

# Biomass burning as the main source of organic aerosol particulate matter in Malaysia during haze episodes

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Received 25 June 2003; received in revised form 31 December 2003; accepted 2 February 2004

## Abstract

The haze episodes that occurred in Malaysia in September–October 1991, August–October 1994 and September–October 1997 have been attributed to suspended smoke particulate matter from biomass burning in southern Sumatra and Kalimantan, Indonesia. In the present study, polar organic compounds in aerosol particulate matter from Malaysia are converted to their trimethylsilyl derivatives and analyzed by gas chromatography–mass spectrometry in order to better assess the contribution of the biomass burning component during the haze episodes. On the basis of this analysis, levoglucosan was found to be the most abundant organic compound detected in almost all samples. The monosaccharides,  $\alpha$ - and  $\beta$ -mannose, the lignin breakdown products, vanillic and syringic acids and the minor steroids, cholesterol and  $\beta$ -sitosterol were also present in some samples. The presence of the tracers from smoke overwhelmed the typical signatures of emissions from traffic and other anthropogenic activities in the urban areas.

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**Keywords:** Levoglucosan; Monosaccharides; Sterols; Biomass burning

## 1. Introduction

The occurrence of localized haze over the urban areas in Malaysia, especially in the Klang valley, has become a common feature for the past two decades. Haze usually occurs during the south-west monsoon season in July to September, when boundary layer convergence of air and moisture over the region reduce atmospheric circulation and advection which normally disperse and dilute the

pollutant concentrations through convective mixing or wash out of the trapped pollutants by widespread heavy rain (MMS, 1995). However, in contrast to localized transient smog conditions, occurring in urban areas from trapping of anthropogenic emissions in a stagnant atmosphere, the haze episodes of April 1983, October 1991, August–October 1994 and September–October 1997 were due to the advection of suspended smoke particulate matter from biomass burning in Southern Sumatra and Kalimantan, Indonesia by the prevailing winds.

Biomass burning is an important primary source of numerous trace compounds which are reactants in atmospheric chemistry and of soot particulate matter that decreases visibility and absorbs incident radiation (e.g., Crutzen and Andreae, 1990; Levine, 1991, 1996; Charlson et al., 1992). Biomass burning, used as a

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method for clearing vegetated (forest and grassland) areas, and wild fires significantly increase the input of organic aerosol components to the atmosphere. The chemical composition data of smoke particulate matter is important for understanding the organic component contribution of biomass burning emissions to atmospheric chemistry and complements existing data on the signatures of direct organic emissions (i.e., particulate matter) from biomass sources (Ramdahl, 1983; Standley and Simoneit, 1987, 1994; Hawthorne et al., 1992; Rogge et al., 1993a, 1998; Simoneit et al., 1993, 1996, 1999; Abas et al., 1995; Schauer et al., 1996; Nolte et al., 2001; Graham et al., 2002). The varying temperature conditions during burning determine the molecular alteration and transformation of the organic compounds emitted from biomass fuels (Simoneit et al., 1993, 1999; Simoneit, 2002). The heat intensity, aeration, and duration of smoldering and flaming conditions determine the distributions and ratios of the natural versus altered compounds present in smoke (Simoneit et al., 1999). The primary chemical reactions which occur under both smoldering and flaming conditions include pyrolysis, depolymerization, water elimination, steam stripping/volatilization of characteristic molecular tracers, fragmentation, oxidation and char formation (Mazurek and Simoneit, 1997). The pyrolysis derivatives from the thermal breakdown of cellulose during burning are the dominant smoke tracers in continental airsheds (Locker, 1988; Simoneit et al., 1999; Fraser and Lakshmanan, 2000; Simoneit and Elias, 2000; Nolte et al., 2001; Graham et al., 2002; Zdráhal et al., 2002). The major tracer compound is levoglucosan(I), the chemical structures cited are given in Appendix A, with lesser amounts of galactosan(II) and mannosan(III). These are the tracers utilized for the assessment of particulate matter from biomass burning in the atmosphere because they cannot be generated by non-combustive processes. Numerous other minor products have also been reported (e.g., Nolte et al., 2001; Graham et al., 2002). In addition, methoxyphenols from lignin breakdown and free lipids and terpenoids, with their thermal alteration products, can be emitted in biomass smoke (Hawthorne et al., 1992; Simoneit et al., 1993). It is therefore useful to apply the identity of the biogenic constituents from both natural and combustion emissions and the anthropogenic emissions as tracers in tropospheric chemistry.

Here we report the use of silylation of total extracts of particulate matter followed by GC-MS analysis to identify and quantify the most polar organic compounds present in the aerosols collected during major haze episodes of August–October 1994 and September–October 1997 in Malaysia. Direct extraction of aerosol particles by a polar organic solvent mixture allows analysis of both neutral and polar components in one step, thus avoiding a double extraction where the

aqueous extract causes hydrolysis of the anhydrosaccharides. The direct extraction technique enables us to make a quick assessment of the biomass burning component superimposed on the anthropogenic and natural constituents of atmospheric particulate matter in Malaysia during those times.

## 2. Experimental methods

### 2.1. Sampling locations

The locales of the sampling stations are shown on the map (Fig. 1). The station in Petaling Jaya was established in 1976 in a highly developed urban region of Malaysia. Traffic and population density are the highest in the country and the region is considered as the most polluted area in Malaysia. Generally, the TSP load for Petaling Jaya fluctuates between 60 and 110  $\mu\text{g}/\text{m}^3$ . The campus of the University of Malaya is situated mid-way between Kuala Lumpur and Petaling Jaya. TSP samples were collected during daytime and nighttime.

The monitoring station in Malacca is located at an airport about 7 km from the center of town. Although quite near the main road with moderate traffic density, this station is surrounded by industrial and housing estates.

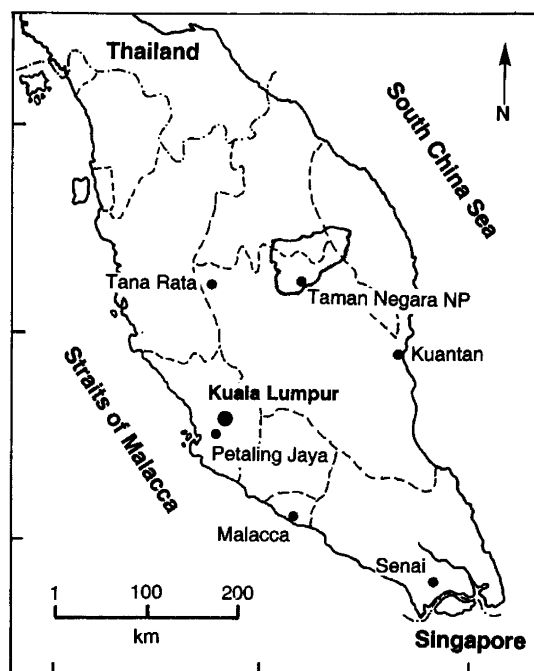


Fig. 1. Map of Peninsular Malaysia showing the sampling locations (all were in urban areas, except Taman Negara National Park).

The Tanah Rata station is about 1500 m above sea level and surrounded by agricultural and forest lands. This station was a locale for the background air pollution monitoring network (BAPMoN, renamed Global Atmospheric Watch, GAW) of the World Meteorological Organization (WMO).

The Senai monitoring station is located at an airport about 5 km away from the main north-south highway. It is one of the fastest developing regions in Malaysia with forest and plantation lands being cleared for housing and industrial development.

The Kuantan station is located at the airport about 13 km from the town center, surrounded by light industries and dense residential areas. The main road past the airport is quite busy.

The Taman Negara National Park, located in the State of Pahang, is the largest park accessible in Peninsular Malaysia and was the first to be established (1939).

## 2.2. Sample acquisition and analysis

Malaysia was under a blanket of smoke (haze) and stagnant atmospheric conditions during the sampling periods of 1994 and 1997. Backward airparcel trajectories are not warranted. Particulate aerosol samples from Senai, Malacca, Kuantan and Tanah Rata were acquired on September 7, 1994, while the one from Petaling Jaya was acquired on September 9, 1994. The samples from the University of Malaya were collected from September 15–19, 1997. Aerosol particulate samples were typically acquired by high volume filtration without particle size preseparator (total suspended particles, TSP) on quartz fiber filters. Sampling was carried out for 12 or 24 h periods and the extraction and analyses procedures are the same as described before (Simoneit, 1977, 1984; Simoneit and Mazurek, 1982; Simoneit et al., 1999). Briefly, the filter samples were extracted, within a few days after collection, using ultrasonic agitation for three 20-min periods using a dichloromethane/methanol mixture (3:1, v/v). The solvent extracts were filtered using a Gelman–Swinney filtration unit containing an annealed glass fiber filter for the removal of insoluble particles and fibers (Simoneit and Mazurek, 1982). The filtrates were then concentrated by use of a rotary evaporator followed by a stream of filtered nitrogen gas. Initial analyses were carried out and the total extracts were subsequently stored in a freezer. For reanalysis, an aliquot of the total extract was derivatized with silylating reagent [*N,O*-bis(trimethylsilyl)trifluoroacetamide, BSTFA] with a trace of pyridine by heating at 60 °C for 2 h in a sealed vial.

The total extracts and the derivatized extracts were then analyzed by capillary gas chromatography–mass spectrometry (GC–MS) using a Hewlett-Packard model 5973 MSD quadrupole mass spectrometer operated in

the electron impact mode at 70 eV and coupled to a Hewlett-Packard model 6890 gas chromatograph. The GC was equipped with a 30 m×0.25 mm i.d. capillary column coated with DB-5 (J&W Scientific, film thickness 0.25 μm). The GC was operated using the temperature program of 65 °C for 2 min, ramp to 300 °C at 6 °C/min, and hold isothermal at 300 °C for 20 min. Helium was the carrier gas. GC–MS data were acquired and processed with a Hewlett-Packard Chemstation. Compounds were identified by GC retention indices, mass spectra and comparison with authentic standards. Levoglucosan (1,6-anhydro-β-D-glucopyranose, CAS number 498-07-7) is available from Fluka/Aldrich Chemical Co. It is typically characterized by its base peak at *m/z* 204 and by *m/z* 217 and 333 in mass fragmentationograms. The mass spectrum of levoglucosan trimethylsilyl ether (MS is shown in Simoneit et al. (1999)) exhibits only a small molecular ion (*m/z* 378) with fragments due to loss of CH<sub>3</sub> (*m/z* 363), CH<sub>5</sub>Si (*m/z* 333), C<sub>6</sub>H<sub>17</sub>OSi<sub>2</sub> (*m/z* 217) and C<sub>7</sub>H<sub>18</sub>OSi<sub>2</sub> (*m/z* 204, base peak). The mass spectra of the other monosaccharide dehydration derivatives (galactosan and mannosan) are similar and were identified by comparison with synthetic standards. A calibration curve for the response factor was prepared for levoglucosan-TMS and the precision of the determination is typically ±10% (Elias and Medeiros, unpublished results).

## 3. Results and discussion

Particulate matter in smoke from biomass burning is the major organic component in the ambient atmospheric samples collected at most of the Malaysian stations during haze episodes. Levoglucosan and other minor monosaccharide derivatives are present in all aerosol samples analyzed. The data for total suspended particles (TSP), total extract yields and key biomarker tracers are listed in Table 1. The particle concentrations during the haze episode of 1994 reflect the proximity of the locations to the source, i.e., the Island of Sumatra, Indonesia. Malacca, being closest, exhibits the highest TSP concentration with 17% being extractable. Based on the same argument, the TSP concentration at Senai is expected to be higher than that of Petaling Jaya, but the result shows otherwise. However, the percent extractable is higher at Senai (18%) compared to that at Petaling Jaya (12%). This is due to the fact that biomass burning is not the only source of TSP at Petaling Jaya (Abas and Simoneit, 1996; Fang et al., 1999; Okuda et al., 2002). Tanah Rata, being a remote northern station has the lowest TSP concentration, while the TSP concentration at Kuantan, on the eastern coast of the Peninsula, is the second lowest. Comparing the TSP concentrations of the 1994 haze episode and that of 1997, it is obvious that

Table 1

Concentrations of particles, extractable organic matter and key tracers from aerosol samples of Malaysia during the 1994 and 1997 haze episodes

Sample	Date/ conditions	Total suspended particles ( $\mu\text{g}/\text{m}^3$ )	Total solvent extract ( $\mu\text{g}/\text{m}^3$ )	Levoglucosan ( $\text{ng}/\text{m}^3$ )	Palmitic acid ( $\text{ng}/\text{m}^3$ )	<i>n</i> -Hen- triacontane ( $\text{ng}/\text{m}^3$ )	<i>n</i> -Octa- cosanol ( $\text{ng}/\text{m}^3$ )
Petaling Jaya	10384 (9/94) haze	227	28	3480	440	90	220
University of Malaya (Petaling Jaya)	Day (9/97) haze Night (9/97) haze	501 297	94 96	18 800 34 000	230 170	88 49	180 136
Malacca	5267 (9/94) haze	250	42	12 100	302	61	180
Tanah Rata	4720 (9/94) haze	40	14	270	168	68	50
Senai	8271 (9/94) haze	140	25	120	35	22	60
Kuantan	8351 (9/94) haze	97	20	910	240	70	126
Taman Negara	(4/96) clear	n.d.	0.1	10	18	7	16
Petaling Jaya	(9/91) haze	300	24	5520	110	900	30
Kuala Lumpur <sup>a</sup>	(5/92) clear	74	5	n.d.	n.d.	n.d.	n.d.

n.d. = not determined.

<sup>a</sup>Data given for comparison purposes (Abas and Simoneit, 1996).

latter haze episode was more severe. The TSP concentration during the haze of 1991 in Kuala Lumpur is also high, but the total solvent extractable material represents only 8% of the particles.

The salient features of the GC–MS data for typical urban and rural stations are shown in Fig. 2. The major resolved compound in all samples is levoglucosan(I). Because of its universal appearance in all wood smokes, levoglucosan has been proposed previously as an important tracer for use in tracking wood smoke in the atmosphere (Simoneit et al., 1999). The typical urban components from traffic (i.e., petroleum derived hydrocarbons, e.g., Simoneit, 1984; Simoneit et al., 1991; Abas and Simoneit, 1996; Zheng et al., 1997) are overwhelmed by the levoglucosan predominance (e.g., Fig. 2a, b and d) and are determined by key ion fragmentograms as discussed in the Experimental section.

The contribution of haze from biomass burning/wildfires to the aerosol of Petaling Jaya in September 1991 could be confirmed by the high levoglucosan concentration (Fig. 2a, Abas and Simoneit, 1996). Levoglucosan, galactosan, and mannosan are sugar anhydrides formed during combustion of cellulose and hemicellulose, and therefore their presence in these samples implies that the particulate matter is impacted by smoke from biomass burning (Simoneit et al., 1999). The series of urban and rural aerosol samples from the haze period during September 1994 and September 1997 also contains high levels of levoglucosan (Table 1),

confirming the smoke overprint on the ambient particulate matter. Some samples also contain minor amounts of galactosan(II) and mannosan(III), as well as other monosaccharide derivatives. The highest levoglucosan concentrations are observed over central western Peninsular Malaysia, intermediate in the south and low in the northern rural and eastern coastal areas. The presence of levoglucosan in the aerosol from Taman Negara during a clear day is suspected to have come from campfires or firewood burning in nearby villages.

The sample from the University of Malaya contains major amounts of  $\alpha$ - and  $\beta$ -mannose (IV) primarily during the day ( $100 \mu\text{g}/\text{m}^3$ ) and traces during the night ( $2.4 \mu\text{g}/\text{m}^3$ , Fig. 2c and d). Mannose is a significant saccharide in many plants and thus is inferred to possibly be derived from the sugar products or beer brewing industries. There is a brewery about 6 km to the northwest of the station at the University of Malaya. No source emission profiles for breweries are known, but based on the saccharide composition a smoke (levoglucosan) to mannose ratio can be calculated as 0.34 during the day and 5.0 during the night.

The next major compound group consists of fatty acids, with palmitic acid as the dominant homolog (Table 1). The  $\text{C}_{16}$  and  $\text{C}_{18}$  fatty acids are the most prominent alkanolic acids found in urban atmospheric fine particulate matter (Rogge et al., 1993b) and interpreted to be of a biogenic origin from both natural and burning emissions. The homologs  $<n\text{-C}_{20}$  may be

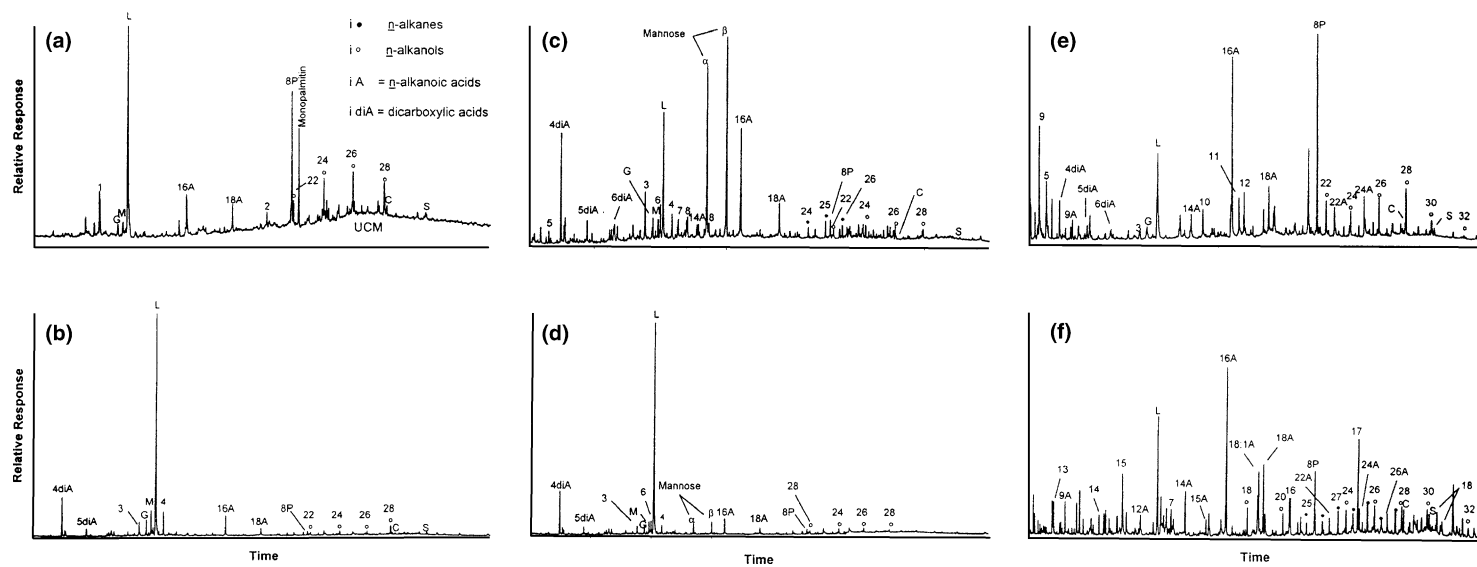


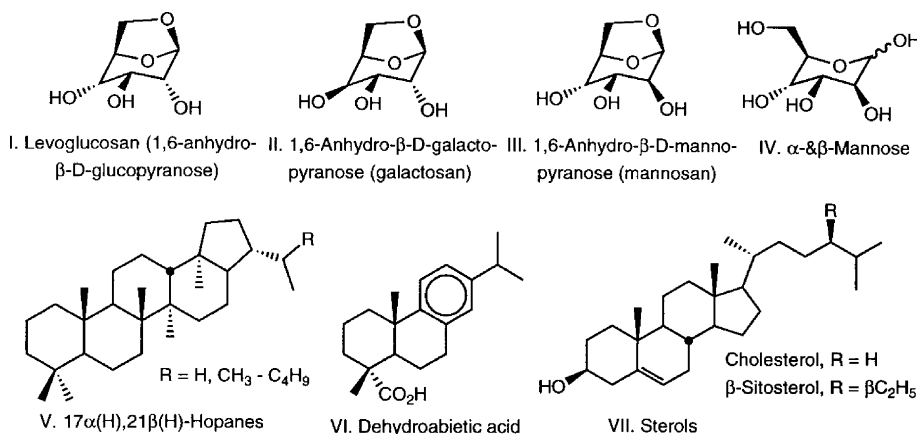
Fig. 2. Annotated total ion current traces of typical GC-MS analyses of total aerosol particle extracts (silylated): (a) Petaling Jaya (9/91) haze, (b) University of Malaya, Station 1, daytime (9/97) haze, (c) University of Malaya, Station 2, daytime (9/97) haze, (d) University of Malaya, Station 2, nighttime (9/97) haze, (e) Senai (9/94) haze, and (f) Taman Negara National Park (4/96) clear, [L=levoglucosan, G=galactosan, M=mannosan, 16M=methyl palmitate, 16A=palmitic acid, C=cholesterol, S= $\beta$ -sitosterol,  $\circ$ =alkanol, UCM=unresolved complex mixture of hydrocarbons, numbers over peaks indicate carbon chain length of alkanes ( $\bullet$ ), alkanols ( $\circ$ ) or alkanolic acids (A)].

derived in part from microbial sources, although these acids are ubiquitous in all biota (Simoneit and Mazurek, 1982). Another source in urban areas is from cooking, grilling and food preparation as well as burning, where these alkanolic acids are volatilized directly into the fumes (Rogge et al., 1991). However, the major resolved compound in smoke from the burning of bamboo leaves and twigs, sugar cane bagasse and forest litter from Malaysia is palmitic acid (Abas, unpublished data). Therefore, the most important source of fatty acids  $<C_{20}$  with a dominance of palmitic acid in the current samples is from biomass burning.

### Acknowledgements

We thank V.O. Elias, A.I. Rushdi and A. Otto for technical assistance and the US Environmental Protection Agency for partial financial assistance (Grant CR823990-01-0). We also acknowledge the assistance of the Director-General of the Malaysian Meteorological Service Department for providing some of the aerosol particulate samples.

### Appendix A. Chemical structures cited in the text



Other representative compounds characteristic of vegetation wax are *n*-hentriacontane and *n*-octacosanol (Table 1). The presence of these wax compounds is indicative of a contribution of particulates from abrasion of vegetation leaf surfaces and from biomass burning. Cholesterol,  $\beta$ -sitosterol, vanillic acid and syringic acid are also detectable as minor components. These compounds are derived from biomass burning and cholesterol is derived from meat cooking or algae (Simoneit, 2002).

### 4. Conclusion

Levoglucosan and palmitic acid are present at high concentrations in atmospheric particulate matter impacted by smoke from biomass burning. Several other compounds emitted from burning, such as mannosan, galactosan, lignin breakdown products and plant wax components, are also found at lower concentrations. The presence of these smoke derived compounds overwhelmed those from common sources normally encountered in urban atmospheres.

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