

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

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## 16 **Abstract.**

17 Tropical peatlands are one of the largest pools of terrestrial organic carbon (OC<sub>terr</sub>); however,  
18 our understanding of the dynamics of OC<sub>terr</sub> in peat-draining rivers remains limited, especially  
19 in Southeast Asia. This study used bulk parameters and lignin phenols concentrations to  
20 investigate the characteristics of OC<sub>terr</sub> in a tropical peat-draining river system (the main channel  
21 of the Rajang and three smaller rivers (the Maludam, Simunjan, and Sebuyau)) in the western  
22 part of Sarawak, Malaysian Borneo. The depleted  $\delta^{13}\text{C}$  levels and lignin composition of the  
23 organic matter indicates that the most important plant source of the organic matter in these  
24 rivers is woody angiosperm C<sub>3</sub> plants, especially in the three small rivers sampled. The diagenetic  
25 indicator ratio (i.e., the ratio of acid to aldehyde of vanillyl phenols ((Ad/Al)<sub>v</sub>) increased with  
26 decreasing mean grain size of sediment from the small rivers. The selective sorption of acid  
27 relative to aldehyde phenols might explain the variations in the (Ad/Al)<sub>v</sub> ratio. Elevated (Ad/Al)<sub>v</sub>  
28 values observed from the Maludam's sediments may be also attributed to source plant variations.  
29 The (Ad/Al)<sub>v</sub> ratio appears to be related to the C/N ratio (the ratio of total organic carbon to  
30 total nitrogen) in the Rajang and small rivers. In small rivers, a quick decline of C/N ratios  
31 responses to the slower modification of (Ad/Al)<sub>v</sub> ratio by the meant of better preservation of  
32 lignin phenols. The accumulation of lignin phenols with higher total nitrogen percentage (TN%)  
33 in the studied systems were observed. Most of the OC<sub>terr</sub> discharged from the Rajang and small  
34 river systems was material derived from woody angiosperm plants with limited diagenetic  
35 alteration before deposition, and so could potentially provide significant carbon to the  
36 atmosphere after degradation.

37

## 38 **1 Introduction**

39 Tropical peatlands are one of the biggest terrestrial organic carbon pools, accounting for about  
40 89,000 Tg (Moore et al., 2013; Rieley et al., 1996, 2008). It is reported that about 77% of the carbon  
41 stored in all tropical peatlands is derived from Southeast Asia, which equals to 11%–14% of the

42 total carbon pool stored in all peat. However, increasing anthropogenic disturbance in the form  
43 of land use change, drainage and biomass burning are converting this peat into a globally  
44 significant source of atmospheric carbon dioxide (Dommain et al., 2014; Miettinen et al., 2016;  
45 Koh et al., 2009; Page et al., 2011). The rivers draining these peatlands are typically rich in lignin  
46 phenols and humic substances, and are often referred to as “blackwater” rivers (Baum et al., 2007;  
47 Cook et al., 2017; Moore et al., 2011). However, knowledge of the fate of terrigenous organic  
48 matter in such peat-draining rivers and estuaries remains limited (Gandois et al., 2014; Hall et al.,  
49 2015; Lourençato et al., 2019).

50 The transport, degradation, and sequestration of OC<sub>terr</sub> in river systems are important because  
51 of their roles in constraining carbon cycle budgets (Aufdenkampe et al., 2011; Battin et al., 2009;  
52 Feng et al., 2016; Spencer et al., 2010; Wu et al., 2018). In terms of transport within fluvial systems,  
53 OC<sub>terr</sub> is subject to various natural processes, such as photo bleaching, microbial degradation,  
54 and selective preservation, as well as anthropogenic activities e.g. dam construction, irrigation  
55 systems, and land use change (Bao et al., 2015; Hernes et al., 2017; Spencer et al., 2010; Wu et al.,  
56 2015, 2018). Thus, it can be difficult to distinguish OC<sub>terr</sub> behavior from dynamics within a fluvial  
57 system. Multiple geochemical approaches have been applied to elucidate the composition and  
58 fate of OC<sub>terr</sub> in riverine and coastal sediments, including C/N ratios,  $\delta^{13}\text{C}$  composition, and the  
59 distribution and composition of specific biomarker compounds such as lignin phenols and plant  
60 wax n-alkanes (Bao et al., 2015; Drenzek et al., 2007; Goñi et al., 2005; Hernes and Benner, 2002;  
61 Jex et al., 2014; Ward et al., 2013). Lignin, which constitutes up to 30% of vascular plant biomass,  
62 is a unique biomarker of OC<sub>terr</sub> although highly degraded soil organic matter may be devoid of  
63 any apparent lignin but as another important contributor to OC<sub>terr</sub> (Burdige, 2005; Goñi and  
64 Hedges, 1995; Hedges and Mann, 1979). The monomeric composition of lignin phenols (S, V, C  
65 series) provides useful information on the biological source (woody *versus* nonwoody and  
66 angiosperm *versus* gymnosperm) and oxidation stage of lignin in natural environments (Benner  
67 et al., 1984; Hedges et al., 1985; Dittmar and Lara, 2001; Tareq et al., 2004; Thevenot et al., 2010).  
68 Most studies designed to understand the sources, compositions and transport of exported

69 OCterr to determine its impact on the carbon cycle have been carried out in large rivers in the  
70 temperate and polar zones (Bao et al., 2015; Bianchi et al., 2002, 2011; Drenzek et al., 2007; Goñi  
71 et al., 1998, 2005; Feng et al., 2016; Wu et al., 2015, 2018). In contrast, lignin signatures from  
72 tropical environments have received less attention, especially in small river systems (Alin et al.,  
73 2008; Alkhatib et al., 2007; Dittmar and Lara, 2001; Goñi et al., 2006; Hedges et al. 1986; Spencer  
74 et al., 2010; Sun et al., 2017; Pradhan et al., 2014).

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

90 Here we present what is, to our knowledge, the first analysis of OCterr concentration and  
91 behavior in four rivers and estuarine regions in the western part of Sarawak, Malaysian Borneo.  
92 We examined the OCterr characteristics using the lignin phenols composition from various  
93 samples (e.g., plants, soils, and sediments) from a major river, the Rajang, and three adjacent  
94 small rivers (the Maludam, Simunjan, and Sebuyau) to resolve the sources and transformation  
95 processes in the wet *versus* dry season. We further compared data among the four rivers to

96 determine the ultimate fate of lignin and the potential controls on its distribution. Our results  
97 also indicate that lignin composition links to sources and modifications along the river–peat/soil–  
98 estuary continuum and reveal its response to peat degradation.

99

## 100 **2 Materials and methods**

### 101 **2.1 Study region and sample collection**

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

106 The Rajang River drainage basin covers an area of about 50,000 km<sup>2</sup>. Elevations exceed 2000 m  
107 and hill slopes are steep, generally in excess of 258 m in the interior highlands and 208 m in  
108 lower areas (Martin et al., 2018). The three small rivers (the Maludam, Simunjan, and Sebuyau)  
109 are blackwater rivers that draining extensive peatlands (Fig. 1). The drainage basin of the  
110 Maludam is about 91.4 km<sup>2</sup> and majority of the river located in the Maludam National Park with  
111 10m thick peat (Muller et al., 2015). The other two rivers are highly human disturbed with intensive  
112 oil palm and sago plantations. For the Rajang, it is separated into two parts by Sibu Town, upper  
113 reaches mainly drain mineral soils, while down reaches develop multiple distributary channels  
114 (e.g., the lower Rajang, Serendeng, Igan; Fig. 1). These channels are also surrounded by broad  
115 peatlands. It is reported that peat greater than 1m thick covered 50% of the delta plain (Staub et  
116 al., 2000). However, Deforestation and changing in land use are accelerating the peatland  
117 degradation (Fig. 1). More than 50% peatland (11% of the catchment size) in Rajang watershed  
118 has been occupied by industry plantation (e.g. oil palm) (Miettinen et al., 2016). Fishery, logging  
119 and timber processing are the traditional supports for local citizens (Miettinen et al., 2016).

120 The climate of the study area is classified as tropical ever-wet, with average rainfall in excess of  
121 3700 mm/year. The average monthly water discharge of the Rajang is about 3600 m<sup>3</sup>/s, with

122 peak discharge (~25,000 m<sup>3</sup>/s) observed during the northeastern monsoon season (December  
123 to March; Staub et al., 2000). However, the amount of suspended sediments delivered from the  
124 Rajang basin to the delta plain demonstrated slightly variation (2.0MT/s dry season *versus* 2.2  
125 MT/s wet season) but changed substantially about the amount of sediment delivered from the  
126 delta plain to the South China Sea (Staub et al., 2000). It is estimated that the annual sediment  
127 discharge of the Rajang is 30 Mt. The turbidity maximum in the lower Rajang channels occurred  
128 during the low or reduced discharge period. It is reported that up to 24 Mt of sediment is  
129 deposited in the delta front with preserved annual sediment layers at the order of one cm thick  
130 (Staub et al., 2000). The water discharge of the Maludam is quite low, only 4.4±0.6 m<sup>3</sup>/s, from  
131 the 91.4 km<sup>2</sup> catchment (Muller et al., 2015). The river length of Maludam is 33 km. For the  
132 Sebuyau and Simunjan, river length is 58 and 54 km, respectively (Martin et al., 2018). However,  
133 hydraulic information for these two rivers is largely unknown. The three sampling periods  
134 resembled the end of this northeastern monsoon (i.e., March, the end of the wettest season of  
135 the year) and were shortly before the beginning of the northeastern monsoon (i.e., August and  
136 September, the end of the drier season).

137 The surface sediments were sampled at the middle stream of river using grab samplers from a  
138 small boat at each station and then 0 - 5 cm subsamples were collected and frozen (-20°C) until  
139 they were dried for subsequent analyses in the laboratory. Soil sampling was conducted at the  
140 same time along the Rajang river bank where the sites have minimal human disturbances and  
141 short soil cores were collected and mixed *in situ* as one composite sample for the depth of 0-  
142 10cm by getting rid of visible roots and detritus. The vegetation of tropical peat swamp forest is  
143 dominated by trees, e.g. the *Anacardiaceae*, *Annonaceae* and *Euphobiaceae* etc. (Page et al.,  
144 2006). Fresh, typical vegetations (listed in Table S2) were separately collected by leave, stem and  
145 roots, some detritus, which floating at the surface layer of the river were also collected for the  
146 comparison study. All botanical samples and soils within the basin were collected at the same  
147 time and stored in a freezer. The hydrological parameters of the surface river water (e.g., salinity,  
148 pH, and temperature) at each station were determined using an Aquaread® multiple parameters

149 probe (AP-2000).

150

## 151 **2.2 Chemical analyses**

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

154 Grain size characteristics were measured directly from aliquots of the surface sediment samples  
155 using a Coulter LS 100Q (Coulter Company, USA), after treatment with 5% H<sub>2</sub>O<sub>2</sub> and 0.2M HCl  
156 to dissolve organic matter and biogenic carbonate. The sediment grain sizes are expressed as  
157 the proportions of clay (<4 μm), silt (4–63 μm), and sand (>63 μm), with a measurement error  
158 of ≤5% for the entire dataset. The remaining sediments were ground to 80 mesh (187.5 μm) for  
159 elemental, isotopic, and lignin analyses.

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

169 Lignin phenols were extracted using the cupric oxide digestion technique (CuO; Hedges and  
170 Ertel, 1982; Yu et al., 2011). Briefly, the powdered samples were weighed and placed in O<sub>2</sub> free  
171 Teflon-lined vessels, and digested in a microwave digestion system (CEM MARS5) at 150°C for  
172 90 min (Goñi and Montgomery, 2000). Samples were then acidified to pH < 2 and phenolic  
173 monomers were extracted into 99:1 (volume ratio) ethyl acetate/petroleum ether, dried, and  
174 stored at –20°C until further analysis. Samples were analyzed as trimethylsilyl derivatives of N,O-  
175 bis(trimethylsilyl)trifluoroacetamide (BSTFA) and trimethylchlorosilane (TMCS; 99:1) by Agilent

176 6890N gas chromatography (DB-1 column, FID). The lignin phenols concentration was quantified  
177 using calibration curves based on commercial standards (Sigma Aldrich). Eleven phenol  
178 monomers were extracted and categorized into five groups: syringyl (S, syringaldehyde,  
179 acetosyringone, syringic acid), vanillyl (V, vanillin, acetovanillone, vanillic acid), cinnamyl (C, *p*-  
180 coumaric acid, ferulic acid), *p*-hydroxyl (P, *p*-hydroxybenzaldehyde, *p*-hydroxyacetophenone,  
181 and *p*-hydroxybenziic acid), and 3,5-dihydroxy benzoic acid (DHBA). Coefficients of analytical  
182 variation associated with phenols values were <10% based on replicate analysis of the same  
183 samples.

184 Ratios of syringyl-to-vanillyl phenols (S/V) and cinnamyl-to-vanillyl phenols (C/V) are often used  
185 to indicate the relative contribution of angiosperm and non-woody tissues versus gymnosperm  
186 wood, respectively (Hedges and Mann, 1979). Since both ratios have been found to decrease  
187 with the preferential degradation of S and C relative to V phenols, lignin phenols vegetation  
188 index (LPVI) was developed to be an alternative approach to evaluate the original of various type  
189 of vegetations (Tareq et al., 2004; Thevenot et al., 2010):

190 Lignin phenols vegetation index (LPVI) =  $\left\{ \frac{S}{S+1} \right\} \times \left\{ \frac{C}{C+1} \right\} \times \left\{ \frac{V}{V+1} \right\} + 1$

191 The ratio of P/(V+S) may reflect the diagenetic state of lignin when the other sources of P phenols  
192 (such as protein and tannin) are relatively constant (Dittmar and Lara 2001). The acid-to-aldehyde  
193 (Ad/Al) ratios of V and S phenols are often used to indicate lignin degradation and increases  
194 with increasing lignin oxidation (Otto and Simpson 2006).

195

### 196 2.3 Statistical analyses

197 All statistical analyses were carried out using SPSS 10.0 (IBM SPSS Inc., USA) and results were  
198 plotted using Origin software (Origin Lab Inc., USA). Multivariate statistical approaches such as  
199 principle component analysis (PCA) and cluster analysis (CA) are among the most widely used  
200 statistical methods in determining the significance of specific parameters (including OC%, TN%,  
201 mean grain size, clay% and silt%, total lignin phenols concentrations, DHBA and the ratios of  
202 vanillic acid to vanillin ((Ad/Al)/V)) within a dataset (Pradhan et al., 2009). Interrelationships among

203 the sampling points in different rivers were characterized by cluster analysis using Ward's  
204 method (linkage between groups) and similarity measurements in terms of Euclidian distance,  
205 illustrated in dendograms. Errors listed in tables represent standard deviations for the analytical  
206 data. Differences and correlations were evaluated as significant at the level of  $p < 0.01$ .

207

## 208 **3 Results**

### 209 **3.1 Hydrological parameters, grain size, and bulk elemental and stable isotopic composition of** 210 **vegetation, soil, and sediment**

211 The hydrological parameters for the study area are summarized in Table S1. The salinity of the  
212 lower Rajang system varied significantly (from 12‰ to 32‰) because of saline water intrusion  
213 in the estuarine region, but there were limited pH variations (6.5–7.9). Dissolved oxygen (DO)  
214 levels showed significant spatial variations, with the lowest values (2–3 mg L<sup>-1</sup>) being recorded in  
215 the Igan channel, where dense peats were observed, and the higher values (4–6 mg L<sup>-1</sup>) recorded  
216 in the other two channels. The salinity of the Simunjan indicated that freshwater dominated,  
217 whereas the two other small rivers showed saline water influences. The variation in pH values  
218 among the three small rivers decreased from the Sebuyau (~6.4), to the Simunjan (~5.1), and the  
219 Maludam (~3.7). The DO concentrations in the three small rivers varied in a low range (average:  
220 2–3 mg L<sup>-1</sup>), with the lowest values in the three systems being around 1.4 mg L<sup>-1</sup>.

221 The compositions of bulk sediments from the Rajang and the three small rivers are presented in  
222 Tables 1 and S1. The mean grain sizes from the upper Rajang (212±47 μm) were much coarser  
223 than those from the lower Rajang (40±38 μm) and the small rivers (22±16 μm). The finest samples  
224 (9±2 μm) were collected from the Maludam in March 2017. Generally, the samples collected  
225 during the dry season were coarser than those from the flood season in the Maludam and  
226 Simunjan, but this was not the case for the Sebuyau. The average organic carbon content shows  
227 a significant negative relationship with mean grain size among these samples ( $r^2 = 0.67$ ,  $p < 0.01$ ).  
228 Mean values of Total organic carbon (TOC) concentrations were higher in the peat-draining  
229 rivers (2.2±0.58%, 2.6±1.23%, and 2.6±0.8% for the Maludam, Sebuyau, and Simunjan,

230 respectively) compared with the lower Rajang ( $1.1 \pm 0.5\%$ ), and the lowest values were observed  
231 in the upper Rajang ( $0.12 \pm 0.02\%$ ). The highest values of OC were measured in plants samples  
232 and varied from 30%–49% (Table S2). The mean TOC value in the soil samples was  $3.6 \pm 0.6\%$   
233 (Table S3).  
234 TN content ranged from 0.02% to 0.17% in the samples collected from the Rajang, from 0.09%  
235 to 0.37% in the small rivers, from 0.73% to 1.65% in the vegetation, and averaged  $0.19 \pm 0.02\%$   
236 for the soil samples (Tables 1, S2, and S3). Although nitrogen was enriched in the samples from  
237 the peat-draining rivers, they still had higher mean C/N values ( $15.8 \pm 3.7$ ) compared with the  
238 lower Rajang ( $11.5 \pm 1.6$ ) while vegetation samples, exhibited low N content and high C/N (C/N =  
239  $56 \pm 34$ ).  
240 The most abundant vegetation collected from the Maludam showed relatively depleted carbon  
241 isotope ratios ( $\delta^{13}\text{C} = -31\text{‰}$ ) that were typical of C3 vegetation (Table S2). The detritus samples  
242 were also relatively depleted in  $^{13}\text{C}$  ( $\delta^{13}\text{C} = -29.2\text{‰}$ ; Table 1). The isotope ratios of the peat-  
243 draining river's sediments (average  $\delta^{13}\text{C}$  varied at  $-28.2$  —  $-27.4\text{‰}$ ) were comparable with the  
244 Rajang's (average  $\delta^{13}\text{C} = -28.6 \pm 0.6\text{‰}$ ) (Tab. 3). The  $\delta^{13}\text{C}$  values of the soil samples are similar to  
245 those of riverine sediments ( $\delta^{13}\text{C} = -28.4\text{‰}$ ).

246

### 247 **3.2 Lignin phenols content**

248 The lignin phenols obtained after CuO oxidation are expressed as  $\Lambda 8$  ( $\text{mg} (100 \text{ mg OC})^{-1}$ ),  
249 except for the lignin yield ( $\Sigma 8$ ), which is the sum of C + S + V and is expressed as  $\text{mg} 10 \text{ g dw}^{-1}$ ,  
250 and are presented in Fig. 2 as well as Tables 2 and S1-3. The highest yields were measured in the  
251 vegetation samples ( $300$ – $900 \text{ mg} 10 \text{ g dw}^{-1}$ ). The lignin yield from the soil samples and the three  
252 small rivers (average of  $\sim 30 \text{ mg} 10 \text{ g dw}^{-1}$ ) is also higher than that from the Rajang samples  
253 (average of  $< 10 \text{ mg} 10 \text{ g dw}^{-1}$ ), with the lowest value observed in the upper Rajang ( $0.16 \text{ mg} 10$   
254  $\text{g dw}^{-1}$ ; Table 2). There are correlations between  $\Sigma 8$  and OC% in each river ( $r^2 > 0.5$ ), with the  
255 slope decreasing in the order of Maludam > Simunjan > Sebuyau > Rajang (Fig. 2a). The variation  
256 in  $\Lambda 8$  from various pools shows a similar distribution as the  $\Sigma 8$  values. The average concentrations

257 for the vegetation, soil, and the four river systems mg (100 mg OC)<sup>-1</sup> approximately 18, 8.3, 5.4  
258 mg (100 mg OC)<sup>-1</sup> (for the Rajang), 6.2 mg (100 mg OC)<sup>-1</sup> (for the Maludam), 7.9 (for the Sebuyau),  
259 and 7.4 mg (100 mg OC)<sup>-1</sup> (for the Simunjan), respectively.

260 The C/V and S/V ratios differ with vegetation type (Fig. 2b). Angiosperm leaves show high S/V  
261 (>1) and C/V ratios (~0.8). Angiosperm wood and root samples show lower C/V ratios (<0.2).  
262 The detritus samples show intermediate S/V ratios (0.6–1.0) and lower C/V ratios (~0.1). Soil  
263 samples have relatively high S/V (~1.1) and low C/V (~0.07) values. The four rivers show limited  
264 variations in S/V (0.4–0.8) and C/V (0.02–0.08) ratios. The LPVI values of the fresh plant material  
265 range from 113 to 2854 for leaves and 192 to 290 for wood. The values for detritus range between  
266 36 and 228, and for soil and sediment range between 30 and 60 (Table 2).

267 The ratios of vanillic acid to vanillin ((Ad/Al)<sub>v</sub>) and syringic acid to syringaldehyde ((Ad/Al)<sub>s</sub>)  
268 increase slightly from the vegetation to river samples (Table 2). The ratios obtained from the  
269 vegetation and soil samples show similar values ((Ad/Al)<sub>s</sub> = ~0.30; (Ad/Al)<sub>v</sub> = ~0.35). The ratios  
270 from the small river samples range from 0.41 to 0.58 for (Ad/Al)<sub>v</sub> and 0.30 to 0.36 for (Ad/Al)<sub>s</sub>.  
271 The values from the lower Rajang are similar to those from the small rivers, but this is not the  
272 case for the upper Rajang, where higher (Ad/Al)<sub>s</sub> and (Ad/Al)<sub>v</sub> values were recorded. The two  
273 ratios are linearly correlated in all sediment samples ( $r^2 = 0.68$ ,  $p < 0.05$ ), except for the samples  
274 collected from the Simunjan.

275 The P/(V + S) ratio is low in the vegetation samples, except for the leaf samples (P/(V + S) = 0.22),  
276 which reflects the low P content in most vegetation (Table 2). However, in some plant samples  
277 (*Elaeis guineensis Jacq.*), we detected relative higher P content (Table S2). The P/(V + S) ratio is  
278  $0.28 \pm 0.03$  for the soil samples,  $0.18 \pm 0.4$  for the small rivers,  $0.17 \pm 0.02$  for the lower Rajang, and  
279  $0.51 \pm 0.04$  for the upper Rajang. DHBA is very low in the upper Rajang (~0.07), but higher in the  
280 Maludam in the dry season (average value of 0.44). Values in the Simunjan in both seasons are  
281 similar to those from the soil samples (~0.38). Higher values of DHBA were measured in the  
282 lower Rajang and the Sebuyau in the dry season than in the wet season.

283

### 284 3.3 Statistical analyses

285 The results of cluster and PCA analyses of both bulk geochemical and lignin phenols proxies for  
286 all sediments are shown in Fig. 3. Four distinct groups were identified based on the cluster  
287 analysis. The Maludam and the tributary of the lower Rajang (Igan) are grouped together, and  
288 the Simunjan and Sebuyau are grouped together. The lower Rajang and upper Rajang are  
289 separated from each other (Fig. 3a). Similar groupings are evident in the results of the PCA  
290 analysis, which was based on the distribution of factors 1 and 2 that represent total loadings of  
291 45% and 32%, respectively (Fig. 3b). The PCA results implied that factor 1 showed close  
292 correlations with the (Ad/Al)<sub>v</sub> ratio and grain size while factor 2 showed a close correlation with  
293  $\Sigma 8$  and OC%.

294

## 295 4 Discussion

### 296 4.1 Comparison with systems worldwide: lignin parameters derived from sediment and peat

297 Table 3 summarizes the distribution of bulk and lignin parameters of sediments from typical  
298 systems worldwide. Although the TOC values of our studied systems are lower than peat  
299 samples but the concentrations of lignin phenols are comparable, which are typically enriched  
300 in lignin phenols compared with other river systems (Table 3; Bianchi et al., 2002; Gandois et  
301 al., 2014; Li et al., 2015; Sun et al., 2017; Pradhan et al., 2014; Winterfeld et al., 2015). The TN  
302 values of our peat samples are between two and four times higher than those seen in other  
303 systems worldwide, as was also observed in small rivers along India's west coast (Pradhan et al.,  
304 2014). The higher values of  $\Lambda 8$  found in our studied systems were linked to vegetation types  
305 (trees dominated) (Zaccone et al., 2008) and partially caused by peat-draining and intense  
306 human activity near the watersheds (e.g. land use change and logging activities), as reported  
307 previously (Milliman and Farnsworth, 2011; Moore et al., 2013; Rieley et al., 2008). Much of the  
308 peatland neighboring the Simunjan and Sebuyau catchments has been changed to palm oil  
309 plantations (Martin et al., 2018). The terrigenous OM has been affected by diagenesis, as  
310 (Ad/Al)<sub>v</sub> varies markedly among the different systems (Table 3). The (Ad/Al)<sub>v</sub> values of the

311 sediments sampled here are comparable to fresh and only low to medium oxidized. Elevated  
312 (Ad/Al)<sub>V</sub> values observed from the Maludam's sediments (March, 2017) may be also attributed  
313 to source plant variations as observed in other study case (Zhu et al., 2019).

314

#### 315 **4.2 Origin of sediment organic matter in tropical peat-draining rivers**

316 The depleted average  $\delta^{13}\text{C}$  values (-31.8 ~ -28.1‰) of our vegetation samples indicate an  
317 insignificant contribution from C4 plants in the study area (Gandois et al., 2014; Sun et al., 2017).

318 The high C/N ratio (64.8) indicates a predominance of terrestrial high plant species (e.g.,  
319 *Nepenthes sp.* and *Avicennia marina Vierh.*). The  $\delta^{13}\text{C}$  and C/N values (-27.2‰ and 12,

320 respectively) obtained from the soil and sediments collected near the rivers suggest that  
321 terrestrial organic matter is the dominant contributor (Table 1). The cluster and PCA analyses

322 suggest that there are no significant seasonal differences in these rivers. Previous study reported  
323 that the sediment load from the basin to the delta was no seasonal pattern, combined with

324 comparable precipitations during our two sampling seasons, our observations matched (Martin  
325 et al., 2018; Staub et al., 2000). The close correlation of factor 2 with OC% and  $\Sigma 8$  in the PCA

326 suggests factor 2 relates to the source of the organic matter (Fig. 3), as also be indicated by the  
327 strong correlation between OC% and  $\Sigma 8$  ( $r^2$ : 0.53-0.85) (Fig. 2). Correlation of OC% and  $\Sigma 8$  of

328 the Maludam ( $r^2 = 0.81$ ) show the highest slope, possibly related to its pristine condition that  
329 promotes better conservation of vegetation in its peat. Furthermore, the differences between

330 the upper and lower Rajang are highlighted by the PCA results (score 1 represents 45% of the  
331 total loading while score 2 is 32%) and bulk parameters; i.e., the upper Rajang drains a mineral

332 soil whereas peat is dominant in the delta region. This also explains why the Rajang data do not  
333 plot with the other small river systems; the linear relationship between  $\delta^{13}\text{C}$  and  $\Sigma 8$  for the Rajang

334 ( $r^2 = 0.92$ ) forms a distinct group separate from the small rivers ( $r^2 = 0.59$ ).

335 The S/V and C/V ratios are often used as indicators of the vegetation origin of the lignin fraction;  
336 e.g., the woody and non-woody parts of gymnosperm and angiosperms (Hedges and Mann,

337 1979). The S/V values (<0.8) of the peat-draining rivers are slightly lower than the values of other

338 peats (<1.5), but the C/V ratios are comparable (Tareq et al., 2004). The differences in these  
339 parameters between the sediments and the vegetation and soils, as illustrated in Fig. 2, suggests  
340 that they are composed mostly of angiosperm wood. This finding is further confirmed by the  
341 LPVI values (Gymnosperm woods: 1, non-woody Gymnosperm tissues, 3-27; Angiosperm woods:  
342 67-415; non Angiosperm tissues: 176-2782), which are commonly less than 60 in these sediment  
343 samples (Tareq et al., 2004). Previous studies have concluded that tropical peats are derived  
344 mainly from wood (Anderson, 1983; Gandois et al., 2014). For the Rajang, the LPVI values show  
345 a positive linear correlation with  $\Lambda 8$  concentrations ( $r^2 = 0.56$ ); however, for the small rivers  
346 (based on mean values, except the samples collected in March 2017 from the Maludam) this  
347 relationship shows a negative correlation ( $r^2 = 0.91$ ). This suggests that the small rivers receive  
348 more lignin derived from woody material, whereas the Rajang has a mixture of sources. The  
349 unusual behavior of the Maludam's samples might be related to the dominance of finer-grained  
350 sediments when compare with the other rivers, because woody material tends to be  
351 concentrated in the coarser fraction (Table 1).

352 P phenols in the Rajang are derived from lignin, as supported by the significant correlation of  
353 the content of P phenols and lignin content ( $r^2 = 0.93$ ). However, there is no correlation between  
354 P phenols and lignin content for the small rivers. All P/V values from the samples (0.13–0.28) are  
355 higher than the average P/V ratio of wood (0.05) but similar to the range observed for leaves  
356 (0.16–6.9; Hedges et al., 1986). Considering this, some non-woody angiosperms are the most  
357 likely source of high P phenols in the small rivers. Combined the composition of P and V in plants  
358 samples listed in Table S2, we find some dominant species, e.g. *Dipterocarpaceae*, *Bruguierag*  
359 *ymnorrhiza(L.) Poir.*, *Elaeis guineensis Jacq.* have a relatively higher P/V ratios in their non-woody  
360 parts.

361

#### 362 **4.3 Transformation of lignin signatures in tropical peat-draining rivers**

363 (Ad/Al)<sub>v</sub> ratios are often used to evaluate the degradation status of terrestrial OM. The (Ad/Al)<sub>v</sub>  
364 ratios for soils reported in previous studies fall within the range 0.16–4.36, 0.1–0.2 for fresh

365 angiosperm wood and 0.2–0.6 for non-woody tissues (Hedges et al., 1988; Opsahl and Benner,  
366 1995; Thevenot et al., 2010). In our study, the variability of the (Ad/Al)<sub>v</sub> ratios obtained from the  
367 vegetation, soil, and sediments was limited, with values between 0.3 and 0.58 except from the  
368 samples from the upper Rajang (~1.0), which suggests the mild degradation of OC<sub>terr</sub> in most  
369 samples. The degradation status of lignin is negatively correlated with the  $\Lambda_8$  values ( $r^2 = 0.73$ )  
370 in the Rajang, and with a higher degradation signal observed in the upper Rajang, which drains  
371 mineral soils with lower lignin levels (Fig. 4a). However, the  $\Lambda_8$  values with (Ad/Al)<sub>v</sub> ratios was  
372 not so significant in the small river systems as we expected, partially resulting from the variation  
373 of (Ad/Al)<sub>v</sub> also could be vegetation source controlled (Fig. 4b). In additional, such a distribution  
374 could be related to the grain size effect, as illustrated in the Rajang with high correlation (Fig. 4c)  
375 and not so convincing but with a certain trend in small rivers (Fig. 4d). Of the sediments sampled  
376 here, the upper Rajang samples contain the largest coarse fraction and the finest sediments are  
377 collected from the Maludam in March 2017. The (Ad/Al)<sub>v</sub> ratios increase with increasing coarse  
378 fraction of the sediments in the Rajang, which is typically observed in other systems (Bianchi et  
379 al., 2002; Li et al., 2015; Sun et al., 2017) (Fig. 4c). The variation of (Ad/Al)<sub>v</sub> ratios with mean size  
380 of the sediments in the small rivers is not so convincing as the Rajang (Fig. 4d). Selective sorption  
381 of acid to aldehyde might affect the variation of the (Ad/Al)<sub>v</sub> ratio in the small river systems  
382 (Hernes et al., 2007). Additionally, the relatively fresh condition of the OM in the Maludam  
383 samples (in September 2017) might be related to the fluvial supply of fresh vegetation.

384 The syringyl and cinnamyl series are preferentially degraded when compared with the vallinyl  
385 series, resulting in a decrease in the S/V and C/V ratios during lignin degradation (Goni et al.,  
386 1995; Opsahl and Benner, 1995). Our samples show a negative linear relationship between the  
387 S/V and (Ad/Al)<sub>v</sub> ratios in the Rajang samples ( $r^2 = 0.85$ ; Fig. 5a). However, the variation of the  
388 S/V and (Ad/Al)<sub>v</sub> ratios in the small rivers is limited, with a scattering decrease trend (Fig. 5b).  
389 Both correlations indicate that the decrease in the S/V ratios is linked to degradation, and this  
390 suggests that we should be cautious when using S/V ratios for source evaluation in this study.

391 Previous studies demonstrated that lignin mineralization in humid tropical forest soils is

392 dominated by methoxyl-C mineralization under aerobic and fluctuating redox conditions (Hall  
393 et al., 2015). Demethylation reduces the yield of methoxylated phenols (V and S phenols) but  
394 does not affect P phenols. Therefore, the P/(S+V) ratio can be used as an indicator of lignin  
395 transformation (Dittmar and Kattner, 2003). However, in this study the ratio of P/(S+V) in most  
396 sediment samples did not vary greatly (~0.2). Although there was a linear correlation between  
397 the P/(S+V) and (Ad/Al)<sub>v</sub> ratios among all the sediments ( $r^2 = 0.89$ ), no clear trend was observed  
398 for the small rivers, which may suggest both parameter's more links to source instead of  
399 diagenetic process in these systems.

400

#### 401 **4.4 Impact of environmental parameters on lignin dynamics**

402 It is well explored that bulk organic matter composition and degradation are influenced by many  
403 environmental factors such as climate, grain size, mineral composition, soil characteristics, land  
404 use changes, logging, and biomass burning (Hernes et al., 2007; Gandois et al., 2014; Sun et al.,  
405 2017; Thevenot et al., 2010). Most Southeast Asian peat-draining rivers are impacted by human  
406 activities such as deforestation, urbanization and damming (Milliman and Farnsworth, 2011). The  
407 PCA analysis revealed that the behavior of lignin in the Rajang is substantially different from that  
408 in the three peat-draining rivers, and especially in the upper Rajang, which drains through a  
409 mineral soil with low  $\Sigma 8$  values and strong degradation (Figs 3 and 4), since it was recently shown  
410 that lignin could decompose as fast as litter bulk carbon in mineral soils (Duboc et al., 2014). In  
411 the delta region, most parameters were quite comparable, except  $\Sigma 8$  and OC% (Table S1). The  
412 higher values of  $\Sigma 8$  and OC% were observed in Simunjan and Sebuyau, where land use and  
413 drainage observed. Usually land use and drainage of tropical peat will accelerate the loss of  
414 vegetation and OC degradation (Kononen, et al., 2016), here it may be explained by the high  
415 content of OC and lignin in oil palm, which is the major plantation in both regions.

416 In this study, the OC content increases with decreasing grain size, implying that fine sediments,  
417 with larger specific surface areas and rich in clay, contain more OM than coarser sediments, as  
418 reported previously (Sun et al., 2017). Increasing (Ad/Al)<sub>v</sub> values are observed in the Rajang with

419 increasing grain size, which suggests that lignin associated with larger mineral particles is more  
420 strongly degraded. This observation indicates the preferential preservation of lignin in finer-  
421 grained sediments, resulting from their ability to provide better protection against further  
422 oxidative degradation (Killops and Killops, 2005). For the small river systems, the  $(Ad/Al)_v$  ratios  
423 inattentively decrease with increasing grain size, corresponding to the increasing  $\Sigma 8$  values (Fig.  
424 4b and 4d). Such kind of trends have been described by Keil et al. (1998) and Tesi et al. (2016),  
425 who found that lower  $(Ad/Al)_v$  values were present in the coarser fractions due to the less  
426 efficient processing of plant remains prior to deposition. The sediments collected from the three  
427 small peat-draining rivers (except samples from the Maludam in September, 2017) could contain  
428 limited amounts of plant debris, in which case fresh plant tissue would have been incorporated  
429 into the coarser sediment fractions, leading to the low  $(Ad/Al)_v$  values. However, the variation in  
430  $\Sigma 8$  values does not support this speculation, and therefore we conclude that the selective  
431 sorption of acid to aldehyde could explain the elevated  $(Ad/Al)_v$  ratios recorded in the fine  
432 fraction. The different grain-size effects on OC<sub>terr</sub> composition, as seen when comparing the  
433 Rajang with the small rivers, suggests that there are other processes (microbial process, logging  
434 etc.) working on OC<sub>terr</sub> in these two systems, which cause post-depositional changes in the  
435 OC<sub>terr</sub> characteristics.

436 Tropical soils are reported naturally poor in N and P, but some studies have shown that with  
437 intensive management (land use/deforestation) they tend to become rich in recalcitrant  
438 compounds, since nitrogen content tends to stimulates decomposition of low-lignin litter by  
439 decomposer microbes, but usually decrease the activity of lignolytic enzymes and inhibit  
440 decomposition of high-lignin litter (Knorr, et al., 2005; Thevenot et al., 2010). In our study, we  
441 found a higher TN% in the small rivers compared with the Rajang. A significant correlation  
442 between  $\Sigma 8$  and TN% ( $r^2= 0.74$ ) is observed in all systems, which might suggest a contribution  
443 from plant litter affecting both parameters (Fig. 6a). The relation of  $(Ad/Al)_v$  ratios with C/N ratios  
444 of the Rajang appears correlated ( $r^2= 0.34$ ). For the comparison, average values were applied to  
445 two systems, we found the average  $(Ad/Al)_v$  ratios had certain correlation with the average C/N

446 ratios, but with different slopes for the Rajang and the small rivers (Fig. 6b). Quicker decline of  
447 C/N ratios related to slower lignin degradation in small rivers, this could be related to the  
448 expected impact of nitrogen on lignin degradation (Dignac et al., 2002; Thevenot et al., 2010). A  
449 high N content will inhibit fungal lignin biodegradation (Fog, 1988; Osono and Takeda, 2001),  
450 and this explains why higher lignin phenols with moderate degraded characteristics was  
451 observed in the small river systems in which higher TN% were recorded. The exceptional data  
452 were collected during September 2017, which was a time of saline water intrusion.

453 Large-scale land reclamation, including deforestation and urbanization, has taken place in  
454 Southeast Asia over the past few decades (Miettinen et al., 2016). Logging activities have had a  
455 significant influence on peat decomposition processes and the quality of organic matter inputs  
456 (Hoscilo et al., 2011; Hooijer et al., 2012; Gandois et al., 2014). Gandois et al. (2013) reported an  
457 increase in the N content at a deforested site and concluded that it was caused by an increase  
458 in the microbial deposition of peat. The lignin yield ( $\Sigma 8$ ) is closely correlated with the OC% in the  
459 different rivers (Fig. 2). The highest yield was observed at the Maludam, which confirmed the  
460 significant contribution of plant litter and better preservation due to the low pH and DO levels,  
461 especially woody carbon. However, the relatively higher yield in the Rajang compared with the  
462 other two disturbed peat-draining rivers (i.e., the Simunjan and Sebuyau) suggests an additional  
463 source of lignin, which might implicate the addition of logging residue to the Rajang systems, as  
464 proposed by Gandois et al. (2014).

465

## 466 **5 Conclusions**

467 We used sediment grain size data, TOC contents, the stable carbon isotopic composition of  
468 organic matter, and lignin phenols concentrations to investigate the characteristics of OC<sub>terr</sub> in  
469 a tropical peat-draining river system, as well as its fate and environmental controls. The depleted  
470  $\delta^{13}\text{C}$  levels of all of the sediment samples demonstrate that contributions from C<sub>3</sub> plants  
471 dominated the OC<sub>terr</sub> in the study region. The lignin composition of the organic matter indicates

472 that the most important plant sources of organic matter were woody angiosperm C3 plants,  
473 especially in the three small rivers. Our cluster and PCA analyses show no distinct seasonal  
474 variations in the bulk and lignin compositional signatures in the study area, although the upper  
475 Rajang receives contributions from mineral soils with unique lignin parameters and a coarser  
476 grain size. Both the bulk organic matter parameters and the lignin compositions were indicated  
477 to be correlated to the grain size of the riverbed sediments. The (Ad/Al)<sub>v</sub> ratios increased with  
478 decreasing mean size of the sediments from the small rivers. Selective sorption of acid to  
479 aldehyde might affect the variation of the (Ad/Al)<sub>v</sub> ratio in the small river systems. Our samples  
480 show a negative linear relationship between the S/V and (Ad/Al)<sub>v</sub> ratios in the Rajang samples,  
481 which implies that the decrease in S/V ratios is linked to degradation. The (Ad/Al)<sub>v</sub> ratios appear  
482 to be related to the C/N ratio in the Rajang and the small rivers. A high N content will inhibit  
483 fungal lignin biodegradation, which might explain higher lignin phenols with moderate degraded  
484 process observed in the small river systems where a higher TN% is recorded. Most of the OC<sub>terr</sub>  
485 discharged from the Rajang and small river systems was composed of woody angiosperm plants  
486 and the terrestrial organic matter undergoes limited diagenetic alteration before deposition, and  
487 could potentially become a significant regional carbon source to the atmosphere after extensive  
488 degradation. This study provides new insights into the amount of terrestrial OC preserved in the  
489 tropical delta region of southeastern Borneo, as well as into the biogeochemical transformation  
490 of OM from terrestrial source to marine sink across this region.

491

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494 collected samples and KZ and YW analyzed the samples. YW, KZ and JZ processed and analyzed  
495 the data. All authors contributed to data interpretation and to the writing of the manuscript.

496

497 **Data availability.** The datasets in the present study are available from the corresponding author  
498 on reasonable request.

499

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

501

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505

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517

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

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

763

764 **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**  
 765 **phenols, P: p-hydroxyl phenols; DHBA: 3,5-dihydroxy benzoic acid; see the main text for definitions of  $\Sigma 8$ ,  $\Lambda 8$ , Ad/Al, and LPVI)**  
 766

Samples	Time	$\Sigma 8$ (mg/10 g dw)	$\Lambda 8$ (mg/100 mg OC)	V	S	C	S/V	C/V	(Ad/Al) <sub>v</sub>	(Ad/Al) <sub>s</sub>	P/(V+S)	DHBA	DHBA/V	LPVI
<b>Angiosperm leaves &amp; grasses (n=10)</b>	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
<b>Angiosperm woods(n=5)</b>	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
<b>Roots(n=3)</b>	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
<b>Lower detritus(n=8)</b>	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
<b>Sebuyau detritus(n=5)</b>	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
<b>Simunjan detritus(n=4)</b>	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
<b>Soil(n=8)</b>	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
<b>Upper Rajang (n=4)</b>	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
<b>Lower Rajang (n=16)</b>	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
<b>Lower Rajang (n=5)</b>	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
<b>Maludam (n=5)</b>	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
<b>Maludam (n=2)</b>	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

<b>Sebuyau (n=6)</b>	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
<b>Sebuyau (n=5)</b>	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
<b>Simunjan (n=6)</b>	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
<b>Simunjan (n=6)</b>	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

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

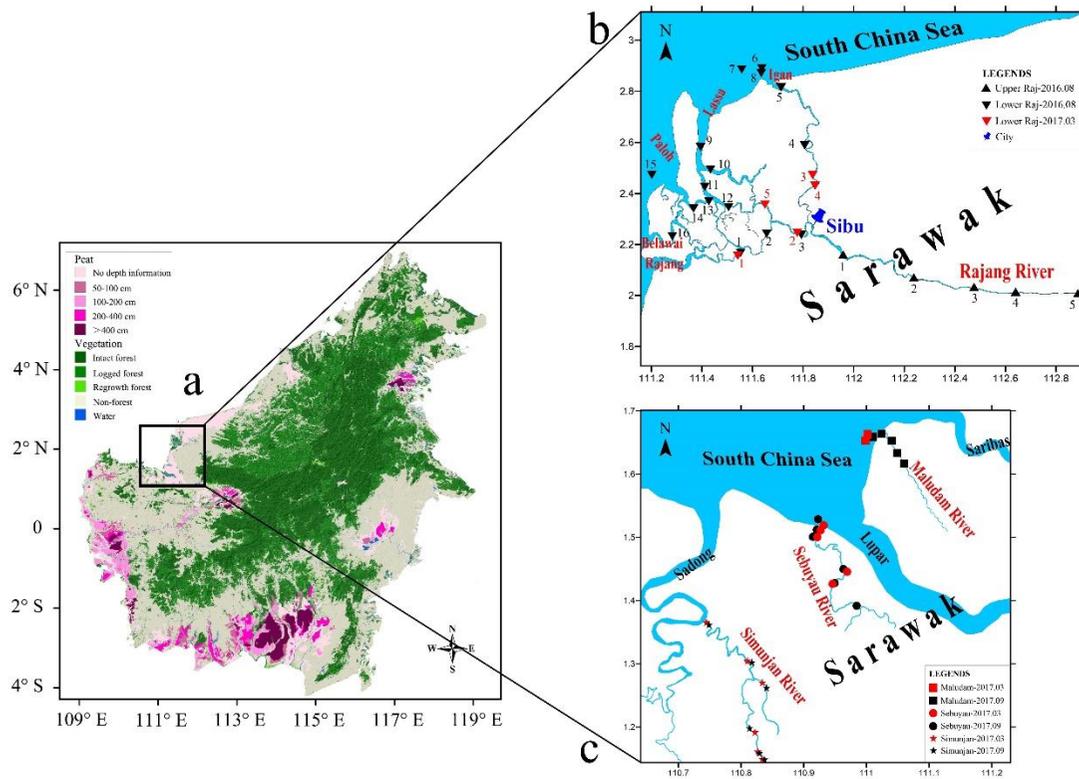
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Samples	Station	OC (%)	TN (%)	C/N	$\delta^{13}\text{C}$ (‰)	$\Sigma 8$ (mg/10g dw)	$\wedge 8$ (mg/100 mg OC)	S/V	C/V	(Ad/Al)v	(Ad/Al)s	P/(S+V)	DHBA/V	References
Amazon River	estuary	0.13~1.44	—	—	-29.4~-27.5	0.10~11.05	0.75~9.27	0.84~1.51	0.12~0.47	0.26~0.61	0.15~0.56	—	—	1
Congo River	submerged delta	0.80~4.20 2.10	—	5.8~10.1 8.3	-23.5~-19.0	—	0.07~0.37 0.15±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	—	—	14.2±1.3	-27.2±1.5	—	—	1.26±0.32	0.19±0.12	0.68±0.11	0.81±0.21	0.57±0.10	—	3
35 Indian rivers	North group	0.61±0.30	0.04±0.01	18.7±6.9	-22.9±0.9	0.11±0.12	1.60±1.00	0.90±0.20	0.20±0.10	0.70±0.20	—	0.40±0.20	0.30±0.20	4
	South group	2.30±0.60	0.12±0.03	19.8±4.1	-26.3±0.8	1.7±0.5	6.70±2.80	1.50±0.50	0.30±0.10	0.50±0.10	—	0.20±0.20	0.10±0.20	
Kapuas River	whole basin	0.55~14.20	0.05~0.55	11.0~34.8	-30.4~-27.3	—	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	11.6±1.7	-28.6±0.6	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	
Maludam River	estuary	2.22±0.69	0.20±0.05	12.8±1.8	-27.4±0.6	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	This research
Sebuyau River	whole basin	2.37±0.69	0.16±0.03	17.4±4.6	-27.8±0.3	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	16.4±3.0	-28.2±0.5	18.45±5.96	7.30±1.04	0.73±0.11	0.08±0.05	0.48±0.10	0.41±0.04	0.20±0.05	0.09±0.01	
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	1.20±0.50	0.1±0.06	13.4±2.8	-23.7±0.8	—	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	—	15.9±3.3	—	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

<b>Pristine peat</b>	Brunei	52.40	1.95	31.4	-30.4±0.8	—	5.65	0.82	0.05			0.28±0.05	0.12	
<b>Disturbed peat</b>	Brunei	50.95	2.09	28.4	-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

773 References: 1. Sun S, Schefuß E, Mulitza S et al., 2017; 2. Holtvoeth J, Wagner T, Schubert C J. 2003; 3. Prasad M B K, Ramanathan A L. 2009; 4. Pradhan U K, Wu Y,  
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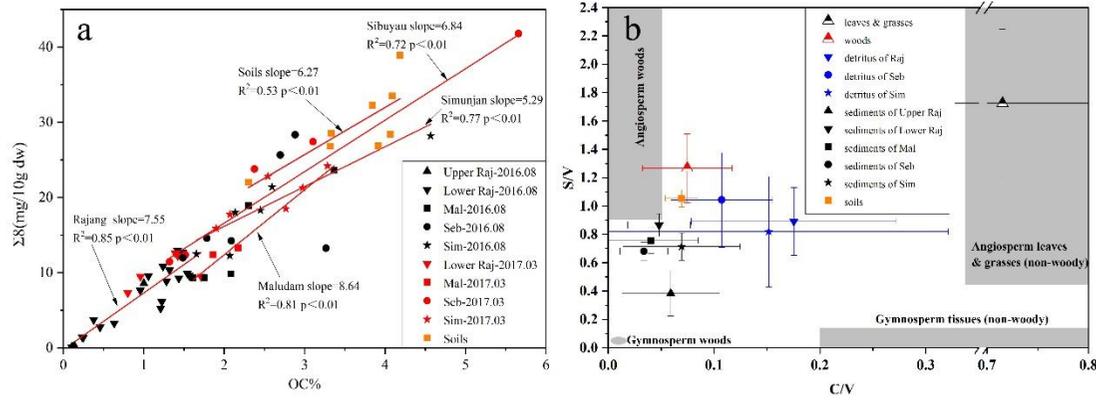
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782 Figure 1 (a) Peat and vegetation distribution in the study region (modified from <https://www.cifor.org/map/atlas/>). (b) Sediment sampling sites along the  
 783 Rajang and tributaries. The city of Sibul divides the river into upper and lower reaches. (c) Sediment sampling sites along the three small rivers. Locations of  
 784 samples collected from the Maludam, Sebuyau, and Simunjan are indicated by squares, circles, and stars, respectively.

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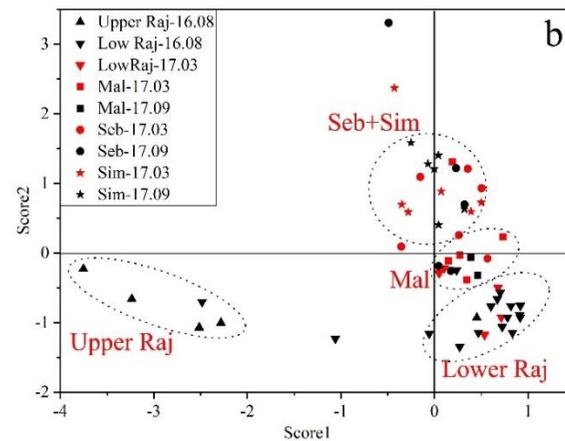
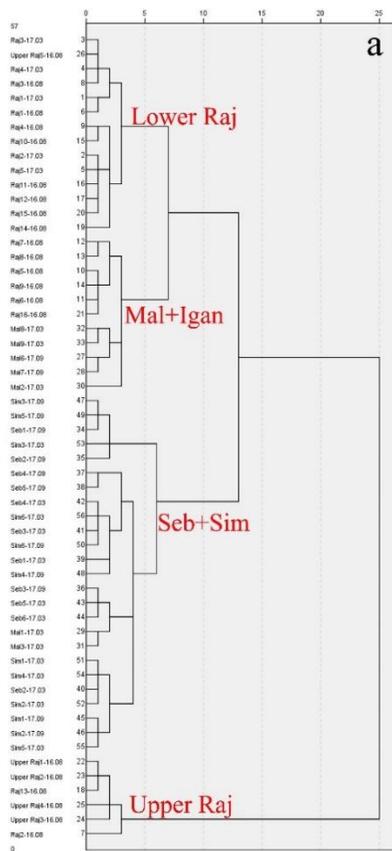
789 Figure 2 (a) Correlation of OC% with  $\Sigma 8$  among the various study systems. (b) Variations of S/V *versus* C/V of different samples from the study systems. Raj:

790 Rajang; Seb: Sebuyau; Sim: Simunjan; Mal: Maludam.

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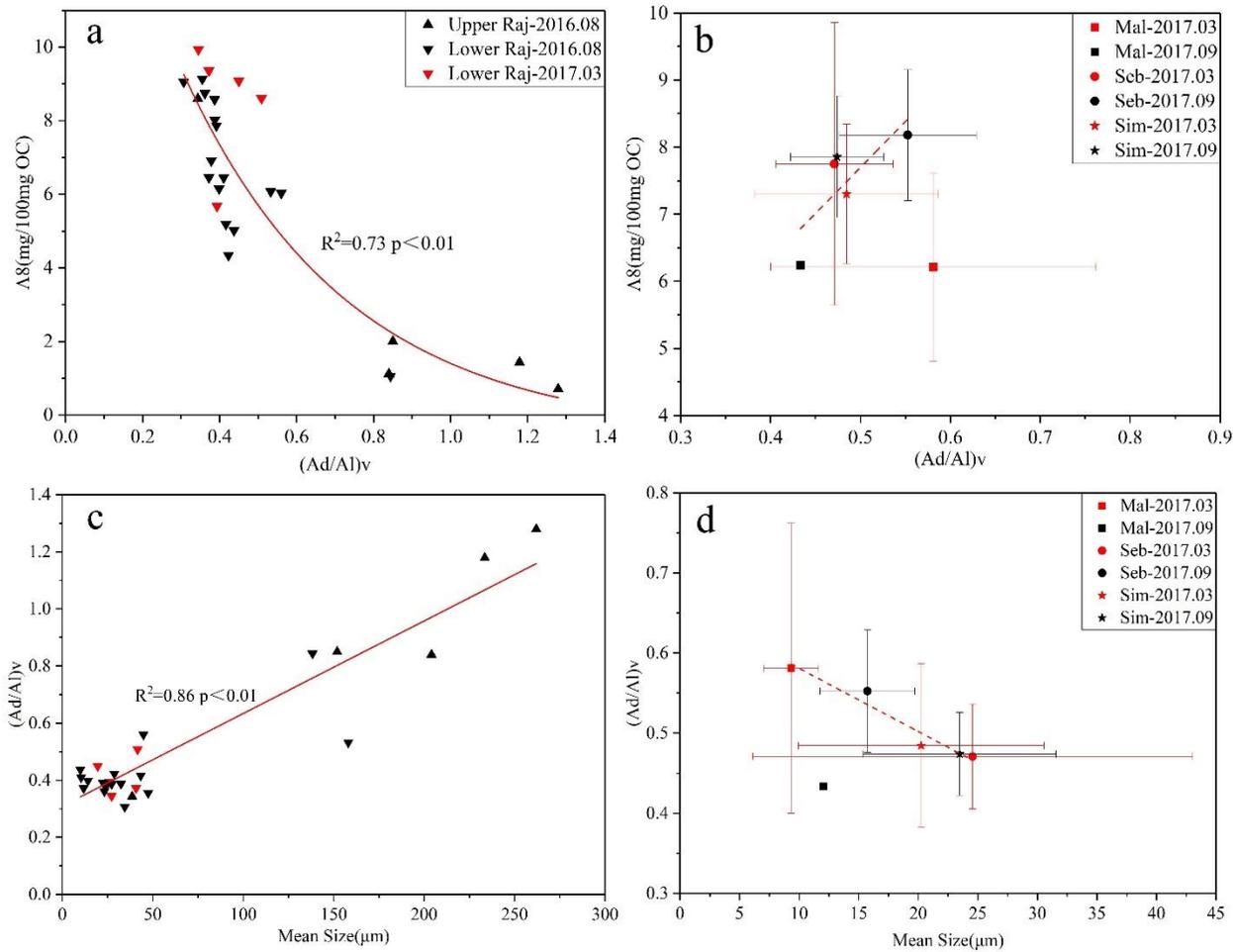
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796 Figure 3 (a) Cluster analysis of the study systems based on bulk and lignin phenols parameters. (b) Plot of PCA results based on the distribution of scores 1

797 and 2. Raj: Rajang; Seb: Sebuyau; Sim: Simunjan; Mal: Maludam.

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800 Figure 4 Variation in  $(Ad/Al)_v$  with  $\Delta 8$  values of sediments from (a) the Rajang and (b) the small river systems. Variation in  $(Ad/Al)_v$  with mean sediment grain  
 801 size for (c) the Rajang and (d) the small river systems.

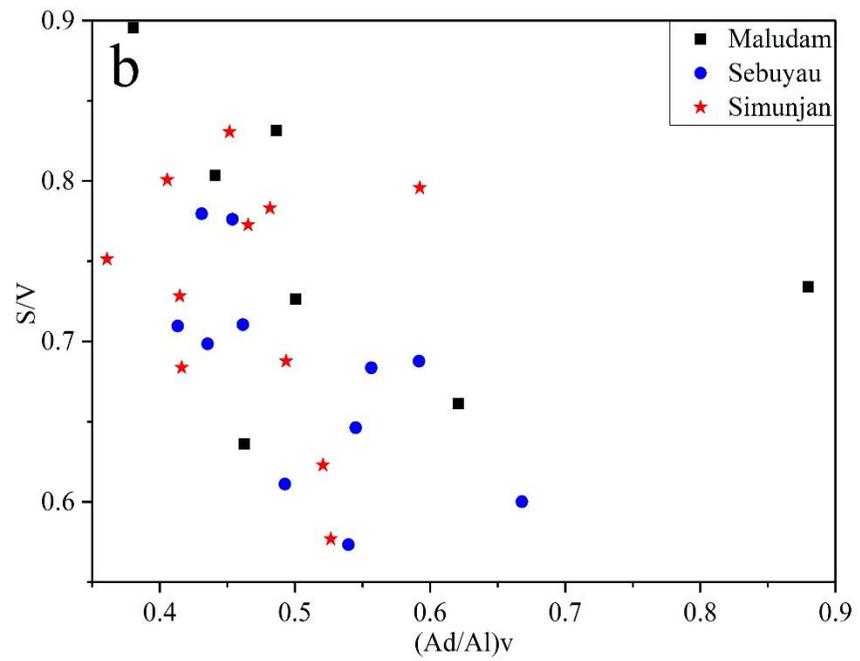
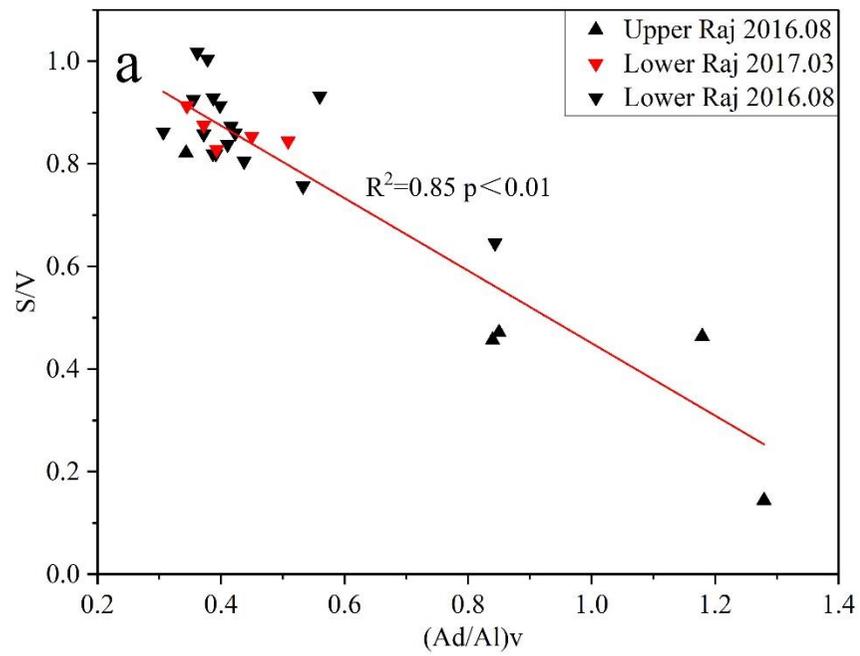


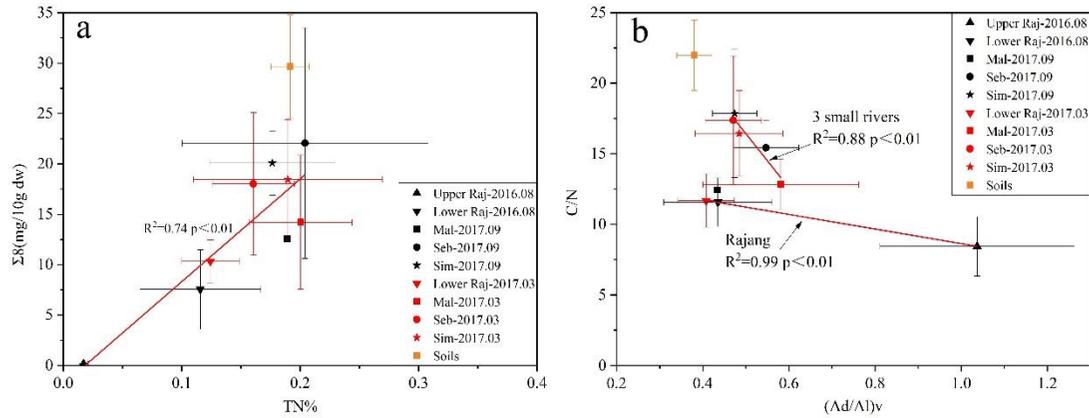
Figure 5 Relationship between  $(Ad/Al)v$  and  $S/V$  ratios based on average values of the various systems for (a) the Rajang and (b) the small river systems.

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814 Figure 6 (a) Correlation of TN% with  $\Sigma 8$  based on average values of the study systems. (b) Correlation of  $(Ad/Al)_v$  with C/N ratio based on average values of

815 the study systems.

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