# 1 Distribution and degradation of terrestrial organic matter in the

- 2 sediments of peat-draining rivers, Sarawak, Malaysian Borneo
- 4 Ying Wu<sup>1</sup>, Kun Zhu<sup>1</sup>, Jing Zhang<sup>1</sup>, Moritz Müller<sup>2</sup>, Shan Jiang<sup>1</sup>, Aazani Mujahid<sup>3</sup>, Mohd
- 5 Fakharuddin Muhamad<sup>3</sup>, Edwin Sien Aun Sia<sup>2</sup>
- 6 1 State Key Laboratory of Estuary and Coastal Research, East China Normal University,
- 7 Shanghai, China.
- 8 2 Faculty of Engineering, Computing and Science Swinburne, University of Technology,
- 9 Sarawak campus, Malaysia.
- 10 3 Faculty of Resource Science & Technology, University Malaysia Sarawak, Sarawak, Malaysia.
- 12 Correspondence,
- 13 Ying Wu, wuying@sklec.ecnu.edu.cn

14

11

# Abstract.

16

17

18

19

20

21

22

23

24

25

26

27

28

29

30

31

32

33

34

35

36

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 (the Maludam, Simunjan, and Sebuyau)) 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 phenols might explain the variations in the (Ad/Al)v ratio. Elevated (Ad/Al)<sub>V</sub> values observed from the Maludam's sediments may be also attributed to source plant variations. 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. In small rivers, a quick decline of C/N ratios responses to the slower modification of (Ad/Al)v ratio by the meant of better preservation of lignin phenols. The accumulation of lignin phenols with higher total nitrogen percentage (TN%) in the studied systems were observed. 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.

37

38

39

40

41

# 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 is derived from Southeast Asia, which equals to 11%–

14% of the total carbon pool stored in all peat. However, increasing anthropogenic disturbance 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; Goñi 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 although highly degraded soil organic matter may be devoid of any apparent lignin but as another important contributor to OCterr (Burdige, 2005; 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;

42

43

44

45

46

47

48

49

50

51

52

53

54

55

56

57

58

59

60

61

62

63

64

65

66

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., 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 proved to be 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

68

69

70

71

72

73

74

75

76

77

78

79

80

81

82

83

84

85

86

87

88

89

90

91

92

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.

100

101

102

103

104

105

106

107

108

109

110

111

112

113

114

115

116

117

118

119

94

95

96

97

98

99

# 2 Materials and methods

# 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<sup>2</sup>. 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). The drainage basin of the Maludam is about 91.4 km<sup>2</sup> and majority of the river located in the Maludam National Park with 10m thick peat (Muller et al., 2015). The other two rivers are highly human disturbed with intensive oil palm and sago plantations. For the Rajang, it is separated into two parts by Sibu Town, upper reaches mainly drain mineral soils, while down reaches develop multiple distributary channels (e.g., the lower Rajang, Serendeng, Igan; Fig. 1). These channels are also surrounded by broad peatlands. It is reported that peat greater than 1m thick covered 50% of the delta plain (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 of the Rajang is about 3600 m<sup>3</sup>/s, with peak discharge (~25,000 m³/s) observed during the northeastern monsoon season (December to March; Staub et al., 2000). However, the amount of suspended sediments delivered from the Rajang basin to the delta plain demonstrated slightly variation (2.0MT/s dry season versus 2.2 MT/s wet season) but changed substantially about the amount of sediment delivered from the delta plain to the South China Sea (Staub et al., 2000). It is estimated that the annual sediment discharge of the Rajang is 30 Mt. The turbidity maximum in the lower Rajang channels occurred during the low or reduced discharge period. It is reported that up to 24 Mt of sediment is deposited in the delta front with preserved annual sediment layers at the order of one cm thick (Staub et al., 2000). The water discharge of the Maludam is quite low, only 4.4±0.6 m³/s, from the 91.4 km<sup>2</sup> catchment (Muller et al., 2015). The river length of Maludam is 33 km. For the Sebuyau and Simunjan, river length is 58 and 54 km, respectively (Martin et al., 2018). However, hydraulic information for these two rivers is largely unknown. 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 sampled at the middle stream of river using grab samplers from a small boat at each station and then 0 - 5 cm subsamples were collected and frozen (-20°C) until they were dried for subsequent analyses in the laboratory. Soil sampling was conducted at the same time along the Rajang river bank where the sites have minimal human disturbances and short soil cores were collected and mixed in situ as one composite sample for the depth of 0-10cm by getting rid of visible roots and detritus. The vegetation of tropical peat swamp forest is dominated by trees, e.g. the Anacardiaceae, Annonaceae and Euphobiaceae etc. (Page et

120

121

122

123

124

125

126

127

128

129

130

131

132

133

134

135

136

137

138

139

140

141

142

143

144

al., 2006). Fresh, typical vegetations (listed in Table S2) were separately collected by leave, stem and roots, some detritus, which floating at the surface layer of the river were also collected for the comparison study. All 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).

152

153

154

155

156

157

158

159

160

161

162

163

164

165

166

167

168

169

170

171

151

146

147

148

149

150

#### 2.2 Chemical analyses

Prior to chemical analyses, 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<sub>2</sub>O<sub>2</sub> 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). 13C/12C 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<sub>2</sub> free Teflon-lined vessels, and digested in a microwave digestion system (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(trimethylsilyI)trifluoroacetamide (BSTFA) and trimethylchlorosilane (TMCS; 99:1) by Agilent 6890N gas chromatography (DB-1 column, FID). The lignin phenols concentration was quantified using calibration curves based on commercial standards (Sigma Aldrich). Eleven phenol monomers were extracted and categorized into five groups: syringyl (S, syringaldehyde, acetosyringone, syringic acid), vanillyl (V, vanillin, acetovanillone, vanillic acid), cinnamyl (C, pcoumaric acid, ferulic acid), p-hydroxyl (P, p-hydroxybenzaldehyde, p-hydroxyacetophenone, and p-hydroxybenziic acid), and 3,5-dihydroxy benzoic acid (DHBA). Coefficients of analytical variation associated with phenols values were <10% based on replicate analysis of the same samples. Ratios of syringyl-to-vanillyl phenols (S/V) and cinnamyl-to-vanillyl phenols (C/V) are often used to indicate the relative contribution of angiosperm and non-woody tissues versus gymnosperm wood, respectively (Hedges and Mann, 1979). Since both ratios have been found to decrease with the preferential degradation of S and C relative to V phenols, lignin phenols vegetation index (LPVI) was developed to be an alternative approach to evaluate the original of various type of vegetations (Tareq et al., 2004; Thevenot et al., 2010): Lignin phenols vegetation index (LPVI) =  $[{S(S + 1)/(V + 1) + 1} \times {C(C + 1)/(V + 1) + 1}]$ The ratio of P/(V+S) may reflect the diagenetic state of lignin when the other sources of P phenols (such as protein and tannin) are relatively constant (Dittmar and Lara 2001). The acidto-aldehyde (Ad/Al) ratios of V and S phenols are often used to indicate lignin degradation and

197

172

173

174

175

176

177

178

179

180

181

182

183

184

185

186

187

188

189

190

191

192

193

194

195

196

increases with increasing lignin oxidation (Otto and Simpson 2006).

#### 2.3 Statistical analyses

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 (including OC%, TN%, mean grain size, clay% and silt%, total lignin phenols concentrations, DHBA and the ratios of vanillic acid to vanillin ((Ad/Al)V)) 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. Errors listed in tables represent standard deviations for the analytical data. Differences and correlations were evaluated as significant at the level of p<0.01.

# 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 showed significant spatial variations, with the lowest values (2–3 mg L<sup>-1</sup>) being recorded in the Igan channel, where dense peats were observed, and the higher values (4–6 mg L<sup>-1</sup>) recorded in the other two channels. The salinity of the Simunjan indicated 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<sup>-1</sup>), with the lowest values in the three systems being around 1.4 mg

224 L<sup>-1</sup>.

225

226

227

228

229

230

231

232

233

234

235

236

237

238

239

240

241

242

243

244

245

246

247

248

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) were 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 were measured in plants samples and varied from 30%-49% (Table S2). The mean TOC value in the soil samples was 3.6±0.6% (Table S3). 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, exhibited low N content and high C/N  $(C/N = 56\pm34).$ The most abundant vegetation collected from the Maludam showed relatively depleted carbon isotope ratios ( $\delta^{13}$ C = -31‰) that were typical of C3 vegetation (Table S2). The detritus samples were also relatively depleted in  ${}^{13}$ C ( $\delta^{13}$ C = -29.2%; Table 1). The isotope ratios of the peatdraining river's sediments (average  $\delta^{13}$ C varied at -28.2 - -27.4%) were comparable with the Rajang's (average  $\delta^{13}$ C = -28.6±0.6‰) (Tab. 3). The  $\delta^{13}$ C values of the soil samples are similar to those of riverine sediments ( $\delta^{13}$ C = -28.4‰).

251

252

253

254

255

256

257

258

259

260

261

262

263

264

265

266

267

268

269

270

271

272

273

274

249

250

#### 3.2 Lignin phenols content

The lignin phenols obtained after CuO oxidation are expressed as  $\Lambda 8$  (mg (100 mg OC)<sup>-1</sup>), except for the lignin yield ( $\Sigma 8$ ), which is the sum of C + S + V and is expressed as mg 10 g 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 g dw-1). The lignin yield from the soil samples and the three small rivers (average of ~30 mg 10 g dw-1) is also higher than that from the Rajang samples (average of <10 mg 10 g dw<sup>-1</sup>), with the lowest value observed in the upper Rajang (0.16 mg 10 g dw<sup>-1</sup>; Table 2). There are correlations between  $\Sigma 8$  and OC% in each river (r<sup>2</sup> > 0.5), with the slope decreasing in the order of Maludam > Simunjan > Sebuyau > Rajang (Fig. 2a). The variation in  $\Lambda 8$  from various pools shows a similar distribution as the  $\Sigma 8$  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)<sup>-1</sup> (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)<sub>V</sub>) and syringic acid to syringaldehyde ((Ad/Al)<sub>S</sub>) increase slightly from the vegetation to river samples (Table 2). The ratios obtained from the 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) $_{\rm V}$  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) $_{\rm S}$  and (Ad/Al) $_{\rm V}$  values were recorded. The two ratios are linearly correlated in all sediment samples ( $_{\rm V}^2$  = 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 low P content in most vegetation (Table 2). However, in some plant samples (*Elaeis guineensis Jacq.*), we detected relative higher P content (Table S2). The P/(V + S) 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. 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 analyses

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 implied that factor 1 showed close correlations with the (Ad/Al)v ratio and grain size while factor 2 showed a close correlation with Σ8 and OC%.

# 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 of sediments from typical systems worldwide. Although the TOC values of our studied systems are lower than 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  $\Lambda 8$  found in our studied systems were linked to vegetation types (trees dominated) (Zaccone et al., 2008) and partially caused by peat-draining and intense human activity near the watersheds (e.g. land use change and logging activities), 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 (Ad/Al)v values of the sediments sampled here are comparable to fresh and only low to medium oxidized. Elevated (Ad/Al)v values observed from the Maludam's sediments (March, 2017) may be also attributed to source plant variations as observed in other study case (Zhu et al., 2019).

320

321

322

323

324

325

326

301

302

303

304

305

306

307

308

309

310

311

312

313

314

315

316

317

318

319

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

The depleted average  $\delta^{13}$ C values (-31.8 ~ -28.1‰) 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 cluster and PCA analyses suggest that there are no significant seasonal differences in these rivers. Previous study reported that the sediment load from the basin to the delta was no seasonal pattern, combined with comparable precipitations during our two sampling seasons, our observations matched (Martin et al., 2018; Staub et al., 2000). The close correlation of factor 2 with OC% and Σ8 in the PCA suggests factor 2 relates to the source of the organic matter (Fig. 3), as also be indicated by the strong correlation between OC% and  $\Sigma 8$  (r<sup>2</sup>: 0.53-0.85) (Fig. 2). Correlation of OC% and  $\Sigma 8$  of the Maludam ( $r^2 = 0.81$ ) show 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 (score 1 represents 45% of the total loading while score 2 is 32%) 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  $\Sigma 8$  for the Rajang (r<sup>2</sup> = 0.92) forms a distinct group separate from the small rivers  $(r^2 = 0.59)$ . 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 (Gymnosperm woods: 1, non-woody Gymnosperm tissues, 3-27; Angiosperm woods: 67-415; non Angiosperm tissues: 176-2782), which are commonly less than 60 in these sediment samples (Tareq et al., 2004). 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  $\Lambda 8$  concentrations ( $r^2 = 0.56$ );

327

328

329

330

331

332

333

334

335

336

337

338

339

340

341

342

343

344

345

346

347

348

349

350

351

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 compare with the other rivers, 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 (0.13–0.28) are higher than the average P/V ratio of wood (0.05) 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. Combined the composition of P and V in plants samples listed in Table S2, we find some dominant species, e.g. *Dipterocarpaceae, Bruguierag ymnorrhiza(L.) Poir., Elaeis guineensis Jacq.* have a relatively higher P/V ration in their non-woody parts.

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

(Ad/Al)v ratios are often used to evaluate 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 is 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 with (Ad/Al)v ratios was not so significant in the small river systems as we expected, partially resulting from the variation

of (Ad/Al)v also could be vegetation source controlled (Fig. 4b). In additional, 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 are collected from the Maludam in March 2017. The (Ad/Al)v ratios increase with increasing coarse fraction of the sediments in the Rajang, which is typically observed in other systems (Bianchi et al., 2002; Li eta I., 2015; Sun et al., 2017) (Fig. 4c). 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 September 2017) might be related to the fluvial supply of fresh vegetation. 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 is 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 (Dittmar and Kattner, 2003). However, in this study the ratio of P/(S+V) in most sediment samples did not vary greatly (~0.2). Although there was a linear correlation between the P/(S+V) and (Ad/Al)v ratios among all the sediments (r<sup>2</sup> = 0.89), no clear trend was observed for the small rivers, which may suggest both parameter's more links to source instead of

379

380

381

382

383

384

385

386

387

388

389

390

391

392

393

394

395

396

397

398

399

400

401

402

403

diagenetic process in these systems.

406

407

408

409

410

411

412

413

414

415

416

417

418

419

420

421

422

423

424

425

426

427

428

429

430

405

#### 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), since it was recently shown that lignin could decompose as fast as litter bulk carbon in mineral soils (Duboc et al., 2014). In the delta region, most parameters were quite comparable, except Σ8 and OC% (Table S1). The higher values of  $\Sigma 8$  and OC% were observed in Simunjan and Sebuyau, where land use and drainage observed. Usually land use and drainage of tropical peat will accelerate the loss of vegetation and OC degradation (Kononen, et al., 2016), here it may be explained by the high content of OC and lignin in oil palm, which is the major plantation in both regions. In this study, the OC content increases with decreasing grain size, implying that fine sediments, with larger specific surface areas and rich in clay, contain more OM than coarser sediments, as reported previously (Sun et al., 2017). Increasing (Ad/Al)v values are 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  $\Sigma$ 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 September, 2017) could contain 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  $\Sigma 8$  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 (microbial process, logging etc.) working on OCterr in these two systems, which cause postdepositional 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, since nitrogen content tends to stimulates decomposition of low-lignin litter by decomposer microbes, but usually decrease the activity of lignolytic enzymes and inhibit decomposition of high-lignin litter (Knorr, et al., 2005; Thevenot et al., 2010). In our study, we found a higher TN% in the small rivers compared with the Rajang. A significant correlation between  $\Sigma 8$  and TN% ( $r^2$ = 0.74) is observed in all systems, which might suggest a contribution from plant litter affecting both parameters (Fig. 6a). 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). Quicker decline of C/N ratios related to slower lignin degradation in small rivers, this could be related 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 higher lignin phenols with moderate degraded characteristics was observed in the small river systems in which higher TN% were recorded.

431

432

433

434

435

436

437

438

439

440

441

442

443

444

445

446

447

448

449

450

451

452

453

454

455

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 ( $\Sigma 8$ ) 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 exceptional data were collected during September 2017, which was a time of saline water

# **5 Conclusions**

the Rajang systems, as proposed by Gandois et al. (2014).

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 demonstrate 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 show no distinct seasonal 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 higher lignin phenols with moderate degraded process observed in the small river systems where a higher TN% is 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. 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.

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.

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

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

Special issue statement. This article is part of the special issue "Biogeochemical processes in highly dynamic peat-draining rivers and estuaries in Borneo". It is not associated with a conference.

512

513

514

515

516

517

518

519

520

521

522

523

509

510

511

# Acknowledgements

The present research was kindly supported by the Newton-Ungku Omar Fund (NE/P020283/1), the Natural Science Foundation of China (41530960), China Postdoctoral Science Foundation (2018M630416), MOHE FRGS 15 Grant (FRGS/1/2015/WAB08/SWIN/02/1) and the SKLEC Open Research Fund (SKLEC-KF201610). The authors would like to thank the Sarawak Forestry Department and Sarawak Biodiversity Centre for permission to conduct collaborative research in Sarawak waters under permit numbers NPW.907.4.4 (Jld.14)-161, Park Permit No WL83/2017, and SBC-RA-0097-MM. Lukas Chin and the *SeaWonder* crew are acknowledged for their support during the cruises. Dr. Zhuoyi Zhu, Ms. Lijun Qi, and the Marine Biogeochemistry Group are especially acknowledged for their contribution and support during the sampling trips and laboratory analysis.

524

525

#### References

- 526 Alin, S. R., Aalto, R., Goñi, M. A., Richey, J. E., and Dietrich, W. E.: Biogeochemical
- 527 characterization of carbon sources in the Strickland and Fly rivers, Papua New Guinea, J. of
- 528 Geophy. Res.: Earth Surface, 113, https://doi:10.1029/2006JF000625, 2008.
- 529 Alkhatib, M., Jennerjahn, T C., Samiaji, J.: Biogeochemistry of the Dumai River estuary,
- 530 Sumatra, Indonesia, a tropical black-water river, Limnol. & Oceanogr., 52, 2410-2417,
- 531 https://doi.org/10.4319/lo.2007.52.6.2410, 2007.
- 532 Anderson, J. A. R.: The tropical peat swamps of western Malesia, Mires: swamp, bog, fen and
- 533 moor: regional studies, 1983.
- Aufdenkampe, A. K., Mayorga, E., Hedges, J. I., Llerena, C., Quay, P. D., Gudeman, J.,

- Krusche, A. V., and Richey, J. E.: Organic matter in the Peruvian headwaters of the Amazon:
- 536 Compositional evolution from the Andes to the lowland Amazon mainstem, Org. Geochem., 38,
- 537 337-364, https://doi.org/10.1016/j.orggeochem.2006.06.003, 2007.
- Aufdenkampe, A. K., Mayorga, E., Raymond, P. A., Melack, J. M., Doney, S. C., Alin, S. R.,
- 539 Aalto, R. E., and Yoo, K.: Riverine coupling of biogeochemical cycles between land, oceans,
- and atmosphere, Front. in Ecol. & the Environ., 9, 53-60, https://doi.org/10.189-0/100014, 2011.
- Bao, H., Lee, T. Y., Huang, J. C., Feng, X., Dai, M., and Kao, S. J.: Importance of Oceanian
- 542 small mountainous rivers (SMRs) in global land-to-ocean output of lignin and modern biospheric
- 543 carbon, Sci. Rep., 5, 16217, https://doi.org/10.1038/srep16217, 2015.
- Battin, T. J., Luyssaert, S., Kaplan, L. A., Aufdenkampe, A. K., Richter, A., and Tranvik, L. J.:
- The boundless carbon cycle, Nat. Geosci., 2, 598-600, https://doi.org/10.1038/ngeo618, 2009.
- 546 Baum, A., Rixen, T., and Samiaji, J.: Relevance of peat draining rivers in central Sumatra for
- the riverine input of dissolved organic carbon into the ocean, Estuar. Coast. Shelf Sci., 73, 563-
- 548 570, https://doi.org/10.1016/j.ecss.2007.02.012, 2007.
- Benner. R., Maccubbin, A. E., and Hodson, R. E.: Anaerobic biodegradation of the lignin and
- 550 polysaccharide components of lignocellulose and synthetic lignin by sediment microflora, Appl.
- 551 Environ. Microbiol., 47, 998-1004, 1984.
- Benner. R., Fogel, M. L., Sprague, E. K., and Hodson, R. E.: Depletion of 13C in lignin and its
- implications for stable carbon isotope studies, Nature, 329, 708, 1987.
- Bianchi, T. S., Mitra, S., and McKee, B. A.: Sources of terrestrially-derived organic carbon in
- 555 lower Mississippi River and Louisiana shelf sediments: implications for differential
- sedimentation and transport at the coastal margin, Mar. Chem., 77, 211-223, https://doi.org/-
- 557 10.1016/S0304-4203(01)00088-3, 2002.
- 558 Bianchi, T. S., Bauer, J. E.: 5.03—particulate organic carbon cycling and transformation,
- Treatise on estuarine and coastal science, 5, 69-117, 2011.
- 560 Burdige, D. J.: Burial of terrestrial organic matter in marine sediments: A re-assessment, Glob.

- 561 Biogeochem. Cyc., 19(4), 2005.
- Cook, S., Peacock, M., Evans, C. D., Page, S. E., Whelan, M. J., Gauci, V., and Kho, L. K.:
- Quantifying tropical peatland dissolved organic carbon (DOC) using UV-visible spectroscopy,
- 564 Water Res., 115, 229-235, https://doi.org/10.1016/j.watres.2017.02.059, 2017.
- 565 Dignac, M. F., Knicker, H., Kögel-Knabner, I.: Effect of N content and soil texture on the
- 566 decomposition of organic matter in forest soils as revealed by solid-state CPMAS NMR
- 567 spectroscopy, Org. Geochem., 33, 1715-1726, 2002.
- Dittmar, T., Lara, R. J.: Molecular evidence for lignin degradation in sulfate-reducing mangrove
- 569 sediments (Amazonia, Brazil). Geochimi. Cosmochim. Acta, 65, 1417-1428, 2001.
- 570 Dittmar, T., Kattner, G.: The biogeochemistry of the river and shelf ecosystem of the Arctic
- 571 Ocean: a review, Mar. chem., 83, 103-120, https://doi.org/10.1016/S0304-4203 (03)00105-1,
- 572 2003.
- 573 Dommain, R., Couwenberg, J., Glaser, P. H., Joosten, H., and Suryadiputra, I. N. N.: Carbon
- 574 storage and release in Indonesian peatlands since the last deglaciation, Quaternary Sci. Rev.,
- 575 97, 1-32, https://doi.org/10.1016/j.quascirev.2014.05.002, 2014.
- 576 Drenzek, N. J., Montluçon, D. B., Yunker, M. B., Macdonald, R. W., and Eglinton, T. I.:
- 577 Constraints on the origin of sedimentary organic carbon in the Beaufort Sea from coupled
- 578 molecular <sup>13</sup>C and <sup>14</sup>C measurements, Mar. Chem., 103, 146-162, https://doi.org/10.1016/j.-
- 579 marchem.2006.06.017, 2007.
- Duboc, O., Dignac, M. F., Djukic, I., Zehetner, F., Gerzabek, M. H., & Rumpel, C.: Lignin
- decomposition along an Alpine elevation gradient in relation to physicochemical and soil
- 582 microbial parameters. Glob. Cha. Biol., 20(7), 2272-2285, 2014.
- Feng, X., Feakins, S. J., Liu, Z., Ponton, C., Wang, R. Z., Karkabi, E., Galy, V., Berelson, W.
- 584 M., Nottingham, A. T., Meir, P., and West, A. J.: Source to sink: Evolution of lignin composition
- in the Madre de Dios River system with connection to the Amazon basin and offshore, J. of
- 586 Geophys. Res.-Biogeo., 121, 1316-1338, https://doi.org/10.1002/2016jg0033-23, 2016.

- Fog, K.: The effect of added nitrogen on the rate of decomposition of organic matter, Biol. Rev.,
- 588 63, 433-462, 1988.
- 589 Gandois, L., Cobb, A. R., Hei, I. C., Lim, L. B. L., Salim, K. A., and Harvey, C. F.: Impact of
- 590 deforestation on solid and dissolved organic matter characteristics of tropical peat forests:
- implications for carbon release, Biogeochem., 114, 183-199, 2013.
- 592 Gandois, L., Teisserenc, R., Cobb, A. R., Chieng, H. I., Lim, L. B. L., Kamariah, A. S., Hoyt, A.,
- 593 and Harvey, C. F.: Origin, composition, and transformation of dissolved organic matter in
- tropical peatlands, Geochimi. Cosmochim. Acta, 137, 35-47, https://doi.org/ 10.1016/j.gca-
- 595 .2014.03.012, 2014.
- 596 Goldsmith, S. T., Carey, A. E., Lyons, W. B., Kao, S.-J., Lee, T. Y., and Chen, J.: Extreme storm
- events, landscape denudation, and carbon sequestration: Typhoon Mindulle, Choshui River,
- 598 Taiwan, Geology, 36, https://doi.org/10.1130/g24624a.1, 2008.
- 599 Goñi, M. A., Hedges, J. I.: Sources and reactivities of marine-derived organic matter in coastal
- sediments as determined by alkaline CuO oxidation, Geochimi. Cosmochim. Acta, 59, 2965-
- 601 2981, 1995.
- 602 Goñi, M. A., Ruttenberg, K. C., Eglinton, T. I.: A reassessment of the sources and importance
- of land-derived organic matter in surface sediments from the Gulf of Mexico, Geochimi.
- 604 Cosmochim. Acta, 62, 3055-3075, 1998.
- Goñi, M. A., Montgomery, S.: Alkaline CuO oxidation with a microwave digestion system: Lignin
- analyses of geochemical samples, Anal. Chem., 72 3116-3121, 2000.
- Goñi, M. A., Yunker, M. B., Macdonald, R. W., and Eglinton, T. I.: The supply and preservation
- of ancient and modern components of organic carbon in the Canadian Beaufort Shelf of the
- 609 Arctic Ocean, Mar. Chem., 93, 53-73, https://doi.org/10.1016/j.marchem.2004.08.001, 2005.
- Goni, M. A., Monacci, N., Gisewhite, R., Ogston, A., Crockett, J., and Nittrouer, C.: Distribution
- and sources of particulate organic matter in the water column and sediments of the Fly River
- 612 Delta, Gulf of Papua (Papua New Guinea), Estuar. Coast. Shelf Sci., 69, 225-245,

- 613 https://doi.org/10.1016/j.ecss.2006.04.012, 2006.
- Hall, S.J., Silver, W., Timokhin, V. I., Hammel, K. E.: Lignin decompositions is sustained under
- 615 fluctuating redox conditions in humid-tropical forest soils, Glob. Chan. Biol., 21, 2818-2828,
- 616 2015.
- 617 Hedges, J. I., Mann, D. C.: The characterization of plant tissues by their lignin oxidation
- 618 products, Geochimi. Cosmochim. Acta, 43, 1803-1807, 1979.
- Hedges, J. I., Ertel, J. R.: Characterization of lignin by gas capillary chromatography of cupric
- oxide oxidation products, Anal. Chem., 54, 174-178, 1982.
- Hedges, J. I., Cowie, G. L., Ertel, J. R., Barbour, R. J., and Hatcher, P. G.: Degradation of
- 622 carbohydrates and lignins in buried woods, Geochimi. Cosmochim. Acta, 49, 701-711, 1985.
- Hedges, J. I., Clark, W. A., Quay, P. D., Richey, J. E., Devol, A. H., and Santos, M.:
- 624 Compositions and fluxes of particulate organic material in the Amazon River<sup>1</sup>, Limnol. &
- 625 Oceanogr., 31, 717-738, 1986.
- Hedges, J. I., Blanchette, R. A., Weliky, K., and Devol, A. H.: Effects of fungal degradation on
- the CuO oxidation products of lignin: a controlled laboratory study, Geochimi. Cosmochim. Acta,
- 628 52, 2717-2726, 1988.
- Hernes, P. J., Benner, R.: Transport and diagenesis of dissolved and particulate terrigenous
- organic matter in the North Pacific Ocean, Deep Sea Res. Part I, 49(12), 2119-2132, 2002.
- Hernes, P. J., Robinson, A. C., and Aufdenkampe, A. K.: Fractionation of lignin during leaching
- and sorption and implications for organic matter "freshness", Geophys. Res. Lett., 34,
- 633 https://doi.org/10.1029/2007gl031017, 2007.
- Hernes, P. J., Dyda, R. Y., and McDowell, W. H.: Connecting tropical river DOM and POM to
- 635 the landscape with lignin, Geochimi. Cosmochim. Acta, 219, 143-159,
- 636 https://doi.org/10.1002/2017JG003935, 2017.
- Holtvoeth, J., Wagner, T., Schubert, C. J.: Organic matter in river-influenced continental margin
- 638 sediments: The land-ocean and climate linkage at the Late Quaternary Congo fan (ODP Site

- 639 1075), Geochem., Geophy., Geosys., 4(12), 1109, 2003.
- Hooijer, A., Page, S., Jauhiainen, J., Lee, W. A., Lu, X. X., Idris, A., and Anshari, G.: Subsidence
- and carbon loss in drained tropical peatlands, Biogeosciences, 9, 1053-1071,
- 642 https://doi.org/10.5194/bg-9-1053-2012, 2012.
- 643 Hoscilo, A., Page, S. E., Tansey, K. J., and Rieley, J. O.: Effect of repeated fires on land-cover
- change on peatland in southern Central Kalimantan, Indonesia, from 1973 to 2005, Inter. J. of
- 645 Wildland Fire, 20, 578-588, 2011.
- Houghton, R. A., Skole, D. L., Nobre, C. A., Hackler, J. L., Lawrence, K. T., and Chomentowski,
- 647 W. H.: Annual fluxes of carbon from deforestation and regrowth in the Brazilian Amazon, Nature,
- 648 403, 301, 2000.
- Jennerjahn, T. C., Ittekkot, V., Klöpper, S., Adi, S., Purwo Nugroho, S., Sudiana, N., Yusmal,
- 650 A., Prihartanto, and Gaye-Haake, B.: Biogeochemistry of a tropical river affected by human
- activities in its catchment: Brantas River estuary and coastal waters of Madura Strait, Java,
- 652 Indonesia, Estuar. Coast. Shelf Sci., 60, 503-514, https://doi.org/10.1016/j.ecss.2004.02.008,
- 653 2004.
- Jennerjahn, T. C., Soman, K., Ittekkot, V., Nordhaus, I., Sooraj, S., Priya, R. S., and Lahajnar,
- N.: Effect of land use on the biogeochemistry of dissolved nutrients and suspended and
- 656 sedimentary organic matter in the tropical Kallada River and Ashtamudi estuary, Kerala, India,
- 657 Biogeochem., 90, 29-47, https://doi.org/10.1007/s10533-008-9228-1, 2008.
- Jex, C. N., Pate, G. H., Blyth, A. J., Spencer, R. G. M., Hernes, P. J., Khan, S. J., and Baker,
- A.: Lignin biogeochemistry: from modern processes to Quaternary archives, Quarter. Sci. Rev.,
- 87, 46-59, https://doi.org/10.1016/j.quascirev.2013.12.028, 2014.
- Kao, S. J., Liu, K. K.: Particulate organic carbon export from a subtropical mountainous river
- 662 (Lanyang Hsi) in Taiwan, Limnol. & Oceanogr., 41, 1749-1757, 1996.
- Keil, R. G., Tsamakis, E., Giddings, J. C., and Hedges, J. I.: Biochemical distributions (amino
- acids, neutral sugars, and lignin phenols) among size-classes of modern marine sediments

- from the Washington coast, Geochimi. Cosmochim. Acta, 62, 1347-1364, 1998.
- 666 Killops, S., Killops, V.: Introduction to Organic Geochemistry, 2nd edition (paperback),
- 667 Geofluids, 5, 236-237, 2005.
- 668 Knorr, M., Frey, S.D. and Curtis, P.S.: Nitrogen additions and litter decomposition: A meta-
- analysis. Ecology 86, 3252–3257, 2005.
- Koh, L. P., Butler, R. A., and Bradshaw, C. J. A.: Conversion of Indonesia's peatlands, Front.
- 671 in Ecol. & the Environ., 7, 238-238, https://doi.org/10.1890/09.WB.013, 2009.
- Kononen, M., Jauhiainen, J., Laiho, R., Spetz, P., Kusin, K., Limin, S., Vasander, H.: Land use
- increases the recalcitrance of tropical peat, Wetlands Ecol. Manage, 24, 717–731, 2016.
- 674 Li, Z., Peterse, F., Wu, Y., Bao, H. Y., Eglinton, T. I., Zhang, J.: Sources of organic matter in
- 675 Changjiang (Yangtze River) bed sediments: preliminary insights from organic geochemical
- 676 proxies, Org. Geochem., 85, 11-21, 2015.
- Loh, P. S., Chen, C. T. A., Anshari, G. Z., Wang, J.T., Lou, J. Y., Wang, S.L.: A comprehensive
- 678 survey of lignin geochemistry in the sedimentary organic matter along the Kapuas River (West
- 679 Kalimantan, Indonesia), J. of Asian Ear. Sci., 43(1), 118-129, 2012.
- 680 Lourençato, L. F., Bernardes, M. C., Buch, A. C., and Silva-Filho, E. V.: Lignin phenols in the
- paleoenvironmental reconstruction of high mountain peatlands from Atlantic Rainforest, SE-
- 682 Brazil, Catena, 172, 509-515, https://doi.org/10.1016/j.catena.2018.09.013, 2019.
- Martin, P., Cherukuru, N., Tan, A. S. Y., Sanwlani, N., Mujahid, A., and Müller, M.: Distribution
- and cycling of terrigenous dissolved organic carbon in peatland-draining rivers and coastal
- waters of Sarawak, Borneo, Biogeosciences, 15, 6847-6865, https://doi.org/10.5194/bg-15-
- 686 6847-2018, 2018.
- 687 Miettinen, J., Shi, C., and Liew, S. C.: Land cover distribution in the peatlands of Peninsular
- 688 Malaysia, Sumatra and Borneo in 2015 with changes since 1990, Glob. Ecol. Conserv., 6, 67-
- 78, https://doi.org/10.1016/j.gecco.2016.02.004, 2016.
- 690 Milliman, J. D., Farnsworth, K. L.: Runoff, erosion, and delivery to the coastal ocean, River

- discharge to the coastal ocean: a global synthesis, Cambridge University Press, Cambridge,
- 692 UK, 13-69, 2011.
- 693 Moore, S., Gauci, V., Evans, C. D., and Page, S. E.: Fluvial organic carbon losses from a
- Bornean blackwater river, Biogeosciences, 8, 901-909, https://doi.org/10.5194/bg-8-901-2011,
- 695 2011.
- Moore, S., Evans, C. D., Page, S. E., Garnett, M. H., Jones, T. G., Freeman, C., Hooijer, A.,
- 697 Wiltshire, A. J., Limin, S. H., and Gauci, V.: Deep instability of deforested tropical peatlands
- 698 revealed by fluvial organic carbon fluxes, Nature, 493, 660-663,
- 699 https://doi.org/10.1038/nature11818, 2013.
- Müller, D., Warneke, T., Rixen, T., Muller, M., Jamahari, S., Denis, N., ... & Notholt, J.: Lateral
- 701 carbon fluxes and CO 2 outgassing from a tropical peat-draining river. Biogeosciences, 12(20),
- 5967-5979, 2015. Opsahl, S., Benner, R.: Early diagenesis of vascular plant tissues: lignin and
- cutin decomposition and biogeochemical implications, Geochimi. Cosmochim. Acta, 59, 4889-
- 704 4904, 1995.
- 705 Osono, T., Takeda, H.: Organic chemical and nutrient dynamics in decomposing beech leaf
- 706 litter in relation to fungal ingrowth and succession during 3-year decomposition processes in a
- cool temperate deciduous forest in Japan, Ecol. Res., 16, 649-670, 2001.
- 708 Otto, A., Simpson, M.J.: Evaluation of CuO oxidation parameters for determining the source
- and stage of lignin degradation in soil, Biogeochem., 80, 121–142, 2006.
- 710 Page, S.E., Reiley, J.O., and Wust, R.: Lowland tropical peatland of Southeast Asia. In:
- 711 Peatlands: Evolution and Records of Environmental and Climate Changes (eds., by Martini,
- 712 I.P., etc.) Elsevier, pp145-171, 2006.
- Page, S. E., Morrison, R., Malins, C., Hooijer, A., Rieley, J. O., and Jauhiainen, J.: Review of
- 714 peat surface greenhouse gas emissions from oil palm plantations in Southeast Asia, White
- 715 paper, 76, 2011.
- 716 Pradhan, U. K., Shirodkar, P., and Sahu, B.: Physico-chemical characteristics of the coastal

- 717 water off Devi estuary, Orissa and evaluation of its seasonal changes using chemometric
- 718 techniques, Curr. Sci., 96(9), 1203-1209, http://www.jstor.org/stable/24105409, 2009.
- 719 Pradhan, U. K., Wu, Y., Shirodkar, P. V., Zhang, J., and Zhang, G.: Sources and distribution of
- 720 organic matter in thirty five tropical estuaries along the west coast of India-a preliminary
- 721 assessment, Estuar. Coast. Shelf Sci., 151, 21-33, https://doi.org/10.1016/j.ecss.2014.09.010,
- 722 2014.
- 723 Prasad, M. B. K., Ramanathan, A. L.: Organic matter characterization in a tropical estuarine-
- 724 mangrove ecosystem of India: preliminary assessment by using stable isotopes and lignin
- 725 phenols, Estu., Coast. &Shel. Sci., 84(4), 617-624, 2009.
- 726 Rieley, J. O., Page, S. E., Setiadi, B.: Distribution of peatlands in Indonesia, Glob. Peat Resour.,
- 727 169-178, 1996.
- 728 Rieley, J. O., Wüst, R. A. J., Jauhiainen, J., Page, S. E., Wösten, J. H. M., Hooijer, A., ... and
- 729 Stahlhut, M.: Tropical peatlands: carbon stores, carbon gas emissions and contribution to
- 730 climate change processes, Peatlands and climate change, Inter.Peat Society, 148-181, 2008.
- 731 Spencer, R. G. M., Hernes, P. J., Ruf, R., Baker, A., Dyda, R. Y., Stubbins, A., and Six, J.:
- 732 Temporal controls on dissolved organic matter and lignin biogeochemistry in a pristine tropical
- 733 river, Democratic Republic of Congo, J. of Geophys. Res.-Biogeo., 115,
- 734 https://doi.org/10.1029/2009jg001180, 2010.
- 735 Staub, J. R., Gastaldo, R. A.: Seasonal sediment transport and deposition in the Rajang River
- delta, Sarawak, East Malaysia, Sci. Sediment. Geol., 133, 249-264, 2000.
- Sun, S., Schefuß, E., Mulitza, S., Chiessi, C. M., Sawakuchi, A. O., Zabel, M., Baker, P. A.,
- Helter, J., Mollenhauer, G.: Origin and processing of terrestrial organic carbon in the Amazon
- 739 system: lignin phenols in river, shelf, and fan sediments, Biogeosciences, 14(9), 2495-2512,
- 740 2017.
- Tareq, S. M., Tanaka, N., and Ohta, K.: Biomarker signature in tropical wetland: lignin phenol
- 742 vegetation index (LPVI) and its implications for reconstructing the paleoenvironment, Sci. Total

- 743 Environ., 324, 91-103, https://doi.org/10.1016/j.scitotenv.2003.10.020, 2004.
- 744 Tesi, T., Semiletov, I., Dudarev, O., Andersson, A., and Gustafsson, Ö.: Matrix association
- 745 effects on hydrodynamic sorting and degradation of terrestrial organic matter during cross-shelf
- transport in the Laptev and East Siberian shelf seas, J. of Geophys. Res.-Biogeo., 121, 731-
- 747 752, 2016.
- 748 Thevenot, M., Dignac, M. F., and Rumpel, C.: Fate of lignins in soils: A review, Soil Biol. &
- 749 Biochem., 42, 1200-1211, https://doi.org/10.1016/j.soilbio.2010.03.017, 2010.
- Ward, N. D., Keil, R. G., Medeiros, P. M., Brito, D. C., Cunha, A. C., Dittmar, T., Yager, P. L.,
- 751 Krusche, A. V., and Richey, J. E.: Degradation of terrestrially derived macromolecules in the
- 752 Amazon River, Nat. Geosci., 6, 530-533, https://doi.org/10.1038/ngeo1817, 2013.
- 753 Winterfeld, M., Goñi, M. A., Just, J., Hefter, J., and Mollenhauer, G.: Characterization of
- 754 particulate organic matter in the Lena River delta and adjacent nearshore zone, NE Siberia –
- 755 Part 2: Lignin-derived phenol compositions, Biogeosciences, 12, 2261-2283, https://doi.org-
- 756 /10.5194/bg-12-2261-2015, 2015.
- 757 Wit, F., Muller, D., Baum, A., Warneke, T., Pranowo, W. S., Muller, M., and Rixen, T.: The
- 758 impact of disturbed peatlands on river outgassing in Southeast Asia, Nat. Commun, 6, 10155,
- 759 https://doi.org/10.1038/ncomms10155, 2015.
- 760 Wu, Y., Bao. H., Yu, H., and Kattner G.: Temporal variability of particulate organic carbon in the
- 761 lower Changjiang (Yangtze River) in the post-Three Gorges Dam period: Links to
- anthropogenic and climate impacts, J. of Geophys. Res.-Biogeo., 120, 2194-2211, https://do-
- 763 i.org/10.1016/j.orggeochem.2015.04.006, 2015.
- Wu, Y., Eglinton, T. I., Zhang. J., and Montlucon, D. B.: Spatiotemporal variation of the quality,
- origin, and age of particulate organic matter transported by the Yangtze River (Changjiang), J.
- 766 of Geophys. Res.- Biogeo., 123, 2908-2921, https://doi.org/10.1029/2017JG004285, 2018.
- 767 Yu, H., Wu, Y., Zhang, J., Deng, B., and Zhu, Z.: Impact of extreme drought and the Three
- 768 Gorges Dam on transport of particulate terrestrial organic carbon in the Changjiang (Yangtze)

- 769 River, J. of Geophys. Res.- Ear. Surf., 116, https://doi.org/116,10.1029/20-11jf002012, 2011.
- Zaccone, C., Said-Pullicino, D., Gigliotti, G., Miano, T.M.: Diagenetic trends in the phenolic
- 771 constituents of Sphagnum-dominated peat and its corresponding humic acid fraction, Org.
- 772 Geochem., 39, 830-838, 2008.
- Zhu, S., Dai, G., MA, T., Chen, L., Chen, D., Lu, X....Feng X.J.: Distribution of lignin phenols in
- comparison with plant-derived lipids in the alpine versus temperate grasslands soils, Plant and
- 775 Soil, 1-14, 2019.

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

Comples	Time	Mean Size	Clay%	Silt%	DO	"U	Salinity	OC	TN	Atomic C/N	δ <sup>13</sup> C	
Samples	Hine	(µm)	Clay 70	SH1 70	(mg/L)	pН	(‰)	(%)	(%)	Atomic C/13	(‰)	
Angiosperm												
leaves & grasses	03/2017	_	_		_	_	_	48.53±2.86	1.65±0.64	40.44±18.95	-31.1±2.5	
(n=10)												
Angiosperm	03/2017	_	_		_	_	_	46.71±4.71	0.52±0.19	117.00±45.32	-31.8±2.3	
woods(n=5)	03/2017							40./1±4./1	0.52±0.17	117.00=+3.32	-31.0±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	08/2016							40.76±13.69	0.94±0.35	47.21±13.03	-29.9±2.1	
detritus(n=8)	06/2010	_	_	_	_	_	_	40.70±13.0>	U.94±u.33	47.21213.03	-29.9±2.1	
Sebuyau	03/2017	_	_	_	_	_	_	30.63±15.00	0.73±0.20	53.39±31.68	-28.1±2.0	
detritus(n=5)	03/2017							30.03±13.00	0.75±0.20	33.37±31.00	-20.1±2.0	
Simunjan	03/2017	_	_		_	_	_	33.46±8.46	1.09±0.35	43.44±29.73	-29.9±0.7	
detritus(n=4)												
Soil(n=8)	09/2017	_	_	_	_	_	_	3.63±0.63	0.19±0.02	21.98±2.50	-28.4±0.2	
Upper Rajang	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	
(n=4)	00/2010	212.7=17.0	).1±±.0	10.7_0.0	T.JJ_1.12	0.74_0.00	Ü	0.12_0.02	0.02_0.00	0.77.2.10	20.120.5	
Lower Rajang	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	
(n=16)	00,200		0210		0.0.	,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,	20	****	V.11	*****	20.0	
Lower Rajang	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	
(n=5)					***	*****	**		*			
Maludam	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	
(n=5)												
Maludam	09/2017	12.1	39.2	58.3	4.96	6.69	11.5	2.02	0.19	12.43	-28.2	
(n=2)												
Sebuyau	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	
(n=6)												
Sebuyau	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	
(n=5)												
Simunjan	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	
(n=6)												
Simunjan	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	
( <b>n=6</b> )											l	

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	Λ8	V	c	_	s/v	c/v	(Ad/Al)v	(Ad/Al)s	P/(V+S)	DHBA	DHBA/V	LPVI
Samples	Time	(mg/10 g dw)	(mg/100 mg OC)	V	S	С	3/ V	C/V	(Au/Ai)v	(Au/Ai)s	P/(V+3)	ИПВА	DIIDAY	LFVI
Angiosperm leaves &														
grasses	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
(n=10)														
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	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
(n=6)	•													
Sebuyau	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
(n=5)	03/201/	22.00±11.44	0.1010.50	4.03±0.00	3.1020.43	0.1710.11	0.00_0.00	0.00.05	0.0020.00	0.0220.12				
Simunjan	03/2017	18.45±5.96	7.30±1.04	4 02±0 E1	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	40+24
(n=6)	03/201/	18.45±5.96	7.3UII.U4	4.03±0.51	2.90±0.00	0.51±0.17	0.73±0.11	0.08±0.05	0.48±0.10	0.41±0.04	0.20±0.05	U.30±U.U5	0.09±0.01	49±24
Simunjan	00/2017	20.00+2.20	7.06+0.04	4.5.4.0.00	2.00+0.24	0.22.0.20	0.6010.00	0.00.00.00	0.47.0.05	0.2610.00	0.47.0.03	0.3710.00	0.0010.03	44.22
(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

786 Table 3 Comparison of bulk and lignin phenols parameters among river systems worldwide787

Samples	Station	OC (%)	TN (%)	C/N	δ <sup>13</sup> 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	0.80~4.20 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
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	research
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
Pristine peat	Brunei	52.40	1.95	31.4	-30.4±0.8		5.65	0.82	0.05			0.28±0.05	0.12	
Disturbed peat	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
788	8 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,													
789	Shirodkar P V et al., 2014; 5.Loh P S, Chen C T A, Anshari G Z et al., 2012; 6. Li Z, Peterse F, Wu Y et al., 2015; 7. Bianchi T S, Mitra S, McKee B A. 2002; 8. Winterfeld M,													
790	Goñi M, Just .	J et al., 2015	; 9. Gando	is L, Teisserenc R,	Cobb A R et a	l., 2014.								
791														

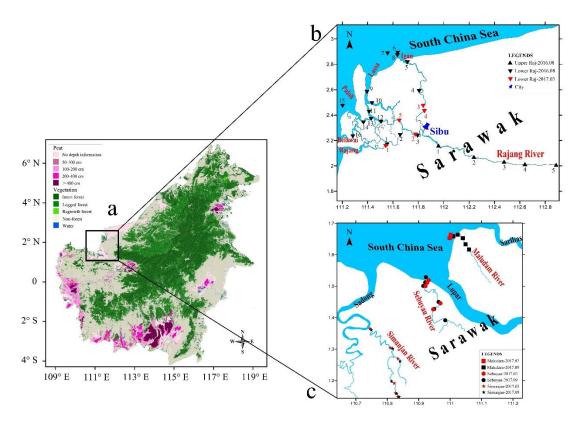


Figure 1 (a) Peat and vegetation distribution in the study region (modified from <a href="https://www.cifor.org/map/atlas/">https://www.cifor.org/map/atlas/</a>). (b) Sediment sampling sites along the Rajang and tributaries. The city of Sibu divides the river into upper and lower reaches. (c) Sediment 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.

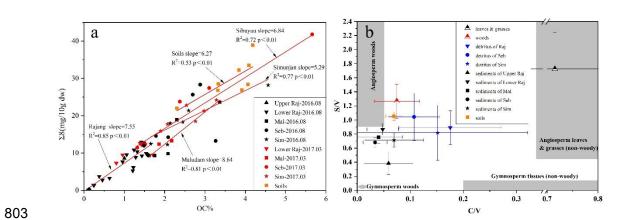


Figure 2 (a) Correlation of OC% with Σ8 among the various study systems. (b) Variations of S/V *versus* C/V of different samples from the study systems. Raj: Rajang; Seb: Sebuyau; Sim: Simunjan; Mal: Maludam.

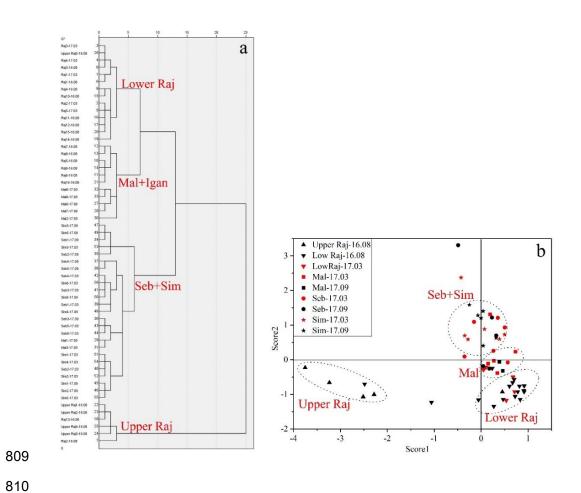


Figure 3 (a) Cluster analysis of the study systems based on bulk and lignin phenols parameters. (b) Plot of PCA results based on the distribution of scores 1 and 2. Raj: Rajang; Seb: Sebuyau; Sim: Simunjan; Mal: Maludam.

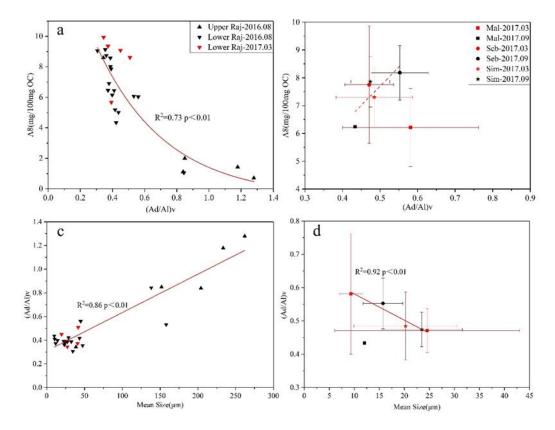


Figure 4 Variation in (Ad/Al)v with  $\Lambda 8$  values of sediments from (a) the Rajang and (b) the small river systems. Variation in (Ad/Al)v with mean sediment grain size for (c) the Rajang and (d) the small river systems.

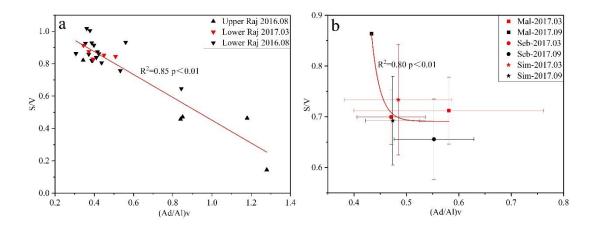


Figure 5 Relationship between (Ad/Al)v and S/V ratios based on average values of the various systems for (a) the Rajang and (b) the small river systems.



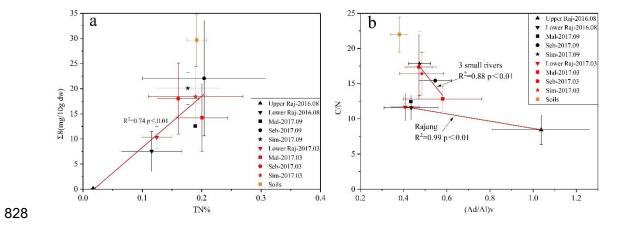


Figure 6 (a) Correlation of TN% with  $\Sigma 8$  based on average values of the study systems. (b) Correlation of  $(Ad/AI)_{v}$  with C/N ratio based on average values of the study systems.