monitored routinely across the globe with remote sensing technologies (e.g. satellite-based active fire observation) that were developed for flaming fires, and thus could be ineffective to detect smouldering peat fires due to their lower temperature and underground burning characteristics (Rein 2013; Wooster *et al.* 2018; Christensen *et al.* 2019; Sofan *et al.* 2019). Being an important layer of wildfire protection of ecosystems, early detection of smouldering

fires is critical in tackling peatland degradation and has attracted research interest to develop specific detection methods for smouldering. For example, there are studies investigating the feasibility using infrared cameras in drones to detect smouldering (Burke *et al.* 2019). In addition, particulate matter and levoglucosan were proposed as potential precursors and tracers of smouldering fires of wood and cotton (Madsen *et al.* 2018; Bluvshtein *et al.* 2020). Modified combustion efficiency (MCE), a signature for determining the importance of flaming or smouldering in a fire, is widely adopted in the literature to classify the combustion regimes of biomass burning (Christian *et al.* 2003; Akagi *et al.* 2011; Stockwell *et al.* 2014, 2016; Smith *et al.* 2018), but the validity of using MCE in differentiating smouldering peat remains incomplete (Hu *et al.* 2018*a*, 2019). In wildland, vegetation is commonly mixed. The co-existence of smouldering and flaming fires and the complex wildland environment pose a challenge of these signatures for fire identification.

In the present study, we study possible gas signatures for early detection of smouldering peat fire and differentiating this type of fire from flaming and the burning of the other wildfire fuels including grassland, forest and agricultural residue. This work consists of systematic exploration and statistical analysis of multiple combinations of gaseous species' EFs collected from the literature.

Methods

Emission factor dataset

To develop gas signatures for smouldering peat, a dataset of EFs of gaseous species from the four biomass fires (peat, savanna and grassland, forest and agricultural residue fires) was first built. When selecting the species included in the dataset, three criteria were considered: (I) the magnitude of the species in terms of EFs; (II) the amount of the species' EF data available in the literature; and (III) the difficulty of measuring the species. There are more than 100 trace gas species present in peat fire smoke plumes (Stockwell *et al.* 2015; Hu *et al.* 2018*a*). Among all the species detected and reported in the literature, carbon dioxide (CO_2), carbon monoxide (CO) and methane (CH_4) are the three most abundant gas species with EFs commonly larger than 10.0 g kg^{-1} (Stockwell *et al.* 2016; Hu *et al.* 2018*b*). Hu *et al.* (2019) found that the EFs of ammonia (NH_3) from smouldering peat were 10–20 times larger than those from flaming biomass, making it an important gas species for smouldering fires. In contrast, the majority of the other species from peat fires are nitric oxide (NO as NO_x), nitrogen dioxide (NO_2), hydrogen cyanide (HCN), sulfur dioxide (SO_2) and the non-methane organic compounds (NMOCs) which commonly have trace levels of EF-values (<1.0 g kg^{-1}) with vast uncertainty and inter-study variability (Akagi *et al.* 2011; Andreae 2019). Furthermore, studies which measured and reported EF data for those trace gas species are very limited ($n < 10$), resulting in an insufficient amount of data for statistical analysis. In terms of measurement difficulty, CO_2, CO, CH_4 and NH_3 can be measured more accurately and feasibly than the NMOCs in most biomass fire emission studies (Stockwell *et al.* 2014, 2016; Hu *et al.* 2018*b*; Smith *et al.* 2018). As a result, the EFs of CO_2, CO, CH_4 and NH_3 were selected for inclusion in the dataset.

The EF data of CO_2, CO, CH_4 and NH_3 from savanna and grassland, forest and agricultural residue fires were adopted directly from an updated compilation of EFs from Andreae (2019) which collected EFs from over 370 studies on various types of biomass burning. The EFs in the peat fire category were a combination from the peat fire EF compilation from Andreae (2019), a review on peat fire EFs (Hu *et al.* 2018*a*) and recent laboratory peat fire emission experiments (Hu *et al.* 2018*b*; Hu *et al.* 2019), jointly forming the most comprehensive up-to-date EFs dataset for peat fire.

In the literature, a carbon balance approach is widely used, especially in field measurements, to calculate $EF_{i,c}$ from biomass burning (Stockwell *et al.* 2016; Smith *et al.* 2018). This method entails the value of the fuel carbon content and assumes all carbon-containing emissions are measured (Eqn 1):

$$EF_{i,c} = F_c \times \frac{MM_i}{MM_c} \times \frac{C_i}{C_T} \quad (1)$$

where $EF_{i,c}$ is the emission factor of species i calculated from the carbon balance approach (g kg^{-1}), F_c is the carbon content of the fuel (%), MM_i is the molar mass of species i (g mol^{-1}), MM_c is the atomic mass of carbon (12 g mol^{-1}), C_i is the number of moles of species i and C_T is the total number of moles of carbon emitted.

For experiments conducted in laboratories where the mass loss of peat sample can be measured, a mass loss approach is commonly used to calculate $EF_{i,m}$ (Eqn 2) (Rein *et al.* 2009; Hu *et al.* 2019):

$$EF_{i,m} = \frac{\dot{m}_i''}{\dot{m}''} \quad (2)$$

where $EF_{i,m}$ is the emission factor of species i calculated from the mass loss approach (g kg^{-1}), $\dot{m}_i''$ is the mass flux of the released species i (g s^{-1} m^{-2}) and $\dot{m}''$ is the mass loss rate (fuel consumption rate) of dry peat (g s^{-1} m^{-2}).

In the compiled EF dataset, 20 independent peat fire studies that reported fire-averaged EFs were included, while 50, 39 and 85 studies were included for savanna and grassland, agricultural residue and forest fires, respectively. In total, 566 EF data of CO_2, CO, CH_4 and NH_3 derived from these four fire fuels originated across tropical, temperate and boreal regions are included in the dataset. Those EFs are either from laboratory experiments or field measurements, which are mutually complementary and are equally important in terms of understanding of accidental

wildfires (Christensen *et al.* 2019). Furthermore, both a carbon balance approach and a mass loss approach for calculating the EFs have been mutually verified in Hu *et al.* (2018*b*), thus all the $EF_{i,c}$ and $EF_{i,m}$ data collected from the literature were included in the dataset indiscriminately.

In some peat fire emission studies, a mixed burning of smouldering and flaming peat was observed in both laboratory experiments and field measurements (Huijnen *et al.* 2016; Stockwell *et al.* 2016; Hu *et al.* 2019). To distinguish pure smouldering peat, two sub-categories (mixed burning ($n = 7$) and pure smouldering ($n = 13$)) were defined and classified in the peat fire group for comparison purposes. Table 1 provides the categorised EFs of peat fires from the literature as well as the information of how these data were obtained. The information of the EFs from savanna and grassland, agricultural residue and forest fires were included and detailed in Andreae (2019) and the literature cited therein, and thus are not shown in this study.

EF comparisons and statistical analysis

Based on the EF dataset, four forms of EF combinations are proposed for systematic comparisons among biomass fires: (I) X (X refers to an individual chemical species (for example, CO_2)); (II) X/Y (Y refers to another single chemical species (for example, CO_2/CO)); (III) X/(X + Y) (for example, $CO_2/(CO_2 + CO)$); and (IV) (Z × Y)/(X × X) (Z refers to a third chemical species different from X and Y. For example, $(CO \times CH_4)/(CO_2 \times CO_2)$).

The rationale of proposing these four formats of EF combinations are described as follows: for (I), individual EF of these four gas species has been complied in multiple review papers (for example, in Andreae (2019), Andreae and Merlet (2001) and Akagi *et al.* (2011)) but has not been compared statistically across fire types; for (II), the format of EF combinations was inspired by an important signature used in combustion, the CO to CO_2 ratio (CO/CO_2), which is regarded as an index of combustion incompleteness (Rein *et al.* 2009;

Table 1. Categorised EFs of peat fires (mean values, g kg^{-1}) from the literature.

	Peat origin	EF determination methods	CO_2	CO	CH_4	NH_3
Pure smouldering peat						
Selimovic *et al.* (2018)	Indonesia	Carbon balance	–	–	10.4	–
Rein *et al.* (2009)	Edinburgh	Mass loss	420	170	–	–
Stockwell *et al.* (2014)	North Carolina	Carbon balance	1066	276	10.9	1.87
Geron and Hays (2013)	North Carolina	Carbon balance	1071	262	–	–
Stockwell *et al.* (2014)	Canada	Carbon balance	1274	197	6.25	2.21
Stockwell *et al.* (2015)	North Carolina, Canada, Indonesia	Carbon balance	1325	256	10.5	1.78
Wilson *et al.* (2015)	Ireland, United Kingdom	nr	1346	218	8.35	0.73
Chakrabarty *et al.* (2016)	Alaska	nr	1418	105.5	–	–
Chakrabarty *et al.* (2016)	Siberia	nr	1565	126.5	–	–
Smith *et al.* (2018)	Malaysia	Carbon balance	1616	257	11.26	8
Stockwell *et al.* (2014)	Kalimantan	Carbon balance	1637	233	12.8	1.39
Christian *et al.* (2003)	Sumatra Island	Carbon balance	1703	210.3	20.8	19.92
Hu *et al.* (2018*b*)	Ireland	Mass loss	1743	264	13.2	12.9
Mixed burning peat						
Black *et al.* (2016)	North Carolina	Carbon balance	861	108	–	–
Black *et al.* (2016)	North Carolina	Carbon balance	922	122	–	–
Yokelson *et al.* (1997)	Alaska and Minnesota	Carbon balance	1395	209	6.85	8.76
Stockwell *et al.* (2016)	Kalimantan	Carbon balance	1564	291	9.51	2.86
Huijnen *et al.* (2016)	Indonesia	Carbon balance	1594	255	7.4	–
Nara *et al.* (2017)	Sumatra Island	Carbon balance	1663	235	7.6	–
Wooster *et al.* (2018)	Indonesia	Carbon balance	1775	279	7.9	–
Flaming peat						
Hu *et al.* (2019)	Ireland	Mass loss	2017	41.7	9.2	0

Peat was categorised into pure smouldering peat, mixed burning peat and pure flaming peat. Peat origin (sampling locations detailed in each study) and the methods used to calculate the EF were given where applicable; nr: not reported.

Rein 2016); for (III), the format of EF combinations was inspired by the MCE whose calculation is built on the excess mole fractions of CO_2 and CO (Ward and Radke 1993) (Eqn 3):

$$\mathrm{MCE} = \frac{\Delta[CO_2]}{\Delta[CO_2] + \Delta[CO]} \quad (3)$$

where $\Delta[CO_2]$ and $\Delta[CO]$ are the excess mole fractions of CO_2 and CO, respectively.

For (IV), flaming wildfire exhibits larger EF-values for CO_2 (a complete combustion product) but smaller values for CO, CH_4 and NH_3 (typical incomplete combustion products) than those from smouldering peat fire (Akagi *et al.* 2011; Rein 2016; Andreae 2019). Inspired by this existing difference in terms of the species' properties, EF combination (IV) was introduced to magnify the EF difference between complete and incomplete species. Because CO_2 is the only complete combustion product among the four gas species selected, as such only CO_2 was used for 'X' in EF combination (III) and (IV).

To conduct a systematic statistical comparison of the EF combinations, the reciprocal functions of types (II) to (IV) were included, resulting in a total number of 28 combinations of inter-category EFs with their expressions shown in Table 2. It is worth noting that even just for CO_2, CO, CH_4 and NH_3, the forms and quantities of EF combinations can be endless. This work focuses on the four main types of EF combinations which have the advantage of simplicity in terms of expression forms and representativeness of biomass combustion characteristics.

Independent sample *t*-tests were conducted to statistically examine: (i) whether EFs from pure smouldering peat were significantly different from those of mixed burning peat and (ii) whether EFs from the combined 'peat fire' group (a combination of mixed burning peat and smouldering peat) were significantly different from the other three types of biomass fires included in this work. Briefly, *t*-test is a type of inferential statistics used to determine a significant difference between the means of two independent groups. It examines the null hypothesis (H_0): all individual batch means are equal ($\mu_1 = \mu_2$) (Ellison *et al.* 2009). Specifically, five *t*-tests (two-tailed) with a significance/alpha level (a, the probability of making the wrong decision when H_0 is true) of 0.05 ($a = 0.05$) were carried out with *t*-values and *P*-values calculated and reported for all EF combinations types (I) to (IV): (i) peat fire vs savanna and grassland fire; (ii) peat fire vs agricultural residue fire; (iii) peat fire vs forest fire; (iv) peat fire vs the combined 'biomass fire' group (a combination of savanna and grassland, forest and agricultural residue fires, excluding peat fire data); and (v) mixed burning peat vs pure smouldering peat.

Calculating *t*-values entails the average EF, the variance and the number of samples within each of the two compared groups. Overall, a *t*-value measures the size of the difference relative to the variation in the sample data (calculated difference represented in units of standard error) (Ellison *et al.* 2009). Generally, the larger the *t*-value is, the higher chance that no relationship exists between/among measured groups. The *P*-value is a measure of the probability that an observed difference could have occurred just by random chance. The *P*-value being less than the significance level (in this work, 0.05) means that the result is significant and therefore, the null hypothesis H_0 can be rejected since the probability of H_0 being true is very low (Ellison *et al.* 2009). In other words, $P < 0.05$ indicates a significant difference between the two compared fire categories. In this work, statistical analysis tool 'QuickCalcs' from GraphPad Software Inc. (2022) was used for calculating *P*-values from *t*-tests.

In addition, one-way analysis of variance (ANOVA) with a significance level of 0.05 was conducted among fire categories. ANOVA is a collection of methods for comparing multiple means across different groups, it examines the following hypotheses. The null hypothesis (H_0): all individual batch means are equal ($\mu_1 = \mu_2 = \ldots\mu_k$); and the alternative hypothesis (H_a): at least one batch mean is not equal to the others. The results of ANOVA determine whether there is any significant difference between the means of three or more independent groups (Ellison *et al.* 2009).

Table 2. Expressions of possible signatures included in this study.

EF combination types				
(I)	(II)		(III)	(IV)
X	X/Y		X/(X + Y)[A,B]	(Z × Y)/(X × X)[A,B]
	CO/NH_3	NH_3/CO	$CO_2/(CO_2 + CH_4)$	$(CO \times CH_4)/(CO_2 \times CO_2)$
CO_2	CH_4/NH_3	NH_3/CH_4	$CO_2/(CO_2 + NH_3)$	$(CH_4 \times NH_3)/(CO_2 \times CO_2)$
CO	CO_2/NH_3	NH_3/CO_2	$CO_2/(CO_2 + CO)$	$(CO \times NH_3)/(CO_2 \times CO_2)$
CH_4	CO_2/CH_4	CH_4/CO_2	$(CO_2 + NH_3)/CO_2$	$(CO_2 \times CO_2)/(CO \times CH_4)$
NH_3	CO/CH_4	CH_4/CO	$(CO_2 + CO)/CO_2$	$(CO_2 \times CO_2)/(CH_4 \times NH_3)$
	CO_2/CO	CO/CO_2	$(CO_2 + CH_4)/CO_2$	$(CO_2 \times CO_2)/(CO \times NH_3)$

[A]Reciprocals of the expressions of EF combinations were also included.
[B]CO_2 is the only complete combustion products among the gas species selected, as such only CO_2 was used for proxy 'X' in EF combinations (III) and (IV).

An ANOVA test computes a test statistic, the *F*-value, which is a ratio of variability between groups and variability within groups. The larger the *F*-value is, the higher chance that no relationship exists among measured groups. Similar to *t*-tests, in ANOVA test, *P*-value being smaller than the significance level (0.05 in this work) means that H_0 can be rejected. In other words, $P < 0.05$ indicates that at least one fire category shows a significant difference in terms of the compared signature among the compared fire categories. 'QuickCalcs' (GraphPad Software Inc. 2022) was used for the determination of *P*-values, *F*-value and degrees of freedom (DF) between groups and within groups from ANOVA tests conducted in this work.

Results and discussion

Individual gas species

Individual gas species (CO_2, CO, CH_4 and NH_3) were first compared among peat, savanna and grassland, agricultural residue and forest fires, forming an overview of the distribution, value range, variability and the mean values of the EFs (Fig. 3). To differentiate smouldering from mixed burning, the peat fire group was divided into three individual sub-groups: pure smouldering, mixed burning and a pure flaming groups. To the best knowledge of the authors, only one source of pure flaming peat EF data is available in the literature (Hu *et al.* 2019), and thus is included for comparison purposes.

The EFs of CO_2, CO, CH_4 and NH_3 showed large variability for all fire types. To quantify the inter-fire EF differences, the number of data available (*n*), mean value and standard deviation (s.d.) of the EFs from the literature are summarised in Table 3. Averagely, peat fire exhibits a ~10% lower mean value of CO_2 EF (1398.8 ± 378.3 g kg^{-1}) and 123, 129 and 410% higher mean values of EFs of CO (205.8 ± 70.6 g kg^{-1}), CH_4 (10.2 ± 3.6 g kg^{-1}) and NH_3 (5.50 ± 6.29 g kg^{-1}) than the other types of 'biomass fires' (the combined dataset of savanna and grassland, forest and agricultural residue fires), respectively. Within the peat fire category, the pure smouldering group showed an 8.5% lower CO_2 EF but 11.4%, 43.6% and 57.8% higher EFs for CO, CH_4 and NH_3 from mixed burning peat, respectively. These findings correspond with the fact that peat fire is dominated by smouldering, which yields more incomplete combustion species than from flaming fires (Rein 2016). The only flaming peat EF data (Hu *et al.* 2019), on the other hand, showed a significantly higher EF of CO_2 (56.2%) and

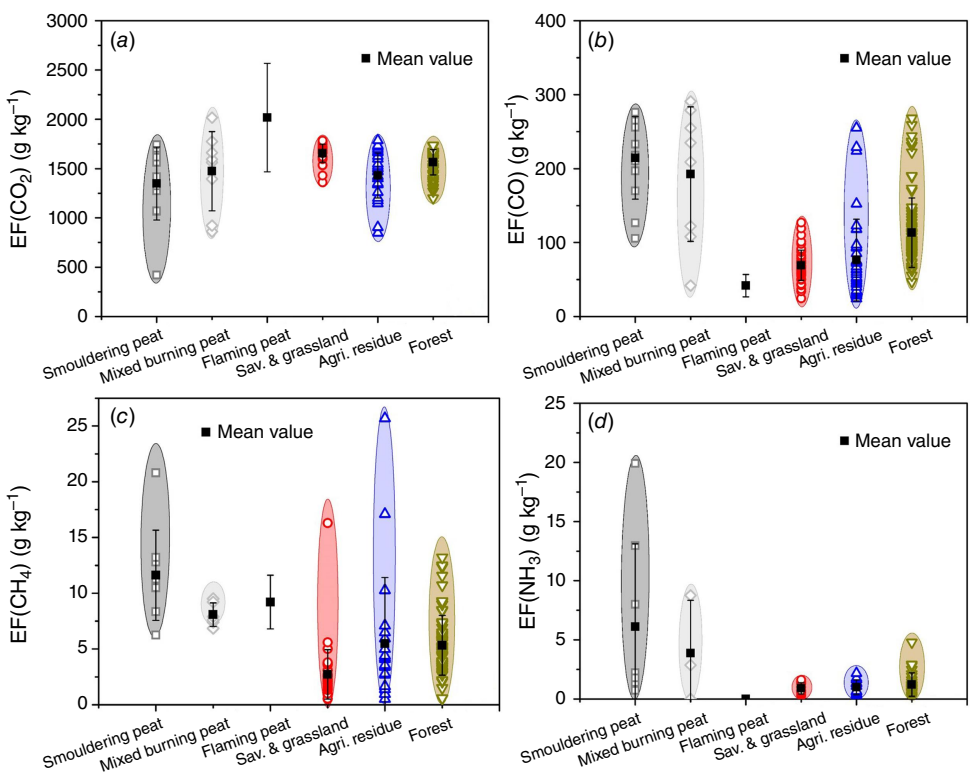

Fig. 3. Scatter plots of EFs of CO_2 (*a*), CO (*b*), CH_4 (*c*) and NH_3 (*d*) (EF combination (I)) from different biomass fire types. Peat fire was divided into a pure smouldering group, a mixed burning group and a pure flaming group for comparison purposes. Mean value from each fire category is shown as a black square with the standard deviation shown as an error bar. The ranges of the EF-values are shown as clouds.

Table 3. Summary of EF data of individual species from different biomass fires.

	CO_2		CO		CH_4		NH_3	
	n	Mean (s.d.)	*n*	Mean (s.d.)	*n*	Mean (s.d.)	*n*	Mean (s.d.)
Smouldering peat	12	1348.7 (370.8)	12	214.6 (55.9)	9	11.61 (4.05)	8	6.11 (7.01)
Mixed burning peat	8	1473.9 (401.9)	8	192.6 (91.0)	6	8.08 (1.05)	3	3.87 (4.47)
Peat fire[A]	20	1398.8 (378.3)	20	205.8 (70.6)	15	10.20 (3.60)	11	5.50 (6.29)
Savanna fire	31	1655.6 (92.1)	50	69.2 (20.5)	49	2.71 (2.21)	16	0.89 (0.49)
Forest fire	63	1565.7 (128.4)	85	113.3 (47.0)	73	5.33 (2.68)	30	1.22 (0.99)
Agri. residue fire	29	1431.5 (229.0)	39	76.3 (55.3)	21	5.49 (5.93)	14	0.98 (0.63)
Biomass fire (peat excluded)[B]	123	1556.7 (169.6)	174	92.3 (47.9)	143	4.46 (3.44)	60	1.08 (0.80)

[A]Peat fire refers to the combination of all peat fire data listed in this paper.
[B]Biomass fire refers to the combination of savanna fire, forest fire and agricultural residue fire listed in this paper.

Table 4. Statistical analysis of individual EF from different biomass fires (EF combination type I).

	CO_2		CO		CH_4		NH_3	
	t-value	*P*-value[A]	*t*-value	*P*-value	*t*-value	*P*-value	*t*-value	*P*-value
Smouldering vs mixed burning peat	0.7039	0.4905	−0.6122	0.5480	−2.4923	0.0270	−0.6244	0.5479
Peat fire vs savanna fire	2.9793	0.0045	−8.5149	<0.0001	−7.6108	<0.0001	−2.4238	0.0229
Peat fire vs forest fire	1.9381	0.0561	−5.5752	<0.0001	−4.9532	<0.0001	−2.2462	0.0304
Peat fire vs agri. residue fire	0.3450	0.7317	−7.1583	<0.0001	−2.9543	0.0057	−2.3712	0.0266
Peat fire vs biomass fire (peat excluded)	1.8371	0.0683	−7.0064	<0.0001	−5.8910	<0.0001	−2.3278	0.0229
	F-value	*P*-value	*F*-value	*P*-value	*F*-value	*P*-value	*F*-value	*P*-value
Peat vs sav. vs agri. vs for.[B]	10.1092	<0.0001	46.3808	<0.0001	21.1044	<0.0001	9.4433	<0.0001

[A]$P < 0.05$ indicates a significant difference between/among compared fire groups.
[B]Peat fire vs savanna fire vs agricultural residue fire vs forest fire.

lower EF of CO (80.0%), CH_4 (20.6%) and NH_3 (~100%)[3] than those from smouldering peat. More studies on flaming peat are needed to improve understanding of the performance and influence of its EF data.

Table 4 summarises the statistical analysis results of the individual EF. CO_2 showed a significantly different EF from peat fire ($n = 20$) than from savanna and grassland fire ($n = 31$) ($P = 0.0045$). However, the EF(CO_2) did not differ between smouldering peat ($n = 12$) and mixed burning peat ($n = 8$) with $P = 0.4905$. Furthermore, CO_2 did not differentiate peat fire from either forest fire or agricultural residue fire ($P > 0.05$). Compared with CO_2, CO performed well in distinguishing peat fire from all the other biomass fire types with $P < 0.0001$. However, EF(CO) did not differ between smouldering peat ($n = 12$) and mixed burning peat ($n = 8$) ($P = 0.5480$). In contrast, CH_4, a gas species with exceptionally high EF from fire compared to natural decomposition of biomass (Akagi *et al.* 2011; Turetsky *et al.* 2015), showed outstanding performance in distinguishing the following fire categories: pure smouldering peat ($n = 9$) from mixed burning peat ($n = 6$) ($P = 0.027$); the combination of peat fire ($n = 15$) from savanna and grassland fire ($n = 49$) ($P < 0.0001$), from forest fire ($n = 73$) ($P < 0.0001$), from agricultural residue fire ($n = 21$) ($P = 0.0057$) and from the other 'biomass fire' ($n = 143$) ($P < 0.0001$). NH_3, a species found to be uniquely high from smouldering peat (Hu *et al.* 2018*b*; Hu *et al.* 2019), showed a significantly different EF from peat fire than from the other biomass fires ($P < 0.05$). Similar to CO, NH_3 failed to differ smouldering peat ($n = 8$) from mixed burning peat ($n = 3$) ($P = 0.5479$), possibly due to its large EF variability within and between the two peat fire groups (Hu *et al.* 2018*a*; Smith *et al.* 2018). The *P*-values from the ANOVA tests were below 0.0001 for all four gas species, indicating that at least one type of fire (not necessarily peat fire) had a significantly different EF to the other fires.

There was significant variability of the EF of individual species within each fire type (Fig. 3). Individual gas species like CH_4 showed ideal performance in differentiating pure smouldering peat from mixed burning peat and the other biomass fires. However, natural variability of the physical properties of the fuels (e.g. compositions, density and moisture content) can greatly affect the values of EFs reported in each study (for example, peats with higher carbon/nitrogen content have higher EFs values for carbonaceous/nitrogenous

[3]In Hu *et al.* (2019), no NH_3 emission was detected from flaming peat, the EF of NH_3 was reported as 0 g kg^{-1}.

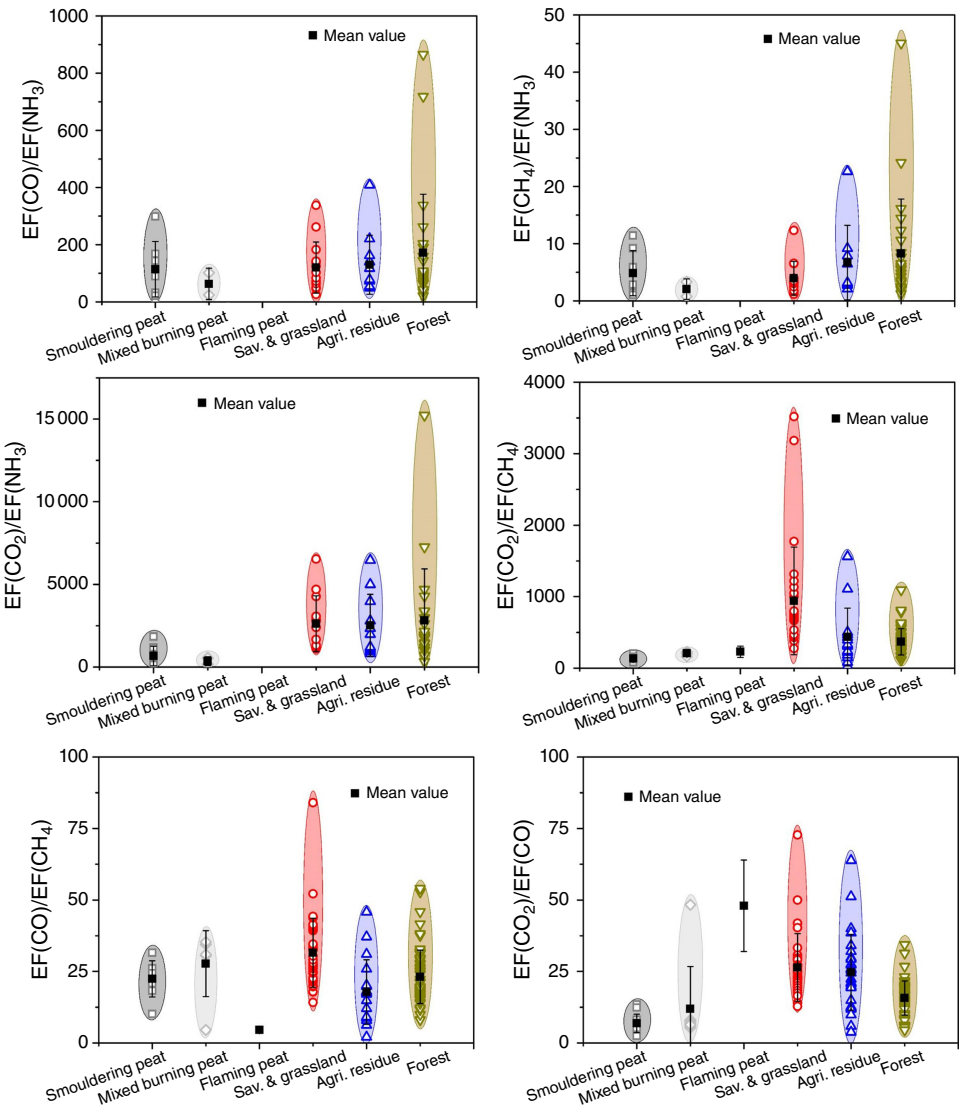

Fig. 4. Scatter plots of six EF ratios between two chemical species (EF combination (II)) from peat fire, savanna fire, forest fire and agricultural residue fire. Mean value from each fire category is shown as a black square with the standard deviation shown as an error bar. The ranges of the ratios are shown as clouds.

species) (Stockwell *et al.* 2016; Hu *et al.* 2018*a*; Smith *et al.* 2018). Furthermore, the wildland fuel map is randomly inhomogeneous and localised transient fire emissions are subjected to complex topography and changing weather (e.g. rainfall and wind) (Smith *et al.* 2018). As a result, large uncertainty can be introduced when using a single species in determining the types of wildland fires, thus individual species are not recommended as smouldering signatures.

EF ratio between two chemical species

Dividing one chemical species by another, a dimensionless EF ratio (EF combination type (II)) is proposed. In this subsection, a total number of 12 EF ratios – EF(CO)/EF(NH_3), EF(CH_4)/EF(NH_3), EF(CO)/EF(CH_4), EF(CO_2)/EF(NH_3), EF(CO_2)/EF(CH_4), EF(CO_2)/EF(CO) and their corresponding reciprocals – were generated, calculated and compared using the EF data from the literature. Fig. 4 shows the original six EF ratios. Qualitatively, the value ranges of EF(CO)/EF(NH_3), EF(CH_4)/EF(NH_3) and EF(CO)/EF(CH_4) from peat fire largely overlapped with savanna and grassland fire, agricultural residue fire and forest fire, while EF(CO_2)/EF(NH_3), EF(CO_2)/EF(CH_4) and EF(CO_2)/EF(CO) from peat fire presented distinct value ranges.

The reciprocals of these six original EF ratios are shown in Fig. 5. Differing from the distribution of the original EF ratios, only the ranges of EF(NH_3)/EF(CO_2) and EF(CO)/EF(CO_2) from peat fire differed significantly among the other fires. Statistical analysis was used to further investigate the performance of the proposed EF ratios.

Table 5 summarises the number of data available, the mean value and the s.d. of 12 EF ratios from each fire category. Compared with the combination of all biomass fires (peat fire excluded) listed in this paper, all six original EF ratios from peat fire showed lower mean values: the percentage difference of mean values ranged from 2.01%

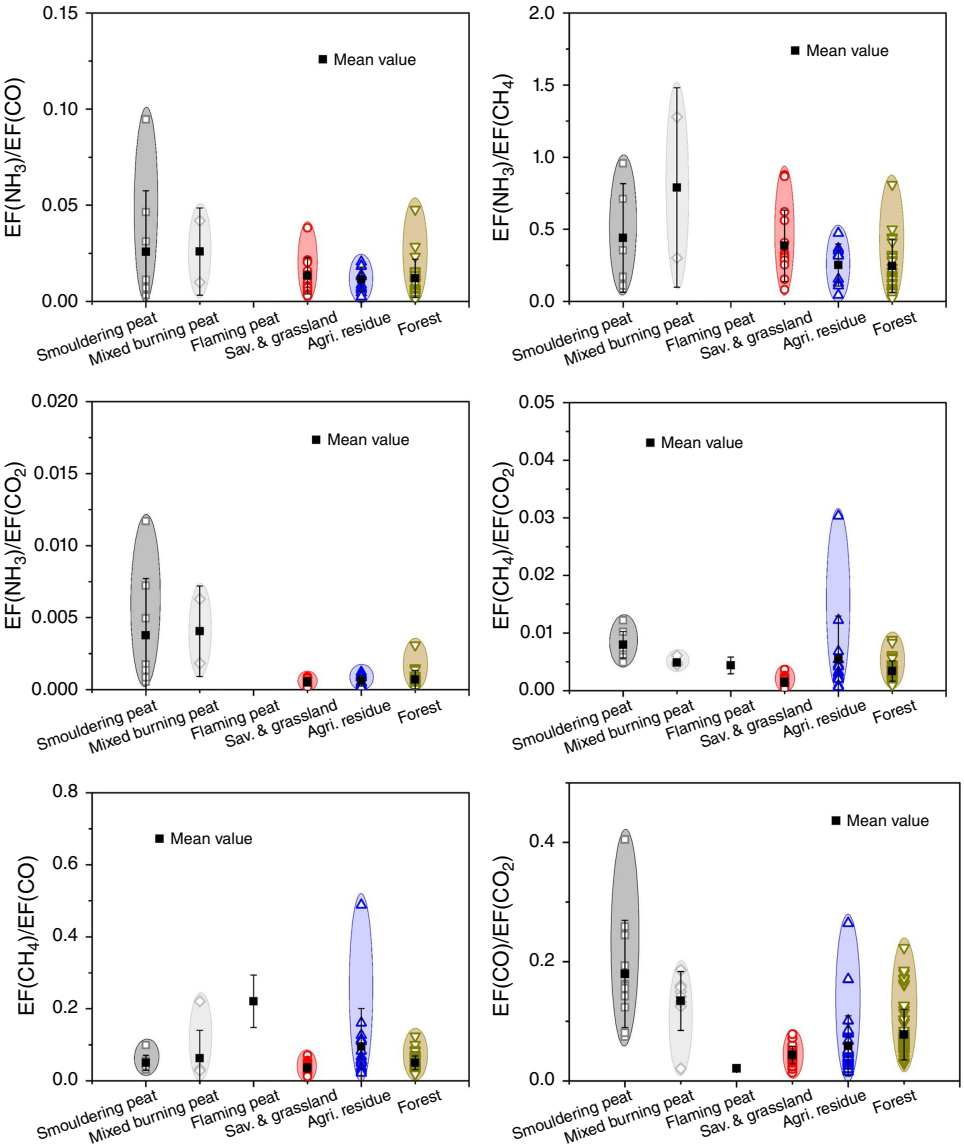

Fig. 5. Scatter plots of the reciprocals of the six EF ratios from EF combination (II) included in Fig. 4 from peat fire, savanna fire, forest fire and agricultural residue fire. Mean value from each fire category is shown as a black square with the standard deviation shown as an error bar. The ranges of the ratios are shown as clouds.

($EF(CO)/EF(CH_4)$) to 126.71% ($EF(CO_2)/EF(NH_3)$). Six corresponding reciprocals of the EF ratios had higher mean values for peat fire than those for the other biomass fires, with percentage differences ranging from 5.61% ($EF(CH_4)/EF(CO)$) to 120.0% ($EF(NH_3)/EF(CO_2)$).

When comparing peat fire with savanna fire, forest fire and agricultural residue fire, the mean values from peat fire were the smallest for $EF(CO)/EF(NH_3)$, $EF(CO_2)/EF(NH_3)$, $EF(CO_2)/EF(CH_4)$ and $EF(CO_2)/EF(CO)$. Peat fire had a larger mean value of $EF(CH_4)/EF(NH_3)$ (4.28 ± 3.73) than for savanna fire (3.96 ± 2.96). The average of $EF(CO)/EF(CH_4)$ from peat fire (24.63 ± 8.99) was larger than those from forest fire (23.07 ± 9.34) and agricultural residue fire (17.78 ± 11.30), while the mean value of its reciprocal, $EF(CH_4)/EF(CO)$ (0.055 ± 0.051), was smaller than from agricultural residue fire (0.095 ± 0.105). The mean values of $EF(NH_3)/EF(CO)$, $EF(NH_3)/EF(CH_4)$, $EF(NH_3)/EF(CO_2)$, $EF(CH_4)/EF(CO_2)$ and $EF(CO)/EF(CO_2)$ from peat fire were the largest among all fire types in this study.

Within the peat fire category, the mean values of $EF(CO)/EF(NH_3)$, $EF(CH_4)/EF(NH_3)$ and $EF(CO_2)/EF(NH_3)$ from the pure smouldering group exceeded those from the mixed burning group with percentage differences of 57.8%, 80.8% and 61.6%, respectively. In contrast, the reciprocals of $EF(CO)/EF(NH_3)$ and $EF(CO_2)/EF(NH_3)$ showed equal mean values between the pure smouldering and mixed burning groups. It is worth noting that the amount of NH_3 data available for mixed burning peat was limited (n = 3), and more experimental studies are needed to understand the rationale of NH_3 emissions (Rein 2016; Hu *et al.* 2019).

Table 6 summarises the results of the statistical analysis of the EF ratios. $EF(CO)/EF(NH_3)$, $EF(CH_4)/EF(NH_3)$, $EF(CO)/EF(CH_4)$ and their three reciprocals, all failed to differentiate peat fire from the biomass fire group (peat

Table 5. Summary of EF ratios between two chemical species (EF combination type II) from different biomass fires.

	CO/NH_3		CH_4/NH_3		CO_2/NH_3		CO_2/CH_4		CO/CH_4		CO_2/CO	
	n	Mean (s.d.)	*n*	Mean (s.d.)	*n*	Mean (s.d.)	*n*	Mean (s.d.)	*n*	Mean (s.d.)	*n*	Mean (s.d.)
Smouldering peat	8	113.9 (96.8)	8	4.83 (3.95)	8	667.3 (598.0)	8	134.6 (37.5)	8	22.41 (6.33)	12	6.825 (3.235)
Mixed burning peat	2	62.8 (55.1)	2	2.05 (1.80)	2	353.0 (274.1)	6	207.7 (22.3)	6	27.73 (11.55)	8	11.952 (14.728)
Peat fire	10	103.5 (90.0)	10	4.28 (3.73)	10	604.1 (551.7)	14	165.7 (48.7)	14	24.63 (8.99)	20	8.884 (9.621)
Savanna fire	14	119.9 (89.2)	15	3.96 (2.96)	13	2624.9 (1688.0)	28	940.2 (754.0)	42	31.51 (12.08)	31	26.367 (11.853)
Forest fire	24	171.1 (205.6)	24	8.26 (9.55)	23	2811.4 (3123.5)	55	368.8 (184.5)	66	23.07 (9.34)	62	15.681 (5.968)
Agri. residue fire	12	129.4 (103.2)	9	6.68 (6.51)	11	2525.2 (1878.8)	15	434.4 (403.4)	18	17.78 (11.30)	29	24.727 (13.294)
Biomass fire (peat exc.)	50	146.8 (157.8)	48	6.62 (7.62)	47	2692.8 (2488.3)	98	542.1 (515.0)	126	25.13 (11.59)	122	20.546 (10.894)
	NH_3/CO		NH_3/CH_4		NH_3/CO_2		CH_4/CO_2		CH_4/CO		CO/CO_2	
	n	Mean (s.d.)	*n*	Mean (s.d.)	*n*	Mean (s.d.)	*n*	Mean (s.d.)	*n*	Mean (s.d.)	*n*	Mean (s.d.)
Smouldering peat	8	0.026 (0.031)	8	0.439 (0.377)	8	0.004 (0.004)	8	0.008 (0.002)	8	0.050 (0.021)	12	0.18 (0.09)
Mixed burning peat	2	0.026 (0.023)	2	0.790 (0.692)	2	0.004 (0.003)	6	0.005 (0.001)	6	0.063 (0.077)	8	0.13 (0.05)
Peat fire	10	0.026 (0.029)	10	0.512 (0.433)	10	0.004 (0.004)	14	0.007 (0.002)	14	0.055 (0.051)	20	0.16 (0.08)
Savanna fire	14	0.013 (0.009)	15	0.385 (0.247)	13	0.001 (0.001)	28	0.001 (0.001)	42	0.036 (0.012)	31	0.04 (0.01)
Forest fire	24	0.012 (0.010)	24	0.244 (0.182)	23	0.001 (0.001)	55	0.003 (0.002)	66	0.050 (0.019)	62	0.08 (0.04)
Agri. residue fire	12	0.012 (0.006)	9	0.252 (0.146)	11	0.001 (0.001)	15	0.005 (0.007)	18	0.095 (0.105)	29	0.06 (0.05)
Biomass fire (peat exc.)	50	0.012 (0.009)	48	0.289 (0.206)	47	0.001 (0.001)	98	00.003 (0.003)	126	0.052 (0.046)	122	0.06 (0.04)

Table 6. Statistical analysis of EF ratios between two chemical species (EF combination type II) from different biomass fires.

	NH_3/CO		NH_3/CH_4		NH_3/CO_2		CH_4/CO_2		CH_4/CO		CO/CO_2	
	t-value	*P*-value	*t*-value	*P*-value	*t*-value	*P*-value	*t*-value	*P*-value	*t*-value	*P*-value	*t*-value	*P*-value
Smouldering vs mixed burning peat	0.0029	0.9978	0.6911	0.5091	0.1113	0.9144	−3.6362	0.0034	0.4036	0.6933	−1.4503	0.1642
Peat vs savanna	−1.3357	0.1939	−0.8393	0.4101	−2.8482	0.0096	−8.0292	<0.0001	−1.4377	0.1556	−6.6739	<0.0001
Peat vs forest	−1.4968	0.1434	−1.8861	0.0684	−2.7025	0.0111	−4.8357	<0.0001	−0.3936	0.6976	−4.5716	<0.0001
Peat vs agri. residue	−1.5511	0.1366	−1.7886	0.0914	−2.7701	0.0122	−0.5750	0.5701	1.4032	0.1708	−5.1728	<0.0001
Peat vs biomass (peat excluded)	−1.4890	0.1416	−1.5856	0.1186	−2.7667	0.0076	−4.8554	<0.0001	−0.2635	0.7953	−5.4150	<0.0001
	F-value	*P*-value	*F*-value	*P*-value	*F*-value	*P*-value	*F*-value	*P*-value	*F*-value	*P*-value	*F*-value	*P*-value
Peat vs sav. vs agri. vs for.	2.6313	0.0590	3.1456	0.0323	11.3995	<0.0001	10.8614	<0.0001	8.1404	<0.0001	28.3642	<0.0001
	CO/NH_3		CH_4/NH_3		CO_2/NH_3		CO_2/CH_4		CO/CH_4		CO_2/CO	
	t-value	*P*-value	*t*-value	*P*-value	*t*-value	*P*-value	*t*-value	*P*-value	*t*-value	*P*-value	*t*-value	*P*-value
Smouldering vs mixed burning peat	−0.9853	0.3533	−1.4734	0.1789	−1.0957	0.3051	4.5435	0.0007	1.0176	0.3289	0.9691	0.3454
Peat vs savanna	0.4399	0.6643	−0.2272	0.8224	4.0448	0.0006	5.4131	<0.0001	2.2638	0.0276	5.7762	<0.0001
Peat vs forest	1.3327	0.1920	1.7492	0.0899	3.2737	0.0026	7.2351	<0.0001	−0.5841	0.5608	2.9799	0.0038
Peat vs agri. residue	0.6279	0.5372	0.9731	0.3442	3.2411	0.0043	2.5602	0.0164	−1.9071	0.0661	4.8383	<0.0001
Peat vs biomass (peat excluded)	1.1947	0.2371	1.4531	0.1518	5.1867	<0.0001	7.0192	<0.0001	0.1921	0.8479	4.9278	<0.0001
	F-value	*P*-value	*F*-value	*P*-value	*F*-value	*P*-value	*F*-value	*P*-value	*F*-value	*P*-value	*F*-value	*P*-value
Peat vs sav. vs agri. vs for.	0.6422	0.5925	1.4520	0.2385	2.2582	0.0921	14.7700	<0.0001	9.0393	<0.0001	19.1054	<0.0001

excluded), as well as smouldering peat from mixed burning peat, with $P > 0.1$. $EF(CO_2)/EF(NH_3)$, $EF(CO_2)/EF(CO)$ and their two reciprocals performed well in distinguishing peat fire from savanna fire, forest fire, agricultural residue fire or their combinations, with $P < 0.05$. These four signatures failed to show significantly different EF ratio values between smouldering peat and mixed burning peat ($P > 0.1$). However, $EF(CH_4)/EF(CO_2)$ differentiated well between smouldering peat and mixed burning peat ($P = 0.0034$), and between peat fire and savanna fire ($P < 0.001$) and forest fire ($P < 0.001$), but failed to differentiate peat fire from agricultural residue fire ($P = 0.5701$).

Differing from the other EF ratios analysed above, $EF(CO_2)/EF(CH_4)$ showed outstanding performance in differentiating smouldering ($n = 8$) from mixed burning ($n = 6$), with $P = 0.0007$. It distinguished peat fire ($n = 14$) well from savanna fire ($n = 28$) with $P < 0.0001$, from forest fire ($n = 55$) with $P < 0.0001$, from agricultural residue fire ($n = 15$) with $P = 0.0164$ and from the combination of all the other biomass fires ($n = 98$) with $P < 0.0001$, making it a promising gas signature for smouldering peat.

The EF ratio between two gaseous species leaves out the carbon content of the fuel, which is integral in the calculation of individual EF using the carbon balance approach detailed in Eqn 1, thus overcoming the limitation in terms of heterogeneous fuel properties (Hu *et al.* 2019). This advantage gives the EF ratio, particularly $EF(CO_2)/EF(CH_4)$, a wide application as an effective gas signature in the wildland environment.

Modified EF ratios

Following the expression form of the MCE, $EF(CO_2)/(EF(CO_2) + EF(CO))$ and its reciprocal were firstly proposed and examined. Replacing CO with CH_4 and NH_3, the two important gas species from smouldering peat (Akagi *et al.* 2011; Hu *et al.* 2018*b*), $EF(CO_2)/(EF(CO_2) + EF(CH_4))$ and $EF(CO_2)/(EF(CO_2) + EF(NH_3))$ were introduced. Together with their corresponding reciprocals, the values from the six forms of modified EF ratios were compared and are shown in Fig. 6. In general, peat fire groups showed distinct value ranges of the six signatures compared to those from savanna and grassland fire, agricultural residue fire and forest fire. Differing from $EF(CO_2)/(EF(CO_2) + EF(CO))$ and $EF(CO_2)/(EF(CO_2) + EF(CH_4))$, which showed small variability, $EF(CO_2)/(EF(CO_2) + EF(NH_3))$ and its reciprocal exhibited significant variability for the ratio values within the peat fire group. This was possibly because the EF of NH_3 differed greatly among peat fire studies, with range 0.73–19.92 g kg^{-1} (Smith *et al.* 2018; Hu *et al.* 2019). Large variability was observed for $EF(CO_2)/(EF(CO_2) + EF(CH_4))$ from agricultural residue fires.

The performance of the six modified EF ratios from EF combinations (IV) differentiating fire types was examined and is shown in Fig. 7. The peat fire group showed value ranges distinct from those from savanna and grassland fire, agricultural residue fire and forest fire. Significant variability was observed for $(EF(CH_4) \times EF(NH_3))/(EF(CO_2) \times EF(CO_2))$ and $(EF(CO) \times EF(NH_3))/(EF(CO_2) \times EF(CO_2))$ ratio values from peat fire. In contrast, when NH_3 was used as part of the denominator, the range of the ratio values of $(EF(CO_2) \times EF(CO_2))/(EF(CH_4) \times EF(NH_3))$ and $(EF(CO_2) \times EF(CO_2))/(EF(CO) \times EF(NH_3))$ decreased. This was possibly due to the higher degree of EF variability for NH_3 among peat fire studies (the maximum $EF(NH_3)$ reported in the literature is ~27 times higher than the minimum $EF(NH_3)$, while for CO_2, CO and CH_4, the multiple difference between the maximum and minimum values in terms of the EFs are 4.0, 2.7 and 3.3, respectively) (Hu *et al.* 2019).

Table 7 summarises the mean values and s.d. from the 12 signatures (six EF ratios from EF combination type (III) and (IV), respectively). For six EF ratios from combination type (III), $EF(CO_2)/(EF(CO_2) + EF(NH_3))$ and its reciprocal showed the same mean values (0.999 and 1.001, respectively) among savanna and grassland fire, agricultural residue fire and forest fire, while the values from peat fire differed between groups. This is because the $EF(NH_3)$ is 2–3 orders of magnitude smaller than for $EF(CO_2)$ (Akagi *et al.* 2011; Andreae 2019). For $EF(CO_2)/(EF(CO_2) + EF(CO))$, the mean value from the peat fire group (0.865) was the lowest among all fire groups and presented a ~8% smaller value than the combined biomass dataset of savanna fire, forest fire and agricultural residue fire. Within the peat fire group, the ratio mean values of $EF(CO_2)/(EF(CO_2) + EF(NH_3))$, $EF(CO_2)/(EF(CO_2) + EF(CH_4))$ and $EF(CO_2)/(EF(CO_2) + EF(CO))$ from pure smouldering peat were slightly smaller (~1.0–3.5%) than those from mixed burning peat.

For EF combination type (IV), the mean values of $(EF(CO) \times EF(CH_4))/(EF(CO_2) \times EF(CO_2))$, $(EF(NH_3) \times EF(CH_4))/(EF(CO_2) \times EF(CO_2))$ and $(EF(CO) \times EF(NH_3))/(EF(CO_2) \times EF(CO_2))$ from the peat fire group were at least one order of magnitude higher, while the reciprocals of these three EF ratios were at least one order of magnitude lower than those from the biomass fire group (peat excluded). Within the peat fire group, $(EF(CO) \times EF(CH_4))/(EF(CO_2) \times EF(CO_2))$ and its reciprocal showed significantly different mean values (by one order of magnitude) between pure smouldering and mixed burning peat.

Table 8 summarises the performance of the 12 modified EF ratios. For EF combination type (III), $EF(CO_2)/(EF(CO_2) + EF(CO))$ and $EF(CO_2)/(EF(CO_2) + EF(NH_3))$ showed significantly different ratio values from peat fire than those from the other biomass fires analysed ($P < 0.05$). However, these EF ratios failed to differentiate smouldering peat from mixed burning peat, with $P > 0.5$. In addition, $EF(CO_2)/(EF(CO_2) + EF(CH_4))$ and its reciprocal differed between smouldering and mixed burning peat ($P = 0.0032$) but failed to distinguish between peat fire and agricultural residue fire, with $P > 0.5$.

For EF combination type (IV), $(EF(CO) \times EF(CH_4))/(EF(CO_2) \times EF(CO_2))$ showed significantly different ratios between pure smouldering peat and mixed burning peat ($P = 0.0169$); however, it failed to differentiate peat fire

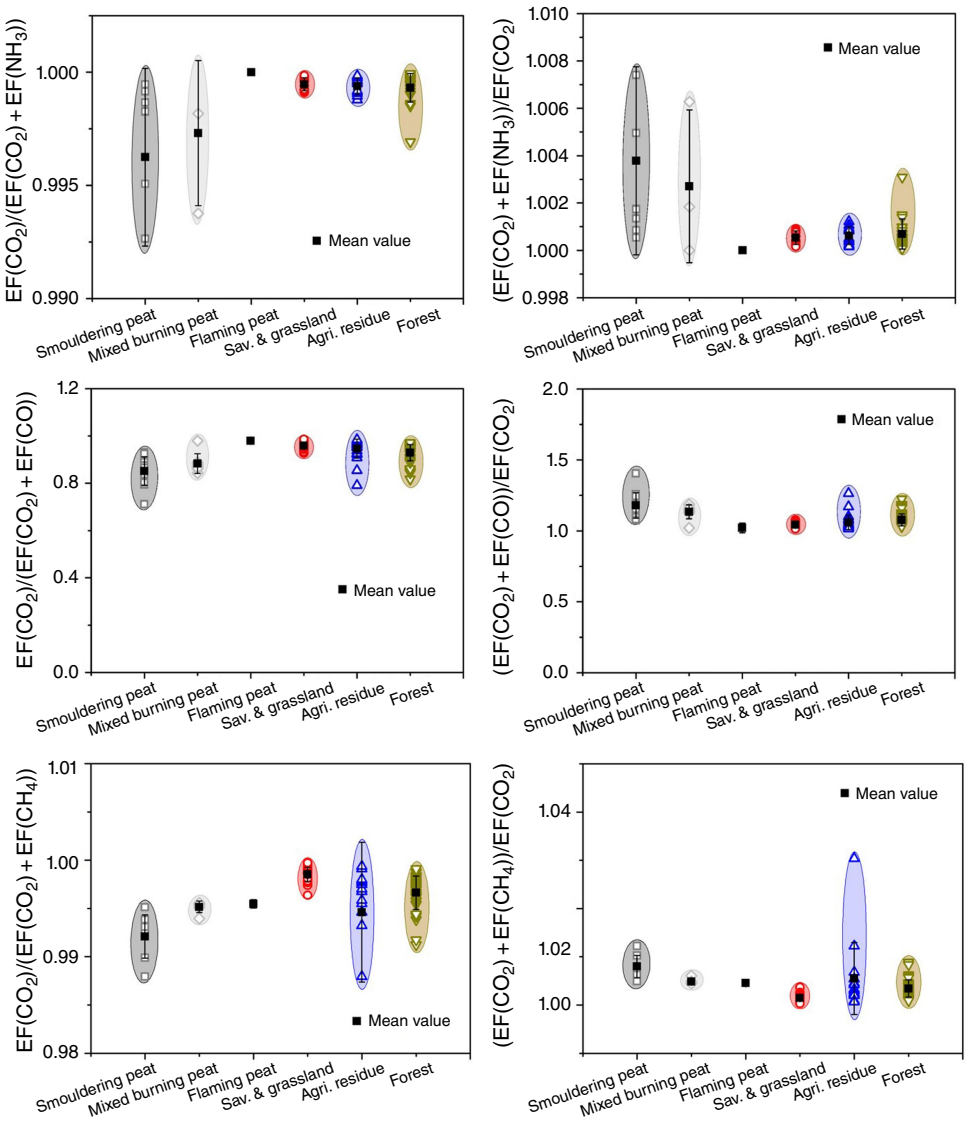

Fig. 6. Scatter plots of modified EF ratio (EF combination type (III)) from peat fire, savanna fire, forest fire and agricultural residue fire. Mean value from each fire category is shown as a black square with the standard deviation shown as an error bar. The ranges of the ratios are shown as clouds.

from agricultural residue fire ($P = 0.5122$). The other five EF modified ratios performed well in distinguishing between peat fire and savanna and grassland fire, agricultural residue fire and forest fire ($P < 0.05$). Nevertheless, no significant difference was found between smouldering peat and mixed burning peat, with $P > 0.05$. As a result, EF ratios included in EF combination type (IV) are not recommended as smouldering peat signatures.

EF combinations for early identification of smouldering peat

As showed above, EF combinations like $EF(CO_2)/EF(CH_4)$, $EF(CO_2)/(EF(CO_2) + EF(CO))$ and $EF(CO_2)/EF(CO)$ showed decent performance in distinguishing peat fires from the other types of biomass fires, while $EF(CO_2)/EF(CH_4)$ was the only signature showing a significant difference between smouldering peat fire and mixed burning peat. This section further investigated the potentiality of proposed signatures for early identification of smouldering peat.

Fig. 8 shows the value ranges of $EF(CO_2)/EF(CH_4)$, $EF(CO_2)/(EF(CO_2) + EF(CO))$, $EF(CO_2)/EF(CO)$ and the widely-adopted signature MCE, from an early ignition and a steady spread stage of smouldering peat. The segmented data are from Hu *et al.* (2018*b*), who experimentally measured transient emissions of smouldering peat. The mean value of $EF(CO_2)/EF(CH_4)$ from the early fire ignition was 38.1 ± 26.7, while fire spreading had an average ratio of

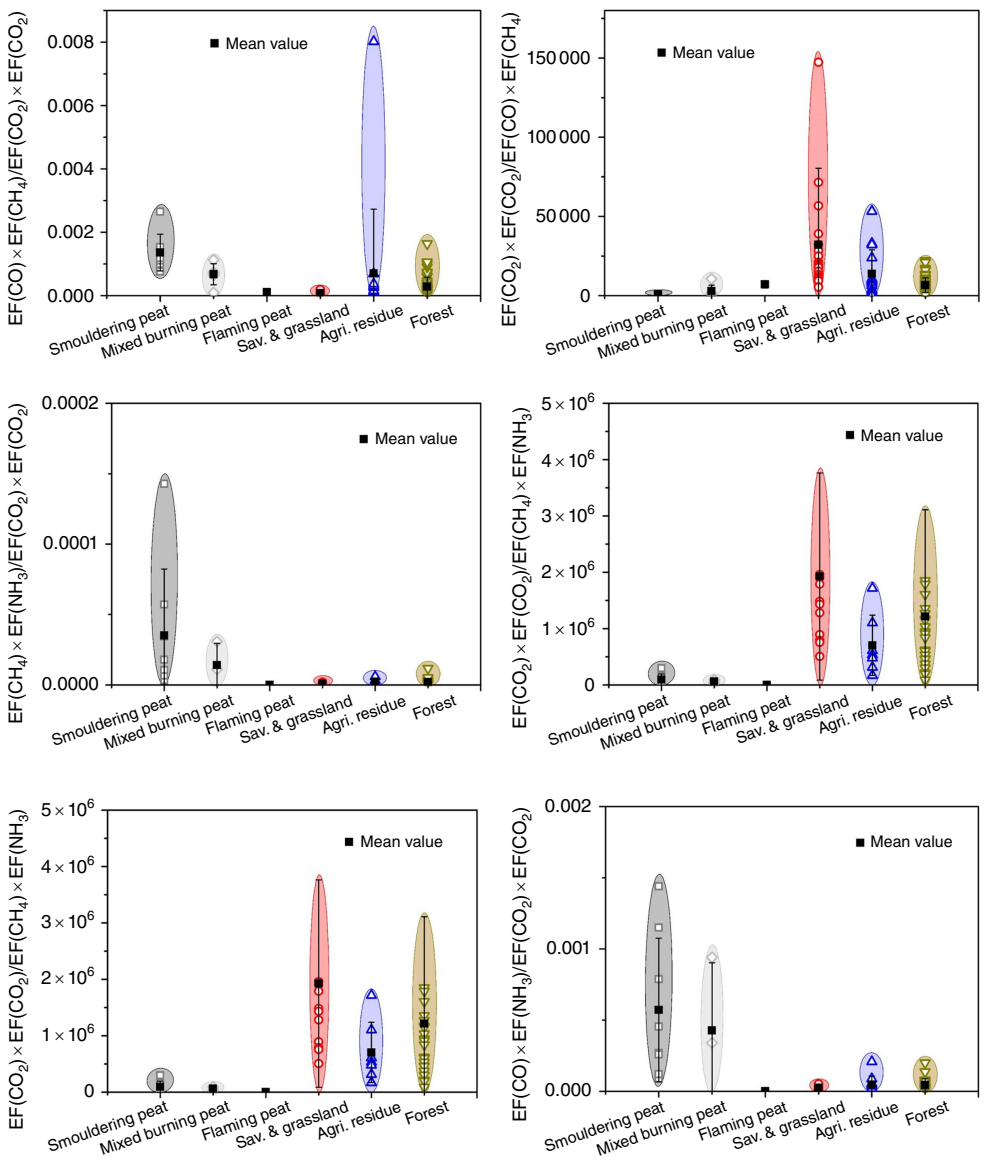

Fig. 7. Scatter plots of EF ratio (EF combination type (IV)) from peat fire, savanna fire, forest fire and agricultural residue fire. Mean value from each fire category is shown as a black square with the standard deviation showed as error bar. The ranges of the ratios are shown as clouds.

130.0 ± 9.5, which lies within the inter-study ratio range (134.6 ± 37.5) of $EF(CO_2)/EF(CH_4)$ shown in Table 4. The percentage difference between the early ignition and fire spreading reached 109%. In contrast, the mean values for the early ignition and spreading for $EF(CO_2)/EF(CO)$ were 4.0 ± 0.7 and 6.4 ± 0.3, respectively (percentage difference 45.8%), while for $EF(CO_2)/(EF(CO_2) + EF(CO))$, the mean values were 0.8 ± 0.03 and 0.9 ± 0.01, respectively (percentage difference 8.1%). For MCE, as reported in Hu *et al.* (2018*b*), the values ranged from 0.68 ± 0.04 (early ignition) to 0.79 ± 0.01 (steady spread), with a percentage difference of 14.0%. This comparison result indicates that $EF(CO_2)/EF(CH_4)$ is the most promising signature in detecting early fire with its largest difference in ratio ranges to the subsequent fire spreading.

One possible reason for the superior performance of $EF(CO_2)/EF(CH_4)$ in distinguishing early fire from fire spreading is that CO_2 and CH_4 show distinct transient emission evolution. In contrast, CO_2 and CO which are included in $EF(CO_2)/(EF(CO_2) + EF(CO))$, $EF(CO_2)/EF(CO)$ and the MCE have similar emission trends. Specifically, heat from the early ignition generates pyrolysis fronts which release alkane species like CH_4 from thermal decomposition (trace levels of CO_2 and CO are emitted at this stage); while, when spreading steadily, exothermic char oxidation dominates the mass loss and emits a large quantity of CO_2 and CO in an

Table 7. Summary of the modified EF ratios (EF combination type III and IV) from different biomass fires.

	$CO_2/(CO_2+CH_4)$		$CO_2/(CO_2+NH_3)$		$CO_2/(CO_2+CO)$		$(CO\times CH_4)/(CO_2\times CO_2)$		$(CH_4\times NH_3)/(CO_2\times CO_2)$		$(CO\times NH_3)/(CO_2\times CO_2)$	
	n	Mean (s.d.)	*n*	Mean (s.d.)	*n*	Mean (s.d.)	*n*	Mean (s.d.)	*n*	Mean (s.d.)	*n*	Mean (s.d.)
Smouldering peat	8	0.992 (0.004)	8	0.996 (0.004)	12	0.852 (0.060)	8	1.362×10^{-3} (0.58×10^{-3})	8	3.52×10^{-5} (4.71×10^{-5})	8	5.71×10^{-4} (5.04×10^{-4})
Mixed burning peat	6	0.995 (0.001)	3	0.997 (0.003)	8	0.883 (0.041)	6	6.75×10^{-4} (3.33×10^{-4})	3	1.40×10^{-5} (1.56×10^{-5})	3	4.27×10^{-4} (4.76×10^{-4})
Peat fire	14	0.993 (0.002)	11	0.997 (0.004)	20	0.865 (0.054)	14	1.06×10^{-3} (0.59×10^{-3})	11	2.93×10^{-5} (4.12×10^{-5})	11	5.30×10^{-4} (4.75×10^{-4})
Savanna fire	28	0.999 (0.001)	13	0.999 (0.0003)	31	0.959 (0.013)	28	7.32×10^{-5} (5.78×10^{-5})	13	9.04×10^{-7} (5.95×10^{-7})	13	2.50×10^{-5} (1.56×10^{-5})
Forest fire	55	0.997 (0.002)	23	0.999 (0.0006)	62	0.929 (0.034)	55	2.83×10^{-4} (2.99×10^{-4})	23	2.09×10^{-6} (2.47×10^{-6})	23	4.35×10^{-5} (4.46×10^{-5})
Agri. residue fire	15	0.995 (0.007)	11	0.999 (0.0003)	29	0.946 (0.040)	15	6.98×10^{-4} (2.03×10^{-3})	7	2.31×10^{-6} (1.79×10^{-6})	11	4.71×10^{-5} (5.88×10^{-5})
Biomass fire (peat exc.)	98	0.997 (0.003)	47	0.999 (0.0005)	122	0.941 (0.034)	98	2.87×10^{-4} (8.29×10^{-4})	43	1.77×10^{-6} (2.02×10^{-6})	47	3.92×10^{-5} (4.30×10^{-5})
	$(CO_2+CH_4)/CO_2$		$(CO_2+NH_3)/CO_2$		$(CO_2+CO)/CO_2$		$(CO_2\times CO_2)/(CO\times CH_4)$		$(CO_2\times CO_2)/(CH_4\times NH_3)$		$(CO_2\times CO_2)/(CO\times NH_3)$	
	n	Mean (s.d.)	*n*	Mean (s.d.)	*n*	Mean (s.d.)	*n*	Mean (s.d.)	*n*	Mean (s.d.)	*n*	Mean (s.d.)
Smouldering peat	8	1.008 (0.002)	8	1.004 (0.004)	12	1.179 (0.090)	8	831.6 (278.2)	8	96 064.2 (95 718.8)	8	4034.4 (3860.0)
Mixed burning peat	6	1.005 (0.001)	3	1.003 (0.003)	8	1.134 (0.049)	6	2859.5 (3793.2)	2	61 182.5 (40 661.5)	2	2001.0 (1326.7)
Peat fire	14	1.007 (0.002)	11	1.003 (0.004)	20	1.161 (0.078)	14	1702.4 (2580.1)	10	89 112.2 (86 730.8)	10	3629.3 (3536.9)
Savanna fire	28	1.001 (0.001)	13	1.001 (0.0002)	31	1.043 (0.015)	28	32 087.4 (48 292.0)	13	1.926E6 (1.839E6)	13	58 815.6 (39 985.9)
Forest fire	55	1.003 (0.002)	23	1.001 (0.0006)	62	1.078 (0.042)	55	6539.7 (4702.0)	23	1.216E6 (1.894E6)	23	48 725.5 (62 978.0)
Agri. residue fire	15	1.005 (0.008)	11	1.001 (0.0004)	29	1.059 (0.050)	15	13 762.2 (15 116.5)	7	7.042E5 (5.341E5)	11	55 607.3 (52 940.2)
Biomass fire (peat exc.)	98	1.003 (0.003)	47	1.001 (0.0005)	122	1.064 (0.042)	98	14 944.5 (28 627.6)	43	1.347E6 (1.752E6)	47	53 127.0 (54 254.6)

Table 8. Statistical analysis of the modified EF ratios (EF combination type III and IV) from different biomass fires

	$CO_2/(CO_2 + CH_4)$		$CO_2/(CO_2 + NH_3)$		$CO_2/(CO_2 + CO)$		$(CO \times CH_4)/(CO_2 \times CO_2)$		$(CH_4 \times NH_3)/(CO_2 \times CO_2)$		$(CO \times NH_3)/(CO_2 \times CO_2)$	
	t-value	*P*-value	*t*-value	*P*-value	*t*-value	*P*-value	*t*-value	*P*-value	*t*-value	*P*-value	*t*-value	*P*-value
Smouldering vs mixed burning peat	3.6769	0.0032	0.4608	0.6559	1.3798	0.1845	−2.7731	0.0169	−1.1208	0.2913	−0.4394	0.6707
Peat vs savanna	8.0783	<0.0001	2.6783	0.0137	7.5709	<0.0001	−6.2785	<0.0001	−2.2914	0.0319	−3.5258	0.0019
Peat vs Forest	4.8562	<0.0001	2.5245	0.0167	4.9951	<0.0001	−4.7968	<0.0001	−2.1940	0.0356	−3.3913	0.0019
Peat vs agri. residue	0.6075	0.5486	2.5957	0.0173	5.7251	<0.0001	−0.6642	0.5122	−2.1754	0.0449	−3.3476	0.0031
Peat vs biomass (peat excluded)	4.9028	<0.0001	2.5922	0.0121	6.0601	<0.0001	−4.3527	<0.0001	−2.2213	0.0307	−3.4249	0.0012
	F-value	*P*-value	*F*-value	*P*-value	*F*-value	*P*-value	*F*-value	*P*-value	*F*-value	*P*-value	*F*-value	*P*-value
Peat vs sav. vs agri. vs for.	0.0195	0.9963	9.0923	<0.0001	30.4788	<0.0001	5.9715	<0.0001	6.5061	<0.0001	16.5755	<0.0001
	$(CO_2 + CH_4)/CO_2$		$(CO_2 + NH_3)/CO_2$		$(CO_2 + CO)/CO_2$		$(CO_2 \times CO_2)/(CO \times CH_4)$		$(CO_2 \times CO_2)/(CH_4 \times NH_3)$		$(CO_2 \times CO_2)/(CO \times NH_3)$	
	t-value	*P*-value	*t*-value	*P*-value	*t*-value	*P*-value	*t*-value	*P*-value	*t*-value	*P*-value	*t*-value	*P*-value
Smouldering vs mixed burning peat	−3.6677	0.0032	−0.4628	0.6545	−1.4495	0.1645	1.3069	0.2157	−0.7855	0.4548	−1.2279	0.2544
Peat vs savanna	−8.0292	<0.0001	−2.6710	0.0140	−6.6739	<0.0001	3.3199	0.0019	3.5967	0.0017	4.9511	<0.0001
Peat vs Forest	−4.8357	<0.0001	−2.5185	0.0385	−4.5716	<0.0001	5.1640	<0.0001	2.8458	0.0078	3.4217	0.0018
Peat vs agri. residue	−0.5750	0.5701	−2.5893	0.0175	−5.1728	<0.0001	3.0427	0.0052	3.0188	0.0086	3.2484	0.0042
Peat vs biomass (peat excluded)	−4.8554	<0.0001	−2.5856	0.0124	−5.4150	<0.0001	4.4543	<0.0001	4.6852	<0.0001	6.1930	<0.0001
	F-value	*P*-value	*F*-value	*P*-value	*F*-value	*P*-value	*F*-value	*P*-value	*F*-value	*P*-value	*F*-value	*P*-value
Peat vs sav. vs agri. vs for.	0.0054	0.9995	9.0568	<0.0001	28.2496	<0.0001	7.6236	<0.0001	2.7586	0.0520	2.7742	0.0505

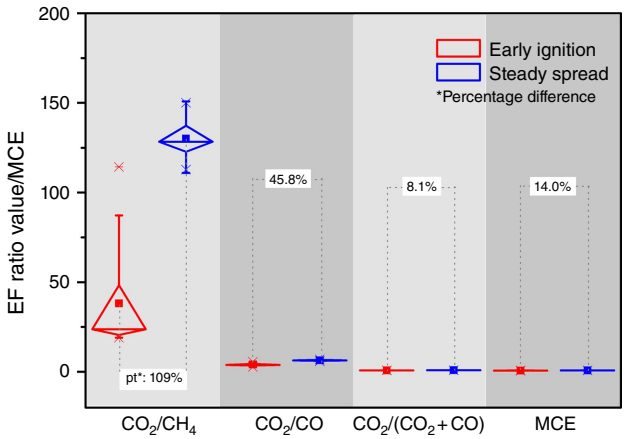

Fig. 8. Box plots of the value ranges of $EF(CO_2)/EF(CH_4)$, $EF(CO_2)/(EF(CO_2) + EF(CO))$, $EF(CO_2)/EF(CO)$ and the MCE from fire ignition and the subsequent fire spreading. The transient EF data are from Hu *et al.* (2018*b*).

almost synchronous fashion (Rein *et al.* 2009; Hu *et al.* 2018*b*).

Eqn 1 for calculating individual EF, $EF(CO_2)/EF(CH_4)$ can be simplified as below (Eqn 4):

$$\frac{EF(CO_2)}{EF(CH_4)} = \frac{MM_{CO_2}}{MM_{CH_4}} \times \frac{C_{CO_2}}{C_{CH_4}} \tag{4}$$

where MM_{CO_2} is the molar mass of species CO_2 (44 g mol^{-1}), MM_{CH_4} is the molar mass of species CH_4 (16 g mol^{-1}) and C_{CO_2} and C_{CH_4} are the mole fractions of CO_2 and CH_4, respectively.

As mentioned above, natural variability of fuel composition (for example, carbon content) could directly affect EF especially in the field (Hu *et al.* 2018*a*; Smith *et al.* 2018). Similar to the various fire ratios used for identifying fire conditions in coal mines (Ray *et al.* 2004) and the MCE (Christian *et al.* 2003; Stockwell *et al.* 2016), mole fraction ratios of CO_2 and CH_4 can be calculated without entailing EF, thus giving this signature advantages over EF-based signatures.

It is worth noting that the performance of gas signatures in early smouldering fire identification is limited to the transient emission data from a single peat fire study (Hu *et al.* 2018*b*). More studies are needed to improve the reliability and applicability of $EF(CO_2)/EF(CH_4)$ and other signatures like MCE (Hu *et al.* 2018*a*). However, the preliminary analysis shown here extends ideas on innovating early detection technologies for smouldering fire, and indicates the value of further research examining its effectiveness in practical use (Rein 2013; Wiggins *et al.* 2018).

Conclusion

Systematic comparisons and statistical analysis of 28 forms of EF combinations of CO_2, CO, CH_4 and NH_3 created from the inspiration of commonly used combustion signatures (for example, CO/CO_2 and the MCE) were carried out for peat, savanna and grassland, agricultural residue and forest fires. This novel framework enables, for the first time, the development of the best gas signature, $EF(CO_2)/EF(CH_4)$, for smouldering peat from three main aspects. First, $EF(CO_2)/EF(CH_4)$ benefits from its statistically distinguished ratio range (134.6 ± 37.5), which has 598.5% and 174.0% lower average than flaming fires and the other wildland fuel fires, respectively. Second, this signature possesses advantages over individual EF signatures in terms of its insusceptibility to fuel composition. Although this paper mainly focuses on EF, $EF(CO_2)/EF(CH_4)$ applies to measurements of gas concentrations in the field, which strengthens its practicality in actual use. Third, it shows the best performance in distinguishing early ignition from the subsequent spreading of a smouldering peat fire among signatures including the widely-adopted MCE. In contrast, the worst three candidates for smouldering peat gas signature from the perspective of statistical difference with the other wildfires are $EF(CO)/EF(NH_3)$, $EF(NH_3)/EF(CO)$ and $EF(CH_4)/EF(NH_3)$.

Limitations exist for the developed gas signature from this work: theoretically, there are endless EF combination types with higher complexity and more gas species which could be created for analysis; $EF(CO_2)/EF(CH_4)$ is not necessarily the perfect gas signature for smouldering peat. A level of uncertainty exists for $EF(CO_2)/EF(CH_4)$, but this is lower than for the MCE and the other analysed signatures. However, the limited peat fire EF data available for statistical analysis could affect the accuracy and reliability of the proposed signatures. This calls for more peat fire emission studies to advance scientific understanding of this little considered but important wildfire. Furthermore, the signature proposed in this work is not the ultimate solution for early smouldering peat detection. Developing a more comprehensive smouldering signature combining not only gas emission footprints but also the other combustion signatures (for example, infrared and particle emission characteristics) could further improve early fire detection technology.

This work provides scientific evidence demonstrating that the by-products of fires, i.e. emissions, when selected properly, can distinguish smouldering peat from the other wildfires and can also be applied for early detection. This is important as flaming wildfires have stronger thermal signatures, while the signals for smouldering fires are typically weak, and traditional wildfire detection technologies developed for flaming wildfires are much less effective for smouldering fires. In wildland where multiple types of wildfires co-exist, challenges remain for efficient early detection of smouldering fires, leading to missed time opportunities for early fire mitigation. The gas signatures developed from this work can contribute to developing tailored detection technologies for smouldering peat, consequently mitigating the smouldering-induced regional haze conundrum.

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Data availability. The data that support this study is freely available in Hu and Rein (2022) Data Compilation of Emission Factors Supporting the Development of Gas Signatures of Smouldering Peat Wildfires (Version 1.0.0) [Dataset]. https://doi.org/10.5281/zenodo.7199335.

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