

# Characteristics of carbonaceous aerosols emitted from peatland fire in Riau, Sumatra, Indonesia

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## Abstract

Biomass burning is a significant source of fine particulate matter (PM<sub>2.5</sub>). Forest, bush, and peat fires in Kalimantan and Sumatra, Indonesia are major sources of transboundary haze pollution in Southeast Asia. However, limited data exist regarding the chemical characteristics of aerosols at sources. We conducted intensive field studies in Riau Province, Sumatra, Indonesia, during the peatland fire and non-burning seasons in 2012. We characterized PM<sub>2.5</sub> carbonaceous aerosols emitted from peatland fire based on ground-based source-dominated sampling. PM<sub>2.5</sub> aerosols were collected with two mini-volume samplers using Teflon and quartz fiber filters. Background aerosols were also sampled during the transition period between the non-burning and fire seasons. We analyzed the carbonaceous content (organic carbon (OC) and elemental carbon (EC)) by a thermal optical reflectance utilizing the IMPROVE\_A protocol and the major organic components of the aerosols by a gas chromatography/mass spectrometry. PM<sub>2.5</sub> aerosols emitted from peatland fire were observed in high concentrations of  $7120 \pm 3620 \mu\text{g}/\text{m}^3$  and were primarily composed of OC ( $71.0 \pm 5.11\%$  of PM<sub>2.5</sub> mass). Levoglucosan exhibited the highest total ion current and was present at concentrations of  $464 \pm 183 \mu\text{g}/\text{m}^3$ . The OC/EC ratios ( $36.4 \pm 9.08$ ), abundances of eight thermally-derived carbon fractions, OC/Levoglucosan ratios ( $10.6 \pm 1.96$ ), and Levoglucosan/Mannosan ratios ( $10.6 \pm 2.03$ ) represent a signature profile that is inherent in peatland fire. These data will be useful in identifying contributions from single or multiple species in atmospheric aerosol samples collected from peatland fires.

**Keywords:** Carbonaceous aerosol; PM<sub>2.5</sub>; Levoglucosan; Mannosan; Biomass Burning

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35 **Highlights**

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- 37 ➤ PM<sub>2.5</sub> aerosols emitted from peatland fire in Indonesia were characterized.
- 38 ➤ PM<sub>2.5</sub> aerosols emitted from peatland fire were primarily composed of OC.
- 39 ➤ We found some source indicators that were inherent in peatland fire.

40

## 41 **1. Introduction**

42

43 Peatland is organic soil that has formed for over thousands of years from decomposed  
44 vegetation and other life forms, and peat deposits can extend up to seven meters in thickness  
45 (Wulandari, 2002). In Southeast Asia, smoke originating from peatland fires in Kalimantan and  
46 Sumatra in Indonesia is a major cause of transboundary haze pollution.

47 In general, approximately 80–90% of the smoke particles produced by biomass burning is in  
48 the PM<sub>2.5</sub> size range, and these particles are primarily composed of organic carbon, which  
49 constitutes 50–60% of the total particle mass (Phuleria *et al.*, 2005; Schollnberger *et al.*, 2002;  
50 Reid *et al.*, 2005). PM<sub>2.5</sub> aerosols present a high risk of deposition in the alveoli of lungs and are  
51 associated with a greater general health risk than coarse aerosols (Federal Register, 2006;  
52 Lippmann, 1998).

53 According to version 3 of the Global Fire Emissions Database (GFED), average PM<sub>2.5</sub>  
54 emissions from fire (including deforestation, savanna, forest, agricultural waste, and peat fires)  
55 from 1997 to 2010 in Indonesia are 2.9 Tg/year, accounting for 9.2% of global fire PM<sub>2.5</sub>  
56 emissions and 62% of Southeast Asian fire emissions. In Indonesia, peatland fire is a dominant  
57 source of PM<sub>2.5</sub> emissions, accounting for 55% of all fire sources. Thus, it is a significant  
58 emission source of PM<sub>2.5</sub> aerosols in Indonesia.

59 Many peatland fires occur on Sumatra and Kalimantan islands during the dry season, emitting

60 gases and smoke aerosols that cause atmospheric pollution (haze) and adversely affect the health  
61 of people living in surrounding areas. For example, an unprecedented Indonesian fire episode  
62 occurred in 1997–1998 due to the El Niño–Southern Oscillation (ENSO) event at that time.  
63 Extensive forest fires, including peatland fires, resulted in the development of a smog blanket that  
64 covered an immense area of 4 million km<sup>2</sup> in Southeast Asia. This smog affected the livelihoods  
65 and health of 75 million people in six countries and completely toppled their lives. Moreover, the  
66 smog resulted in the closure of airports; it was cited as the possible cause of an air crash near  
67 Medan, Sumatra, and a tanker collision in the Straits of Malacca (Stolle and Tomich, 1999). Air  
68 pollution attained previously unknown levels in East Kalimantan, Singapore, and Kuala Lumpur,  
69 with daily average particulate matter reaching extremely hazardous levels (4000 µg/m<sup>3</sup>; Heil *et*  
70 *al.*, 1998). Although ENSO events considerably contribute to the occurrence of Indonesian fires,  
71 pollution from smoke haze is a recurrent problem in Indonesia and neighboring countries, even in  
72 non-ENSO years (Tacconi, 2003).

73 Riau Province in Sumatra is one of the primary hotspots for peatland fire during the dry season,  
74 and the smoke aerosols generated there cause haze in Riau and in neighboring countries such as  
75 Malaysia and Singapore (Harahap, 2012; Hong, 2012). However, limited data exist regarding the  
76 chemical characteristics of these smoke aerosols (Othman and Latif, 2013; See *et al*, 2007) and  
77 the effects of aerosols from peatland fires on the atmospheric environment and human health. To

78 investigate these effects, the chemical characterization of fresh smoke aerosols from peatland fire  
79 is necessary. In this study, the carbonaceous species of PM<sub>2.5</sub> aerosols emitted from peatland fire  
80 were characterized by directly sampling PM<sub>2.5</sub> aerosols at fire hotspots in Riau Province.  
81 Moreover, we determined source indicators of carbonaceous species of smoke from peatland fires  
82 for source apportionment. These data can help in identifying single or multiple species in  
83 atmospheric aerosol samples that contribute to peatland fires.

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## 86 **2. Materials and methods**

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### 88 ***2.1. Sampling locations***

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90 The sampling locations in this study are illustrated in Fig. 1. The burning site and background  
91 site were located at Sepahat Village and Sukajadi Village, respectively, in Bengkalis Regency,  
92 Riau Province. The burning site was surrounded by peatland and forest, and the background site  
93 was located ~50 km away from the burning site and housing estates. Bengkalis Regency lies on  
94 the east coast of Sumatra Island and consists of several islands. This district covers an area of  
95 1,204,423 km<sup>2</sup>, in which nearly 85% of the land exhibits low topography and is covered with  
96 tropical forests, with an average elevation of only 2.0–6.1 m above sea level. Most of the soils are  
97 organosols or peat soils; that is, they contain abundant organic substances. The temperature in a  
98 Bengkalis is strongly influenced by the tropical marine climate and is typically 26–32°C. The

99 rainy season generally lasts from September to January, with an average rainfall of 809–4078  
100 mm/year, and the dry season usually spans February to August. Fig. 2 illustrates monthly hotspot  
101 counts in Riau in 2011 and 2012 based on Indofire datasets, confirming that many hotspots are  
102 detected in the area during the dry season.

103

## 104 ***2.2. Sample collection and analysis***

105

106 In 2012, background and peatland fire PM<sub>2.5</sub> aerosol samples were collected on May 16–17  
107 and June 13–17, respectively. Two mini-volume samplers (MiniVol™ TAS, Airmetrics) were  
108 utilized to continuously collect PM<sub>2.5</sub> aerosols on Teflon and quartz fiber filters for 24 h  
109 (background) and 2.5–5.2 h (peatland fire), respectively, at a flow rate of 5 L/min. PM<sub>2.5</sub> aerosols  
110 were collected at several burning sites, about 1.5 m away from peatland fire hotspots and a  
111 background site on 7 and 4 occasions, respectively. Wind speeds in the peatland fire samplings  
112 ranged from 0.610 to 6.21 m/s (average: 2.50 m/s) and aerosols were collected in smoke plumes.  
113 Quartz fiber filters were heat-treated at 900°C for 4 h before sampling to remove any absorbed  
114 organic materials.

115 Filter samples were analyzed to determine PM<sub>2.5</sub> mass concentrations, carbonaceous content  
116 (organic carbon (OC) and elemental carbon (EC)), and the quantity of key biomarkers.

117 The Teflon filter was weighed using a microbalance (ME5-F, Sartorius) with a sensitivity of ±1  
118 µg in a stable environment of 25.7 ± 0.432°C (average ± standard deviation) and 26.0 ± 0.607

119 RH% before and after sampling, respectively, to determine PM<sub>2.5</sub> mass concentrations.

120 The carbonaceous contents of the aerosols collected in the quartz fiber filters were quantified  
121 using a DRI Model 2001 OC/EC Carbon Analyzer, which employs thermal optical reflectance  
122 following the IMPROVE\_A protocol. The IMPROVE\_A temperature defines temperature  
123 plateaus for thermally-derived carbon fractions as follows: 140°C for OC<sub>1</sub>, 280°C for OC<sub>2</sub>,  
124 480°C for OC<sub>3</sub>, and 580°C for OC<sub>4</sub> in helium (He) carrier gas; 580°C for EC<sub>1</sub>, 740°C for EC<sub>2</sub>,  
125 and 840°C for EC<sub>3</sub> in a mixture of 98% He and 2% oxygen (O<sub>2</sub>) carrier gas (Chow *et al.*, 2007).

126 OC, EC, and total carbon (TC) were calculated from the eight carbon fractions as follows:

127  
128 
$$\text{OC} = \text{OC}_1 + \text{OC}_2 + \text{OC}_3 + \text{OC}_4 + \text{OP} \quad (1)$$

129  
130 
$$\text{EC} = \text{EC}_1 + \text{EC}_2 + \text{EC}_3 - \text{OP} \quad (2)$$

131  
132 
$$\text{TC} = \text{OC} + \text{EC} \quad (3)$$

133  
134 where OP (the amount of pyrolyzed OC) is defined as the carbon content measured after the  
135 introduction of O<sub>2</sub> until reflectance returns to its initial value at the start of analysis.

136 Two key biomarkers, levoglucosan and mannosan, obtained from the quartz fiber filters were  
137 quantified by gas chromatography/mass spectrometry (GC/MS). Organic compound speciation  
138 was basically accomplished, following the procedures of Fabbri *et al.*, 2009 and Pashynska *et al.*,  
139 2002. Aliquots from the quartz fiber filter were spiked with the internal standard of methyl  
140 β-L-arabinopyranoside before extraction. Each spiked filter was extracted by ultrasonic agitation

141 for 3 × 20 min periods using 3 mL of a dichloromethane/methanol mixture (3/1, v/v)  
142 (dichloromethane: Wako, purity >99.5%; methanol: Wako, purity >99.7%). The combined  
143 extracts were reduced to approximately 100 µL using a rotary evaporator (250 hPa, 40°C).  
144 Subsequently, the concentrated extract was filtered through a Teflon syringe filter (pore size 0.1  
145 µm) and dried completely under a nitrogen stream. Prior to analysis, the total extracts were  
146 converted to trimethylsilyl derivatives by reaction with 150 µL of  
147 N,O-bis-(trimethylsilyl)-trifluoroacetamide with 1% trimethylchlorosilane and 90 µL of pyridine  
148 for 3 h at 70°C.

149 The derivatized samples were analyzed on a Shimadzu GC/MS system (GCMS-QP2010-Plus,  
150 Shimadzu) equipped with an Rtx-5Sil MS column (with selectivity similar to that of a mixture of  
151 5% diphenyl and 95% dimethyl polysiloxane, 30 m × 0.25 mm I.D., 0.25 µm, RESTEK). Helium  
152 (purity 99.9995%) with an average velocity of 35.3 cm/s was used as the carrier gas. The GC  
153 oven temperature program was as follows: isothermal at 80°C for 5 min, 80–180°C at 3°C/min,  
154 then 180–300°C at 20°C/min, and maintained at 300°C for 5 min. The injection port and transfer  
155 line were maintained at 300°C. The data for quantitative analysis were acquired in the electron  
156 impact mode (70 eV).

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### 159 **3. Results and discussion**

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161 **3.1. *PM<sub>2.5</sub> mass concentration***

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163 Background and peatland fire  $PM_{2.5}$  concentrations determined by gravimetric analysis were  
164  $23.9 \pm 2.53 \mu\text{g}/\text{m}^3$  and  $7120 \pm 3620 \mu\text{g}/\text{m}^3$ , respectively; that is, peatland fire  $PM_{2.5}$   
165 concentrations were ~300 times higher than background concentrations. These extremely high  
166  $PM_{2.5}$  concentrations present a great risk to human health, particularly in the case of fire fighters.  
167 Since the high concentration is due to the sampling at 1.5 m away from the fire, a different value  
168 would be obtained in the different sampling distance.

169

170 **3.2. *OC and EC***

171

172 The average OC concentrations from peatland fire and background were  $4970 \pm 3620 \mu\text{g}/\text{m}^3$   
173 and  $9.42 \pm 1.97 \mu\text{g}/\text{m}^3$ , respectively, and average EC concentrations were  $133 \pm 46.6 \mu\text{g}/\text{m}^3$  and  
174  $3.23 \pm 0.795 \mu\text{g}/\text{m}^3$ , respectively. In terms of the carbonaceous fractions in  $PM_{2.5}$  aerosols,  $PM_{2.5}$   
175 aerosols emitted from peatland fire were composed of  $71.0 \pm 5.11\%$  OC and  $2.05 \pm 0.509\%$  EC.  
176 Conversely, background  $PM_{2.5}$  aerosols were composed of  $39.1 \pm 5.08\%$  OC and  $13.5 \pm 3.04\%$   
177 EC. The remaining undetermined mass can be attributed to the various functional groups in  
178 organic aerosols, inorganic ions, metals, soil and particle bound water.

179 OC to EC mass ratios (OC/EC) provide some indication of the origins of carbonaceous  $PM_{2.5}$   
180 (Cao *et al.*, 2005; Chow *et al.*, 1996; Gray *et al.*, 1986; Turpin and Huntzicker, 1991). In this  
181 study, the average peatland fire and background OC/EC ratios were  $36.4 \pm 9.08$  and  $2.99 \pm 0.738$ ,

182 respectively. Few differences in the OC/EC ratio were observed, regardless of PM<sub>2.5</sub> mass  
183 concentrations, and the coefficient of variance for samples from peatland fire was 24.9%. See *et*  
184 *al.*, 2007 reported that the average OC/EC ratio was 2.42 for PM<sub>2.5</sub> aerosols collected in the open  
185 field within a 100 m circumference from the boundary of the Indonesian peatland fires in 2005.  
186 This value is much smaller than our result of 36.4. The discrepancy is due to the different  
187 measurement techniques for the OC-EC split. See *et al.*, 2007 adopted thermal method whose  
188 temperature of OC-EC split was 350°C. The method would cause significant overestimation of  
189 EC concentration in aerosol (Gelencsér, 2004). On the other hand, we utilized thermal-optical  
190 reflectance method with pyrolysis correction. OC/EC ratios from our peatland fire and other  
191 vegetative burning sources are illustrated in Fig. 3. OC/EC ratios ranged from 4.34 to 79.7; this  
192 discrepancy can be partly attributed to variations in the type and moisture content of the burning  
193 material.

194 Abundances of eight thermally-derived carbon fractions differ by carbon sources (Cao *et al.*,  
195 2005; Chow *et al.*, 2004; Watson *et al.*, 1994). Fig. 4 illustrates the abundances of eight  
196 thermally-derived carbon fractions at both the peatland fire and background sites by mass  
197 percentage of total carbon. Distinct differences in carbon fractions were observed between the  
198 two sites. OC<sub>1</sub> accounted for  $31.7 \pm 2.21\%$  of TC in peatland fire samples but only  $0.710 \pm$   
199  $0.721\%$  of TC in background samples. OC<sub>2</sub> accounted for  $47.5 \pm 0.948\%$  of TC in peatland fire

200 samples and  $13.9 \pm 1.64\%$  of TC in background samples. OC<sub>3</sub> accounted for  $7.44 \pm 1.57\%$  of TC  
201 in peatland fire samples and  $40.7 \pm 2.62\%$  of TC in background samples. OC<sub>4</sub> accounted for  $1.22$   
202  $\pm 0.412\%$  of TC in peatland fire samples and  $15.7 \pm 1.27\%$  of TC in background samples. OP  
203 accounted for  $9.28 \pm 1.39\%$  of TC in peatland fire samples and  $3.95 \pm 4.13\%$  of TC in  
204 background samples. Lower EC fractions were observed in the peatland fire than the background  
205 samples. Thus, it was found that TC in PM<sub>2.5</sub> aerosols derived from peatland fires was primarily  
206 composed of OC<sub>1</sub> and OC<sub>2</sub>.

207 Fig. 5 illustrates the average percentages of eight fractions in PM<sub>2.5</sub> for the peatland fire  
208 samples and those from other sources. The vegetation burning profiles used as reference were  
209 obtained by ground-based source-dominated sampling of the plumes of small controlled burns of  
210 wood debris at the Big Bend National Park in the US (see Chow *et al.*, 2004). The carbon fraction  
211 abundances differ by emission source: OC<sub>1</sub> is enriched ( $23.9 \pm 12.4\%$ ) in the vegetative burning  
212 profile; OC<sub>2</sub> is enriched ( $34.7 \pm 2.16\%$ ) in the peatland fire profile; OC<sub>3</sub> is enriched ( $37.6 \pm$   
213  $15.9\%$ ) in the cooking profile; EC<sub>2</sub> is enriched ( $23.3 \pm 10.4\%$ ) in the motor vehicle profile. In  
214 terms of the coefficient of variance, the carbon fraction obtained from peatland fire is more  
215 consistent than any source presented in Fig. 5. Thus, the peatland profile will be useful for source  
216 discrimination.

217

### 218 **3.3. Key biomarkers**

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220 The organic compounds produced by peatland fires were analyzed by GC/MS, and an example  
221 of typical GC/MS total ion current (TIC) tracers for the total extract of peatland fire samples is  
222 presented in Fig. 6. Levoglucosan clearly exhibits the highest TIC peak in peatland fire samples,  
223 although mannosan and palmitic acid were also detected. Palmitic acid is one of the most basic  
224 units of plant fats, oils, and phospholipids (Simoneit, 2002). The major tracers in smoke emitted  
225 from biomass burning are the thermal degradation (pyrolysis) products of the biopolymers of  
226 cellulose and lignin in woody tissue. The thermal degradation of cellulose (also hemicelluloses)  
227 yields dehydromonosaccharide derivatives, which are predominant compounds in the smoke  
228 (Simoneit *et al.*, 1999). The major compound is levoglucosan, with minor and variable amounts  
229 of galactosan and mannosan. These compounds cannot be formed by hydrolysis or by the  
230 microbial alteration of carbohydrates; thus, they are specific to burning (Simoneit *et al.*, 1999). In  
231 addition, levoglucosan is considerably resistant to atmospheric degradation (Fraser and  
232 Lakshmanan, 2000). Therefore, levoglucosan and mannosan are regarded as key biomarkers in  
233 peatland fire aerosols like other biomass burnings.

234 The average levoglucosan concentrations from peatland fire and background samples were 464  
235  $\pm 183 \mu\text{g}/\text{m}^3$  and  $0.278 \pm 0.155 \mu\text{g}/\text{m}^3$ , respectively and the average mannosan concentration  
236 were  $47.5 \pm 25.6 \mu\text{g}/\text{m}^3$  and  $0.0190 \pm 0.0108 \mu\text{g}/\text{m}^3$ , respectively. Levoglucosan and mannosan in  
237 background samples may have originated from wood burning for cooking, agricultural use,

238 among others. However, concentrations of levoglucosan and mannosan emitted in association  
239 with peatland fire events were much higher than their corresponding background concentrations.

240 OC/Levoglucosan ratios vary to some extent for different burning conditions and wood types  
241 (Pio, *et al.*, 2008). In this study, the average OC/Levoglucosan ratio in peatland fire was  $10.6 \pm$   
242  $1.96 \mu\text{gC}/\mu\text{g}$ . For comparison of our peatland fire samples with biomass burning samples, the  
243 average OC/Levoglucosan ratios for various source samples are presented in Fig. 7, which shows  
244 significant differences in OC/Levoglucosan ratios between the peatland fire and other emission  
245 sources. Thus, this ratio may be useful for source discrimination for peatland fire. However,  
246 because levoglucosan is emitted from other biomass burning sources and there are other types of  
247 biomass burning in Riau, Sumatra, Indonesia, selection of only OC/Levoglucosan ratio for source  
248 apportionment of peatland fire can lead to overestimation of the contribution of peatland fire.  
249 Hence, other indicators for peatland fire are needed.

250 The relative amounts of the individual anhydrosaccharides in biomass smoke aerosols can be  
251 used for further source assignment of specific biofuels (Fabbri *et al.*, 2009; Alves *et al.*, 2010).  
252 Here, we investigated the Levoglucosan/Mannosan ratios and compared those ratios from  
253 peatland fire and other biomass burning sources. In this study, the average  
254 Levoglucosan/Mannosan ratios in peatland fire and background were  $10.6 \pm 2.03$  and  $14.5 \pm 3.39$ ,  
255 respectively. For comparison of our peatland fire samples with other biomass burning samples,

256 the average Levoglucosan/Mannosan ratios for various source samples are presented in Table 1,  
257 which shows the significant differences in Levoglucosan/Mannosan ratios between the peatland  
258 fire and other emission sources. Thus, this ratio should be more useful for source discrimination  
259 for peatland fire than OC/Levoglucosan ratio.

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## 262 **4. Conclusions**

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264  $PM_{2.5}$  carbonaceous aerosols were collected at a peatland fire hotspots and a background site  
265 on 7 and 4 occasions, respectively.  $PM_{2.5}$  aerosols emitted from peatland fire were observed in  
266 very high concentrations ( $7120 \pm 3620 \mu\text{g}/\text{m}^3$ ) and were primarily composed of OC ( $71.0 \pm$   
267  $5.11\%$  of  $PM_{2.5}$  mass).

268 The OC/EC ratios ( $36.4 \pm 9.08$  for peatland fire), abundances of eight thermally-derived  
269 carbon fractions, OC/Levoglucosan ratios ( $10.6 \pm 1.96$  for peatland fire), and  
270 Levoglucosan/Mannosan ratios ( $10.6 \pm 2.03$  for peatland fire) observed here represent a signature  
271 profile that seems to be inherent in peatland fire emissions. Therefore, it is suggested that such  
272 profiles are useful for peatland fire source discrimination.

273 In future studies, additional chemical speciation for fresh and aged smoke from peatland fires  
274 will be required to estimate atmospheric environmental impacts and/or adverse health effects (e.g.,  
275 humic-like substances, polycyclic aromatic hydrocarbons).

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277

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**Table Captions**

376

377 Table 1. Range of Levoglucosan/Mannosan ratios in PM<sub>2.5</sub> emitted from several sources and  
378 background.

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	<b>Levoglucosan/Mannosan</b>	<b>Reference</b>
<b>Peatland fire</b>	7.09–14.0	this study
<b>Background</b>	10.7–18.9	this study
<b>Hardwoods burning</b>	13.8–52.3	Engling <i>et al.</i> , 2006
<b>Softwoods burning</b>	2.6–5.0	Engling <i>et al.</i> , 2006
<b>Grasses burning</b>	108–203	Engling <i>et al.</i> , 2006

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Table 1.

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## Figure Captions

399

400 Fig. 1. Map of Bengkalis showing the sampling sites.

401 Fig. 2. Monthly hotspot counts in Riau in 2011 and 2012.

402 Fig. 3. Comparison of OC/EC ratios in PM<sub>2.5</sub> emitted from peatland fire and other burning

403 sources. Error bar in this study indicates standard deviation.

404 Fig. 4. Abundances (as mass percentage of total carbon) of eight thermally-derived carbon

405 fractions of aerosols sampled at the peatland fire and background sites. Error bars indicate

406 standard deviations.

407 Fig. 5. Carbon fraction composition of PM<sub>2.5</sub> combustion source profiles. Error bars indicate

408 standard deviations.

409 Fig. 6. GC/MS total ion current (TIC) chromatograph of total extract showing major compounds

410 of peatland fire aerosols.

411 Fig. 7. Comparison of OC/Levoglucosan ratios of PM<sub>2.5</sub> obtained from peatland fire and other

412 burning sources. Error bar in this study indicates standard deviation.

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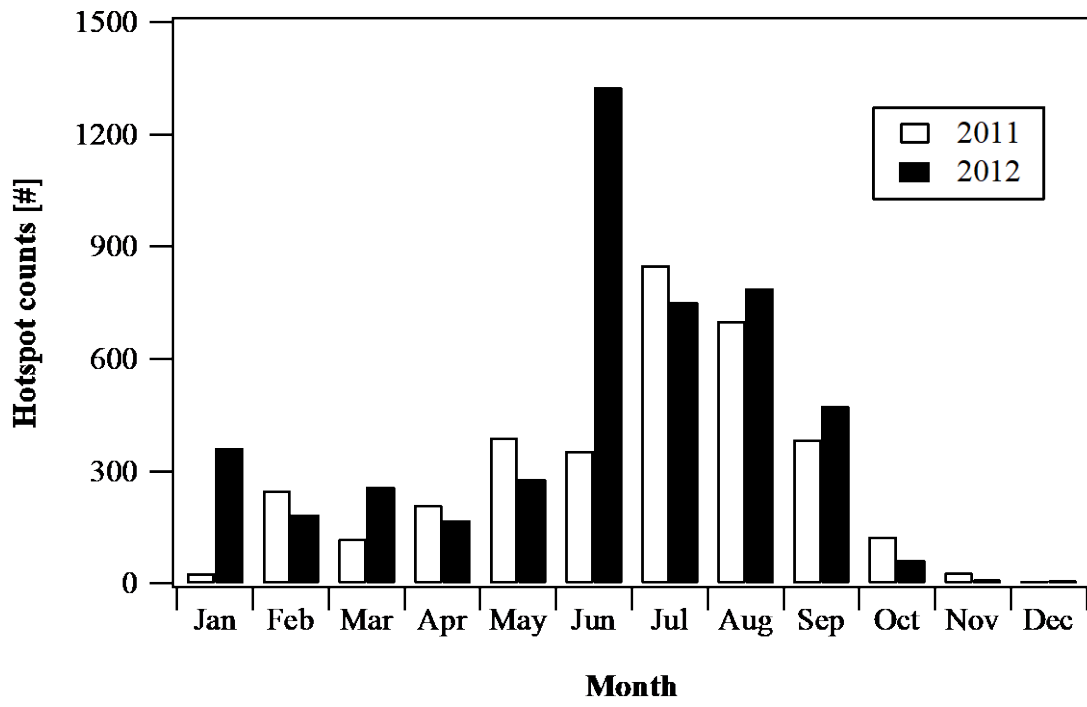
Fig. 1.



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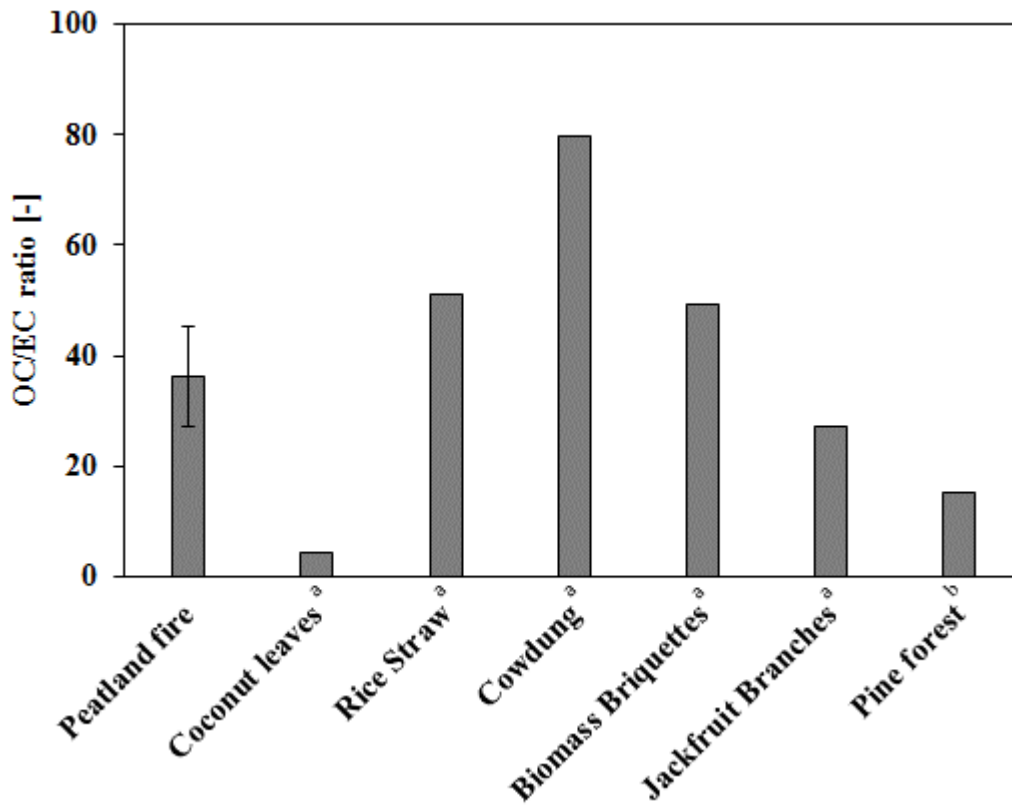
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Fig. 2.

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444 <sup>a</sup> data from Sheesley and Schauer, 2003, <sup>b</sup> data from Lee *et al.*, 2005.

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Fig. 3.

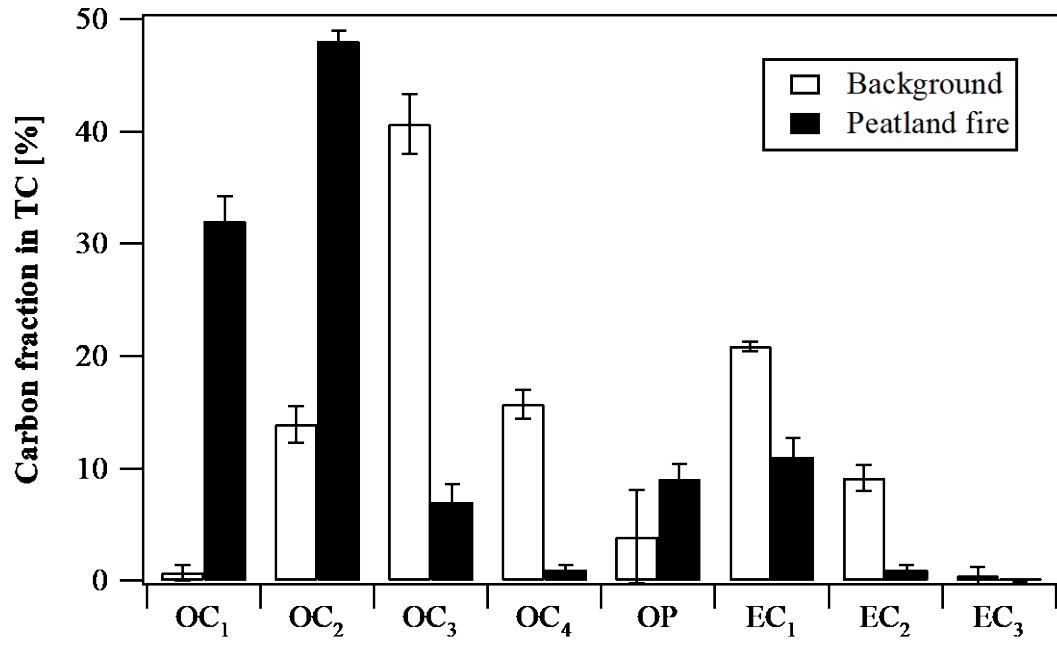
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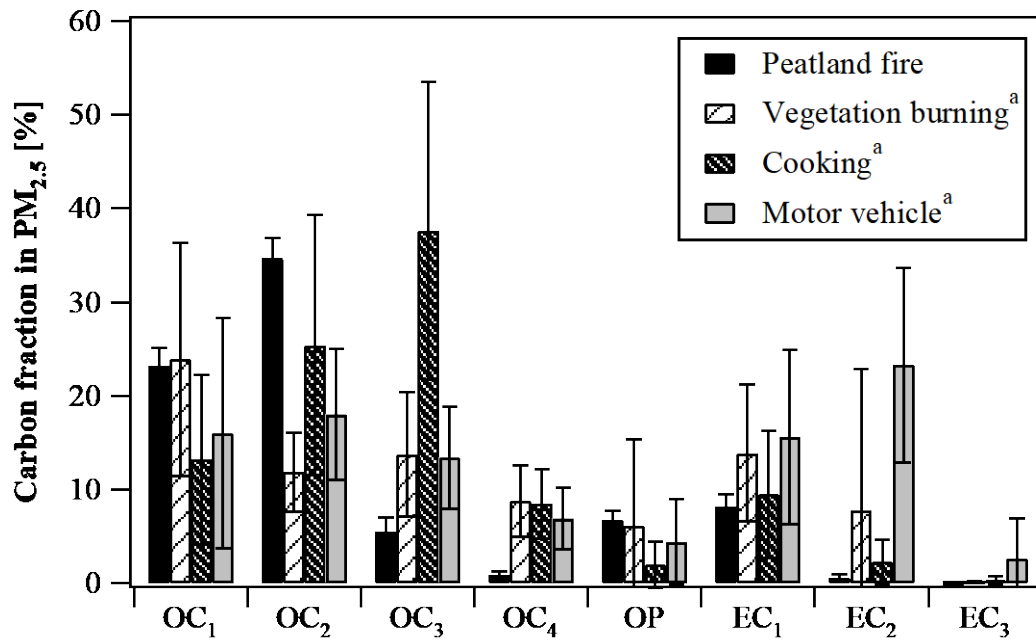
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Fig. 4.

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465 <sup>a</sup> data from Chow *et al.*, 2004.

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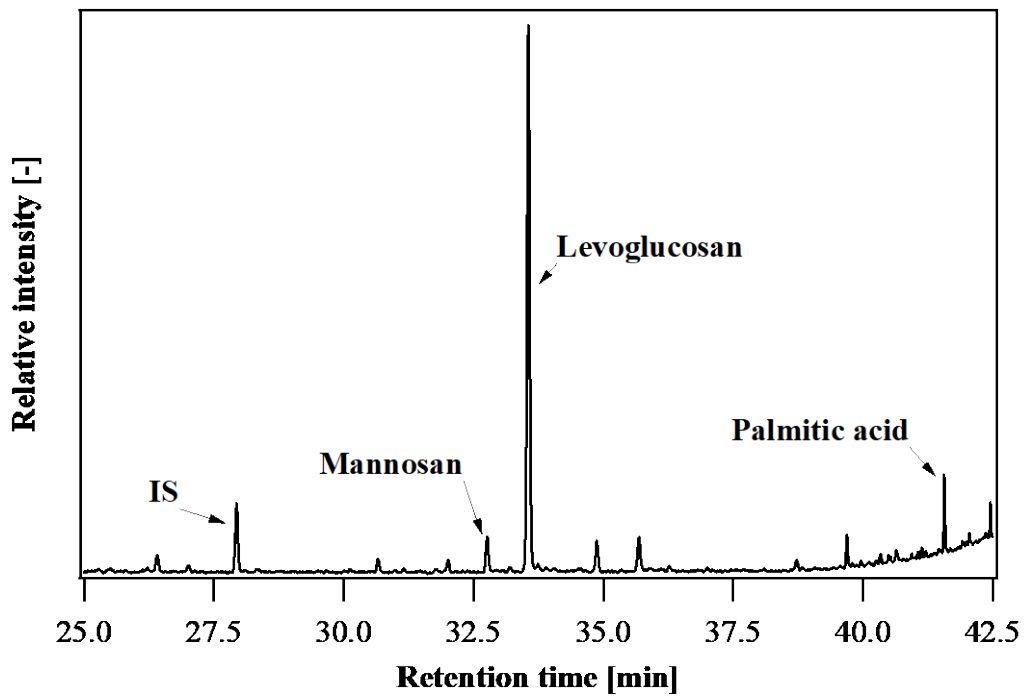
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Fig. 5.

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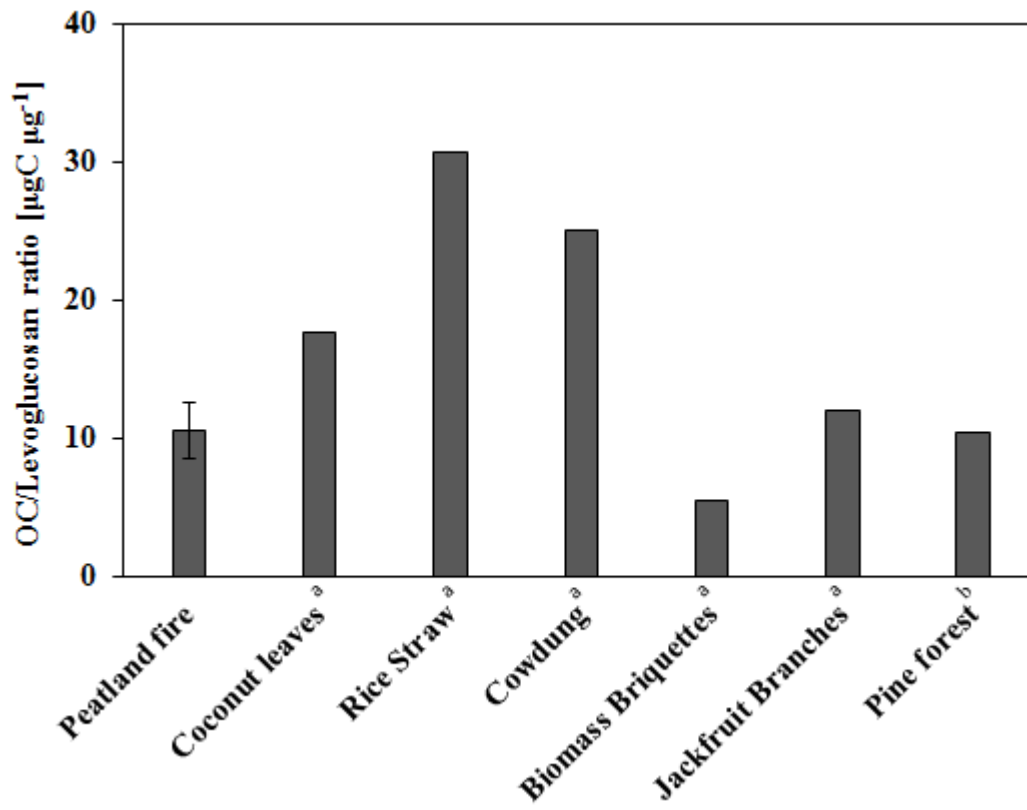
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Fig. 6.

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489 <sup>a</sup> data from Sheesley and Schauer, 2003, <sup>b</sup> data from Lee *et al.*, 2005.

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Fig. 7.

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