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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_{2.5}). 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_{2.5} carbonaceous aerosols emitted from peatland fire based on ground-based source-dominated sampling. PM_{2.5} 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_{2.5} 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_{2.5} 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 ± 9.08), abundances of eight thermally-derived carbon fractions, OC/Levoglucosan ratios (10.6 ± 1.96), and Levoglucosan/Mannosan ratios (10.6 ± 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_{2.5}; Levoglucosan; Mannosan; Biomass Burning

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35 **Highlights**

36

- 37 ➤ PM_{2.5} aerosols emitted from peatland fire in Indonesia were characterized.
- 38 ➤ PM_{2.5} 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_{2.5} 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_{2.5} 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_{2.5}
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_{2.5}
56 emissions and 62% of Southeast Asian fire emissions. In Indonesia, peatland fire is a dominant
57 source of PM_{2.5} emissions, accounting for 55% of all fire sources. Thus, it is a significant
58 emission source of PM_{2.5} 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 km² 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/m³; 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_{2.5} aerosols emitted from peatland fire
80 were characterized by directly sampling PM_{2.5} 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.

84

85

86 **2. Materials and methods**

87

88 ***2.1. Sampling locations***

89

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 km², 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_{2.5} 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_{2.5} 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_{2.5} 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_{2.5} 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_{2.5} 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 OC₁, 280°C for OC₂,
124 480°C for OC₃, and 580°C for OC₄ in helium (He) carrier gas; 580°C for EC₁, 740°C for EC₂,
125 and 840°C for EC₃ in a mixture of 98% He and 2% oxygen (O₂) 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 O₂ 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).

157

158

159 **3. Results and discussion**

160

161 **3.1. *PM_{2.5} mass concentration***

162

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 ± 9.08 and 2.99 ± 0.738 ,

182 respectively. Few differences in the OC/EC ratio were observed, regardless of PM_{2.5} 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_{2.5} 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. OC₁ 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. OC₂ 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. OC₃ 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. OC₄ 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_{2.5} aerosols derived from peatland fires was primarily
206 composed of OC₁ and OC₂.

207 Fig. 5 illustrates the average percentages of eight fractions in PM_{2.5} 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: OC₁ is enriched ($23.9 \pm 12.4\%$) in the vegetative burning
212 profile; OC₂ is enriched ($34.7 \pm 2.16\%$) in the peatland fire profile; OC₃ is enriched ($37.6 \pm$
213 15.9%) in the cooking profile; EC₂ 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**

219

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 ± 2.03 and 14.5 ± 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.

260

261

262 **4. Conclusions**

263

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 ± 9.08 for peatland fire), abundances of eight thermally-derived
269 carbon fractions, OC/Levoglucosan ratios (10.6 ± 1.96 for peatland fire), and
270 Levoglucosan/Mannosan ratios (10.6 ± 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).

276

277

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279

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283

284

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286

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Table Captions

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377 Table 1. Range of Levoglucosan/Mannosan ratios in PM_{2.5} emitted from several sources and
378 background.

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

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Table 1.

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Figure Captions

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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_{2.5} 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_{2.5} 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_{2.5} obtained from peatland fire and other

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

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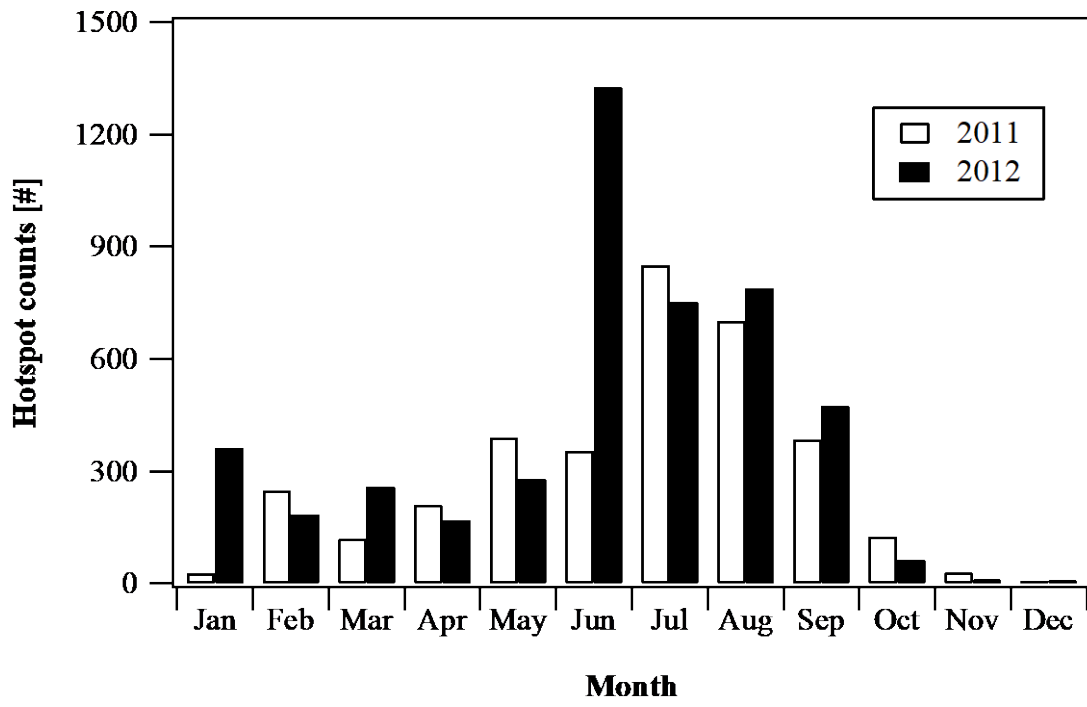
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Fig. 1.

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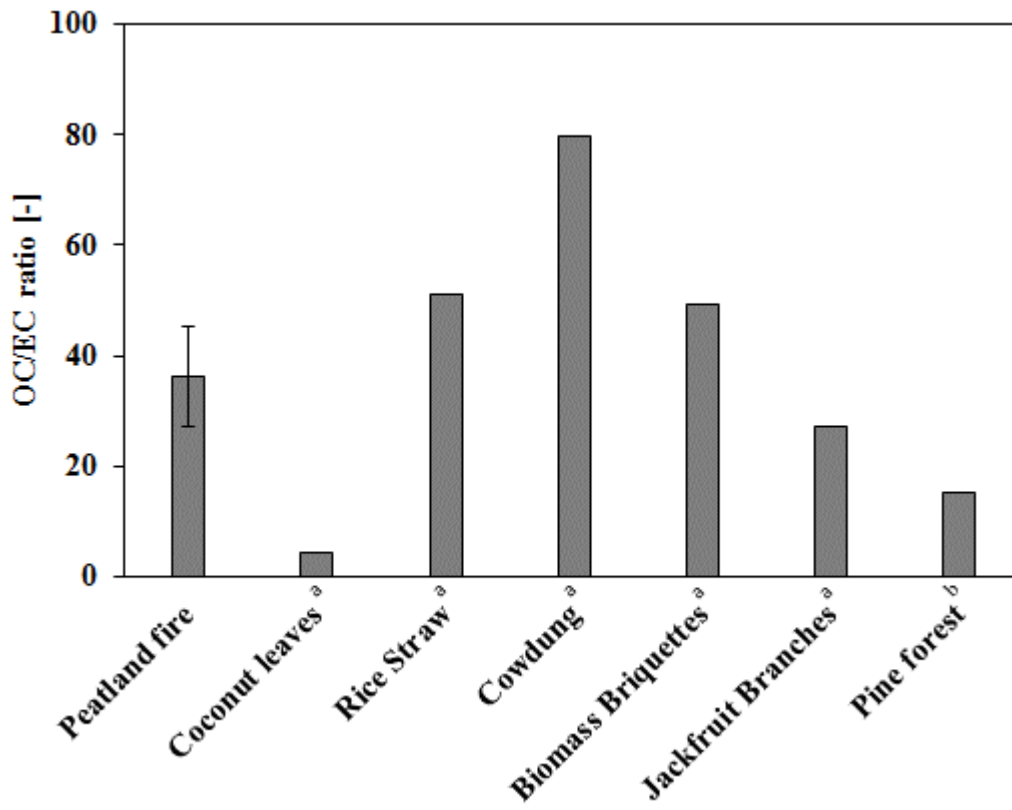
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Fig. 2.

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444 ^a data from Sheesley and Schauer, 2003, ^b data from Lee *et al.*, 2005.

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Fig. 3.

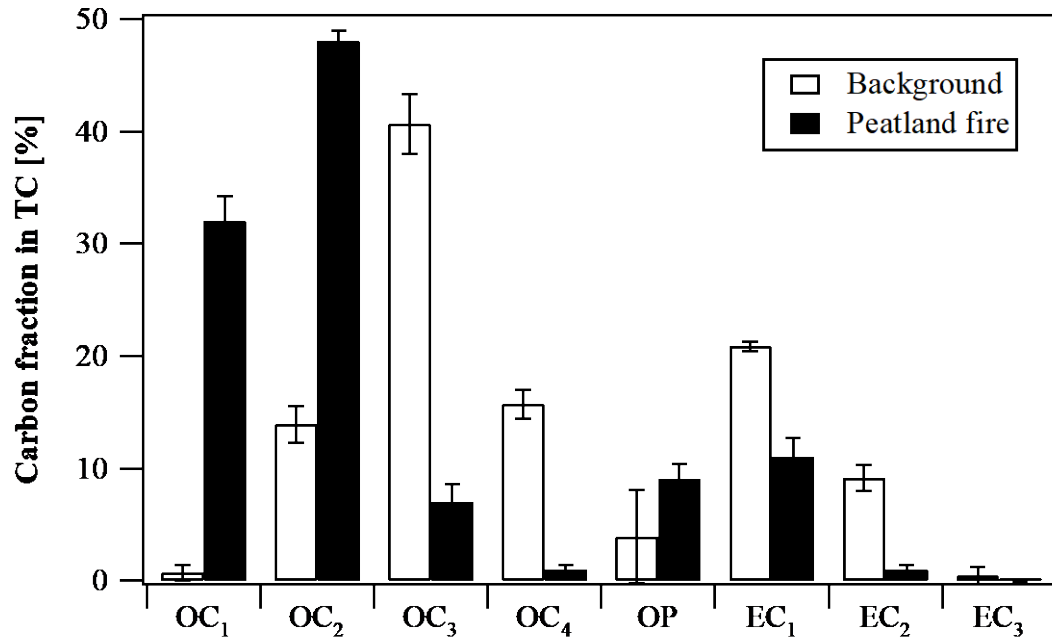
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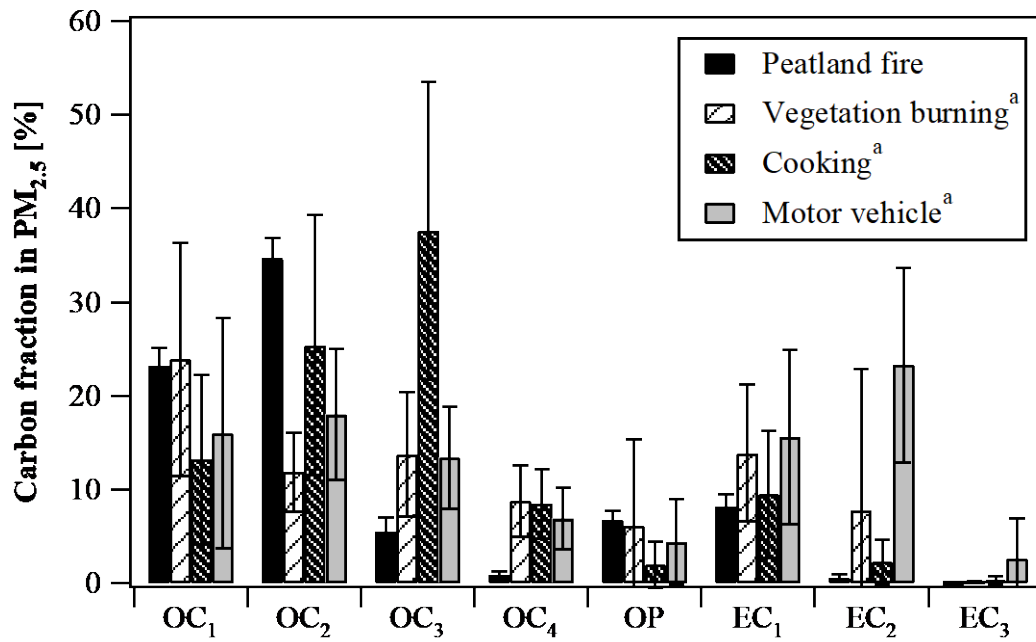
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Fig. 4.

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465 ^a data from Chow *et al.*, 2004.

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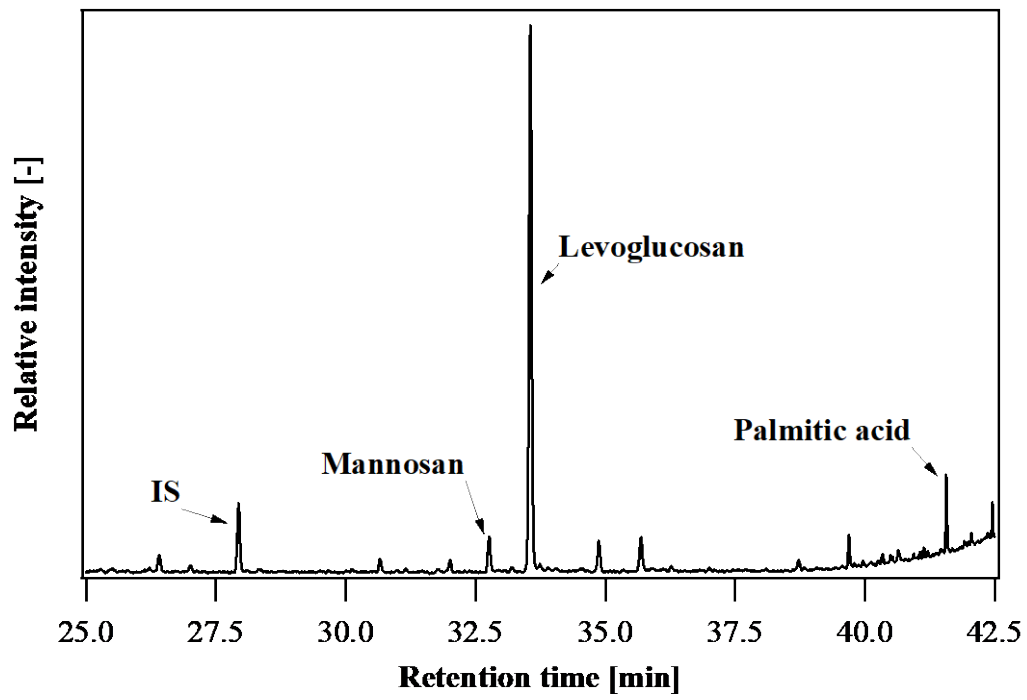
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Fig. 5.

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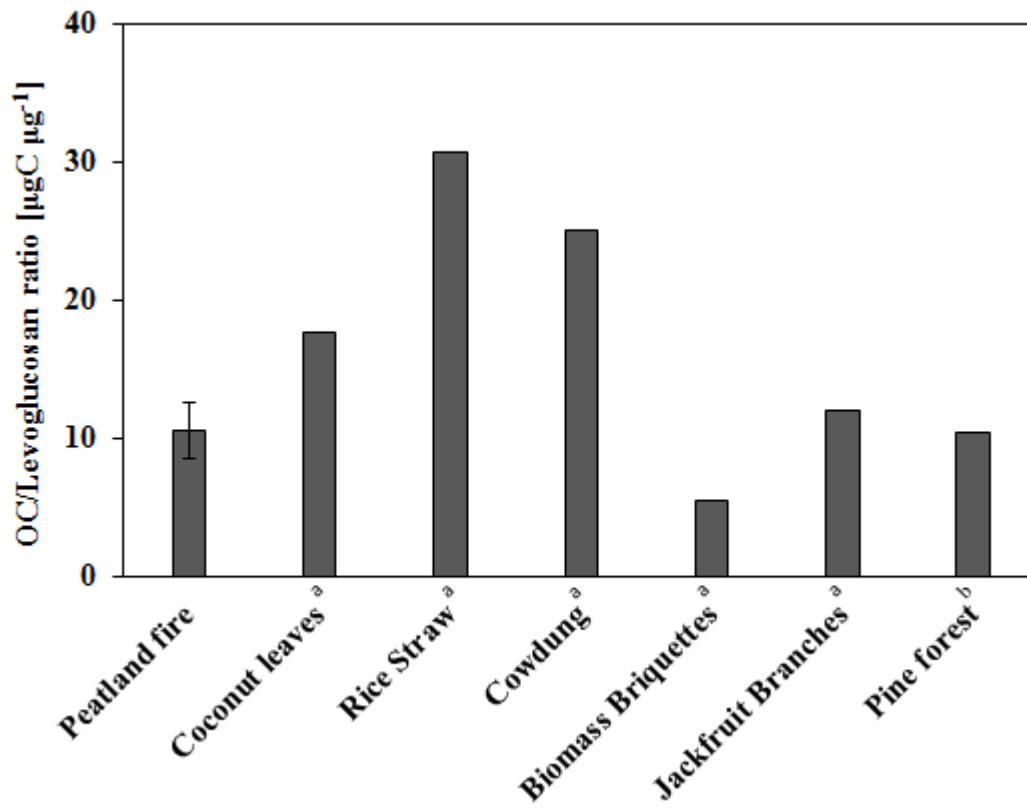
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Fig. 6.

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489 ^a data from Sheesley and Schauer, 2003, ^b data from Lee *et al.*, 2005.

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Fig. 7.

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