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Water uptake by fresh Indonesian peat burning particles is limited by water-soluble organic matter

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Abstract. The relationship between hygroscopic properties and chemical characteristics of Indonesian biomass burning (BB) particles, which are dominantly generated from peatland fires, was investigated using a humidified tandem differential mobility analyzer. In addition to peat, acacia (a popular species at plantation) and fern (a pioneering species after disturbance by fire) were used for experiments. Fresh Indonesian peat burning particles are almost non-hygroscopic (mean hygroscopicity parameter, $\kappa < 0.06$) due to predominant contribution of water-insoluble organics. The range of κ spans from 0.02 to 0.04 (dry diameter = 100 nm, hereinafter) for Riau peat burning particles, while that for Central Kalimantan ranges from 0.05 to 0.06. Fern combustion particles are more hygroscopic ($\kappa = 0.08$), whereas the acacia burning particles have a mediate κ value (0.04). These results suggest that κ is significantly dependent on biomass types. This variance in κ is partially determined by fractions of water-soluble organic carbon (WSOC), as demonstrated by a correlation analysis ($R = 0.65$). κ of water-soluble organic matter is also quantified, incorporating the 1-octanol–water partitioning method. κ values for the water extracts are high, especially for peat burning particles (A_0 (a whole part of the water-soluble fraction): $\kappa = 0.18$, A_1 (highly water-soluble fraction): $\kappa = 0.30$). This result stresses the importance of both the WSOC fraction and κ of the water-soluble fraction in determining the hygroscopicity of organic aerosol particles. Values of κ correlate positively ($R = 0.89$) with the fraction of m/z 44 ion signal quantified using a mass spectrometric technique, demonstrating the importance of highly oxygenated organic compounds to the water uptake by Indonesian BB particles. These results provide an experimentally validated reference for hygroscopicity of organics-dominated particles, thus contributing to more accurate estimation of environmental and climatic impacts driven by Indonesian BB particles on both regional and global scales.

1 Introduction

In Southeast Asia, tropical peatland fires, which occur by combustion of both peat and vegetation, have become frequent during the last few decades (van der Werf et al., 2010; Reddington et al., 2014; Marlier et al., 2015; Spracklen et al., 2015; Stockwell et al., 2016). As one of the most important biomass burning types, the peatland fires keep smoldering for months, releasing huge amounts of greenhouse gases and fine particles to the atmosphere, impacting atmospheric radiation (Levine et al., 1999; Page et al., 2002; van der Werf et al., 2010). In addition, the peatland burning particles cause transboundary air pollution, influencing visibility and human health (Kunii et al., 2002; Wang et al., 2004; Marlier et al., 2013; Crippa et al., 2016; Koplitz et al., 2016). These regional and global climate impacts have been commonly evaluated using different models (e.g., large-eddy simulation and global climate models) but with large uncertainties likely due to the inaccurate estimation of cloud condensation nuclei (CCN) number concentration and subsequent cloud formation and precipitation processes (Mircea et al., 2005; Rose et al., 2010; Bougiatioti et al., 2016). One of the essential uncertainty contributors to the complex aerosol–cloud–precipitation interactions is water uptake properties of peatland burning particles, as water vapor alters aerosol physical and chemical characteristics, such as particle diameter.

Water uptake properties of biomass burning particles, including those emitted from peatlands, have been explored in a laboratory through measurements of hygroscopic growth and CCN activity (Chand et al., 2005; Dusek et al., 2005, 2011; Day et al., 2006; Petters et al., 2009; Carrico et al., 2010). In general, freshly emitted biomass burning particles have been found to be hygroscopic. For instance, the reported range of hygroscopicity parameter, κ, which serves as a metric for water uptake properties, varies from weakly ($\kappa = 0.02$) to highly hygroscopic ($\kappa = 0.80$) (Day et al., 2006; Petters and Kreidenweis, 2008; Petters et al., 2009; Carrico et al., 2010). A field observation of water uptake properties of Indonesian biomass burning plumes also demonstrated that these particles are hygroscopic, with a median hygroscopic growth in light scattering (f(RH)) of 1.65 between 20 and 80 % relative humidity (RH) (Gras et al., 1999). On the other hand, freshly emitted Indonesian peat burning particles generated in a laboratory were suggested to be non-hygroscopic with respect to quite a low $f(\mathrm{RH}) = 1.05$ at 90 % RH (Chand et al., 2005), and they were almost CCN inactive especially for particles larger than 150 nm (equivalent to $\kappa = 0.05$ for 100 nm particles, calculated with an assumed surface tension of $0.072\,\mathrm{N\,m^{-1}}$ at 25 °C) (Dusek et al., 2005). The uniqueness of water uptake properties of freshly emitted Indonesian peatland burning particles as well as the discrepancy between the previously reported laboratory and field data needs to be consistently understood based on their chemical compositions for accurate evaluation on the environmental impacts.

Chemical composition of biomass burning particles, including those from Indonesian peatland fires, is dominated by a complex mixture of organic species (Jimenez et al., 2009; Ng et al., 2010; Cubison et al., 2011; Stockwell et al., 2016). The complexity in chemical composition inhibits understanding their water uptake properties at molecular levels (Asa-Awuku et al., 2008; Psichoudaki and Pandis, 2013; Riipinen et al., 2015). To overcome this difficulty, classification of organic compounds using multiple solvents (Carrico et al., 2008; Polidori et al., 2008; Chen et al., 2016), liquid–liquid extraction using 1-octanol and water (Kuwata and Lee, 2017), and solid-phase extraction (Asa-Awuku et al., 2008) has been conducted. Functional-group analysis of segregated organic matter has also been demonstrated as a strong tool to characterize complex mixture of organic compounds (Chen et al., 2016). For instance, chemical characteristics of water-soluble organic matter (WSOM) have been intensively investigated, revealing that WSOM predominantly consists of levoglucosan-like species, carboxylic acids, aldehydes, ketones, aliphatic alcohols, and polyacids (Decesari et al., 2000; Peng et al., 2001; Suzuki et al., 2001; Mayol-Bracero et al., 2002; Chan et al., 2005; Psichoudaki and Pandis, 2013). Recently, the important roles of functional groups in water uptake properties were also investigated by both theoretical and experimental approaches (Suda et al., 2014; Petters et al., 2016).

In this study, hygroscopic growth of Indonesian peatland burning particles was investigated in a series of laboratory experiments to understand the relationships between water uptake properties and chemical characteristics. Hygroscopic growth of various types of fresh peat burning particles, along with those originating from combustion of dried peatland plants, was measured using a humidified tandem differential mobility analyzer (HTDMA) for the first time. Chemical characterization was also conducted using the Aerodyne Time-of-Flight Aerosol Chemical Speciation Monitor (ToF-ACSM). In addition, ratios of water-soluble organic carbon (WSOC) to organic carbon (OC) were quantified. The concurrent HTDMA and ToF-ACSM measurements were also performed for the bulk WSOM and its highly hydrophilic fraction classified with the 1-octanol–water partitioning method in terms of water solubility (Kuwata and Lee, 2017). This method provides a new angle (i.e., water solubility) to characterize chemical properties of WSOM, facilitating a more detailed investigation of particle water uptake properties with the first application of the method in HTDMA measurements of a highly hydrophilic organic fraction. These data were synergistically combined to provide a detailed picture of water uptake properties of freshly emitted Indonesian peatland burning particles.

2 Experiment

2.1 Combustion setup

Figure 1a shows the experimental setup. Peat and biomass samples were collected at peatlands in Riau and Central Kalimantan provinces in Indonesia (Table 1). The sampling sites include both burnt and undisturbed forest areas. In this region, peatland fire frequently recurs, and vast areas experience regeneration of vegetation after fire events. The peat samples were segregated for different sampling depths, as detailed in Table 1. Two other types of biomasses from Riau, including *Pteridium aquilinum* (called fern here) and leaves of *Acacia mangium* (abbreviated as acacia), were also employed for the experiment. Fern is one of the major pioneer species after peatland fires (Aswin et al., 2004). Acacia is one of the representative trees for plantations over drained peatland. Both acacia and fern samples were dried at ambient temperature after sampling. Further detailed information on the biomass samples is available in Budisulistiorini et al. (2017).

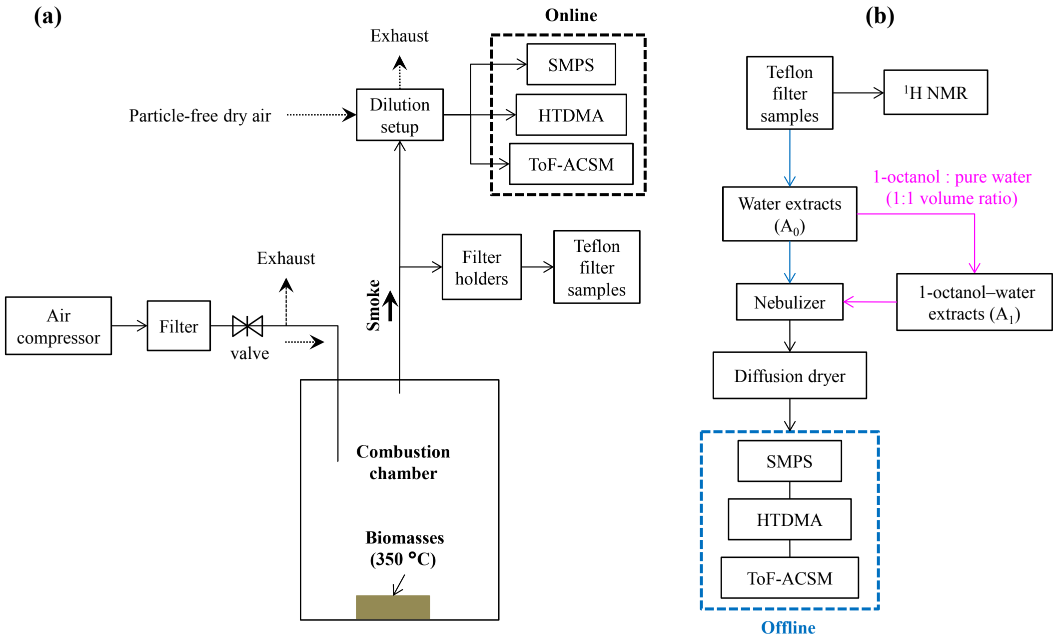

Figure 1. Schematic diagrams of the laboratory experimental setups. **(a)** The experimental setup for combustion experiment and subsequent online measurements, and **(b)** experimental setup for offline measurements of water extracts (A_0, blue arrows) and 1-octanol–water extracts (A_1, magenta arrows) from the filter samples.

Table 1. Summary of Indonesian peat and biomasses used for the laboratory combustion experiments. Samples 9, 15, and 16 were also used for offline experiments of their aqueous extracts (A_0 and A_1).

Exp. no.	Sample name	Sample depth (cm)	Type/species	Location	Description
1	Riau-1	Surface ∼ 10	Peat	Riau, Sumatra	D&B[a] peatland
2	Riau-1.1	10–20	Peat	Riau, Sumatra	D&B[a] peatland
3	Riau-1.2	30–40	Peat	Riau, Sumatra	D&B[a] peatland
4	Riau-2	Surface ∼ 10	Peat	Riau, Sumatra	D&B[a] peatland
5	Riau-2.1	10–20	Peat	Riau, Sumatra	D&B[a] peatland
6	Riau-2.2	30–40	Peat	Riau, Sumatra	D&B[a] peatland
7	Riau-3	Surface ∼ 10	Peat	Riau, Sumatra	D&B[a] peatland
8	Riau-3.1	10–20	Peat	Riau, Sumatra	D&B[a] peatland
9	Riau-4[c]	Surface ∼ 10	Peat	Riau, Sumatra	D&B[a] peatland
10	Riau-4.1	10–20	Peat	Riau, Sumatra	D&B[a] peatland
11	Riau-SF	Surface ∼ 5	Peat	Riau, Sumatra	Secondary forest
12	Riau-Zam	Surface ∼ 5	Peat	Riau, Sumatra	Undisturbed peat forest
13	C.K.-DF	Surface ∼ 5	Peat	Palangkaraya, Central Kalimantan	D&UB[b] peat forest
14	C.K.-DB	Surface ∼ 5	Peat	Palangkaraya, Central Kalimantan	D&B[a] peat forest
15	Acacia[c]	n/a	*Acacia mangium*	Riau, Sumatra	Dried leaves
16	Fern[c]	n/a	*Pteridium aquilinum*	Riau, Sumatra	Dried leaves

[a] D&B stands for the drained and burnt condition.
[b] D&UB represents the drained but unburnt case.
[c] Teflon filter samples were collected during online combustion experiments.
n/a: not applicable.

The biomass samples were used without desiccation. Approximately 1.0 g of biomass sample was combusted in a sealed 100 L stainless-steel container using a crucible, which was heated at 350 °C by a ribbon heater, thermocouple, and proportional–integral–derivative (PID) controller (Kuwata et al., 2017). The target heating temperature was normally achieved within 2–3 min. Visual inspection confirmed that the combustion condition was mostly smoldering, consistent with a previous report (Usup et al., 2004). Particle-free air was continuously supplied to the container. Excess amount of particle-free air was released to the laboratory, allowing the experiments to be conducted at room pressure. Particles generated by the burning experiments were diluted by a two-stage dilution system. Size distributions of biomass burning particles were measured using the Scanning Mobility Particle Sizer (SMPS, TSI Inc.). The measurement range of the SMPS was set as 14.6–685.4 nm, and time resolution was 3 min. Chemical compositions of particles were quantified using the Aerodyne ToF-ACSM (Fröhlich et al., 2013), while water uptake properties were measured using the HTDMA (Massling et al., 2003, 2007; Duplissy et al., 2009). Online instruments such as SMPS, ToF-ACSM, and HTDMA were all operated following the dilution. Each combustion experiment lasted for ~ 1 h. Detailed descriptions of the ToF-ACSM and HTDMA are provided in the following sections.

Two filter samples were also collected simultaneously for each of the experiments. Particles were collected onto two 47 mm diameter filters housed in stainless-steel filter holders (BGI Inc.) for half an hour at flow rates of $5.0\,\mathrm{L\,min^{-1}}$. Teflon filters (0.2 µm pore size, Fluoropore™, Sigma Aldrich) were used for WSOM samples, while quartz filter samples were employed for carbon analysis by the thermal-optical method. The collected samples were stored under refrigeration at −20 °C until analysis.

2.2 Extraction and nebulization of WSOM

Filter samples were extracted using approximately 20 mL of ultrapure water (Type I) by sonicating them for 30 min at room temperature. The resulting solutions were filtered through 0.2 µm PTFE syringe filters (514-0070, VWR), yielding water extracts (denoted as A_0, i.e., a whole part of or the bulk water-soluble fraction). An aliquot of A_0 was mixed with the same volume (5 mL) of 1-octanol (Wako first grade, Wako) using a separatory funnel for classification by 1-octanol–water extraction (Valvani et al., 1981). The aqueous phase following the 1-octanol–water extraction is denoted as A_1 (slightly less than 5 mL), corresponding to the highly water-soluble fraction. Details of the extraction method are provided in Kuwata and Lee (2017).

The aqueous solutions were nebulized using a glass nebulizer. A mass flow controller (MC-20 SLPM-D, Alicat Scientific, Inc.) was used to regulate the flow rate ($3.5\,\mathrm{L\,min^{-1}}$) of particle-free air supplied to the nebulizer. Following nebulization, the sample was desiccated by a diffusion dryer (model 42000, Brechtel Manufacturing, Inc.) filled with silica gel (Chameleon 83000.360, VWR International). The desiccated particles were measured using the HTDMA, ToF-ACSM, and SMPS. The analysis of A_0 and A_1 was conducted only for peat (sampled from a burnt area, Riau-4), acacia, and fern samples.

2.3 HTDMA

Hygroscopic growth of particles was measured using the HTDMA (Massling et al., 2003, 2007; Duplissy et al., 2009; Gysel et al., 2009). The HTDMA system consists of three major components: (1) the first DMA (TSI Inc. model 3081) to select monodisperse particles of a specific diameter, (2) the humidification unit for hydrating the classified particles at a target RH, and (3) the second DMA (TSI Inc. model 3081) and a condensation particle counter (CPC, TSI Inc. model 3775) to detect humidified size distributions (Fig. S1 in the Supplement).

Aerosol particles were dried using a diffusion dryer (model 42000, Brechtel Manufacturing, Inc.), and introduced to the first DMA at a flow rate of $0.3\,\mathrm{L\,min^{-1}}$. The first DMA selected 50, 100, and 200 nm particles. Both the first and second DMAs were operated at sheath-to-sample flow ratios of 10 : 1. The resulting monodisperse particles were exposed to a predefined RH environment using two Nafion membrane tubes (Permapure Inc. model MD-110-12S-4) in series. The target RH, which was set at 90 %, was regulated by controlling the flow ratio of humidified and dry airflows via PID controlling software (LabVIEW, National Instruments Inc.). The particle residence time between the humidifier and the second DMA was approximately 10 s. The RH-controlled humid air was used as the sheath flow for the second DMA. The RH and temperature of the second DMA were continuously monitored at both the sample inlet and sheath outlet using two capacitive RH and temperature probes (Rotronic Inc. Hygroclip HC2-S). The RH differences between the sample and sheath flows were less than 2 %. RH of humidified sample air was slightly higher than that of the sheath outflow for the setup shown in Fig. S1.

The operating conditions of the DMAs were checked using 100 ± 3 nm polystyrene latex particles (PSL, Thermo Scientific Inc., cat. no. 3100A). Hygroscopic growth of particles in the HTDMA was calibrated by measuring growth factor (g), which is defined as a diameter ratio of humidified (D (RH)) and dry particles (D_0) ($g = (D(\mathrm{RH})/D_0)$, of ammonium sulfate particles. The measured value of g at RH = 85 % was 1.71 ($D_0 = 150$ nm), which is comparable to literature data ($g = 1.69$) (Tang and Munkelwitz, 1994). The stability of detected RH of the HTDMA system was within ±1.0 % RH (peak to peak) for the target RH of 90 % (see Fig. S2). A volume mean growth factor (GF) was calculated using measured probability density function of g (i.e., $c(g, D_0)$) as $\mathrm{GF} = (\int_0^\infty g^3 c(g, D_0)\,dg)^{1/3}$.

2.4 Chemical characterization

The Aerodyne ToF-ACSM was utilized to measure chemical compositions of non-refractory submicron particles ($\mathrm{NR\text{-}PM_1}$) (Fröhlich et al., 2013). Five specific chemical components – including organic matter (OM), sulfate ($\mathrm{SO_4^{2-}}$), nitrate ($\mathrm{NO_3^-}$), ammonium ($\mathrm{NH_4^+}$), and chloride ($\mathrm{Cl^-}$) – were quantitatively detected (Allan et al., 2003), with a time resolution of 3 min.

Bulk OC, elemental carbon (EC), and WSOC contents were also analyzed for 10 samples (Table 3) using quartz-fiber filter samples (47 mm in diameter). All the quartz-fiber filters were prebaked at 900 °C for 3 h before sampling. The filter samples were stored in a refrigerator (−20 °C) until analysis. For each sampling, a backup quartz-fiber filter was used to account for potential influence of adsorption of gas phase organic components (Turpin et al., 1994). OC loading on the backup filter was subtracted from that on the front filter to estimate particulate OC (i.e., corrected OC).

OC and EC were analyzed by thermal-optical reflectance analysis (Chow et al., 1993) using a Sunset Laboratory OC–EC analyzer, following the IMPROVE-A protocol. WSOC was quantified with a Sievers 800 total organic carbon (TOC) analyzer after extraction of biomass burning filter samples by water. A portion (8 mm φ) of each WSOC sample was extracted using 10 mL of HPLC-grade water. The samples were shaken by an orbital shaker for 21 h. The extracted sample solutions were filtered with syringe filters (pore size of 0.2 µm) prior to injection to the TOC analyzer. The particulate WSOC of peat burning particles was also corrected following a similar procedure to that of particulate OC.

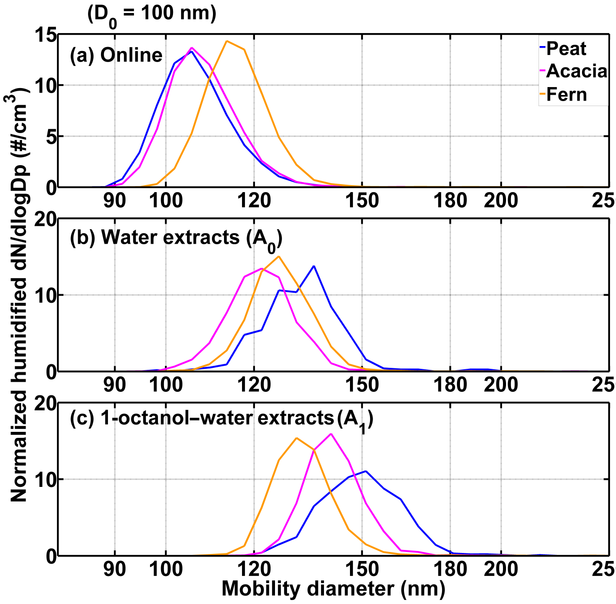

Figure 2. Normalized number size distributions measured by the HTDMA ($D_0 = 100$ nm; RH = 90 %) of peat, fern, and acacia burning particles. **(a)** Online data, **(b)** A_0, and **(c)** A_1. For online data, peat burning particles are nearly non-hygroscopic, while fern burning particles are more hygroscopic. Aqueous extracts of peat burning particles are the most hygroscopic among the three types of biomasses. The x axis is on a logarithmic scale.

3 Results and discussion

Both the HTDMA and chemical analysis data are summarized in Tables 2 and 3. The ToF-ACSM and OC–EC data (Table 3) demonstrate that chemical composition of submicron biomass burning particles is dominated by organic species, accounting for approximately 99 % in mass (Budisulistiorini et al., 2017). Contributions of other species, including inorganic ionic species and EC, were minimal. In the following, the relationships between hygroscopic properties and chemical characteristics of organic species are discussed.

3.1 Hygroscopic growth factor

Figure 2 shows normalized particle number size distributions of peat sampled from a burnt area (Riau-4), acacia leaves, and fern burning particles following hygroscopic growth at RH = 90 % ($D_0 = 100$ nm). The data shown in Fig. 2 include both online (a) and offline (b: A_0; c: A_1) measurements. In all cases, narrow monodisperse distributions were observed (see Fig. S3), demonstrating that chemical compositions of particles were uniform (Gysel et al., 2007; Carrico et al., 2010). For online measurements, diameter change induced by hygroscopic growth was minimal for the peat and acacia burning particles ($g = 1.05$–1.09), while diameter of fern burning particles significantly increased following exposure to high RH ($g = 1.17$). The variation in hygroscopic properties is attributed to differences in organic chemical composition, as these biomass burning particles contain negligible fractions of inorganic ionic species (Budisulistiorini et al., 2017).

Table 2 summarizes all the values of GF. Values of GF for most of peat samples from burnt peatland in Riau were less than 1.1. Sampling depths of peat did not significantly affect GF. There was no clear size dependence of GF. For instance, GF values of particles from combustion of peat at drained and burnt areas in Riau were 1.07 ± 0.04 ($D_0 = 50$ nm), 1.06 ± 0.02 ($D_0 = 100$ nm), and 1.07 ± 0.02 ($D_0 = 200$ nm). Particles emitted from the undisturbed forest area in Riau (i.e., Riau-Zam) were more hygroscopic (GF = 1.11 for $D_0 = 100$ nm) than those generated from other samples from Riau, while GF of particles emitted by combustion of a peat sample from the secondary forest in Riau (i.e., Riau-SF) was very similar to those from Riau peat samples from burnt areas (i.e., Riau-1–4). The similarity is probably due to the short distance between the two sampling sites (less than 10 km). Particles emitted from peat samples collected in Central Kalimantan (i.e., C.K.-DB and C.K.-DF) were relatively more hygroscopic (GF > 1.11) than those from Riau.

Hygroscopic growth of the bulk water-soluble fraction (A_0) is much more significant than that of the online measurements. Specifically, the mean diameter growth factors were 1.34 (peat sampled from a burnt area, Riau-4), 1.23 (acacia), and 1.28 (fern) for 100 nm particles. The significant hygroscopic growth of A_0 from peat burning particles

Table 2. Volume weighted mean GF and κ values (average ±1 standard deviation, SD) of different types of Indonesian peat and biomasses. The results of their aqueous extracts (A_0 and A_1) are also shown.

	Sample name		Mean GF (RH = 90 %)			Mean κ (RH = 90 %)		
			50 nm	100 nm	200 nm	50 nm	100 nm	200 nm
Online	Sumatra	Riau-1	1.17 ± 0.07	1.09 ± 0.06	1.04 ± 0.01	0.089 ± 0.042	0.039 ± 0.028	0.016 ± 0.002
		Riau-1.1	1.15 ± 0.06	1.05 ± 0.01	1.04 ± 0.01	0.080 ± 0.036	0.021 ± 0.005	0.014 ± 0.002
		Riau-1.2	1.00 ± 0.002	1.08 ± 0.04	1.06 ± 0.01	0.001 ± 0.001	0.036 ± 0.017	0.025 ± 0.002
		Riau-2	–	1.07 ± 0.01	1.12 ± 0.06	–	0.029 ± 0.005	0.052 ± 0.032
		Riau-2.1	1.06 ± 0.04	1.06 ± 0.01	1.06 ± 0.01	0.029 ± 0.019	0.023 ± 0.005	0.024 ± 0.005
		Riau-2.2	1.05 ± 0.02	1.09 ± 0.02	1.10 ± 0.02	0.024 ± 0.010	0.037 ± 0.009	0.042 ± 0.009
		Riau-3	1.07 ± 0.04	1.05 ± 0.01	1.08 ± 0.01	0.035 ± 0.022	0.022 ± 0.006	0.033 ± 0.006
		Riau-3.1	1.05 ± 0.02	1.05 ± 0.02	1.08 ± 0.01	0.024 ± 0.012	0.022 ± 0.010	0.031 ± 0.006
		Riau-4	1.04 ± 0.01	1.08 ± 0.01	1.05 ± 0.002	0.017 ± 0.003	0.034 ± 0.003	0.019 ± 0.001
		Riau-4.1	1.07 ± 0.10	0.99 ± 0.01	1.02 ± 0.01	0.059 ± 0.063	n/a	0.007 ± 0.003
	(Burnt peatland)	Riau D&B[b]	1.07 ± 0.04	1.06 ± 0.02	1.07 ± 0.02	0.040 ± 0.023	0.029 ± 0.010	0.026 ± 0.007
	(Secondary forest)	Riau-SF	1.04 ± 0.05	1.07 ± 0.004	1.09 ± 0.01	0.025 ± 0.028	0.028 ± 0.002	0.034 ± 0.003
	(Undisturbed area)	Riau-Zam	1.10 ± 0.07	1.11 ± 0.04	1.08 ± 0.004	0.053 ± 0.038	0.048 ± 0.017	0.032 ± 0.002
	Kalimantan	C.K.-DF	1.11 ± 0.06	1.13 ± 0.01	1.11 ± 0.01	0.057 ± 0.033	0.058 ± 0.005	0.046 ± 0.004
		C.K.-DB	1.11 ± 0.05	1.12 ± 0.02	1.13 ± 0.01	0.055 ± 0.028	0.054 ± 0.011	0.056 ± 0.005
		Acacia	1.05 ± 0.01	1.09 ± 0.01	1.09 ± 0.01	0.026 ± 0.005	0.039 ± 0.006	0.037 ± 0.006
		Fern	1.08 ± 0.02	1.17 ± 0.02	1.20 ± 0.03	0.039 ± 0.011	0.078 ± 0.010	0.088 ± 0.014
Aqueous extracts: A_0 (the water extracts), A_1 (the 1-octanol–water extracts)								
Offline	A_0	Peat[a]	1.29 ± 0.05	1.34 ± 0.06	1.38 ± 0.09	0.168 ± 0.039	0.179 ± 0.038	0.198 ± 0.058
		Acacia	1.17 ± 0.03	1.23 ± 0.04	1.28 ± 0.05	0.090 ± 0.019	0.110 ± 0.025	0.135 ± 0.027
		Fern	1.18 ± 0.03	1.28 ± 0.03	1.32 ± 0.05	0.100 ± 0.016	0.141 ± 0.020	0.157 ± 0.031
	A_1	Peat[a]	1.47 ± 0.06	1.50 ± 0.09	1.47 ± 0.11	0.311 ± 0.052	0.302 ± 0.074	0.262 ± 0.083
		Acacia	1.32 ± 0.04	1.42 ± 0.03	1.44 ± 0.07	0.195 ± 0.027	0.237 ± 0.023	0.239 ± 0.049
		Fern	1.28 ± 0.04	1.33 ± 0.05	1.39 ± 0.05	0.162 ± 0.026	0.177 ± 0.034	0.205 ± 0.038

– Data are unavailable due to low particle number concentration.

[a] Riau D&B represents the averages of all the D&B peat samples collected from different depths of the Sumatran peatlands (i.e., samples used for Exp. 1–10 in Table 1).

[b] Peat in Table 2 refers to the Riau-4 sample collected from burnt peatlands in Sumatra (see Sect. 2 for details).

could be due to high water uptake by the highly water-soluble fraction, A_1 (GF = 1.50). The GFs of A_1 for acacia and fern burning particles were 1.42 and 1.33, respectively. Although water uptake by fresh peat burning particles was much less than that of vegetation burning particles, the water-soluble fraction of peat burning particles was the most hygroscopic. This result stresses the importance of understanding hygroscopic properties of WSOM as well as the WSOC fraction in total OC.

3.2 Hygroscopicity parameter (κ)

Hygroscopicity parameter (κ) was calculated using the κ–Köhler theory (Petters and Kreidenweis, 2007):

$$\kappa = (\mathrm{GF}^3 - 1) \cdot \left(\frac{\exp\left(\frac{4\sigma_{\mathrm{s/a}} \cdot M_{\mathrm{w}}}{\rho_{\mathrm{w}} \cdot R \cdot T \cdot D_0 \cdot \mathrm{GF}}\right)}{\mathrm{RH}} - 1 \right), \tag{1}$$

where $\sigma_{\mathrm{s/a}}$ is the surface tension of the solution–air interface (0.0718 N m^{-1} at 25 °C), M_{w} is the molecular weight of water (18 g mol^{-1}), ρ_{w} is the density of water (1.0 g cm^{-3}), R is the universal gas constant (8.31 J K^{-1} mol^{-1}), and T is temperature (298 K). The calculated κ results for our HTDMA measurements are summarized in Fig. 3 and Table 2. It is worth noting that κ is related to the molar volume of water-soluble compounds ($M_{\mathrm{s}}/\rho_{\mathrm{s}}$), which is calculated from both the molecular weight (M_{s}) and density (ρ_{s}) by the following equation (Rose et al., 2008):

$$\kappa = i_{\mathrm{s}} \frac{\rho_{\mathrm{s}} M_{\mathrm{w}}}{\rho_{\mathrm{w}} M_{\mathrm{s}}}, \tag{2}$$

where i is the van't Hoff factor. Eq. (2) was mainly employed to derive κ from the experimental data of Asa-Awuku et al. (2008), who calculated the mean molar volume of WSOM extracted from biomass burning particles with a CCN measurement.

The range of κ for peat burning particles in Riau (sampled from burnt areas) is 0.02 to 0.04, while that for Central Kalimantan samples is 0.05 to 0.06 (100 nm). These values may be compared with CCN activity of peat burning particles reported by Dusek et al. (2005). Based on the experimental data by Dusek et al. (2005), the critical supersaturation for CCN activation of Indonesian peat burning particles is derived as 0.53 % for 100 nm particles. This value can be converted to

Table 3. Summary of chemical characteristics of different types of Indonesian peat and biomass burning particles.

Sample name		Mean κ	f_{44}	f_{HMW}	OC	EC	WSOC / OC
		(100 nm)	(%)	(%)	(mg C)	(mg C)	(%)
Sumatra	Riau-1	0.039	0.4	42.9	12.69	0.13	0.93
	Riau-2	0.029	0.7	26.9	14.08	0.12	1.80
	Riau-3	0.022	0.7	31.7	13.58	0.13	1.63
	Riau-4	0.034	0.5	29.0	18.86	0.13	1.51
	Riau-SF	0.028	1.7	21.1	7.64	0.07	4.15
	Riau-Zam	0.048	1.6	23.8	2.58	0.03	6.08
Kalimantan	C.K.-DF	0.058	2.0	19.5	5.58	0.05	4.16
	C.K.-DB	0.054	1.9	19.3	7.51	0.05	2.03
	Acacia	0.039	1.1	27.0	14.61	0.05	3.42
	Fern	0.078	1.9	21.5	13.34	0.07	6.56
A_0	Peat[a]	0.179	9.5	15.8	n/a	n/a	n/a
	Acacia	0.110	4.7	21.8	n/a	n/a	n/a
	Fern	0.141	5.2	17.4	n/a	n/a	n/a
A_1	Peat[a]	0.302	9.3	16.0	n/a	n/a	n/a
	Acacia	0.237	6.6	11.6	n/a	n/a	n/a
	Fern	0.177	7.9	8.2	n/a	n/a	n/a

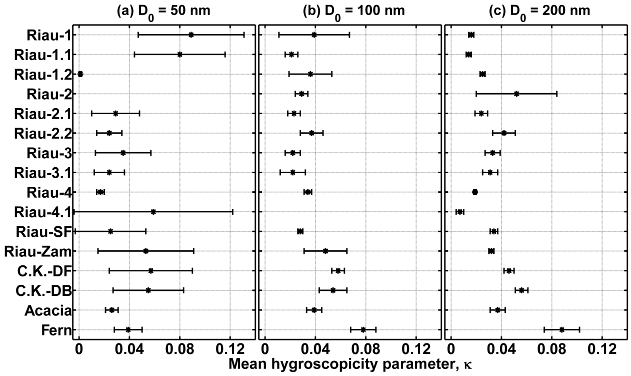

Figure 3. Mean κ values of fresh Indonesian biomass burning particles measured for **(a)** $D_0 = 50$ nm, **(b)** $D_0 = 100$ nm, and **(c)** $D_0 = 200$ nm particles. In all cases, κ values are lower than 0.1. The largest κ values were measured for fern burning particles ($D_0 = 100$ nm and $D_0 = 200$ nm). Particles emitted from combustion of peat in Central Kalimantan are generally more hygroscopic than those from Riau, Sumatra. Error bars denote the corresponding standard deviations.

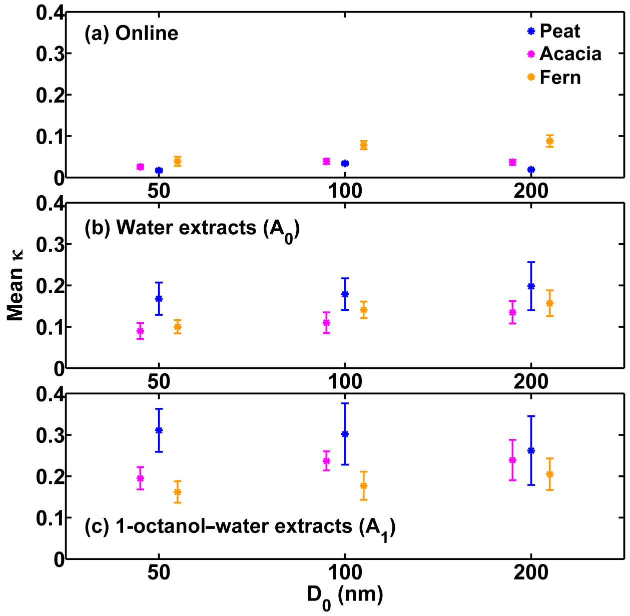

Figure 4. Comparison of κ values for **(a)** online, **(b)** water extracts (A_0), and **(c)** 1-octanol–water extracts (A_1). The data for peat (Riau-4), acacia, and fern are shown. Indonesian peat burning particles are almost non-hygroscopic for online data, while their water-soluble organic fractions are highly hygroscopic.

κ of 0.05, which is very similar to the values summarized in Fig. 3 and Table 2. The consistently low values of κ suggest that water uptake by freshly emitted peat burning particles is minimal. The range of κ observed for acacia and fern burning particles ($\kappa = 0.04$–0.08) is comparable to that observed for a less hygroscopic mode by previous laboratory experiments on biomass burning particles (Carrico et al., 2010).

The values of κ observed for water extracts (A_0) span from 0.11 (acacia, 100 nm) to 0.18 (peat sampled from a burnt area, Riau-4, 100 nm) (Fig. 4 and Table 2). The κ value for peat burning particles (A_0) is significantly higher than those emitted from the acacia and fern leaves, highlighting the importance of understanding hygroscopicity of the WSOM as well as the water-soluble fraction in order to quantitatively understand water uptake properties. The value of κ for acacia burning particles is similar to what was measured for WSOM extracted from a prescribed forest fire experiment in Georgia (USA) ($\kappa = 0.10$), which was estimated using a molar volume of $1.6 \times 10^{-4}\,\mathrm{m^3\,mol^{-1}}$ (Asa-Awuku et al., 2008).

The κ values for A_1 are higher than those for A_0. Namely, κ values observed for A_1 were 0.30 (peat sampled from a burnt area, Riau-4), 0.24 (acacia), and 0.18 (fern). Although it is the first hygroscopic measurement for WSOM classified with the 1-octanol–water liquid–liquid extraction technique, the value could be compared with those for hydrophilic fractions classified by the XAD-8 column. Kuwata and Lee (2017) demonstrated that classification of WSOM by the XAD-8 column, which is one of the most frequently used materials for solid-phase extraction of WSOM, has a strong relationship with the 1-octanol–water partitioning coefficient (K_{OW}). Namely, the XAD-8 column selectively traps hydrophobic chemical species in WSOM, which tend to partition to the 1-octanol phase ($K_{\mathrm{OW}} > 1$). Thus, the hydrophilic fraction separated by XAD-8 is dominantly composed of chemical species with $K_{\mathrm{OW}} < 1$. The WSOM in A_1 is also dominantly composed of organic compounds with lower values of K_{OW} ($K_{\mathrm{OW}} < 1$), suggesting that A_1 and WSOM classified by XAD-8 are comparable. The κ value of biomass burning WSOM separated by XAD-8 is estimated as 0.29, using molar volume ($6.2 \times 10^{-5}\,\mathrm{m^3\,mol^{-1}}$) estimated from a CCN measurement by Asa-Awuku et al. (2008). The comparison provides a typical range of κ for the hydrophilic ($K_{\mathrm{OW}} < 1$) fraction of WSOM emitted from biomass burning as 0.2–0.3. Our results of water uptake by organic compounds (e.g., bulk organic material, bulk WSOM, and highly hydrophilic WSOM) could be further employed to verify a theoretical framework which uses distributions of water solubility as input parameters (Riipinen et al., 2015).

3.3 κ (online) and WSOC / OC

WSOC / OC ratios of Indonesian peat and vegetation burning particles are summarized in Table 3. In general, WSOC / OC ratios for peat burning particles from the burnt area in Riau are small, ranging from 0.93 to 1.80 %. Particles emitted from combustion of peat collected in other areas tend to contain higher fractions of WSOC (WSOC / OC = 2.03–6.08 %). The variability in WSOC / OC ratios could be due to differences in chemical composition of peat sampled at different areas (Hikmatullah and Sukarman, 2014). These values are an order of magnitude lower than the experimental data by Iinuma et al. (2007), which reported WSOC / OC ratios for Indonesian peat burning particles from South Sumatra as 39 %. The significant difference in WSOC / OC ratios could stem from the variations in chemical compositions of peat as well as combustion conditions. Both a systematic laboratory experiment and chemical analysis of freshly emitted peat burning particles are needed to address the difference in the data. The WSOC fractions for acacia and fern burning particles were relatively higher (WSOC / OC = 3.42–6.56 %) than those from peat combustion.

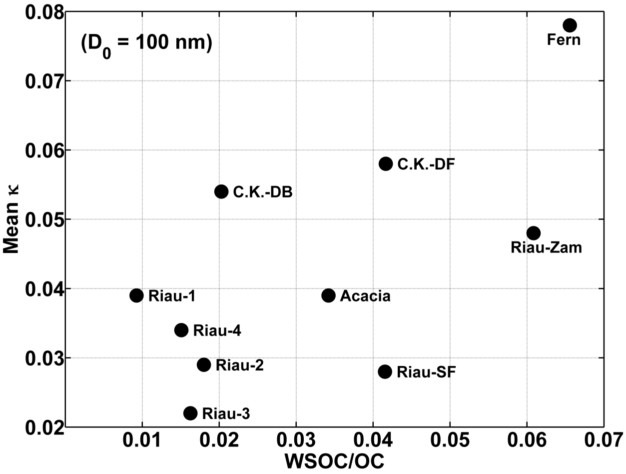

Figure 5. Correlation of κ and WSOC / OC ratios for different types of Indonesian peat and biomasses.

Figure 5 compares κ and WSOC / OC ratios. κ and WSOC / OC correlate to some extent ($R = 0.65$), although the variation ranges for both variables are small. Fern burning particles contain a significantly higher fraction of WSOM than other samples, providing an explanation for the higher κ value for fern burning particles. Nevertheless, the correlation between these two parameters is not tight. This result suggests that other factors, such as chemical composition and hygroscopic properties of the water-soluble fraction, should also be considered to quantitatively understand water uptake properties.

3.4 κ and OM mass spectra

Figure 6 shows the ToF-ACSM mass spectra for online, A_0, and A_1 particles, including those from peat (sampled at a burnt area, Riau-4), acacia, and fern burning. The online mass spectra have intense signals at m/z 41 ($C_3H_5^+$), 43 (most likely $C_3H_7^+$), 55 ($C_4H_7^+$), and 57 ($C_4H_9^+$), suggesting that these particles are highly hydrogenated (Canagaratna et al., 2015). On the other hand, fractions of ions at m/z 44 (f_{44}, mostly CO_2^+) are limited ($f_{44} < 0.02$), especially for peat burning particles. This result signifies that the freshly emitted Indonesian biomass burning particles, especially those from peat, are not highly oxygenated (Ng et al., 2011). This is in accordance with a previous study, which showed that f_{44} values for primary hydrocarbon-like organic compounds are usually less than 0.05 (Ng et al., 2011). In addition, m/z 60 and 73 (mainly from $C_2H_4O_2^+$ and $C_3H_5O_2^+$, respectively), marker ions of levoglucosan-like species (a tracer for cellulose in biomass burning particles) (Simoneit et al., 1999;

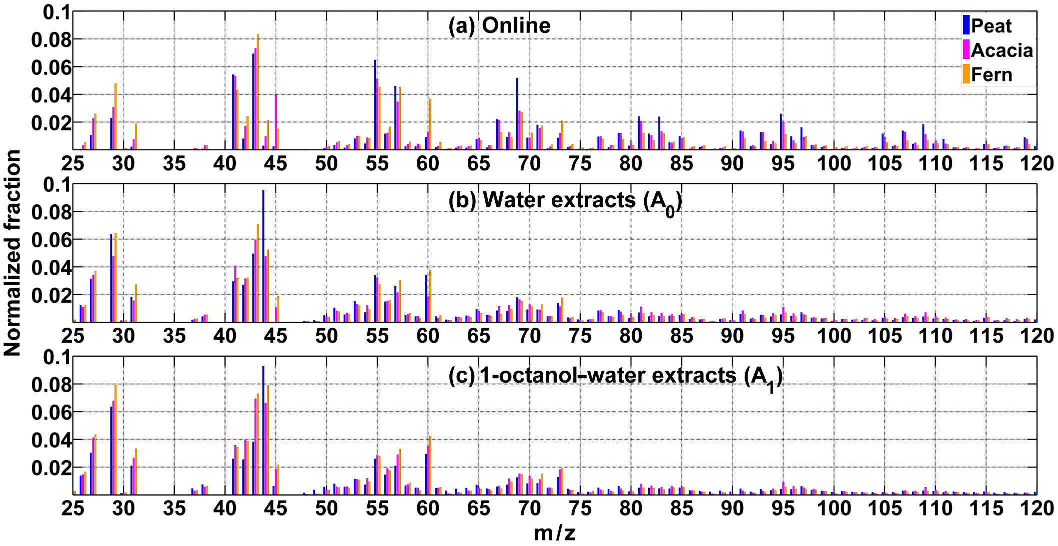

Figure 6. Mass spectra of organics in Indonesian biomass burning particles measured for peat, acacia, and fern samples. **(a)** Online, **(b)** A_0, and **(c)** A_1 data are shown. Ion signals (m/z) from hydrocarbon-like organic compounds (e.g., m/z 41, 43, 55, 57) are prominent for online data, while intensities of ions for oxygenated organics (e.g., m/z 44) and biomass burning tracers (e.g., m/z 60, 73) are relatively less intense. On the other hand, both m/z 44 and m/z 60 signals are significant for mass spectra of A_0 and A_1. See the text for details.

Cubison et al., 2011), were especially pronounced for fern burning particles.

The mass spectra of A_0 are significantly different from those of online measurements. The most abundant ion in the mass spectra of A_0 is m/z 44. Hydrocarbon peaks – such as m/z 41, 43, 55, and 57 – are still significant, yet they are less abundant than those of the online measurements. In addition, contributions of m/z 60 and 73 are also enhanced. These results consistently support the idea that the A_0 fraction is highly oxygenated. Especially, the A_0 fraction for peat burning particles is much more oxidized than those of fern and acacia samples.

The mass spectra of A_1 from acacia and fern burning show that the A_1 fraction is more oxidized than A_0, as indicated by higher values of f_{44}. For instance, f_{44} of A_1 from fern burning particles is 0.08, while that of A_0 is 0.05. Another notable characteristic of A_1 mass spectra is the smaller fraction of high-molecular-weight (HMW) ions, which is observed for the region of $m/z > 100$. The HMW fractions (f_{HMW}) for A_0 and A_1 are 15.8 and 16.0 % (peat sampled at a burnt area, Riau-4), 21.8 and 11.6 % (acacia), and 17.4 and 8.2 % (fern), which are significantly lower than the corresponding values for online measurements (Table 3). These results suggest that A_1 contains lower fractions of high-molecular-weight species, although decomposition during ionization process makes the estimation of actual contributions of these compounds difficult.

Figure 7 displays f_{44}, f_{60}, and mean κ for different types of Indonesian biomass burning particles. The data points in Fig. 7 distribute to two different regions. Low f_{44} and f_{60} values are observed for particles emitted from Sumatran peat burning (i.e., Riau-1, -2, -3, -4). The f_{44} of acacia burning particles is slightly higher, but f_{60} is low. By contrast, distinctly higher f_{44} and f_{60} are observed for fern, undisturbed peat (Riau), and peat (Central Kalimantan) burning particles. This is also in accordance with the proton nuclear magnetic resonance (^{1}H NMR) analysis, which suggests that functional-group distributions of peat and acacia burning particles are significantly different from those emitted from fern burning particles. Namely, the peat and acacia samples contain a significantly higher fraction of the saturated aliphatic group (i.e., H–C; 71.7 % for peat, and 64.0 % for acacia) than the fern sample (38.6 %; see Fig. S4a), which readily prohibits the bulk hygroscopic growth of fresh peat burning particles. Besides, the highly polar structure (i.e., H–C–O) in the

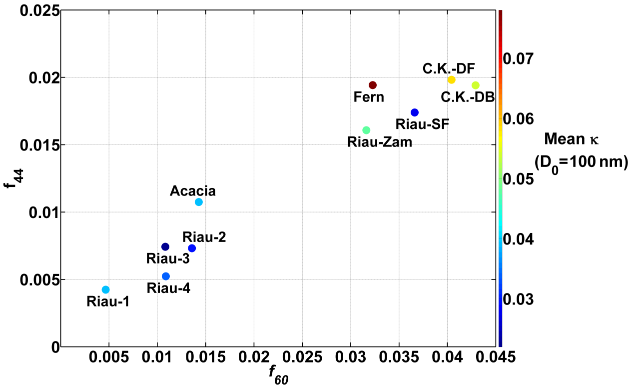

Figure 7. Correlation of f_{44} and f_{60} for different types of Indonesian peat and biomasses.

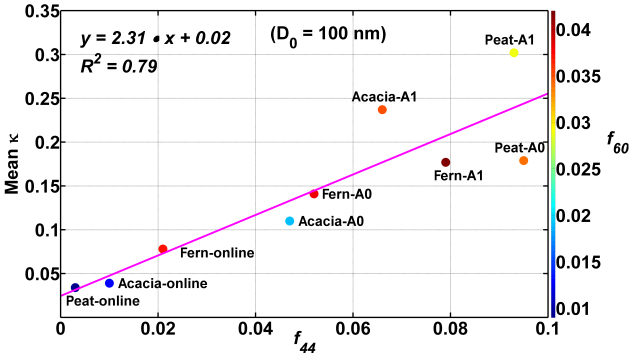

Figure 8. Correlation of κ and f_{44}. The magenta line shows the result of fitting by the least-squares method.

peat (6.0 %) and acacia (8.1 %) samples is distinctly lower than that in the fern sample (15.5 %, Fig. S4a). This likely contributes to the higher WSOC fraction of fern burning particles, and the corresponding higher κ values could be related to the higher f_{44} and f_{60}.

Figure 8 shows a correlation of κ with f_{44} for both online (i.e., bulk organic matter) and offline (i.e., bulk WSOM and highly hydrophilic WSOM fraction) measurements of peat sampled at a burnt area (Riau-4), as well as acacia and fern burning particles. Similar correlations for all the biomass burning samples are displayed in Fig. S5 of the Supplement. The correlation of these two variables is represented as $\kappa = 2.31 \times f_{44} + 0.02$ ($R = 0.89$). The slope is very close to that reported for the relationship between the hygroscopicity of organics (κ_{org}) and f_{44} ($\kappa_{\mathrm{org}} = 2.2 \times f_{44} - 0.13$; Duplissy et al., 2011). The correlation demonstrates that the degree of oxidation, which is represented by f_{44}, is the key controlling parameter in determining hygroscopicity of Indonesian peatland burning particles. As discussed above, f_{44} of peat burning particles (Riau-4) is extremely small (Table 3). Minimal water uptake by peat burning particles (Riau-4) could be associated with a low water-soluble fraction (Table 3), considering that both κ and f_{44} for the corresponding A_0 or A_1 sample are high. One notable difference in the correlation found in this work from previous studies is the inclusion of a highly soluble fraction in the analysis. Although the κ–f_{44} correlations have been related to enhanced water solubility, the relationships among these three parameters (i.e., κ, f_{44}, and water solubility) have not been shown prior to this study.

The correlation shown in Fig. 8 has a significant divergence, especially at the region for high f_{44} and κ values (upper right corner of the figure), suggesting that degree of oxidation is not the only parameter that controls water uptake properties. Especially, comparison of A_0 and A_1 for peat burning particles highlights the limitation of correlating f_{44} and κ. The values of f_{44} for these two fractions are almost the same, while κ for A_1 is significantly higher than that for A_0. As discussed in Sect. 3.2, the difference of A_0 and A_1 could be related to that of hydrophobic and hydrophilic WSOM separated by XAD-8 (Graber and Rudich, 2006; Sullivan and Weber, 2006). The hydrophobic fraction separated by XAD-8 is typically considered as humic-like substances (HULIS), which has high molecular weight (e.g., fulvic and humic acids; Gysel et al., 2004; Graber and Rudich, 2006; Fan et al., 2013). These results suggest that quantification of HULIS as well as evaluation of their water uptake properties will be important for understanding hygroscopicity of biomass burning particles, including those emitted from Indonesian peatland fires.

The present study demonstrates the importance of water-soluble organic fractions, which include a highly soluble one, in quantifying the hygroscopic growth of freshly emitted biomass burning particles. Addition of these different water-soluble organic fractions could provide an accurate estimation of hygroscopic growth, which is based on a theoretical background. Only size-unresolved bulk chemical data were employed for the present study. However, chemical characteristics of actual atmospheric particles could depend on both particle size and mixing state. These factors would also need to be considered when applying the laboratory data to future studies.

4 Conclusions

Hygroscopic growth of freshly emitted Indonesian biomass burning particles was investigated in the laboratory using a humidified tandem differential mobility analyzer (RH = 90 %). The biomass samples included peat, fern, and acacia leaves collected in Riau and Central Kalimantan in Indonesia. Hygroscopicity was measured for the freshly emitted particles (online), the bulk water-soluble fraction (A_0), and the highly water-soluble fraction (i.e., fraction with lower K_{OW} values) classified by the 1-octanol–water partitioning method (A_1). Hygroscopicity parameter κ was derived from the growth factor data. Chemical compositions of these particles were also quantified using both online and offline techniques.

Hygroscopicity of fresh Indonesian peat burning particles is highly dependent on the origin and condition (e.g., pristine and disturbed) of peat. Particles emitted from combustion of disturbed peat in Riau were nearly non-hygroscopic (mean $\kappa = 0.02$–0.04), while those from undisturbed areas were more hygroscopic ($\kappa = 0.03$–0.05). Particles emitted from Central Kalimantan peat samples were generally more hygroscopic ($\kappa = 0.05$–0.06) than those from Riau. For biomass samples, acacia burning particles were slightly hygroscopic ($\kappa = 0.03$–0.04), and fern burning particles were the most hygroscopic ($\kappa = 0.04$–0.09) among all samples. These values loosely correlated with the ratio of WSOC to OC ($R = 0.65$). This result suggests that the WSOC fraction could play an important role in determining κ, yet other factors, such as difference in hygroscopicity of slightly and highly water-soluble fractions, should also be considered.

Hygroscopicity data of A_0 and A_1 fractions were significantly different from those of online measurements. The values of κ for A_0 were 0.17–0.20 (Riau disturbed peat), 0.09–0.14 (acacia), and 0.10–0.16 (fern). These values were even higher for A_1 ($\kappa = 0.26$–0.31 for Riau disturbed peat, $\kappa = 0.19$–0.24 for acacia, and $\kappa = 0.16$–0.20 for fern). These results demonstrate that the low hygroscopicity of Riau peat burning particles (online) is due to the small water-soluble fraction.

The variation in κ was related to aerosol mass spectra of organics. f_{44}, which is an indicator for degree of oxygenation, correlated well with κ ($R = 0.89$), demonstrating that oxygenated functional groups are important for water uptake. In addition, comparison of A_0 and A_1 data suggested the importance of high-molecular-weight species, such as humic-like substances, in determining the magnitude of hygroscopicity for water-soluble fractions.

Our experimental results are consistent with previous laboratory studies, which have reported insignificant water uptake by fresh Indonesian peat burning particle (Chand et al., 2005; Dusek et al., 2005). However, Gras et al. (1999) showed that particles observed in a wildfire plume from Kalimantan were hygroscopic. The differences between field observation and laboratory experiments could be caused by atmospheric processes (e.g., secondary formation and chemical aging of particles) and likely resulted from the differences of burnt materials, their origins, and combustion conditions in practical situations as well. In the future, observations of both chemical composition and hygroscopic growth of particles emitted from peatland fires need to be conducted both locally and in downstream regions to address the question. Last but not the least, our results can provide an experimentally validated reference for organics-dominated particle hygroscopicity, thus lowering uncertainties in current climate models and contributing to more accurate estimations of climate impacts caused by Indonesian peatland burning particles on both regional and global scales.

Data availability. The data set for this publication is available upon contacting the corresponding authors.

The Supplement related to this article is available online at https://doi.org/10.5194/acp-17-11591-2017-supplement.

Competing interests. The authors declare that they have no conflict of interest.

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