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Distribution and degradation of terrestrial organic matter in the sediments of peat-draining rivers, Sarawak, Malaysian Borneo

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Abstract.

Tropical peatlands are one of the largest pools of terrestrial organic carbon (OCterr); however, our understanding of the dynamics of OCterr in peat-draining rivers remains limited, especially in Southeast Asia. This study used bulk parameters and lignin phenols concentrations to investigate the characteristics of OCterr in a tropical peat-draining river system (the main channel of the Rajang and three smaller rivers) in the western part of Sarawak, Malaysian Borneo. The depleted $\delta^{13}C$ levels and lignin composition of the organic matter indicates that the most important plant source of the organic matter in these rivers is woody angiosperm C3 plants, especially in the three small rivers sampled. The diagenetic indicator ratio (i.e., the ratio of acid to aldehyde of vanillyl phenols (Ad/Al)v) increased with decreasing mean grain size of sediment from the small rivers. The selective sorption of acid relative to aldehyde might explain the variations in the (Ad/Al)v ratio. The (Ad/Al)v ratio appears to be related to the C/N ratio (the ratio of total organic carbon to total nitrogen) in the Rajang and small rivers, where slower degradation of OCterr and a higher total nitrogen percentage (TN%) were observed, compared to other river systems. Most of the OCterr discharged from the Rajang and small river systems was material derived from woody angiosperm plants with limited diagenetic alteration before deposition, and so could potentially provide significant carbon to the atmosphere after degradation.

1 Introduction

Tropical peatlands are one of the biggest terrestrial organic carbon pools, accounting for about 89,000 Tg (Moore et al., 2013; Rieley et al., 1996, 2008). It is reported that about 77% of the carbon stored in all tropical peatlands derived from Southeast Asia, which equals to 11%–14% of the total carbon pool stored in all peat. However, increasing anthropogenic disturb in the form of land use change, drainage and biomass burning are converting this peat into a globally significant source of atmospheric carbon dioxide (Dommain et al., 2014; Miettinen et al., 2016;

Koh et al., 2009; Page et al., 2011). The rivers draining these peatlands are typically rich in lignin phenols and humic substances, and are often referred to as "blackwater" rivers (Baum et al., 2007; Cook et al., 2017; Moore et al., 2011). However, knowledge of the fate of terrigenous organic matter in such peat-draining rivers and estuaries remains limited (Gandois et al., 2014; Hall et al., 2015; Lourençato et al., 2019).

The transport, degradation, and sequestration of OCterr in river systems are important because of their roles in constraining carbon cycle budgets (Aufdenkampe et al., 2011; Battin et al., 2009; Feng et al., 2016; Spencer et al., 2010; Wu et al., 2018). In terms of transport within fluvial systems, OCterr is subject to various natural processes, such as photo bleaching, microbial degradation, and selective preservation, as well as anthropogenic activities e.g. dam construction, irrigation systems, and land use change (Bao et al., 2015; Hernes et al., 2017; Spencer et al., 2010; Wu et al., 2015, 2018). Thus, it can be difficult to distinguish OCterr behavior from dynamics within a fluvial system. Multiple geochemical approaches have been applied to elucidate the composition and fate of OCterr in riverine and coastal sediments, including C/N ratios, $\delta^{13}C$ composition, and the distribution and composition of specific biomarker compounds such as lignin phenols and plant wax n-alkanes (Bao et al., 2015; Drenzek et al., 2007; Goni et al., 2005; Hernes and Benner, 2002; Jex et al., 2014; Ward et al., 2013). Lignin, which constitutes up to 30% of vascular plant biomass, is a unique biomarker of OCterr (Goñi and Hedges, 1995; Hedges and Mann, 1979). The monomeric composition of lignin phenols (S, V, C series) provides useful information on the biological source (woody *versus* nonwoody and angiosperm *versus* gymnosperm) and oxidation stage of lignin in natural environments (Benner et al., 1984; Hedges et al., 1985; Dittmar and Lara, 2001; Tareq et al., 2004; Thevenot et al., 2010). Most studies designed to understand the sources, compositions and transport of exported OCterr to determine its impact on the carbon cycle have been carried out in large rivers in the temperate and polar zones (Bao et al., 2015; Bianchi et al., 2002, 2011; Drenzek et al., 2007; Goñi et al., 1998, 2005; Feng et al., 2016; Wu et al., 2015, 2018). In contrast, lignin signatures from tropical environments have received less attention, especially in small river systems (Alin et al., 2008; Alkhatib et al., 2007; Dittmar and Lara, 2001; Goñi et al.,

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2006; Hedges et al. 1986; Spencer et al., 2010; Sun et al., 2017; Pradhan et al., 2014).

The export of OCterr in tropical river systems is typically constrained by natural rainfall, typhoons, floods, and tectonic activity (Alin et al., 2008; Aufdenkampe et al., 2007; Bao et al., 2015). Elevated soil turnover rates, coupled with short water residence times in small tropical river catchments, lead to the accelerated transformation of terrestrial organic matter (OM), especially during high-discharge events (Bao et al., 2015; Goldsmith et al., 2008; Kao and Liu, 1996). Anthropogenic processes such as deforestation have been a major cause of altered hydrology and OM compositions in tropical river systems (Houghton et al., 2000; Jennerjahn et al., 2004, 2008; Pradhan et al., 2014). The current paucity of information on OCterr characteristics and its export by rivers from tropical peat-draining rivers remains a major gap in our understanding of OCterr biogeochemical cycling in rivers from tropical Southeast Asia. Previous studies have reported that peatland-draining rivers in Sumatra and Borneo contained the highest values of dissolved organic carbon (DOC) in rivers globally (3000–5500 μmol L^{-1}), and most of the terrestrial DOC delivered into the sea (Wit et al., 2015). To understand the biogeochemical processing of OCterr in Southeast Asia, more work is needed on the dynamics of OCterr in the fluvial systems of this region.

Here we present what is, to our knowledge, the first analysis of OCterr concentration and behavior in four rivers and estuarine regions in the western part of Sarawak, Malaysian Borneo. We examined the OCterr characteristics using the lignin phenols composition from various samples (e.g., plants, soils, and sediments) from a major river, the Rajang, and three adjacent small rivers (the Maludam, Simunjan, and Sebuyau) to resolve the sources and transformation processes in the wet *versus* dry season. We further compared data among the four rivers to determine the ultimate fate of lignin and the potential controls on its distribution. Our results also indicate that lignin composition links to sources and modifications along the river–peat/soil–estuary continuum and reveal its response to peat degradation.

2 Materials and methods

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2.1 Study region and sample collection

Samples were collected during three field expeditions to Sarawak in August 2016 (only the Rajang), early March 2017 (the Rajang and the three small rivers), and September 2017 (only the small rivers; Fig. 1). During the 2017 expeditions, typical plants (Table S2) and soil samples were also collected for the comparison study.

The Rajang River drainage basin covers an area of about 50,000 km^2. Elevations exceed 2000 m and hill slopes are steep, generally in excess of 258 m in the interior highlands and 208 m in lower areas (Martin et al., 2018). The three small rivers (the Maludam, Simunjan, and Sebuyau) are blackwater rivers that draining extensive peatlands (Fig. 1). For the Rajang, it is separated into two parts by Sibu Town, upper reaches mainly drains mineral soils, while down reaches develops multiple distributary channels (e.g., the lower Rajang, Serendeng, Igan; Fig. 1). These channels are also surrounded by broad peatlands (Staub et al., 2000). However, Deforestation and changing in land use are accelerating the peatland degradation (Fig. 1). More than 50% peatland (11% of the catchment size) in Rajang watershed has been occupied by industry plantation (e.g. oil palm) (Miettinen et al., 2016). Fishery, logging and timber processing are the traditional supports for local citizens (Miettinen et al., 2016).

The climate of the study area is classified as tropical ever-wet, with average rainfall in excess of 3700 mm/year. The average monthly water discharge is about 3600 m^3/s, with peak discharge during the northeastern monsoon season (December to March; Staub et al., 2000). The three sampling periods resembled the end of this northeastern monsoon (i.e., March, the end of the wettest season of the year) and were shortly before the beginning of the northeastern monsoon (i.e., August and September, the end of the drier season).

The surface sediments were collected using grab samplers from a small boat at each station and then 0 - 5 cm subsamples were removed and frozen (–20°C) until they were dried for subsequent analysis in the laboratory. The dominant botanical samples and soils within the basin were collected at the same time and stored in a freezer. The hydrological parameters of the surface river water (e.g., salinity, pH, and temperature) at each station were determined using an

Aquaread® multiple parameters probe (AP-2000).

2.2 Chemical analysis

Prior to chemical analysis, all botanical samples as well as the soil and sediment samples, were dried at 55 °C and disaggregated in an agate mortar to form a homogeneous sample.

Grain size characteristics were measured directly from aliquots of the surface sediment samples using a Coulter LS 100Q (Coulter Company, USA), after treatment with 5% H_2O_2 and 0.2M HCl to dissolve organic matter and biogenic carbonate. The sediment grain sizes are expressed as the proportions of clay (<4 μm), silt (4–63 μm), and sand (>63 μm), with a measurement error of ≤5% for the entire dataset. The remaining sediments were ground to 80 mesh (187.5 μm) for elemental, isotopic, and lignin analyses.

The concentrations of organic carbon and total nitrogen (TN) were analyzed using a CHNOS Elemental Analyzer (Vario EL III) with a relative precision of ±5%. The weight percentages of organic carbon were analyzed after removing the carbonate fraction by vapor phase acidification. The weight percentages of TN were also analyzed following the same procedure but without acidification. The stable carbon isotopic composition of the decarbonated sediments was determined by a Flash EA1112 Elemental Analyzer connected to an Isotope Ratio Mass Spectrometer (MAT Delta Plus/XP, Finnigan). $^{13}C/^{12}C$ ratios are expressed relative to the PDB standard using conventional δ notation. The analytical precision, determined by replicate analysis of the same sample, was ±0.2‰.

Lignin phenols were extracted using the cupric oxide digestion technique (CuO; Hedges and Ertel, 1982; Yu et al., 2011). Briefly, the powdered samples were weighed and placed in O_2 free Teflon-lined vessels, and digested in a microwave (CEM MARS5) at 150°C for 90 min (Goñi and Montgomery, 2000). Samples were then acidified to pH < 2 and phenolic monomers were extracted into 99:1 (volume ratio) ethyl acetate/petroleum ether, dried, and stored at –20°C until further analysis. Samples were analyzed as trimethylsilyl derivatives of N,O-bis(trimethylsilyl)trifluoroacetamide (BSTFA) and trimethylchlorosilane (TMCS; 99:1) by Agilent

6890N gas chromatography (DB–1 column, FID). The lignin phenols concentration were quantified using calibration curves based on commercial standards (Sigma Aldrich). Eleven phenol monomers were extracted and categorized into five groups: syringyl (S), vanillyl (V), cinnamyl (C), p-hydroxyl (P), and 3,5-dihydroxy benzoic acid (DHBA).

These phenol monomers are detected by gas chromatography in their trimethylsilated forms. The S, V, and P groups further contain three monomers with aldehyde (–CHO), ketone (C=O), and carboxylic acid (–CO_2H) functional groups. Any complications are usually related to conventional diagenesis indicators such as the S/V and cinnamyl/vanillyl phenols (C/V) ratios (see Table 1), and these were also solved by normalizing V, S, and C to total lignin phenols as defined below:

Lignin phenols vegetation index (LPVI) = $[\{S(S + 1)/(V + 1) + 1\} \times \{C(C + 1)/(V + 1) + 1\}]$

2.3 Statistical analysis

All statistical analyses were carried out using SPSS 10.0 (IBM SPSS Inc., USA) and results were plotted using Origin software (Origin Lab Inc., USA). Multivariate statistical approaches such as principle component analysis (PCA) and cluster analysis (CA) are among the most widely used statistical methods in determining the significance of specific parameters within a dataset (Pradhan et al., 2009). Interrelationships among the sampling points in different rivers were characterized by cluster analysis using Ward's method (linkage between groups) and similarity measurements in terms of Euclidian distance, illustrated in dendograms.

3 Results

3.1 Hydrological parameters, grain size, and bulk elemental and stable isotopic composition of vegetation, soil, and sediment

The hydrological parameters for the study area are summarized in Table S1. The salinity of the lower Rajang system varied significantly (from 12‰ to 32‰) because of saline water intrusion in the estuarine region, but there were limited pH variations (6.5–7.9). Dissolved oxygen (DO) levels show significant spatial variations, with the lowest values (2–3 mg L^{-1}) being recorded in the Igan channel, where dense peats were observed, and the higher values (4–6 mg L^{-1}) recorded in the other two channels. The salinity of the Simunjan indicates that freshwater dominated, whereas the two other small rivers showed saline water influences. The variation in pH values among the three small rivers decreased from the Sebuyau (~6.4), to the Simunjan (~5.1), and the Maludam (~3.7). The DO concentrations in the three small rivers varied in a low range (average: 2–3 mg L^{-1}), with the lowest values in the three systems being around 1.4 mg L^{-1}.

The compositions of bulk sediments from the Rajang and the three small rivers are presented in Tables 1 and S1. The mean grain sizes from the upper Rajang (212±47 μm) are much coarser than those from the lower Rajang (40±38 μm) and the small rivers (22±16 μm). The finest samples (9±2 μm) were collected from the Maludam in March 2017. Generally, the samples collected during the dry season were coarser than those from the flood season in the Maludam and Simunjan, but this was not the case for the Sebuyau. The average organic carbon content shows a significant negative relationship with mean grain size among these samples (r^2 = 0.67, $p < 0.01$). Mean values of Total organic carbon (TOC) concentrations were higher in the peat-draining rivers (2.2±0.58%, 2.6±1.23%, and 2.6±0.8% for the Maludam, Sebuyau, and Simunjan, respectively) compared with the lower Rajang (1.1±0.5%), and the lowest values were observed in the upper Rajang (0.12±0.02%). The highest values of OC for the vegetation were measured in plants samples (30%–49%). The mean TOC value in the soil samples was 3.6±0.6%.

TN content ranged from 0.02% to 0.17% in the samples collected from the Rajang, from 0.09% to 0.37% in the small rivers, from 0.73% to 1.65% in the vegetation, and averaged 0.19±0.02% for the soil samples (Tables 1, S2, and S3). Although nitrogen was enriched in the samples from the peat-draining rivers, they still had higher mean C/N values (15.8±3.7) compared with the lower Rajang (11.5±1.6) while vegetation samples, which exhibited low N content and high C/N (C/N = 56±34).

The most abundant vegetation collected from the Maludam showed relatively depleted carbon isotope ratios ($\delta^{13}C$ = −31‰) that are typical of C3 vegetation (Table S2). The detritus samples

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were also relatively depleted in ^{13}C ($\delta^{13}C$ = −29.2‰; Table 1). The isotope ratio of the peat-draining river samples was slightly enriched in ^{13}C (average $\delta^{13}C$ = −28.0±0.4‰) compared with the Rajang (average $\delta^{13}C$ = −28.7±0.6‰). The $\delta^{13}C$ values of the soil samples are similar to those of the small rivers ($\delta^{13}C$ = −28.4‰).

3.2 Lignin phenols content

The lignin phenols obtained after CuO oxidation are expressed as Λ8 (mg (100 mg OC)$^{-1}$) , except for the lignin yield (Σ8), which is the sum of C + S + V and is expressed as mg 10 mg dw^{-1}, and are presented in Fig. 2 as well as Tables 2 and S1-3. The highest yields were measured in the vegetation samples (300–900 mg 10 mg dw^{-1}). The lignin yield from the soil samples and the three small rivers (average of ~30 mg 10 mg dw^{-1}) is also higher than that from the Rajang samples (average of <10 mg 10 mg dw^{-1}), with the lowest value observed in the upper Rajang (0.16 mg 10 mg dw^{-1}; Table 2). There are good correlations between Σ8 and OC% in each river, with the slope decreasing in the order of Maludam > Simunjan > Sebuyau > Rajang (Fig. 2a). The variation in Λ8 from various pools shows a similar distribution as the Σ8 values. The average concentrations for the vegetation, soil, and the four river systems mg (100 mg OC)$^{-1}$ approximately 18, 8.3, 5.4 mg (100 mg OC)$^{-1}$ (for the Rajang), 6.2 mg (100 mg OC)$^{-1}$ (for the Maludam), 7.9 (for the Sebuyau), and 7.4 mg (100 mg OC)$^{-1}$ (for the Simunjan), respectively.

The C/V and S/V ratios differ with vegetation type (Fig. 2b). Angiosperm leaves show high S/V (>1) and C/V ratios (~0.8). Angiosperm wood and root samples show lower C/V ratios (<0.2). The detritus samples show intermediate S/V ratios (0.6–1.0) and lower C/V ratios (~0.1). Soil samples have relatively high S/V (~1.1) and low C/V (~0.07) values. The four rivers show limited variations in S/V (0.4–0.8) and C/V (0.02–0.08) ratios. The LPVI values of the fresh plant material range from 113 to 2854 for leaves and 192 to 290 for wood. The values for detritus range between 36 and 228, and for soil and sediment range between 30 and 60 (Table 2).

The ratios of vanillic acid to vanillin ($(Ad/Al)_V$) and syringic acid to syringaldehyde ($(Ad/Al)_S$) increase slightly from the vegetation to river samples (Table 2). The ratios obtained for the

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vegetation and soil samples show similar values ($(Ad/Al)_S$ = ~0.30; $(Ad/Al)_V$ = ~0.35). The ratios from the small river samples range from 0.41 to 0.58 for $(Ad/Al)_V$ and 0.30 to 0.36 for $(Ad/Al)_S$. The values from the lower Rajang are similar to those from the small rivers, but this is not the case for the upper Rajang, where higher $(Ad/Al)_S$ and (Ad/Al)v values were recorded. The two ratios are linearly correlated in all sediment samples ($r^2 = 0.68$, $p < 0.05$), except for the samples collected from the Simunjan.

The P/(V + S) ratio is low in the vegetation samples, except for the leaf samples (P/(V + S) = 0.22), which reflects the very low V content (Table 2). The ratio is 0.28±0.03 for the soil samples, 0.18±0.4 for the small rivers, 0.17±0.02 for the lower Rajang, and 0.51±0.04 for the upper Rajang. The 3,5-dihydroxybenzoic (DBHA) levels determined from the soil and sediments are plotted in Fig. 2d. DHBA is very low in the upper Rajang (~0.07), but higher in the Maludam in the dry season (average value of 0.44). Values in the Simunjan in both seasons are similar to those from the soil samples (~0.38). Higher values of DHBA were measured in the lower Rajang and the Sebuyau in the dry season than in the wet season.

3.3 Statistical analysis

The results of cluster and PCA analyses of both bulk geochemical and lignin phenols proxies for all sediments are shown in Fig. 3. Four distinct groups were identified based on the cluster analysis. The Maludam and the tributary of the lower Rajang (Igan) are grouped together, and the Simunjan and Sebuyau are grouped together. The lower Rajang and upper Rajang are separated from each other (Fig. 3a). Similar groupings are evident in the results of the PCA analysis, which was based on the distribution of factors 1 and 2 that represent total loadings of 45% and 32%, respectively (Fig. 3b). The PCA results show a close relationship between Σ8 and OC% in factor 2, whereas the (Ad/Al)v ratio is related to grain size in factor 1.

4 Discussion

4.1 Comparison with systems worldwide: lignin parameters derived from sediment and peat

Table 3 summarizes the distribution of bulk and lignin parameters from typical systems worldwide. Although the TOC values of our studied systems are compared lower with peat samples but the concentrations of lignin phenols are comparable, which are typically enriched in lignin phenols compared with other river systems (Table 3; Bianchi et al., 2002; Gandois et al., 2014; Li et al., 2015; Sun et al., 2017; Pradhan et al., 2014; Winterfeld et al., 2015). The TN values of our peat samples are between two and four times higher than those seen in other systems worldwide, as was also observed in small rivers along India's west coast (Pradhan et al., 2014). The higher values of Λ8 found in our studied systems were potentially caused by peat-draining and intense human activity near the watersheds, as reported previously (Milliman and Farnsworth, 2011; Moore et al., 2013; Rieley et al., 2008). Much of the peatland neighboring the Simunjan and Sebuyau catchments has been changed to palm oil plantations (Martin et al., 2018). The terrigenous OM has been affected by diagenesis, as $(Ad/Al)_V$ varies markedly among the different systems (Table 3). The fluvial matter across watersheds within the Arctic region is strongly degraded (Winterfeld et al., 2015). The $(Ad/Al)_V$ values of the sediments sampled here are comparable to fresh and only low to medium oxidized. This study provides new insights into the amount of terrestrial OC preserved in the tropical delta region of southeastern Borneo, as well as into the biogeochemical transformation of OM from terrestrial source to marine sink across this region.

4.2 Origin of sediment organic matter in tropical peat-draining rivers

The depleted average $\delta^{13}C$ values (–28.5‰) of our vegetation samples indicate an insignificant contribution from C4 plants in the study area (Gandois et al., 2014; Sun et al., 2017). The high C/N ratio (64.8) indicates a predominance of terrestrial high plant species (e.g., *Nepenthes sp.* and *Avicennia marina Vierh.*). The $\delta^{13}C$ and C/N values (–27.2‰ and 12, respectively) obtained from the soil and sediments collected near the rivers suggest that terrestrial organic matter is the dominant contributor (Table 1). The enriched $\delta^{13}C$ values obtained from the peat-draining rivers when compared with the Rajang could be the result of higher contribution of peatland

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vegetation (Benner et al., 1987; Gandois et al., 2014). The cluster and PCA analyses suggest that there were no significant seasonal differences in these rivers. This might be because of the similar precipitation levels during our sampling seasons and sediments samples related to long-term records compared with particulate phase (Martin et al., 2018). The close relationship between the OC% and Σ8 in the PCA suggests factor 2 relates to the source of the organic matter (Fig. 3), as also indicated by the strong correlation between OC% and Σ8 (Fig. 2). Correlation of OC% and Σ8 of the Maludam showed the highest slope, possibly related to its pristine condition that promotes better conservation of vegetation in its peat. Furthermore, the differences between the upper and lower Rajang are highlighted by the PCA results and bulk parameters; i.e., the upper Rajang drains a mineral soil whereas peat is dominant in the delta region. This also explains why the Rajang data do not plot with the other small river systems; the linear relationship between $\delta^{13}C$ and Σ8 for the Rajang (r^2 = 0.92) forms a distinct group separate from the small rivers (r^2 = 0.59; Fig. 3).

The S/V and C/V ratios are often used as indicators of the vegetation origin of the lignin fraction; e.g., the woody and non-woody parts of gymnosperm and angiosperms (Hedges and Mann, 1979). The S/V values (<0.8) of the peat-draining rivers are slightly lower than the values of other peats (<1.5), but the C/V ratios are comparable (Tareq et al., 2004). The differences in these parameters between the sediments and the vegetation and soils, as illustrated in Fig. 2, suggests that they are composed mostly of angiosperm wood. This finding is further confirmed by the LPVI values, which are commonly less than 60 in these sediment samples. Previous studies have concluded that tropical peats are derived mainly from wood (Anderson, 1983; Gandois et al., 2014). For the Rajang, the LPVI values show a positive linear correlation with Λ8 concentrations (r^2 = 0.56); however, for the small rivers (based on mean values, except the samples collected in March 2017 from the Maludam) this relationship shows a negative correlation (r^2 = 0.91). This suggests that the small rivers receive more lignin derived from woody material, whereas the Rajang has a mixture of sources. The unusual behavior of the Maludam's samples might be related to the dominance of finer-grained sediments when compared with the other rivers,

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because woody material tends to be concentrated in the coarser fraction (Table 1). P phenols in the Rajang are derived from lignin, as supported by the significant correlation of the content of P phenols and lignin content (r^2 = 0.93). However, there is no correlation between P phenols and lignin content for the small rivers. All P/V values from the samples (0.13–0.72) are higher than the average P/V ratio of wood (0.05) and are similar to the range observed for leaves (0.16–6.9; Hedges et al., 1986). Considering this, non-woody angiosperms are the most likely source of additional lignin.

4.3 Transformation of lignin signatures in tropical peat-draining rivers

(Ad/Al)v ratios are often used to estimate the degradation status of terrestrial OM. The (Ad/Al)v ratios for soils reported in previous studies fall within the range 0.16–4.36, 0.1–0.2 for fresh angiosperm wood and 0.2–0.6 for non-woody tissues (Hedges et al., 1988; Opsahl and Benner, 1995; Thevenot et al., 2010). In our study, the variability of the (Ad/Al)v ratios obtained from the vegetation, soil, and sediments was limited, with values between 0.3 and 0.58 except from the samples from the upper Rajang (~1.0), which suggests the mild degradation of OCterr in most samples. The degradation status of lignin was negatively correlated with the $\Lambda 8$ values (r^2 = 0.73) in the Rajang, and with a higher degradation signal observed in the upper Rajang, which drains mineral soils with lower lignin levels (Fig. 4a). However, the $\Lambda 8$ values are positively correlated with the (Ad/Al)v ratios (r^2 = 0.50) for the small rivers, except for the samples collected from the Maludam in March 2017 (Fig. 4b). Such a distribution could be related to the grain size effect, as illustrated in Fig. 4c and 4d. Of the sediments sampled here, the upper Rajang samples contain the largest coarse fraction and the finest sediments were collected from the Maludam in March 2017. The (Ad/Al)v ratios increase with decreasing mean size of the sediments in the small rivers. Selective sorption of acid to aldehyde might affect the variation of the (Ad/Al)v ratio in the small river systems (Hernes et al., 2007). However, the relatively fresh condition of the OM in the Maludam samples (in March 2017) might be related to the fluvial supply of fresh vegetation during the flood season.

The syringyl and cinnamyl series are preferentially degraded when compared with the vallinyl series, resulting in a decrease in the S/V and C/V ratios during lignin degradation (Goni et al., 1995; Opsahl and Benner, 1995). Our samples show a negative linear relationship between the S/V and (Ad/Al)v ratios in the Rajang samples (r^2 = 0.85; Fig. 5a). However, the variation of the S/V and (Ad/Al)v ratios in the small rivers was limited, and a non-linear correlation is evident (Fig. 5b). Both correlations indicate that the decrease in the S/V ratios is linked to degradation, and this suggests that we should be cautious when using S/V ratios for source evaluation in this study. Previous studies demonstrated that lignin mineralization in humid tropical forest soils is dominated by methoxyl-C mineralization under aerobic and fluctuating redox conditions (Hall et al., 2015). Demethylation reduces the yield of methoxylated phenols (V and S phenols) but does not affect P phenols. Therefore, the P/(S+V) ratio can be used as an indicator of lignin transformation (Ditmar and Kattner, 2003). However, in this study the ratio of P/(S+V) in most samples did not vary greatly (~0.2), and no clear trend was observed for the small rivers, although there was a linear correlation between the P/(S+V) and (Ad/Al)v ratios (r^2 = 0.89).

4.4 Impact of environmental parameters on lignin dynamics

It is well explored that bulk organic matter composition and degradation are influenced by many environmental factors such as climate, grain size, mineral composition, soil characteristics, land use changes, logging, and biomass burning (Hernes et al., 2007; Gandois et al., 2014; Sun et al., 2017; Thevenot et al., 2010). Most Southeast Asian peat-draining rivers are impacted by human activities such as deforestation, urbanization and damming (Milliman and Farnsworth, 2011). The PCA analysis revealed that the behavior of lignin in the Rajang is substantially different from that in the three peat-draining rivers, and especially in the upper Rajang, which drains through a mineral soil with low $\Lambda 8$ values and strong degradation (Figs 3 and 4).

In this study, the OC content increases with decreasing grain size, implying that fine particles, with larger specific surface areas and rich in clay, contain more OM than coarser particles, as reported previously (Sun et al., 2017). However, other studies found that coarse particulate OM in the Amazon basin has a higher content of lignin phenols than fine POM, and also that coarse POM is composed of fresher lignin derived from wood debris (Hedges et al., 1986). Nevertheless, increasing $(Ad/Al)_V$ values were observed in the Rajang with increasing grain size, which suggests that lignin associated with larger mineral particles is more strongly degraded. This observation indicates the preferential preservation of lignin in finer-grained sediments, resulting from their ability to provide better protection against further oxidative degradation (Killops and Killops, 2005). For the small river systems, the $(Ad/Al)_V$ ratios decrease with increasing grain size, corresponding to the increasing $\Lambda 8$ values (Fig. 4a and b). Our observations of $(Ad/Al)_V$ values are similar to the trends described by Keil et al. (1998) and Tesi et al. (2016), who found that lower $(Ad/Al)_V$ values were present in the coarser fractions due to the less efficient processing of plant remains prior to deposition. The sediments collected from the three small peat-draining rivers (except samples from the Maludam in March, 2017) could have contained limited amounts of plant debris, in which case fresh plant tissue would have been incorporated into the coarser sediment fractions, leading to the low $(Ad/Al)_V$ values. However, the variation in $\Lambda 8$ values does not support this speculation, and therefore we conclude that the selective sorption of acid to aldehyde could explain the elevated $(Ad/Al)_V$ ratios recorded in the fine fraction. The different grain-size effects on OCterr composition, as seen when comparing the Rajang with the small rivers, suggests that there are other processes working on OCterr in these two systems, which cause post-depositional changes in the OCterr characteristics.

Tropical soils are reported naturally poor in N and P, but some studies have shown that with intensive management (land use/deforestation) they tend to become rich in recalcitrant compounds, and recalcitrant substrates will continue to decompose, when the conditions microbial preferred (Thevenot et al., 2010). In our study, we found a higher TN% in the small rivers compared with the Rajang. A good correlation between $\Sigma 8$ and TN% was observed in all systems, which might suggest a contribution from plant litter affecting both parameters (Fig. 6a). However, the (Ad/Al)v ratios appear to be related to the C/N ratios, but with different slopes obtained for the Rajang and the small rivers (Fig. 6b). This could be relative to the expected impact of nitrogen on lignin degradation (Dignac et al., 2002; Thevenot et al., 2010). A high N content will inhibit fungal lignin biodegradation (Fog, 1988; Osono and Takeda, 2001), and this explains why slower degradation was observed in the small river systems in which higher TN% were recorded. The exceptional data were collected during September 2017, which was a time of saline water intrusion.

Large-scale land reclamation, including deforestation and urbanization, has taken place in Southeast Asia over the past few decades (Miettinen et al., 2016). Logging activities have had a significant influence on peat decomposition processes and the quality of organic matter inputs (Hoscilo et al., 2011; Hooijer et al., 2012; Gandois et al., 2014). Gandois et al. (2013) reported an increase in the N content at a deforested site and concluded that it was caused by an increase in the microbial deposition of peat. The lignin yield (Σ8) is closely correlated with the OC% in the different rivers (Fig. 2). The highest yield was observed at the Maludam, which confirmed the significant contribution of plant litter and better preservation due to the low pH and DO levels, especially woody carbon. However, the relatively higher yield in the Rajang compared with the other two disturbed peat-draining rivers (i.e., the Simunjan and Sebuyau) suggests an additional source of lignin, which might implicate the addition of logging residue to the Rajang systems, as proposed by Gandois et al. (2014).

5 Conclusions

We used sediment grain size data, TOC contents, the stable carbon isotopic composition of organic matter, and lignin phenols concentrations to investigate the characteristics of OCterr in a tropical peat-draining river system, as well as its fate and environmental controls. The depleted $\delta^{13}C$ levels of all of the sediment samples demonstrates that contributions from C3 plants dominated the OCterr in the study region. The lignin composition of the organic matter indicates that the most important plant sources of organic matter were woody angiosperm C3 plants, especially in the three small rivers. Our cluster and PCA analyses showed no distinct seasonal

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variations in the bulk and lignin compositional signatures in the study area, although the upper Rajang receives contributions from mineral soils with unique lignin parameters and a coarser grain size. Both the bulk organic matter parameters and the lignin compositions were indicated to be correlated to the grain size of the riverbed sediments. The (Ad/Al)v ratios increased with decreasing mean size of the sediments from the small rivers. Selective sorption of acid to aldehyde might affect the variation of the (Ad/Al)v ratio in the small river systems. Our samples show a negative linear relationship between the S/V and (Ad/Al)v ratios in the Rajang samples, which implies that the decrease in S/V ratios is linked to degradation. The (Ad/Al)v ratios appear to be related to the C/N ratio in the Rajang and the small rivers. A high N content will inhibit fungal lignin biodegradation, which might explain the slower degradation observed in the small river systems where a higher TN% was recorded. Most of the OCterr discharged from the Rajang and small river systems was composed of woody angiosperm plants and the terrestrial organic matter undergoes limited diagenetic alteration before deposition, and could potentially become a significant regional carbon source to the atmosphere after extensive degradation.

Author contributions. YW, JZ, MM, and AM conceptualized the research project and planned the field expeditions. JZ, MM, AM obtained research funding. JZ, KZ, JS, MM, MFM, EA and AM collected samples and KZ and YW analyzed the samples. YW, KZ and JZ processed and analyzed the data. All authors contributed to data interpretation and to the writing of the manuscript.

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Table 1 Average values of bulk geochemical parameters for plants, soils, and sediments collected from the study systems

Samples	Time	Mean Size (μm)	Clay%	Silt%	DO (mg/L)	pH	Salinity (‰)	OC (%)	TN (%)	Atomic C/N	$\delta^{13}C$ (‰)
Angiosperm leaves & grasses (n=10)	03/2017	—	—	—	—	—	—	48.53±2.86	1.65±0.64	40.44±18.95	-31.1±2.5
Angiosperm woods(n=5)	03/2017	—	—	—	—	—	—	46.71±4.71	0.52±0.19	117.00±45.32	-31.8±2.3
Roots(n=3)	03/2017	—	—	—	—	—	—	38.60±4.80	1.06±0.64	50.10±19.58	-28.3±0.4
Lower Rajang detritus(n=8)	08/2016	—	—	—	—	—	—	40.76±13.69	0.94±0.35	47.21±13.03	-29.9±2.1
Sebuyau detritus(n=5)	03/2017	—	—	—	—	—	—	30.63±15.00	0.73±0.20	53.39±31.68	-28.1±2.0
Simunjan detritus(n=4)	03/2017	—	—	—	—	—	—	33.46±8.46	1.09±0.35	43.44±29.73	-29.9±0.7
Soil(n=8)	09/2017	—	—	—	—	—	—	3.63±0.63	0.19±0.02	21.98±2.50	-28.4±0.2
Upper Rajang (n=4)	08/2016	212.9±47.0	9.7±2.5	10.4±3.0	4.53±4.42	6.74±0.05	0	0.12±0.02	0.02±0.00	8.44±2.10	-28.1±0.5
Lower Rajang (n=16)	08/2016	41.9±43.3	32.3±11.7	45.4±14.8	3.64±0.66	7.33±0.52	15.4±10.8	1.07±0.46	0.11±0.05	11.44±1.69	-28.6±0.6
Lower Rajang (n=5)	03/2017	30.9±9.8	29.3±3.1	54.9±2.8	5.82±0.78	6.66±0.26	0.1±0.2	1.26±0.37	0.12±0.02	11.68±1.90	-29.1±0.2
Maludam (n=5)	03/2017	9.3±2.3	39.6±2.7	59.3±2.0	3.24±2.24	4.93±1.71	7.2±10.0	2.22±0.69	0.20±0.05	12.83±1.80	-27.4±0.6
Maludam (n=2)	09/2017	12.1	39.2	58.3	4.96	6.69	11.5	2.02	0.19	12.43	-28.2
Sebuyau (n=6)	03/2017	24.6±18.5	31.6±6.5	58.8±8.3	3.07±1.92	5.40±5.48	5.5±6.5	2.37±0.69	0.16±0.03	17.37±4.56	-27.8±0.3
Sebuyau (n=5)	09/2017	15.7±4.0	30.4±3.6	66.1±3.1	4.30±1.36	7.45±0.22	2.3±4.5	2.79±1.75	0.20±0.10	15.42±1.96	-28.2±0.4
Simunjan (n=6)	03/2017	20.2±10.3	22.0±5.3	71.0±6.5	1.85±0.65	5.22±0.61	0	2.58±1.03	0.19±0.08	16.44±3.03	-28.2±0.5
Simunjan (n=6)	09/2017	23.5±8.10	20.9±4.8	71.0±3.1	4.00±1.15	5.04±0.57	0	2.59±0.53	0.18±0.05	17.86±4.56	-28.4±0.5

Table 2 Average values of lignin phenols parameters for plants, soils, and sediments from the study systems (V: vallinyl phenols; S: syringyl phenols; C: cinnamyl phenols, P: p-hydroxyl phenols; DHBA: 3,5-dihydroxy benzoic acid; see the main text for definitions of Σ8, Λ8, Ad/Al, and LPVI)

Samples	Time	Σ8 (mg/10 g dw)	Λ8 (mg/100 mg OC)	V	S	C	S/V	C/V	(Ad/Al)v	(Ad/Al)s	P/(V+S)	DHBA	DHBA/V	LPVI
Angiosperm leaves & grasses (n=10)	03/2017	317.94 ±160.00	6.64±3.38	2.08±1.29	3.31±2.09	1.11±0.54	1.73±0.52	0.72±0.39	0.38±0.14	0.28±0.09	0.22±0.11	0.16±0.04	0.13±0.07	1420±910
Angiosperm woods(n=5)	03/2017	817.58±270.00	17.54±5.66	7.65±2.75	9.31±2.90	0.58±0.43	1.27±0.24	0.07±0.04	0.33±0.07	0.24±0.13	0.04±0.00	0.10±0.06	0.01±0.01	87±34
Roots(n=3)	03/2017	312.98±44.51	8.24±1.96	2.63±0.82	5.15±1.21	0.46±0.10	2.01±0.41	0.18±0.05	0.34±0.04	0.37±0.07	0.30±0.45	0.11±0.13	0.05±0.07	18±6
Lower Rajang detritus(n=8)	08/2016	418.98±151.87	11.57±6.47	5.40±2.60	5.35±3.73	0.86±0.56	0.89±0.24	0.18±0.10	0.35±0.09	0.27±0.12	0.24±0.10	0.26±0.18	0.08±0.13	10±55
Sebuyau detritus(n=5)	03/2017	638.41±373.55	20.39±3.15	9.63±2.01	9.70±2.29	1.05±0.64	1.04±0.33	0.11±0.05	0.34±0.13	0.37±0.09	0.15±0.09	0.16±0.11	0.02±0.01	85±34
Simunjan detritus(n=4)	03/2017	534.62±277.93	15.51±5.88	7.79±2.42	6.72±4.37	1.00±0.95	0.82±0.39	0.15±0.17	0.32±0.06	0.25±0.09	0.08±0.07	0.14±0.02	0.02±0.00	80±54
Soil(n=8)	09/2017	29.67±5.13	8.25±0.96	3.89±0.45	4.10±0.53	0.27±0.05	1.05±0.06	0.07±0.02	0.38±0.04	0.30±0.06	0.28±0.03	0.37±0.05	0.10±0.02	69±10
Upper Rajang (n=4)	08/2016	0.16±0.08	1.32±0.55	0.89±0.29	0.37±0.22	0.06±0.05	0.38±0.16	0.06±0.05	1.04±0.23	0.39±0.15	0.51±0.04	0.07±0.05	0.07±0.03	18±11
Lower Rajang (n=16)	08/2016	7.55±3.96	6.57±2.09	3.42±1.05	3.01±1.00	0.14±0.12	0.87±0.09	0.04±0.03	0.43±0.13	0.26±0.10	0.16±0.07	0.29±0.13	0.09±0.03	48±11
Lower Rajang (n=5)	03/2017	10.33±2.12	8.54±1.67	4.42±0.82	3.83±0.80	0.29±0.10	0.86±0.03	0.07±0.02	0.41±0.07	0.30±0.11	0.17±0.02	0.23±0.11	0.05±0.02	52±7
Maludam (n=5)	03/2017	14.21±6.66	6.21±1.40	3.62±0.99	2.53±0.46	0.07±0.05	0.71±0.07	0.02±0.02	0.58±0.18	0.30±0.18	0.20±0.01	0.44±0.13	0.12±0.04	33±6
Maludam (n=2)	09/2017	12.55	6.24	3.21	2.76	0.27	0.8	0.09	0.43	0.30	0.16	0.18	0.06	62

Sebuyau (n=6)	03/2017	18.02±7.07	7.75±2.10	4.50±1.33	3.12±0.82	0.13±0.108	0.70±0.05	0.03±0.02	0.47±0.07	0.34±0.06	0.17±0.04	0.33±0.08	0.08±0.01	33±6
Sebuyau (n=5)	09/2017	22.06±11.44	8.18±0.98	4.85±0.68	3.16±0.43	0.17±0.11	0.66±0.08	0.04±0.03	0.55±0.08	0.32±0.12	0.16±0.02	0.18±0.09	0.04±0.02	31±9
Simunjan (n=6)	03/2017	18.45±5.96	7.30±1.04	4.03±0.51	2.96±0.60	0.31±0.17	0.73±0.11	0.08±0.05	0.48±0.10	0.41±0.04	0.20±0.05	0.36±0.05	0.09±0.01	49±24
Simunjan (n=6)	09/2017	20.09±3.20	7.86±0.91	4.54±0.80	3.09±0.31	0.23±0.20	0.69±0.09	0.06±0.06	0.47±0.05	0.36±0.08	0.17±0.03	0.37±0.09	0.08±0.02	41±22

Table 3 Comparison of bulk and lignin phenols parameters among river systems worldwide

Samples	Station	OC (%)	TN (%)	C/N	δ^{13}C (‰)	Σ8 (mg/10g dw)	⍰8 (mg/100 mg OC)	S/V	C/V	(Ad/Al)v	(Ad/Al)s	P/(S+V)	DHBA/V	References
Amazon River	estuary	0.13~1.44	——	——	-29.4~-27.5	0.10~11.05	0.75~9.27	0.84~1.51	0.12~0.47	0.26~0.61	0.15~0.56	——	——	1
Congo River	submerged delta	0.8~4.2 2.1	——	5.8~10.1 8.3	-23.5~-19.0	——	0.066~0.373 0.151±16%	0.47~1.38 0.87±7%	0.15~0.39 0.28±13%	0.47~1.74 0.72±17%	0.26~1.94 0.46±14%	——	——	2
Pichavaram River	estuary	——	——	14.17±1.33	-27.15±1.53	——	——	1.26±0.32	0.19±0.12	0.68±0.11	0.81±0.21	0.57±0.097	——	3
35 Indian rivers	North group	0.61±0.3	0.04±0.01	18.7±6.9	-22.9±0.9	0.11±0.12	1.6±1.0	0.9±0. 2	0.2±0. 1	0.7±0. 2	——	0.4±0. 2	0.3±0. 2	4
	South group	2.3±0.6	0.12±0.03	19.8±4.1	-26.3±0.8	1.7±0.5	6.7±2.8	1.5±0. 5	0.3±0. 1	0.5±0. 1	——	0.2±0. 2	0.1±0. 2	
Kapuas River	whole basin	0.55~14.20	0.05~0.55	11.0~34.8	-30.39~-27.29	——	0.13~3.70	0.34~1.18	0.28~1.40	0.71~2.01	0.72~2.12	——	——	5
Rajang River	estuary	1.12±0.50	0.12±0.05	11.57±1.72	-28.6±0.60	7.55±3.96	6.57±2.09	0.87±0.09	0.04±0.03	0.43±0.13	0.26±0.10	0.16±0.07	0.09±0.03	This research
Maludam River	estuary	2.22±0.69	0.20±0.05	12.83±1.80	-27.4±0.61	14.21±6.66	6.21±1.40	0.71±0.07	0.02±0.02	0.58±0.18	0.30±0.18	0.20±0.01	0.12±0.04	
Sebuyau River	whole basin	2.37±0.69	0.16±0.03	17.37±4.56	-27.8±0.27	18.02±7.07	7.75±2.10	0.70±0.05	0.03±0.02	0.47±0.07	0.34±0.06	0.17±0.04	0.08±0.01	
Simunjan River	whole basin	2.58±1.03	0.19±0.08	16.44±3.03	-28.2±0.48	18.45±5.96	7.30±1.04	0.73±0.1	0.08±0.05	0.48±0.10	0.41±0.04	0.20±0.05	0.09±0.01	

								1						
Yangtze River	whole basin	0.64±0.06	——	——	-25.0±0.1	3.60±0.18	5.66±0.33	1.16±0.05	0.37±0.01	——	——	——	——	6
Mississippi River	estuary	1.2±0.5	0.1±0.06	13.4±2.8	-23.7±0.80	——	1.64±0.53	0.93±0.30	0.03±0.01	0.27±0.14	0.20±0.07	——	——	7
Lena River	delta	2.06±0.33	——	15.88±3.33	——	0.41±0.19	1.96±0.81	0.43±0.02	0.42±0.36	1.28±0.30	1.04±0.24	0.30±0.03	——	8
Pristine peat	Brunei	52.4	1.95	31.35	-30.4±0.8	——	5.65	0.82	0.05			0.28±0.05	0.12	
Disturbed peat	Brunei	50.95	2.09	28.44	-29.5±0.6	——	10.29	0.97	0.05	0.42±0.10	0.40±0.01	0.22±0.10	0.07	9

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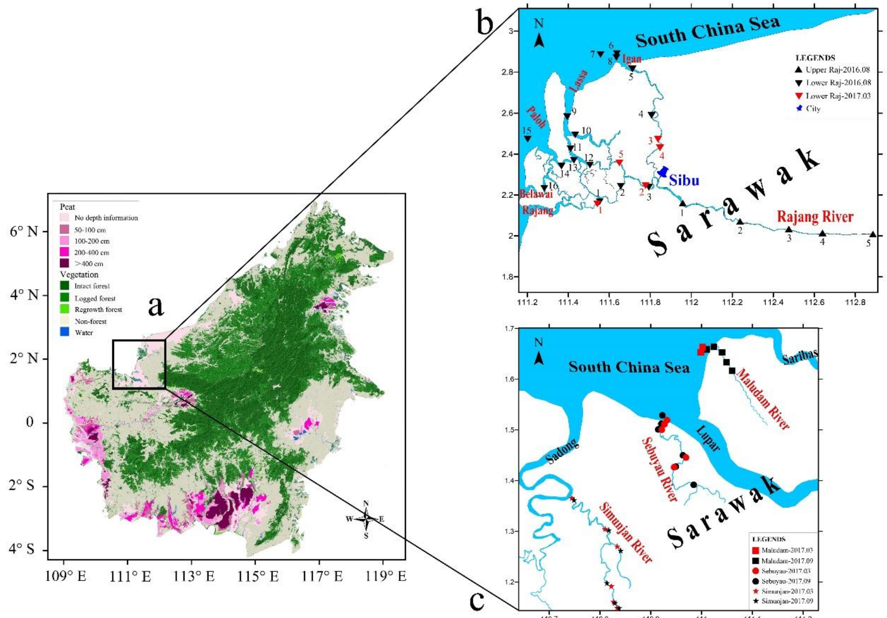

Figure 1 (a) Peat and vegetation distribution in the study region (modified from https://www.cifor.org/map/atlas/). (b) Sediment sampling sites along the Rajang and tributaries. The city of Sibu divides the river into upper and lower reaches. (c) Sediment sampling sites along the three small rivers. Locations of samples collected from the Maludam, Sebuyau, and Simunjan are indicated by squares, circles, and stars, respectively.

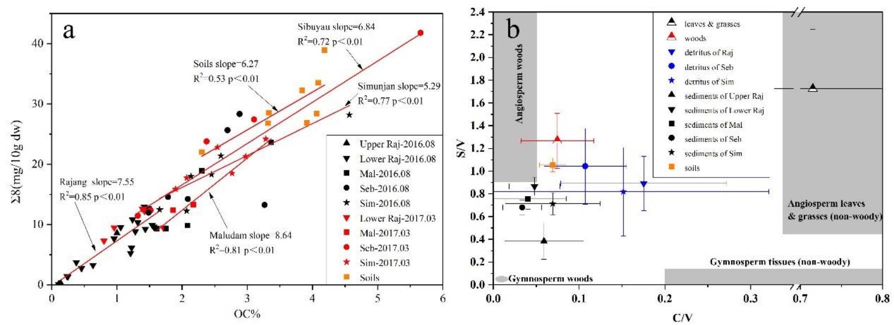

Figure 2 (a) Correlation of OC% with Σ8 among the various study systems. (b) Variations of S/V *versus* C/V of different samples from the study systems. Raj: Rajang; Seb: Sebuyau; Sim: Simunjan; Mal: Maludam.

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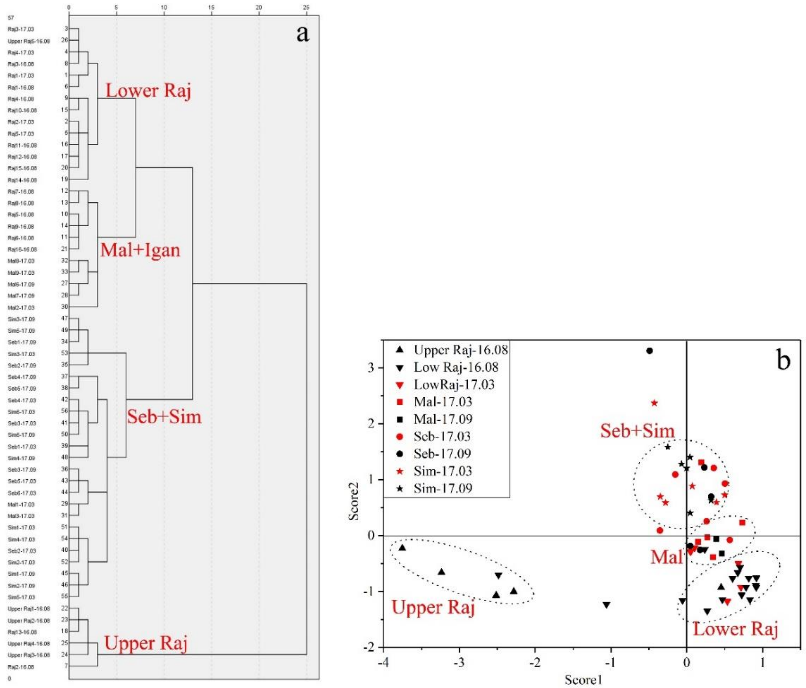

Figure 3 (a) Cluster analysis of the study systems based on bulk and lignin phenols parameters. (b) Plot of PCA results based on the distribution of scores 1 and 2. Raj: Rajang; Seb: Sebuyau; Sim: Simunjan; Mal: Maludam.

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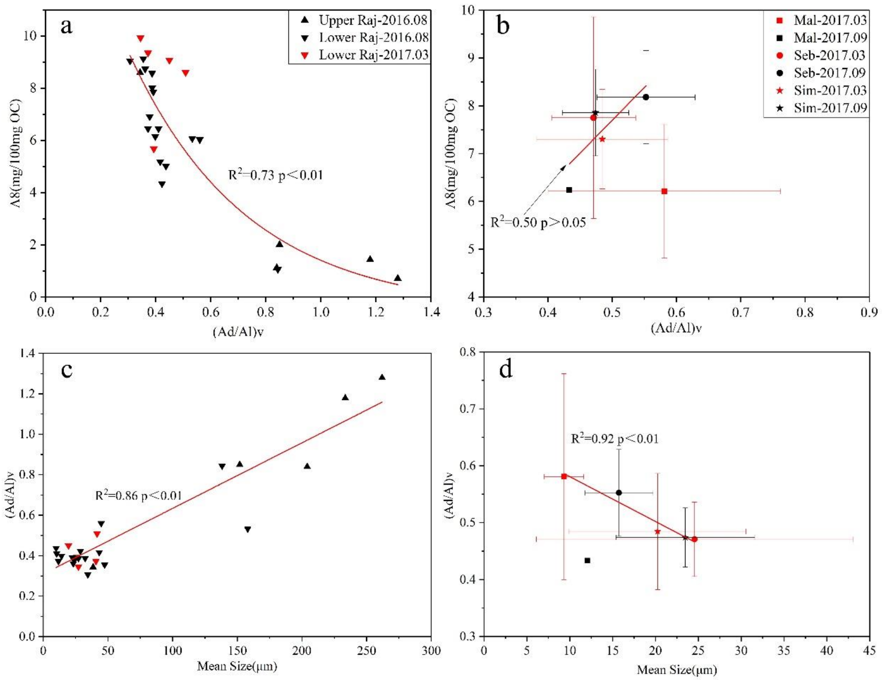

Figure 4 Variation in (Ad/Al)v with Λ8 values of sediments from (a) the Rajang and (b) the small river systems. Variation in (Ad/Al)v with mean sediment grain size for (c) the Rajang and (d) the small river systems.

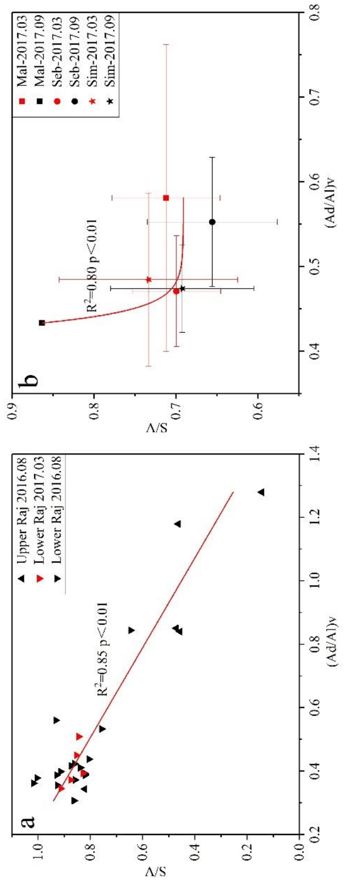

Figure 5 Relationship between (Ad/Al)v and S/V ratios based on average values of the various systems for (a) the Rajang and (b) the small river systems.

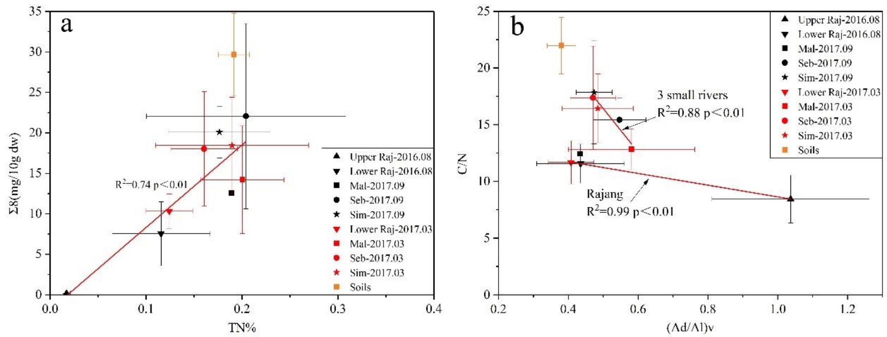

Figure 6 (a) Correlation of TN% with Σ8 based on average values of the study systems. (b) Correlation of $(Ad/Al)_v$ with C/N ratio based on average values of the study systems.