Characteristics of organic components in PM2.5 emitted from peatland fires on Sumatra in 2015: Significance of humic-like substances

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Characteristics of organic components in $PM_{2.5}$ emitted from peatland fires on Sumatra in 2015: Significance of humic-like substances

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ABSTRACT

We characterize fine particulate matter ($PM_{2.5}$) emitted from Indonesian peatland fires using ground-based source-dominated samplings of $PM_{2.5}$ near peatland fire sources at two Regencies in Riau, Sumatra, Indonesia (number of samples = 13). Organic carbon (OC), elemental carbon, water-soluble OC (WSOC), the carbon content of humic-like substances (HULIS-C), and biomass burning tracers are determined. The carbon mass ratios of WSOC to OC (0.085 ± 0.015) and HULIS-C to WSOC (0.55 ± 0.085) are fairly constant and independent of the peatland fire sampling sites. By comparing diagnostic ratios using OC, WSOC, and HULIS-C at the peatland fire source and the receptor site (Malaysia) during peatland fire-induced haze periods, secondary WSOC and HULIS-C formation during transport from the source to the receptor site is highly possible. Interestingly, the mass ratio of syringic acid to levoglucosan (0.045 ± 0.0075) is fairly constant at Indonesian peatland fire sources. Because syringic acid is less stable than levoglucosan, this ratio is an aging indicator for Indonesian peatland fires at receptor sites. By comparing the mass fraction of each organic compound in the present study and previous studies, it is evident that the source profile for the coburning of peat with surface vegetation is significantly different compared with the burning of peat alone. Further knowledge of peat burning emissions is needed, particularly with respect to burning conditions, peat composition, and the effects of vegetative burning on peatland. Improved knowledge of these factors would lead to more reliable speciated emission inventories of Indonesian peatland fires, advancing chemical transport and radiative forcing modeling, as well as health risk assessment.

1. Introduction

Indonesia has the largest area of tropical peatland (2.7×10^5 km^2) in the world that has been drained and cleared of natural vegetation, making said area susceptible to fire (Joosten, 2010; Kiely et al., 2019). Peat stores large quantities of carbon in the form of partially decayed organic matter. Indonesian peatland in particular has a carbon store of ~60 PgC, which is ~65% of the tropical peatland carbon reservoir (Nechita-Banda et al., 2018; Page et al., 2011). Peatland fires on Kalimantan and Sumatra in Indonesia emit considerable amounts of carbon dioxide (CO_2), carbon monoxide (CO), methane (CH_4), and fine particulate matter ($PM_{2.5}$) (Huijnen et al., 2016; Kiely et al., 2019; Nechita-Banda et al., 2018; Page et al., 2002). The generated smoke is dense and can cause domestic and transboundary haze pollution in Southeast Asia (Balasubramanian et al., 2003; Behera and Balasubramanian, 2014; Fujii et al., 2013, 2014; Fujii et al., 2015b, 2016, 2017, 2019; Keywood et al., 2003; Khan et al., 2016; Reddington et al., 2014; See et al., 2006, 2007). The El Niño dry season in 2015 was the strongest Indonesian peatland fire episode since 1997, and the total carbon amount released from September to October in 2015 is estimated to be 227 ± 67 TgC, of which 83% was in the form of CO_2, 16% was in the form of CO, and 1% was in the form of CH_4 (Huijnen et al., 2016). Jayarathne et al. (2018) reported that from the 2015 fire event, the total amount of $PM_{2.5}$ released into the atmosphere is estimated to be 6.0 ± 5.5 Tg, with major contributions from organic carbon (OC; 4.3 TgC). Wiggins et al. (2018) reported that the peat smoke accounted for approximately 85% of smoke plumes reaching Singapore, based on the radiocarbon content of carbonaceous $PM_{2.5}$ samples collected in Singapore from September

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2014 to October 2015.

The $PM_{2.5}$ source profile (the mass fractions of designated chemical species in $PM_{2.5}$) of Indonesian peatland fire is essential to create speciated emission inventories for source-oriented models, such as chemical transport models, to conduct source apportionment with receptor models (e.g., chemical mass balance model), and to estimate toxic/hazardous pollutant emissions. Additionally, the source profile is important with respect to understanding the aging process of smoke aerosols, such as chemical reaction, outgassing, coagulation, and condensation during transport. Several studies have been made to clarify the chemical characteristics of $PM_{2.5}$ with regard to OC, elemental carbon (EC), black carbon, water-soluble OC (WSOC), polycyclic aromatic hydrocarbons, *n*-alkanes, organic aerosol tracers (particularly for biomass burning), water-soluble ions, and trace metals at Indonesian peatland fire sources through intensive field observations (Betha et al., 2013; Fujii et al., 2014, 2015a, 2019; Jayarathne et al., 2018; See et al., 2007; Stockwell et al., 2016; Tham et al., 2019) and controlled burning experiments in the laboratory (Chen et al., 2017; Christian et al., 2003; Das et al., 2019; Iinuma et al., 2007; Lestari et al., 2020; Watson et al., 2019). Previous field studies demonstrate that organic matter is substantially important because OC constitutes approximately 70 wt% of $PM_{2.5}$ from Indonesian peatland fire sources, whereas EC constitutes only 1–2 wt.% (Fujii et al., 2014; Jayarathne et al., 2018). Regarding water-soluble ions, which are minor components at Indonesian peatland fire sources, Fujii et al. (2019) mentioned that the $PM_{2.5}$ source profile of typical water-soluble ions (chloride, sulfate, and ammonium) differs depending on the location of peatland fire. The source profiles of organic components may be also different among peatland fire sources based on the reported data for Sumatra (Fujii et al., 2015a) and Kalimantan (Jayarathne et al., 2018). Variations in the type and moisture content of the burning material as well as combustion temperature may also attribute this difference (Chen et al., 2010; Huang and Rein, 2014). However, a few papers have reported the source profiles of detailed organic compounds with regard to biomass burning tracers based on field observations near Indonesian peatland fire sources (Fujii et al., 2015a; Jayarathne et al., 2018), and the lab experiments have also been conducted for controlled peat burning (Iinuma et al., 2007; Watson et al., 2019). Additionally, the field study for Sumatra was based on only one site (Bengkalis Regency). Aerosol-phase humic-like substances (HULIS) are also emitted from biomass burning (Hoffer et al., 2006), but there is a lack of information regarding particulate HULIS at Indonesian peatland fire sources. Indeed, HULIS are an important substance class in the hydrophobic WSOC fraction and have been the subject of increasing attention due to their universal ambient presence, active participation in atmospheric chemistry, radiative effect, and adverse health implications (Fujii et al., 2021; Zheng et al., 2013). Very recently, Fujii et al. (2021) reported that the concentrations of the carbon content of HULIS (HULIS-C) and WSOC in total suspended particulates (TSP) in Malaysia during Indonesian peatland fire-induced haze periods are significantly higher than non-haze periods. They also mentioned that a comprehensive study of HULIS at Indonesian peatland fire sources is required to clarify the chemical evolution of organic aerosols and the process of secondary aerosol formation during transport to receptor sites. Therefore, the representative real-world source profiles (including HULIS) should be investigated by source-dominated samplings at multiple sites and compared with the reported data on Sumatra and Kalimantan.

In this study, we characterize $PM_{2.5}$ emitted from Indonesian peatland fires at different sites based on ground-based source-dominated samplings in Riau, Sumatra, Indonesia, in 2015. We focus on the following carbonaceous components: OC, EC, WSOC, HULIS-C, and biomass burning tracers such as cellulose and lignin pyrolysis products. Then, we compare the speciated source profiles and indicators obtained in the present study with existing reports for Indonesia as well as with those obtained from previous peat-burning lab experiments to investigate the potential for providing representative source profiles.

2. Materials and methods

Table S1 shows the details of the samples taken from peatland fire sources at the Siak and Kampar districts of the Riau province (Fig. 1). The sampling locations partially overlapped with those reported in our former paper, which focused on the characteristics of water-soluble ionic components (Fujii et al., 2019). The sampling sites at Siak were located on plantations where palm trees are the dominant plant species on the peatlands. The other sampling sites at Kampar were located on wildland, and wild plant-like grass is dominant plant species on the peatlands. We used a $PM_{2.5}$ sampler (ChemComb model 3500 speciation sampling cartridge, Thermo) equipped with two volatile organic compound denuders to reduce the organic gas adsorption artifacts and continuously collect $PM_{2.5}$ on 47 mm diameter quartz fiber filters for 3–4 h at a flow rate of 10 L min^{-1}. Before sampling, the filters were heat-treated at 900 °C for 4 h. We collected $PM_{2.5}$ at several peatland fire sites on 13 occasions. As all samples were collected ~5 m away from peatland fire hotspots, we regarded our samples as exclusive source samples. All fires were regarded as smoldering combustion, because there were no flames during the samplings in this study. After sampling, we determined the concentrations of OC, EC, WSOC, HULIS-C, and biomass burning tracers using the quartz fiber filter samples.

The carbonaceous content (OC and EC) of $PM_{2.5}$ was quantified using a DRI Model 2001 OC/EC carbon analyzer, which employs the thermal/optical reflectance method using the IMPROVE_A protocol. Detailed information on the quantification procedure for the IMPROVE_A protocol and comparisons with the previous version (IMPROVE protocol) are provided elsewhere (Chow et al., 2007). Blank corrections were performed on the OC and EC data by subtracting the blank filter values from the loaded filter values. Here, blank filter values were based on the data from heat-treated laboratory blank filters.

The methods used to quantify WSOC and HULIS-C followed the procedure outlined by Fujii et al. (2021). The detailed information of WSOC and HULIS-C analyses is provided in the supplementary material.

Biomass burning tracers obtained from the quartz fiber filters were quantified by gas chromatography–mass spectrometry. In this study, we focused on levoglucosan (LG), mannosan (MN), galactosan, *p*-hydroxybenzaldehyde, *p*-hydroxybenzoic acid, vanillin, vanillic acid (VA), homovanillic acid, syringaldehyde, syringic acid (SA), homosyringic acid, and *β*-sitosterol. The detailed information of biomass burning tracers' analyses is provided in the supplementary material.

3. Results and discussion

3.1. OC and EC

The OC and EC concentrations for all samples are 920 ± 760 (average ± standard deviation) and 8.1 ± 9.9 μgC m^{-3}. Obviously, the OC concentration is much higher than that of EC. In this study, a relatively high OC/EC carbon mass ratio (210 ± 190 for all samples) was observed compared with the reported ratios at peatland fire sources on Sumatra and Kalimantan (Fujii et al., 2014; Jayarathne et al., 2018; See et al., 2007; Tham et al., 2019), as shown in Table 1. The discrepancy among samples can be attributed to the differences in the combustion conditions such as fuel load, moisture content (Chen et al., 2010), smoldering kinetics, and the reaction-zone structure of peatland fires (Huang and Rein, 2014). Because OC and EC measurements are protocol dependent, different protocols such as NIOSH and IMPROVE for determination may also lead to uncertainties (Wu et al., 2016). Thus, to show the representative OC/EC ratio for Indonesian peatland fires is extremely difficult.

Fig. 2 shows the abundances of five thermally-derived OC fractions (OC1, OC2, OC3, OC4, and OP [pyrolyzed OC]) by OC mass percentage at each peatland fire source. OC1 and OC2 account for 42 ± 5.4 and 32 ± 5.8 wt% of OC, respectively, for all samples collected at Siak and Kampar, both of which are the predominant OC fractions. Others are

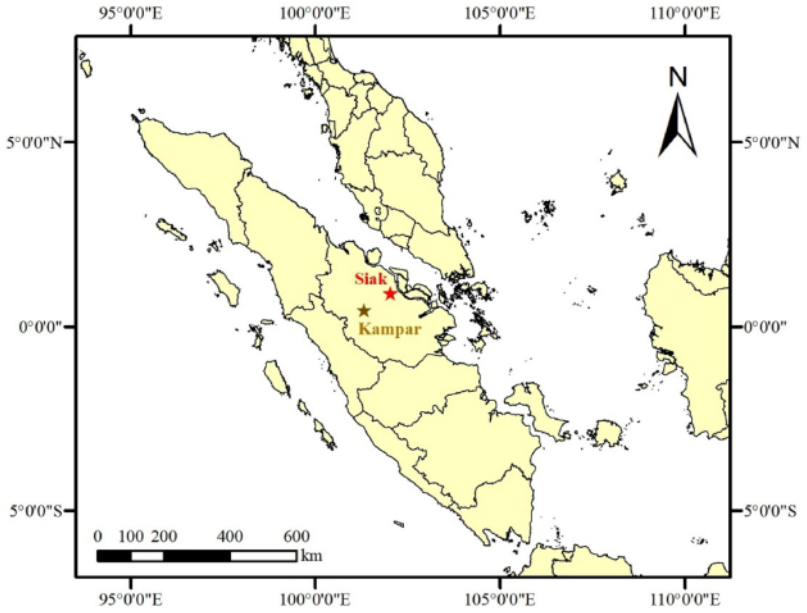

Fig. 1. Sampling site of peatland fire sources (Siak and Kampar).

Table 1
Diagnostic ratios (OC/EC and WSOC/OC) for ground-based measurements during Indonesian peatland fire events and laboratory studies of Indonesian peat burning (average ± standard deviation). TOR = thermal-optical reflectance method; TOT = thermal-optical transmission method.

Location	PM size	Method for OC and EC	OC/EC	WSOC/OC	Number of samples	Reference
Indonesia (peatland fire source)	$PM_{2.5}$	TOR (IMPROVE_A)	220 ± 120 (Siak) 190 ± 280 (Kampar) 210 ± 190 (All)	0.089 ± 0.015 (Siak) 0.079 ± 0.015 (Kampar) 0.085 ± 0.015 (All)	8 (Siak) 5 (Kampar) 13 (All)	This study
	$PM_{2.5}$	CHN analyzer (EC: >350 °C)	2.4	–	3*	See et al., (2007)
	$PM_{2.5}$	TOR (IMPROVE_A)	36.4 ± 9.08	–	7**	Fujii et al., (2014)
	$PM_{2.5}$	TOR (IMPROVE_A)	18	–	6***	Tham et al., (2019)
	$PM_{2.5}$	TOT (NIOSH 5040)	67 ± 26	0.16 ± 0.11	21****	Jayarathne et al., (2018)
Singapore (haze by peatland fires)	$PM_{2.5}$	CHN analyzer TC, IC (1 M HCl) EC (resultant mass after 350 °C for 24 h) OC = TC-IC-EC	6.0 ± 3.1	0.30 ± 0.10	31	Balasubramanian et al., (2003)
	TSP	TOT (Modified NIOSH)	15.5	–	No data	Engling et al. (2014)
	$PM_{2.5}$	TOR (IMPROVE_A)	4.75	0.64	8	Budisulistiorini et al. (2018)
	$PM_{2.5}$	TOR (IMPROVE_A)	14.9 ± 2.9 (episodic smoke) 4.8 ± 2.5 (smoke-dominant)	–	6 (episodic smoke) 131 (smoke-dominant)	Tham et al., (2019)
Malaysia (haze by peatland fires)	$PM_{2.5}$	TOR (IMPROVE_A)	4.2 ± 1.5	–	15	Fujii et al., (2015b)
	TSP	TOR (IMPROVE_A)	9.8 (strong haze) 4.2 ± 0.72 (light haze)	0.34 (strong haze) 0.39 ± 0.034 (light haze)	2 (strong haze) 3 (light haze)	Fujii et al., 2016; 2021
Lab experiment (Indonesian peat)	$PM_{2.5}$	Not specified	150	–	1	Christian et al., (2003)
	PM_{10}	Thermographic method (C-mat 5500)	14	0.39	1	Iinuma et al., (2007)
	TSP	TOR (IMPROVE_A)	86–145 (Sumatra) 112–150 (Kalimantan)	0.0093–0.0608 (Sumatra) 0.0203–0.0416 (Kalimantan)	6 (Sumatra) 2 (Kalimantan)	Chen et al., (2017)
	$PM_{2.5}$	TOR (IMPROVE_A)	73 (fresh) 35 (aged)	0.20 (fresh) 0.36 (aged)	4 (fresh) 4 (aged)	Watson et al., 2019 (sampled at Borneo, Malaysia)
	$PM_{2.5}$	TOR (IMPROVE_A)	160 ± 55 (surface) 100 ± 70 (sub surface)	–	5 (surface) 5 (sub surface)	Lestari et al., (2020)

Note: sampling location) *Sungai Sembilan (Sumatra), **Bengkalis (Sumatra), ***Jambi (Sumatra), ****Central Kalimantan (Kalimantan).

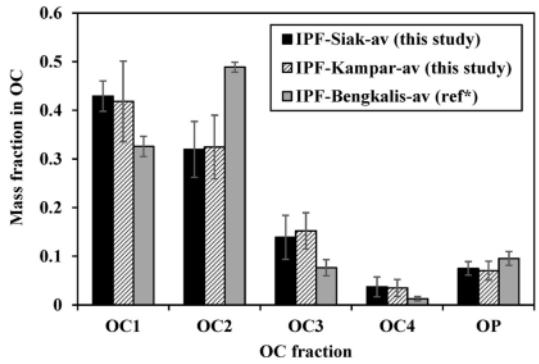

Fig. 2. Abundances of five thermally derived OC carbon fractions of $PM_{2.5}$ as mass percentage of OC at each peatland fire site. Error bars indicate standard deviations. *Data from Fujii et al. (2014).

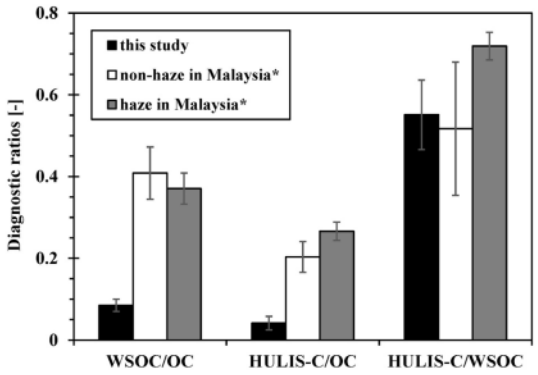

Fig. 3. Comparison of WSOC/OC, HULIS-C/OC, and HULIS-C/WSOC ratios in $PM_{2.5}$ at Indonesian peatland fire sources and TSP at the receptor site during non-haze and Indonesian peatland fire-induced haze periods in Malaysia. *Data from Fujii et al. (2021).

minor OC fractions: 14 ± 4.1 and 3.6 ± 1.9 wt% for OC3 and OC4, respectively, and 7.3 ± 1.6 wt% for OP. From Fig. 2, it is evident that no significant differences of OC mass fractions exist in the sampling sites between Siak and Kampar. For the present study, the OC2 fraction (Siak: 32 ± 5.8 wt%; Kampar: 32 ± 6.5 wt%) is ~1.5 times smaller than that obtained from the Indonesian peatland fire source in Bengkalis, Riau, Sumatra (49 ± 1.0 wt%), in 2012 (Fujii et al., 2014) as shown in Fig. 2.

3.2. WSOC and HULIS-C

The concentrations of WSOC for all samples and HULIS-C for all samples except for SIAK-4 and KMP-5 are 78 ± 66 and 50 ± 43 μgC m^{-3}, respectively. HULIS-C accounts for 55 ± 8.5 wt% of the WSOC, and only 8.5 ± 1.5 and 4.8 ± 0.75 wt% of the OC are WSOC and HULIS-C, respectively. Therefore, water insoluble OC (WIOC) is the dominant OC in $PM_{2.5}$ at the peatland fire source (92 ± 1.5 wt% of OC for all samples).

The WSOC/OC carbon mass ratio from Indonesian peatland fires in the present study (0.085 ± 0.015) is much lower compared with that of other biomass burning sources: ~0.38 from corn burning, ~0.68 from wheat burning (Wang et al., 2020), 0.43 ± 0.06 from rice straw burning, 0.42 ± 0.05 from pine needle burning, and 0.57 ± 0.09 from sesame stem burning (Park and Yu, 2016). The WSOC/OC ratio from Indonesian peatland fire on Kalimantan (0.16 ± 0.11) reported by Jayarathne et al. (2018) is higher than that obtained in the present study. It should be noted that Jayarathne et al. (2018) applied NIOSH thermal optical transmittance protocol for OC determination although we applied IMPROVE_A thermal optical reflectance protocol. The difference in EC/OC protocol may cause the discrepancy between the two ratios. However, it is still lower than that obtained from other biomass burning sources. Moreover, the WSOC/OC ratio obtained from lab peat-burning experiments (Chen et al., 2017; Iinuma et al., 2007; Watson et al., 2019) is lower (Table 1). Thus, the WSOC/OC ratio is consistently smaller at Indonesian peatland fire sources compared with other biomass burning sources. By contrast, differences in the HULIS-C/WSOC carbon mass ratio cannot be observed between our results (0.55 ± 0.085) and other biomass burning sources: ~0.53 from corn burning, ~0.46 from wheat burning (Wang et al., 2020), 0.63 ± 0.05 from rice straw burning, 0.36 ± 0.08 from pine needle burning, and 0.51 ± 0.08 from sesame stem burning (Park and Yu, 2016).

Fig. 3 compares the WSOC/OC, HULIS-C/OC, and HULIS-C/WSOC ratios obtained in the present study with those of reported data (Fujii et al., 2016, 2021) for Malaysian haze induced by Indonesian peatland fires on Sumatra Island. It should be noted that the obtained ratios are based on TSP (Fujii et al., 2016, 2021), whereas our ratios are based on $PM_{2.5}$ samples. However, mutual comparisons are still possible because, in general, 80 %–90% of aerosols produced by biomass burning are in the size range of $PM_{2.5}$ (Reid et al., 2005). The average WSOC/OC, HULIS-C/OC, and HULIS-C/WSOC ratios for haze in Malaysia are 4.4, 6.4, and 1.3 times higher than those for Indonesian peatland fire sources, respectively, which suggests significant differences particularly for the WSOC/OC and HULIS-C/OC ratios. However, the effects of other sources on haze samples in Malaysia should be evaluated. To remove those effects, Δ(WSOC/OC), Δ(HULIS-C/OC), and Δ(HULIS-C/WSOC) in "aged" peatland fire TSP were calculated by subtracting the average concentrations of OC, WSOC, and HULIS-C during non-haze periods from those of haze periods:

$$\Delta(WSOC \Big/ OC) = \frac{WSOC|_{haze} - WSOC|_{non-haze}}{OC|_{haze} - OC|_{non-haze}}$$

$$\Delta(HULIS-C \Big/ OC) = \frac{HULIS-C|_{haze} - HULIS-C|_{non-haze}}{OC|_{haze} - OC|_{non-haze}}$$

$$\Delta(HULIS-C \Big/ WSOC) = \frac{HULIS-C|_{haze} - HULIS-C|_{non-haze}}{WSOC|_{haze} - WSOC|_{non-haze}}$$

As a result, Δ(WSOC/OC) = 0.36 ± 0.055, Δ(HULIS-C/OC) = 0.30 ± 0.044, and Δ(HULIS-C/WSOC) = 0.86 ± 0.11 were obtained, all of which are still higher than those obtained for peatland fire sources. This result suggests that secondary WSOC and HULIS-C formation occurs during transport from the Indonesian peatland fire sources to the receptor site in Malaysia. Based on peat-burning lab experiments, Chen et al. (2017) reported that fresh Indonesian peat burning aerosols are almost non-hygroscopic due to predominant contribution of water-insoluble organics, and emphasized the importance of both the WSOC fraction and hygroscopicity parameter of the water-soluble fraction in determining the hygroscopicity of organic aerosols. They also mentioned that quantification of HULIS as well as evaluation of their water uptake properties will be important for understanding hygroscopicity of Indonesian peatland fire aerosols. Thus, we infer that properties of hygroscopic growth and cloud condensation nuclei activity linked to cloud formation, precipitation, and regional radiative forcing should be changed during transport from Indonesian peatland fire sources to receptors. In addition, generated HULIS during transport can also affect regional radiative forcing by light absorption properties, as HULIS are major components of brown carbon (Graber and Rudich, 2006; Laskin et al., 2015; Zheng et al., 2013).

3.3. Biomass burning tracers

LG is a typical pyrolysis product of cellulose and has been widely

accepted as a useful biomass burning tracer (Simoneit et al., 1999). Among the quantified organic compounds in this study, LG is the most abundant at 13 ± 7.2 mg g-OC^{-1}, followed by syringaldehyde at 2.2 ± 1.0 mg g-OC^{-1} and VA at 1.4 ± 0.77 mg g-OC^{-1} (Table 2). The OC mass fraction of each organic compound at Siak is comparable to that at Kampar. However, Table 2 shows that the mass fractions for compounds differ from those of previous studies conducted in Sumatra (Fujii et al., 2015a) and Kalimantan (Jayarathne et al., 2018). In particular, the mass fractions for most compounds in Bengkalis, Sumatra (Fujii et al., 2015a), are roughly an order of magnitude greater than those obtained in the present study. Jayarathne et al. (2018) reported that the OC fraction of LG from the coburning of peat with surface vegetation (360 mg g-OC^{-1}) differs from the subsurface burning of peat (46 ± 40 mg g-OC^{-1}). Additionally, Watson et al. (2019) reported that the average mass fraction of LG in OC (fresh $PM_{2.5}$) from peat-burning lab experiments is 43 mg g-OC^{-1}. Because the former study by Fujii et al. (2015a) concerned large-scale peatland fires, including the burning of palm trees, fallen trees, and bushes, with large $PM_{2.5}$ concentrations (7120 ± 3620 μg m^{-3}), said fires may be regarded as the coburning of peat with surface vegetation. Thus, $PM_{2.5}$ source profiles of biomass burning tracers for Indonesian peatland fires must be selected with extreme caution if applied to a receptor model.

The MN/LG mass ratio is used for the source assignment of specific biofuels (Alves et al., 2010; Fabbri et al., 2009; Fujii et al., 2014, 2015b; Kuo et al., 2011; Myers-Pigg et al., 2016; Sullivan et al., 2014; Yang et al., 2016). The MN/LG mass ratio is a useful tracer with respect to distinguishing the effects of hardwood burning from softwood burning. On the basis of the report by Fabbri et al. (2009), the differences in the MN/LG mass ratio between hardwood burning (0.042–0.077) and softwood burning (0.15–0.26) can be shown. In this study, the MN/LG mass ratio for all samples is 0.091 ± 0.023, which is similar to the characteristics of hardwood burning. A comparable ratio was observed between the two sampling sites of Siak (0.095 ± 0.025) and Kampar (0.076 ± 0.012). The MN/LG mass ratio obtained in the present study is also consistent with the ratio (0.070 ± 0.014) reported by Fujii et al. (2015a) for peatland fire on Sumatra; for Kalimantan (Jayarathne et al., 2018), the ratios are 0.023 ± 0.011 for subsurface burning of peat and 0.034 for coburning of peat with surface vegetation, which are significantly smaller than the ratios obtained from this study and previous studies. However, the MN/LG mass ratios for Indonesian peatland fires on Sumatra and Kalimantan are consistently similar to those obtained from hardwood burning, irrespective of burning types (coburning of peat with surface vegetation and subsurface burning of peat).

The SA/VA mass ratio is suggested as an indicator for Indonesian peatland fires (Fujii et al., 2015a). In contrast to the MN/LG mass ratio, significant differences can be observed for the SA/VA mass ratio between samples collected at Siak (0.33 ± 0.18) and Kampar (0.81 ± 0.57). The SA/VA mass ratios are different from previously obtained ratios for Sumatra (0.96 ± 0.14) by Fujii et al. (2015a) and for Kalimantan (0.48 ± 0.14) by Jayarathne et al. (2018), as shown in Fig. 4. The SA/VA mass ratio from the lab experiment of Indonesian peat burning is 0.11 (Iinuma et al., 2007), which is smaller than that obtained from field observations. The SA/VA mass ratios range widely from 0.1 to 12 for different burning sources over the world, and there is a large discrepancy in the ratio even for the same source such as hardwood (Wan et al., 2019). Then, this discrepancy may be due to the effects of other vegetative burning on real-world peatland fires, and further experiments for testing are necessary.

Fig. 5 shows the relationship between LG and SA concentrations. Interestingly, the results show a fairly good linear relationship with a slope of 0.045 ± 0.0075. Note that the SA/LG mass ratio is fairly constant and independent of the sampling sites of the Indonesian peatland fires. It is also comparable to the SA/LG mass ratio obtained from the Indonesian peatland fire sources on Sumatra (0.061) reported by Fujii et al. (2015a) and on Kalimantan (0.078) reported by Jayarathne et al. (2018). The SA/LG mass ratio obtained from lab experiments of peat burning is 0.044 (Iinuma et al., 2007), which is consistent with the present study's results. SA is less stable than VA in air (Fujii et al., 2015b), and VA is less stable than LG based on the OH-initiated reaction experiments (Lai et al., 2014; Liu and Zeng, 2018). Therefore, SA is less stable than LG. Thus, the SA/LG mass ratio is expected to decrease after long-range transport from Indonesian peatland fire sources, which suggests that it is an aging indicator for Indonesian peatland fires. However, it should be noted that possible sources of SA except for Indonesian peatland fires are local biomass burning specific to

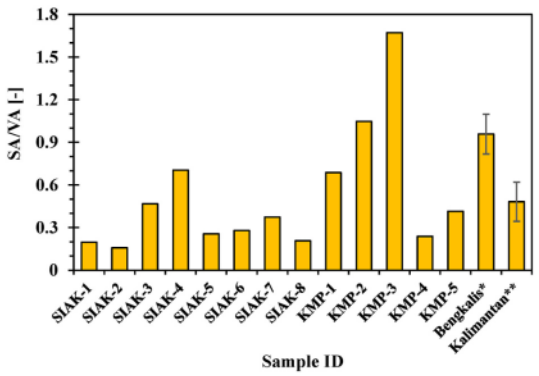

Fig. 4. Comparison of SA/VA mass ratios in $PM_{2.5}$ at several Indonesian peatland fire sources on Sumatra and Kalimantan. Error bars indicate standard deviations. *Data from Fujii et al. (2015a). **Data from Jayarathne et al. (2018).

Table 2
Bulk chemical compositions of OC in $PM_{2.5}$ emitted from Indonesian peatland fires (average ± standard deviation). N = Number of samples.

Organic compounds [mg g-OC^{-1}]	Sumatra Island				Kalimantan Island[3] (N = 21)
	Siak[1] (N = 8)	Kampar[1] (N = 5)	All[1] (N = 13)	Bengkalis[2] (N = 7)	
Levoglucosan	10 ± 5.5	17 ± 8.5	13 ± 7.2	130 ± 1.7	46 ± 40
Mannosan	0.88 ± 0.30	1.3 ± 0.48	1.0 ± 0.41	8.6 ± 1.1	0.93 ± 0.76
Galactosan	0.43 ± 0.18	0.61 ± 0.28	0.50 ± 0.23	2.8 ± 0.39	0.14 ± 0.13
Vanillin	0.29 ± 0.081	0.38 ± 0.093	0.33 ± 0.093	0.93 ± 0.12	0.03 ± 0.044
Vanillic acid	1.5 ± 0.76	1.3 ± 0.86	1.4 ± 0.77	7.9 ± 1.7	3.7 ± 2.2
Homovanillic acid	0.097 ± 0.032	0.12 ± 0.070	0.11 ± 0.052	0.61 ± 0.10	ND
Syringaldehyde	2.0 ± 1.1	2.6 ± 0.99	2.2 ± 1.0	3.2 ± 0.53	0.83 ± 0.46
Syringic acid	0.43 ± 0.23	0.80 ± 0.39	0.57 ± 0.34	7.5 ± 1.6	34 ± 0.91
Homosyringic acid	0.044 ± 0.031	0.095 ± 0.030	0.069 ± 0.039	0.36 ± 0.080	ND
p-Hydroxybenzaldehyde	0.25 ± 0.056	0.19 ± 0.018	0.23 ± 0.054	ND	ND
p-Hydroxybenzoic acid	0.42 ± 0.27	0.54 ± 0.28	0.46 ± 0.27	2.4 ± 0.32	ND
β-Sitosterol	0.47 ± 0.14	0.78 ± 0.34	0.59 ± 0.27	ND	0.53 ± 0.34

Data from [1] this study, [2] Fujii et al. (2015a), and [3] Jayarathne et al. (2018). ND = No Data.

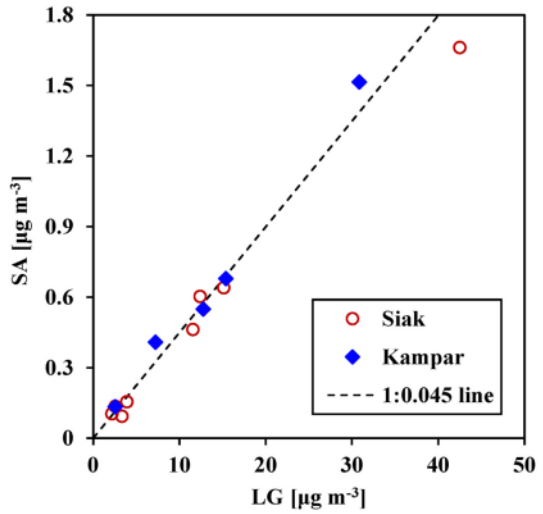

Fig. 5. LG vs. SA concentrations at two Indonesian peatland fire sources (Siak and Kampar).

hardwood burning (Wan et al., 2019).

As the same calculation procedure of Δ(WSOC/OC), Δ(HULIS-C/OC), and Δ(HULIS-C/WSOC), Δ(SA/LG) in "aged" peatland fire TSP based on the data reported by Fujii et al. (2016) were calculated by subtracting the average concentrations of SA and LG during non-haze periods from those of haze periods:

$$\Delta(SA/LG) = \frac{SA|_{haze} - SA|_{non-haze}}{LG|_{haze} - LG|_{non-haze}}$$

As a result, Δ(SA/LG) ranged from 0.0071 to 0.029, all of which are lower than that for peatland fire sources obtained in this study (0.045 ± 0.0075), suggesting that an aging process occurs during transport from the Indonesian peatland fire sources to the receptor site in Malaysia. Furthermore, Δ(SA/LG) at the receptor tend to decreases as Δ(WSOC/OC) and Δ(HULIS-C/OC) increase as shown in Fig. S1. This supports that Δ(SA/LG) has a potential aging indicator of Indonesian peatland fires. However, our data points are so limited (only five points) that the availability of Δ(SA/LG) as an aging indicator should be further investigated based on the long-term observation data.

4. Conclusion

We conducted a field study to characterize carbonaceous species in $PM_{2.5}$ emitted from Indonesian peatland fires using ground-based samplings at fire sources in Riau, Sumatra, Indonesia. The OC, EC, WSOC, and HULIS-C concentrations as well as biomass burning tracers, such as cellulose and lignin pyrolysis products, were determined. The main conclusions are outlined below.

1. WSOC accounted for only 8.5 ± 1.5 wt% of OC, which suggests that WIOC was the dominant OC in $PM_{2.5}$ (92 ± 1.5 wt% of OC). The WSOC/OC ratios for Indonesian peatland fires were consistently smaller than those obtained from other biomass burning sources in other reports, such as rice straw and wheat burning. HULIS-C accounted for 55 ± 8.5 wt% of WSOC, which is consistent with other biomass burning aerosols. In particular, it should be noted that the WSOC/OC and HULIS-C/WSOC ratios are fairly constant and independent of the sampling sites.
2. By comparing the diagnostic ratios using OC, WSOC, and HULIS-C at Indonesian peatland fire sources and the receptor site in Malaysia during fire-induced haze periods, we found that the formation of secondary WSOC and HULIS-C is highly possible during transport from Indonesian peatland fire sources to receptor sites.
3. Interestingly, the SA/LG mass ratio (0.045 ± 0.0075) is fairly constant and independent of the sampling sites. Because SA is less stable than LG under atmospheric conditions, the SA/LG mass ratios is an aging indicator for Indonesian peatland fires at receptor sites.
4. Among the quantified organic compounds, LG was the most abundant compound at 13 ± 7.2 mg g-OC^{-1}. The MN/LG mass ratio for Indonesian peatland fires is similar to that for hardwood burning, irrespective of burning types (coburning of peat with surface vegetation and subsurface burning of peat). By comparing the mass fractions of each organic compound in this study and previous studies, it was suggested that the source profile is highly influenced by burning types (the coburning of peat and surface vegetation, and peat burning alone). Further knowledge of peat burning emissions is needed, particularly with respect to burning conditions, peat composition, and the effects of vegetative burning on peatland. Improved knowledge of these factors would lead to more reliable speciated emission inventories of Indonesian peatland fires, advancing chemical transport and radiative forcing modeling, as well as health risk assessment.

CRediT authorship contribution statement

Yusuke Fujii: Conceptualization, Validation, Formal analysis, Methodology, Investigation, Data curation, Writing – original draft, Visualization, Supervision, Funding acquisition. **Susumu Tohno:** Formal analysis, Resources, Writing – review & editing, Project administration, Funding acquisition. **Hiroki Kurita:** Formal analysis, Investigation, Data curation. **Haryono Setiyo Huboyo:** Investigation, Field data collection and arrangement, Funding acquisition. **Badrus Zaman:** Investigation, Sample preparation and treatment in Indonesia.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.aeaoa.2021.100116.

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