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Molecular characterization of urban organic aerosol in tropical India: contributions of biomass/biofuel burning, plastic burning, and fossil fuel combustion

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Abstract

Organic molecular composition of PM_{10} samples, collected at Chennai in tropical India, was studied using capillary gas chromatography/mass spectrometry. Twelve organic compound classes were detected in the aerosols, including aliphatic lipids, sugar compounds, lignin products, terpenoid biomarkers, sterols, aromatic acids, phthalates, hopanes, and polycyclic aromatic hydrocarbons (PAHs). At daytime, phthalates was found to be the most abundant compound class; while at nighttime, fatty acids was the dominant one. Concentrations of total quantified organics were higher in summer (611–3268 ng m^{-3}, average 1586 ng m^{-3}) than in winter (362–2381 ng m^{-3}, 1136 ng m^{-3}), accounting for 11.5±1.93% and 9.35±1.77% of organic carbon mass in summer and winter, respectively. Di-(2-ethylhexyl) phthalate, C_{16} fatty acid, and levoglucosan were identified as the most abundant single compounds. The nighttime maxima of most organics in the aerosols indicate a land/sea breeze effect in tropical India, although some other factors such as local emissions and long-range transport may also influence the composition of organic aerosols. The abundances of anhydrosugars (e.g., levoglucosan), lignin and resin products, hopanes and PAHs in the Chennai aerosols suggest that biomass burning and fossil fuel combustion are significant sources of organic aerosols in tropical India. Interestingly, terephthalic acid was maximized at nighttime, which is different from those of phthalic and isophthalic acids. A positive correlation was found between the concentration of 1,3,5-triphenylbenzene (a tracer for plastic burning) and terephthalic acid, suggesting that field burning of municipal solid wastes including plastics is a significant source of terephthalic acid. This study demonstrates that, in addition to biomass burning and fossil fuel combustion, the open-burning of plastics also contributes to the organic aerosols in South Asia.

1 Introduction

Primary organic aerosol (POA, particle mass directly emitted from sources such as plant material, soil dust, biomass and fossil fuel burning) and secondary organic aerosol (SOA, particle mass formed in the atmosphere from the oxidation of gas-phase precursors) are ubiquitous in the atmosphere (Robinson et al., 2007). They account for up to 70% of the fine aerosol mass, and potentially control the physicochemical properties of atmospheric particles (Kanakidou et al., 2005). Organic aerosols are highlighted for the past decade because they are important environmental issues related to global and regional climate, chemistry of the atmosphere, biogeochemical cycling, and people's health (Crutzen and Andreae, 1990; Kanakidou et al., 2005; Pöschl, 2005; Andreae and Rosenfeld, 2008).

India has experienced serious air pollution problem due to the rapid economic growth and urbanization in the past decade. It is regarded as a major source region to the Indo-Asian haze, due to significant industrial emissions, coal burning, vehicle exhaust emission, and waste incineration (Lelieveld et al., 2001). Indo-Asian haze is also known as atmospheric brown clouds (ABCs), which consists of a persistent and large-scale layer of air pollutants containing a mixture of black carbon (BC), organic carbon (OC), and dust. ABCs significantly absorb and scatter solar radiation (Lelieveld et al., 2001; Ramanathan et al., 2005; Seinfeld, 2008; Szidat, 2009). It can impact on South Asian climate and hydrological cycle (Ramanathan et al., 2005). Efforts to reduce the extent of ABCs require the knowledge of their sources (Szidat, 2009). Biomass/biofuel (including wood, agricultural residues, and dried animal manure) burning and fossil fuel combustion are considered as the major sources of carbonaceous aerosols in this region (Lelieveld et al., 2001; Venkataraman et al., 2005; Stone et al., 2007). Recently, Gustafsson et al. (2009) used radiocarbon (^{14}C) as a tracer to quantify biomass and fossil fuel contributions to the ABCs. Chowdhury et al. (2007) reported the organic speciation and source apportionment of fine particles in four Indian cities using a receptor-based method. Furthermore, it is important to understand the changes in the organic aerosol composition due to photochemical oxidation at a molecular level (Robinson et al., 2006; Rudich et al., 2007). Tropical region may provide a unique site to study the photochemical aging of organic aerosols becuase of its high ambient temperature and strong sunligh irradiation. However, knowledge about the organic molecular composition of atmospheric aerosols in tropical India is still limited.

In this study, we present the concentrations of 155 organic compounds in tropical Indian aerosols. Based on the molecular distributions, their possible sources and seasonal/diurnal differences are reported. Contributions of each compound class to OC and water-soluble organic carbon (WSOC) in the samples are also discussed. Other water-soluble organic compounds such as low molecular weight dicarboxylic acids (Pavuluri et al., 2009a), hydroxy-/polyacids (e.g., glycolic, salicylic, and tricarballylic acids) and biogenic SOA tracers (e.g., 2-methyltetrols, pinic acid, and β-caryophyllinic acid) (Fu et al., 2009b) are discussed elsewhere.

2 Experimental

2.1 Aerosol sampling

Day- and night-time PM_{10} samples were collected on the campus of the Indian Institute of Technology Madras (IITM), Chennai (13.04° N; 80.17° E), tropical India during winter (23 January–6 February, n=29) and summer (23–31 May, n=20) 2007. IITM campus is located in a natural forest, covered with vegetations, and is ca. 3 km away from the coast. Air sampler was set-up on a rooftop of the Mechanical Sciences building (18 m, above ground level). Sampling was conducted during daytime (06:00–18:00, local time) and nighttime (18:00–06:00) using a high volume air sampler (Envirotech APM 460 DX, India) and pre-combusted (450°C, 4 h) quartz fiber filters (Pallflex 2500QAT-UP, 20×25 cm). The sample was placed in a pre-combusted glass jar with a Teflon-lined screw cap and stored in a dark freezer room at –20°C prior to analysis.

2.2 Extraction, derivatization, and GC/MS determination

Detailed analytical method has been described elsewhere (Fu et al., 2008). Briefly, filter aliquots were extracted with dichloromethane/methanol (2:1, v/v), followed by concentration, and derivatization with 50 µl of N,O-bis-(trimethylsilyl)trifluoroacetamide (BSTFA) with 1% trimethylsilyl chloride and 10 µl of pyridine at 70°C for 3 h. After reaction, the derivatives were diluted by the addition of 140 µl of *n*-hexane with 1.43 ng μl^{-1} of the internal standard (C_{13} *n*-alkane) prior to GC/MS injection. GC/MS analyses of the samples were performed on a Hewlett-Packard model 6890 GC coupled to Hewlett-Packard model 5973 mass-selective detector (MSD). The GC instrument was equipped with a split/splitless injector and a DB-5MS fused silica capillary column (30 m×0.25 mm i.d., 0.25 µm film thickness). The mass spectrometer was operated in the electron impact (EI) mode at 70 eV and scanned from 50 to 650 Da. Data were acquired and processed with the Chemstation software. GC/MS response factors were determined using authentic standards. Recoveries of the quantified organic compounds were generally better than 80%. Field blank filters were treated as real samples for quality assurance. The results showed no significant contamination (less than 5% of real samples). The data reported here were corrected for the field blanks but not for recoveries.

Detailed procedures for the measurement of OC and WSOC are described elsewhere (Pavuluri et al., 2009b). Briefly, OC was determined using a Sunset Lab EC/OC Analyser following the Interagency Monitoring of Protected Visual Environments (IMPROVE) thermal evolution protocol. An aliquot of each filter was also analysed for WSOC. The filter aliquot was first extracted with 10 ml organic-free pure water by ultrasonication for 10 min. The water extracts were then filtered using a pre-rinsed syringe filter (Millex-GV with 0.22 µm pore size, Millipore). WSOC in the water extracts was measured using a TOC-5000A (Shimadzu).

3 Results and discussion

3.1 Meteorology and air mass back trajectories

Detailed weather information has been mentioned elsewhere (Pavuluri et al., 2009a). In brief, the weather in Chennai is generally hot and humid. Ambient temperature during the campaigns varied from 14.2–34.9°C (average 23°C) in winter and 28.3–41°C (32°C) in summer. No rain was recorded during the campaigns. A clear diurnal oscillation in wind speed and wind direction was found in Chennai due to a strong land-sea thermal gradient. The onset of sea breeze at daytime that introduce cool marine air passing over a warmer land surface results in a thermal internal boundary layer (TIBL) below the planetary boundary layer (PBL). In contrast, the onset of land breeze at nighttime may remove the TIBL and the PBL moves down.

Air mass trajectory analysis (HYSPLIT, NOAA) showed that most of the air masses were transported long distances from North India and the Middle East in early winter (23–28 January) and from Southeast Asia over the Bay of Bengal in late winter (29 January–6 February). In contrast, the Arabian Sea, Indian Ocean and South Indian continent are suggested as major source regions in summer (22–31 May) (Pavuluri et al., 2009a). Back trajectory analysis also showed that the air masses originated from mixed regions (North India and Southeast Asia) between 30 January and 2 February.

3.2 Speciation of particulate organic compounds

Homologous of 12 organic compound classes, i.e., *n*-alkanes, fatty acids, fatty alcohols, anhydrosugars, sugars/sugar alcohols, lignin products, terpenoid biomarkers, sterols, aromatic acids, phthalate esters, hopanes, and polycyclic aromatic hydrocarbons (PAHs) were detected in the tropical Indian aerosols. Table 1 presents the concentrations of more than 150 organic compounds detected in this study. Among the detected organic compounds, fatty acids and phthalates are the major compound classes, followed by *n*-alkanes and anhydrosugars. Other compound classes are relatively minor. Figure 1 presents a typical GC/MS traces at total ion current (TIC) for the samples. As a single compound detected (on average), di-(2-ethylhexyl) phthalate (DEHP) was the most abundant one during summer and winter, followed by levoglucosan, diisobutyl phthalate (DiBP), and $C_{16:0}$ fatty acid in winter (See Fig. 1 and Appendix A for chemical structures). However in summer, levoglucosan became less abundant than those of $C_{16:0}$ fatty acid and DiBP.

3.3 Day-/nighttime variations in the concentrations: Effect of land/sea breeze

As shown in Figs. 2 and 3, most of the organic compound classes were more abundant at nighttime than at daytime. For example, *n*-alkanes, fatty acids, fatty alcohols, terpenoid biomarkers, and sterols showed clear nighttime maxima. This feature may be associated with the land/sea breeze circulation in Chennai (Pavuluri et al., 2009a). However, during late winter, the differences between day- and nighttime concentrations of most of the compound classes were minor. This is reasonable because the land/sea breeze effect was less important during late winter when the air masses originated from Southeast Asia over the Bay of Bengal. As seen in Fig. 2, anhydrosugars (levoglucosan and its two isomers, galactosan (II) and mannosan (III), the tracers for biomass burning (Simoneit, 2002), showed higher daytime concentrations (average 235 ng m^{-3}) than nighttime (156 ng m^{-3}) in late winter. This demonstrates that biomass burning events in Southeast Asia are very active and the atmospheric transport of smoke aerosols from this region is more significant than those of local emissions in tropical India during late winter.

3.4 Biomass burning tracers

Levoglucosan, formed during the pyrolysis of cellulose, has been detected in urban (Wang et al., 2006a; Wang et al., 2006b; Zheng et al., 2006; Chowdhury et al., 2007; Yttri et al., 2007), marine (Simoneit and Elias, 2000; Simoneit et al., 2004; Wang et al., 2009), and polar regions (Stohl et al., 2007; Fu et al., 2009a). In this study, levoglucosan was found to be one of the most abundant species among the individually identified compounds with a concentration range of 50.7–213 ng m^{-3} (average 111 ng m^{-3}) in summer (Table 1), which are similar to those reported in Delhi (210±40 ng m^{-3}) and Kolkata aerosols (75±15 ng m^{-3}) (Chowdhury et al., 2007). While in winter, the concentrations of levoglucosan ranged from 4.30 to 361 ng m^{-3} (112 ng m^{-3}), which are much lower than those reported in Delhi (5300±1100 ng m^{-3}), Kolkata (5500±1100 ng m^{-3}), and Mumbai aerosols (910±180 ng m^{-3}) (Chowdhury et al., 2007). However, our results are 1–2 orders of magnitude higher than those reported in the Indian Ocean aerosols collected at the Maldives climate observatories in Gan (12.3–25.4 ng m^{-3}) and Kanimaadhoo (1.97–8.62 ng m^{-3}) sites in the ABC monitoring network (Stone et al., 2007).

Seven lignin products (vanillin, shonanin, and 3-hydroxylbenzoic, 4-hydroxybenzoic, 3,4-dihydroxybenzoic, vanillic, and syringic acids) were detected in the aerosols with a total average concentration of 19.3±11.2 ng m^{-3} in winter and 12.3±3.49 ng m^{-3} in summer (Fig. 3f). 4-Hydroxybenzoic acid (IV) was the dominant species in both winter and summer, followed by 3,4-dihydroxybenzoic acid. 3,4-Dihydroxybenzoic acid (protocatechuic acid) is a antioxidant that has a polyphenolic structure. In human health, polyphenol antioxidants are thought to be instrumental in combating oxidative stress, a process associated with some neurodegenerative diseases and some cardiovascular diseases (http://en.wikipedia.org/wiki/Polyphenol_antioxidant). These hydroxy acids have been reported in continental aerosols (Fu et al., 2008) and smoke particles (Oros and Simoneit, 2001a, b; Simoneit, 2002). Vanillic acid (V) is a source specific tracer for conifers (Simoneit, 2002). Vanillin (VI) and syringic acid (VII) are detectable in pine wood smoke (Simoneit, 2002). Shonanin (VIII) is a predominant lignan of soft wood smoke (e.g., pine). These tracers showed temporal trends similar to each other with higher concentrations in winter than in summer (Fig. 4d–i).

Terpenoid biomarkers are present in vegetation smoke, both as natural and thermally altered products (Medeiros and Simoneit, 2008). Dehydroabietic acid (IX) was the dominant diterpenoid in the aerosols, followed by 7-oxodehydroabietic acid (X), indicating the predominance of softwood burning. Regular diterpenoid acids such as abietic acid (XI) or pimaric acid (XII) were also detectable (Table 1). α-/β-Amyrins (XIV, XV) have been reported in soft wood smokes (Oros and Simoneit, 2001b; Simoneit, 2002; Medeiros and Simoneit, 2008), Amazonian smoke aerosol (bin Abas et al., 1995), and urban aerosols in China (Simoneit et al., 1991). They are triterpenoids that are important biomarker constituents of many vascular plants, especially in the gums and mucilages of angiosperms. The concentration ranges of $\alpha+\beta$-amyrin were 0.07–5.96 ng m^{-3} (average 1.93 ng m^{-3}) in winter and 0.34–3.60 ng m^{-3} (1.22 ng m^{-3}) in summer with a temporal pattern similar to those of dehydroabietic acid or shonanin (Fig. 4h–j).

In India, biofuel is a major domestic energy source for cooking and heating. Cholesterol (XVI) is a tracer for smoke particles generated from meat cooking and also has been proposed as a source marker of marine organisms (Simoneit and Elias, 2000). Stigmasterol (XVII) is a tracer used to identify cow dung smoke (Sheesley et al., 2003). β-Sitosterol (XVIII), together with stigmasterol, is present in terrestrial higher plants and emitted to the air via biomass burning (Simoneit, 2002; Kawamura et al., 2003). These sterols were more abundant at nighttime than at daytime, especially in winter (Fig. 4k–m). Their total concentrations were 1.55–195 ng m^{-3} (55.0 ng m^{-3}) in winter and 2.14–119 ng m^{-3} (24.8 ng m^{-3}) in summer (Fig. 3h). The abundance of sterols, lignin products, and terpenoid biomarkers in the troposphere over tropical India, as well as anhydrosugars, indicates that biomass/biofuel burning is an important source of organic aerosols in this region, especially during nighttime in winter.

3.5 Plastics emission

Plastics are versatile polymeric materials produced and used worldwide (Simoneit et al., 2005). Phthalate esters (phthalates) are used as plasticizers in resins and polymers. They can be released into the air from the matrix by evaporation because they are not chemically bonded to the polymer. Attention has been paid to phthalates due to their potential carcinogenic and endocrine disrupting properties (Sidhu et al., 2005;

Swan, 2005). Five phthalates were detected in this study, i.e., dimethyl, diethyl, diisobutyl, di-*n*-butyl, and di-(2-ethylhexyl) (XIX) phthalates (Fig. 5). The concentrations of phthalates were 295–857 ng m^{-3} (553 ng m^{-3}) in summer versus 175–598 ng m^{-3} (303 ng m^{-3}) in winter (Table 1). Higher concentrations observed in summer (Fig. 3j) may be caused by enhanced emission of phthalates from plastics because of the ambient temperature. Similarly, Wang et al. (2006a) also reported that summertime concentrations of phthalates were higher than those in wintertime in China. However in Chennai, nighttime concentrations of DEHP were relatively higher than those at daytime (Fig. 5). This feature is different from other studies and will be discussed below.

In India, most of the municipal solid wastes are generally disposed into open landfills, within which there are a large amount of plastics. Plastics are readily combustible and under open-fire conditions generate numerous compounds into the atmosphere (Simoneit et al., 2005). Simoneit et al. (2005) reported that 1,3,5-triphenylbenzene (XXI) can be used as specific tracer for open-burning of plastics, especially when coupled with the presence of the antioxidant tris(2,4-di-*tert*-butyl-phenyl)phosphate (TBPP, XX). A good correlation between 1,3,5-triphenylbenzene and TBPP was found (Fig. 6a). In this study, we detected TBPP in most of the samples with higher concentrations at nighttime than at daytime (Fig. 7), indicating an enhanced plastic burning at night, which has been also reported in Algiers metropolitan area (Yassaa et al., 2001). Such a refuse burning event may explain the higher concentrations of DEHP at nighttime than at daytime, because land breeze transports the burning products over the sampling site at night.

3.6 Fossil fuel combustions

Hopanes (hopanoid hydrocarbons, XXIII) are specific biomarkers of petroleum and coal (Simoneit et al., 1991; Rogge et al., 1993; Schauer et al., 1999, 2002). They may be emitted into the atmosphere from internal combustion engines and the use of coal. A series of hopanes (C_{27}–C_{35}, but no C_{28}, see Table 1 and Fig. 8) were detected in the Chennai aerosols with the dominance of $C_{29}\alpha\beta$. Such a molecular distribution has been reported in urban aerosols in China (Simoneit et al., 1991). Their average concentrations were 14.4±9.09 ng m^{-3} in winter and 4.97±1.91 ng m^{-3} in summer, which are higher than those reported in Chinese mega-cities (3.1±4.6 ng m^{-3}) (Wang et al., 2006a), Tokyo (0.7–15 ng m^{-3}, average 5.5 ng m^{-3}) in Japan (Kawamura et al., 1995), Auckland (5.7±4.3 ng m^{-3}) and Christchurch (2.0±2.4 ng m^{-3}) in New Zealand (Wang et al., 2006b), suggesting a severe air pollution in India. It should be noted that hopanes are expected to react with OH radical in the atmosphere, causing a loss of these compounds in ambient particles, especially in summer (Robinson et al., 2006).

Twenty PAHs (3- to 7-ring) were detected in the Chennai aerosols ranged from phenanthrene to dibenzo(a,e)pyrene. Their total concentrations were 35.7± 18.7 ng m^{-3} in winter versus 16.5±12.3 ng m^{-3} in summer, which are similar to those observed in Chinese mega-cities (28±4.8 ng m^{-3} in summer) (Wang et al., 2006a). The sources of PAHs include coal and natural gas combustion, automobile emissions, and biomass burning. The diagnostic ratios of IP/(IP+BghiP) were 0.44±0.04 at daytime and 0.45±0.03 at nighttime, suggesting a mixed source of traffic and coal burning. Among the PAHs detected, benzo(b)fluoranthene (BbF) was the dominant species in winter (Table 1 and Fig. 9). However in summer, 1,3,5-triphenylbenzene became the most abundant, especially at nighttime (Figs. 4t and 9). This suggests that severe plastic burning activities frequently happened during summer nighttime in tropical India as mentioned above.

3.7 Aromatic acids

Seven aromatic acids, including benzoic acid, three toluic acid (*o*-, *m*-, and *p*-isomers), and three phthalic acids (*o*-, *m*-, and *p*-isomers) were detected in the samples. They can play an important role to enhance the atmospheric new particle formation (Zhang et al., 2004). The total concentrations of aromatic acids in average were 36.5±21.9 ng m^{-3} in winter versus 51.8±33.9 ng m^{-3} in summer. Except terephthalic acid (Fig. 4s), all the aromatic acids showed higher concentrations at daytime than at nighttime (Fig. 10), indicating a photochemical source.

Benzoic acid is proposed to be a primary pollutant in the exhaust of motor vehicles (Kawamura and Kaplan, 1987; Rogge et al., 1993; Kawamura et al., 2000) and a secondary product from photochemical degradation of aromatic hydrocarbons emitted by automobiles (Suh et al., 2003). The concentration ranges of benzoic acid were 0.54–2.16 ng m^{-3} (average 1.25 ng m^{-3}) in winter and 0.95–2.56 ng m^{-3} (1.44 ng m^{-3}) in summer (Table 1). The concentration ranges of total toluic acids were 0.15–1.18 ng m^{-3} (0.62 ng m^{-3}) in winter and 0.33–1.09 ng m^{-3} (0.59 ng m^{-3}) in summer. They may be derived from the oxidation of xylene (Forstner et al., 1997) or directly emitted from motor vehicle exhausts (Kawamura et al., 2000). The relatively low detection of benzoic and toluic acids in the tropical Indian aerosols may be explained by their presence mainly in the gaseous phase (Kawamura et al., 2000; Fraser et al., 2003). The molecular distribution of phthalic acids was characterized by a predominance of terephthalic acid (Fig. 10). This pattern is different from those reported in aerosols from other studies that phthalic acid was generally found to be the dominant one (Wang et al., 2006a; Fu et al., 2008). Interestingly, a good correlation was found between 1,3,5-triphenylbenzene and terephthalic acid, while no correlation between 1,3,5-triphenylbenzene and phthalic acid in this study (Fig. 6b), suggesting that terephthalic acid can be produced by the burning of plastics as well.

3.8 Aliphatic lipids

Homologous *n*-alkanes were detected in a range of C_{18}–C_{40} with higher concentrations at nighttime (Fig. 11). Their concentration ranges were 30.9–727 ng m^{-3} (average 187 ng m^{-3}) in summer and 15.2–437 ng m^{-3} (141 ng m^{-3}) in winter. Their molecular distributions are characterized by weak odd-carbon-numbered predominance with a maximum at C_{29} (CPI ranged from 1.17–2.34). Such a molecular distribution suggests that they are mainly derived from the incomplete combustion of fossil fuels and petroleum residue, especially for lower molecular weight *n*-alkanes such as C_{20}–C_{26}. However, higher molecular weight *n*-alkanes were likely derived from higher plant waxes, in which C_{27}, C_{29}, C_{31} and C_{33} are dominant species (Gagosian et al., 1982).

Interestingly, the carbon number of *n*-alkanes was up to 40 (Fig. 11). Such a feature has been reported in smokes from landfill plastic burning test (Simoneit et al., 2005). This again suggests that the open-burning of municipal wastes is an important source of organic aerosols in tropical India.

Plant wax *n*-alkanes are attributable to vascular plant waxes. They are calculated as the excess odd homologues compared to adjacent even homologues (Simoneit et al., 2004) and shown in Table 1. In summer, the concentration range of total plant wax *n*-alkanes was 4.85–76.3 ng m^{-3} (22.8 ng m^{-3}), which are comparable to those in winter (2.26–49.6 ng m^{-3}, 19.8 ng m^{-3}), suggesting that there is no significant difference in higher plant emissions between cold and warm seasons in tropical India. This feature may be associated with the vegetation types and relative high ambient temperature even in winter (14.2–34.9°C, av. 23°C) during the campaign.

A homologous series of straight chain fatty acids, including $C_{8:0}$–$C_{34:0}$ saturated and unsaturated acids (e.g., $C_{16:1}$ and $C_{18:1}$) were detected in the aerosols with higher nighttime concentrations (Figs. 3b and 4n). Concentration ranges were 40.5–657 ng m^{-3} (302 ng m^{-3}) in winter and 141–1448 ng m^{-3} (504 ng m^{-3}) in summer, which are higher than those of *n*-alkanes and fatty alcohols. Their distribution is characterized by a strong even carbon number predominance with two maxima at $C_{16:0}$ and $C_{24:0}$ (CPI ranged from 2.80–6.40, Table 1 and Fig. 12). A similar bimodal molecular distribution has been reported in continental and marine aerosols (Mochida et al., 2002; Kawamura et al., 2003; Fu et al., 2008).

The concentration ratios of lower molecular weight fatty acids (LFAs, $<C_{20:0}$) to higher molecular weight fatty acids (HFAs, $C_{20:0}$–$C_{34:0}$) were 5.3±1.8 in summer versus 1.4±0.8 in winter. These values are higher than those reported in Mt. Tai aerosols (average ratio of LFAs/HFAs was 1.02±0.80) in Central East China (Fu et al., 2008). HFAs are derived from terrestrial higher plant wax, while LFAs have multiple sources including vascular plants, microbes, marine phytoplankton, and kitchen emissions (Rogge et al., 1991; Schauer et al., 2001). Thus, our results indicate that much more LFAs may be emitted from microbial sources in tropical India due to high temperature and

humidity, especially in summer.

Unsaturated fatty acids are indicative of recent biogenic inputs from higher plants as well as microbial/marine sources. In urban environments, cooking, motor vehicles, and biomass burning can also be the major anthropogenic sources for these acids (Rogge et al., 1993, 1996). They can be rapidly oxidized once emitted to the atmosphere (Kawamura and Gagosian, 1987). Oleic acid ($C_{18:1}$) and linoleic acid ($C_{18:2}$) were detected as dominant species in the Chennai aerosols. Their total concentrations showed a clear diurnal trend peaked at nighttime (Fig. 4n), suggesting an enhanced photodegradation in the atmosphere at daytime. Moreover, significant concentrations of unsaturated fatty acids were observed at nighttime during late winter when the air mass mainly originated from Southeast Asia over the Bay of Bengal, indicating that marine air masses enriched with unsaturated fatty acids should be transported to Chennai.

Oleic acid is also a good proxy for unsaturated organic matter in atmospheric aerosols and a good model compound for studying aerosol reactivity (Rudich et al., 2007). The ratios of oleic acid to stearic acid ($C_{18:1}/C_{18:0}$) ranged from 0.01–0.18 (average 0.05) and 0.04–1.05 (0.61) during daytime and nighttime in winter, respectively. On the other hand, the ratios of $C_{18:1}/C_{18:0}$ decreased in summer and ranged from 0–0.05 (0.01) and 0.04–0.35 (0.10) during daytime and nighttime, respectively. This again suggests an enhanced photochemical degradation of oleic acid during daytime, especially in summer. Wang et al. (2006a) reported that the ratios of unsaturated fatty acids ($C_{16:1}+C_{18:1}$) to saturated fatty acids ($C_{16:0}+C_{18:0}$) in urban aerosols from fourteen Chinese megacities were 1.14 ± 0.98 in winter versus 0.43 ± 0.09 in summer. In the Chennai aerosols, these values (0.12 ± 0.15 in winter versus 0.01 ± 0.02 in summer) are about one order of magnitude lower than those reported in Chinese megacities, indicating that the photooxidation of unsaturated fatty acids in tropical region occurrs much more quickly under stronger radiation conditions than those in mid-latitudal regions.

Normal C_{14}–C_{34} fatty alcohols were detected in the aerosols. Their distribution are characterized with strong even carbon number predominance (CPIs are 9.75 ± 2.94 in winter and 10.9 ± 6.77 in summer) with C_{max} at C_{28} or C_{30} (Fig. 13). Concentrations of all identified *n*-alcohols were 11.0–116 ng m^{-3} (67.9 ng m^{-3}) in winter and 21.0–155 ng m^{-3} (70.4 ng m^{-3}) in summer (Fig. 3c). Long-chain fatty alcohols are abundant in higher plant waxes, soils, and loess deposits, whereas the homologues of $<C_{20}$ are abundant in soil microbes and marine biota (Simoneit et al., 1991). It should be noted that biomass burning can also produce a large amount of fatty alcohols, together with *n*-alkanes and fatty acids (Simoneit, 2002). The CPI values together with the molecular distributions of fatty alcohols suggested that they are mainly derived from waxes of higher plants and partly from microbial and/or marine emissions both in winter and in summer.

3.9 Sugars/sugar alcohols

Eleven sugar compounds (glycerol, erythritol, xylose, arabitol, fructose, glucose, mannitol, inositol, sucrose, trehalose, and maltose) were detected in the tropical Indian aerosols (Fig. 14). These compounds are completely water-soluble and thus contribute to WSOC in aerosol. They have been proposed as tracers for resuspension of surface soil and unpaved road dust, which contain biological materials including pollen, fungi and bacteria (Graham et al., 2003; Simoneit et al., 2004; Yttri et al., 2007). Concentrations of total sugars/sugar alcohols were more abundant in summer (29.5±7.83 ng m^{-3}) than in winter (20.0±11.8 ng m^{-3}) (Table 1), suggesting higher biological activities in summer.

Most of them were found to be more abundant at daytime than nighttime (Figs. 3e, 4o–q, and 14). Fructose and glucose, together with sucrose (XXV), have been proposed to be released as pollen, fern spores, and other "giant" bioaerosol particles during daytime (Graham et al., 2003). The direct emission from developing leaves is also an important source of sugars and sugar alcohols. It should be noted that sugar compounds can further be emitted through biomass burning. For example, maltose and sucrose have been reported in wood smoke (Nolte et al., 2001). Medeiros and Simoneit (2008) reported that a large amount of sugars (e.g., maltose) and sugar alcohols (e.g., erythritol (XXIV), arabitol, and mannitol) can be emitted upon combustion of green vegetation from temperate forests.

In order to get further insight to the sources of sugars in the tropical Indian aerosols, principal component analysis (PCA) was performed using the concentrations of various sugars (including anhydrosugars) as variables. PCA is a useful approach for verifying the sources of aerosols (Hopke, 1985) and has been successfully used for a series of dicarboxylic acids (DCAs) and other atmospheric trace species (Wolff and Korsog, 1985; Kawamura and Sakaguchi, 1999; Mochida et al., 2003; Wan and Yu, 2007; Fu et al., 2008). The datasets of sugars for winter (n=29) and summer (n=20) aerosol samples were subjected to PCA analysis based on their correlation matrix, followed by the varimax rotation of the eigenvectors. Principal component loadings, which are correlation coefficients between the concentrations of individual saccharides with principal components, are shown in Table 2. Three components were set for both winter and summer samples by the scree tests.

For the winter dataset, three components were found to account for 87.2% of the total variance, with the first component corresponding to 48.1%. Levoglucosan, galactosan, mannosan, erythritol, xylose, inositol, and maltose showed loadings of >0.87 in component 1, which is mainly associated with the emissions from biomass burning. In contrast, mannitol, sucrose, and trehalose showed loadings of >0.91 in component 2 (29.1%). Arabitol, fructose, and glucose also showed major loadings of >0.56 in component 2, suggesting a common biological origin. These sugar polyols are abundantly produced by many fungi. They are also the major soluble carbohydrates in the bark of trees, branches and leaves. Trehalose (glucose+glucose) is present in a large variety of microorganisms (fungi, bacteria and yeast), and a few higher plants and invertebrates (Medeiros et al., 2006). Sucrose is the dominant sugar in the phloem of plants and is important in developing flower buds (Bieleski, 1995). They are used to trace the resuspension of surface soil and unpaved road dust (Simoneit et al., 2004). Arabitol and fructose also show loadings of 0.62 and 0.55 in component 1, respectively. This suggests that during winter these compounds are emitted by biomass burning as well, as mentioned earlier in this section. Glycerol showed a loading of >0.89 in component 3 (10.0%). It is a reduced sugar primarily produced by fungal metabolism in soils and is resuspended into the atmosphere by wind motion (Simoneit et al., 2004). However, the differences of the estimated sources between component 2 and 3 can not be elucidated at this moment.

In summer, three components were found to account for 85.5% of the total variance, with the first component corresponding to 40.8%. Levoglucosan, galactosan, mannosan, erythritol, and xylose showed loadings of >0.91 in component 1 that is associated with biomass burning. Inositol and maltose also showed loadings of >0.74 in component 1, suggesting a significant contribution from biomass burning. Arabitol, glucose, mannitol, sucrose, and trehalose showed loadings of >0.86 in component 2 (31.8%). As mentioned above, these primary saccharides and sugar polyols could be derived from biological sources or suspended soil dust. Bauer et al. (2008) proposed arabitol and mannitol as tracers for the quantification of airborne fungal spores. Graham et al. (2003) reported that glucose and fructose, together with sucrose, showed higher daytime concentrations, which were explained by the specific daytime release of pollen, fern spores and other bioaerosols in summer. Glycerol and fructose showed loadings of 0.78 and 0.84 in component 3 (12.9%), respectively, suggesting that these two compounds are derived from similar sources in tropical India in summer.

3.10 Contributions to OC and WSOC

To better understand the chemial composition of organic aerosols in tropical India, contributions of each compound class to OC and WSOC in the samples were examined (Table 3). Total organics indentified in the tropical Indian aerosols accounted for 6.04–13.8% (average 9.35%) of OC in winter and 7.70–15.0% (11.5%) in summer. Sugar compounds (including anhydrosugars and sugar alcohols) accounted for 0.09–1.67% (0.69%) of OC in winter and 0.40–1.06% (0.73%) of OC in summer. They are lower than those reported in urban aerosols from Hong Kong (0.3–3.6%, average 1.3%) (Wan and Yu, 2007). Although anhydrosugars comprise a similar fraction in OC (average

0.59% in winter and summer), contributions of other biomass burning tracers such as lignin products, terpenoid biomarkers, sterols to OC are 2–3 times higher in winter than in summer. This indicates that intensive biomass/biofuel burning can more significantly affect the chemical composition of organic aerosols in tropical India during winter. In contrast, summertime samples showed more contributions from fatty acids and aromatic acids to OC. This suggests that both biological emission and photochemical production are more important in summer than winter. Similarly, phasticizers/antioxidants showed higher contributions to OC in summer due to the serious evaporation.

The percentage of levoglucosan to WSOC ranged from 0.07–3.49% (average 1.17%) in winter versus 0.70–1.97% (1.20%) in summer. These values are comparable to those reported in Mt. Tai aerosols in Central East China (average 1.50% at daytime versus 1.13% at nighttime) (Fu et al., 2008). The percentage of sugars/sugar alcohols to WSOC were 0.21±0.12% in winter and 0.30±0.09% in summer. The higher contribution of primary saccharides to WSOC in summer than in winter indicates that atmospheric sugar compounds may be associated with the enhanced release of primary bioaerosols or soil resuspention during summertime. Total water-soluble organics listed in Table 3 accounted for 1.90±1.10% and 1.87±0.46% of WSOC in winter and summer, respectively.

4 Conclusions

Concentrations of total quantified organic compounds in the tropical Indian aerosols were higher in summer (611–3268 ng m^{-3}, average 1586 ng m^{-3}) than in winter (362–2381 ng m^{-3}, 1136 ng m^{-3}). These organics accounted for 11.5±1.93% and 9.35±1.77% of OC in summer and winter, respectively. This suggests that the major portion of organic aerosols is still not revealed in this study in terms of chemical structure, which may include dicarboxylic acids (Pavuluri et al., 2009a), biogenic SOA, humic-like substances (Kanakidou et al., 2005; Graber and Rudich, 2006), and others.

The abundances of anhydrosugars, lignin products, terpenoid biomarkers, sterols,

hopanes, and PAHs suggest that both biomass/biofuel burning and fossil fuel combustion are important sources of organic aerosols in tropical India. Meanwhile, the detection of 1,3,5-triphenylbenzene, tris(2,4-di-*tert*-butyl-phenyl)phosphate, together with higher concentrations of terephthalic acid at nighttime than daytime suggests that the open burning of municipal solid wastes including plastics is also a significant source for atmospheric aerosols in this region. Such information may help to better understand the sources of atmospheric aerosols that cause the atmospheric brown clouds. These primary anthropogenic particles in South Asia or other developing regions should be constrained as environmental concerns.

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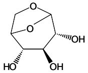
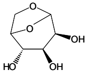
Appendix A

Chemical structure cited in the text



I. Levoglucosan



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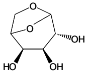

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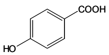
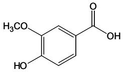


IV. 4-Hydroxybenzoic acid



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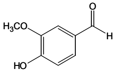
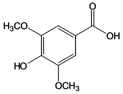
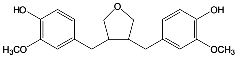


VI. Vanillin



VII. Syringic acid



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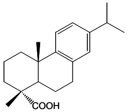
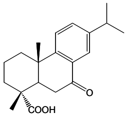
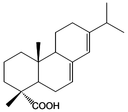


IX. Dehydroabietic acid



X. 7-Oxodehydroabietic acid



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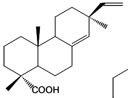

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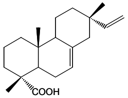

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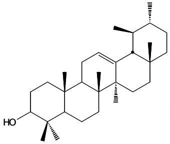
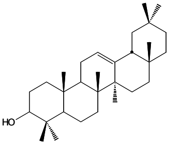
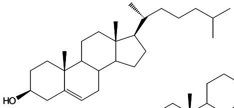


XIV. α-Amyrin



XV. β-Amyrin



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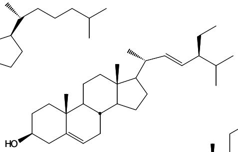
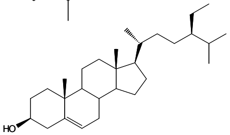
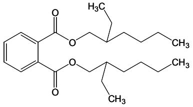


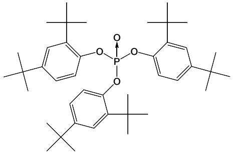
XVII. Stigmasterol



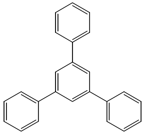
XVIII. β-Sitosterol



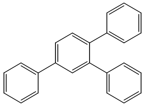
XIX. Di-(2-ethylhexyl) phthalate (DEHP)



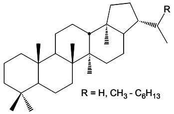
XX. Tris(2,4-*di*-*tert*-butyl-phenyl)phosphate (TBPP)



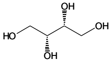
XXI. 1,3,5-Triphenylbenzene



XXII. 1,2,4-Triphenylbenzene



XXIII. 17α(H)-Hopanes



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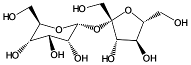

XXV. Sucrose

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Table 1. Concentrations of organic compounds detected in the atmospheric aerosols from Chennai, tropical India (ng m^{-3}).

Compounds	Winter (n=29)				Summer (n=20)			
	Min	Max	Average	SD[a]	Min	Max	Average	SD
n-Alkanes								
C_{18}	0.07	0.85	0.46	0.20	0.07	2.15	0.55	0.49
C_{19}	0.42	2.12	1.15	0.46	0.17	4.41	1.66	1.05
C_{20}	0.40	3.26	1.45	0.71	0.06	5.67	1.34	1.36
C_{21}	0.84	7.89	3.50	1.84	1.10	32.1	8.37	9.12
C_{22}	0.29	7.34	2.25	1.88	0.39	4.63	2.04	1.31
C_{23}	0.60	15.2	4.02	3.87	1.25	11.9	3.99	2.64
C_{24}	0.59	26.7	6.13	6.81	1.34	21.6	5.72	5.05
C_{25}	0.71	39.3	10.4	9.59	2.67	41.9	11.8	10.9
C_{26}	0.48	37.1	11.0	9.59	2.25	46.8	13.0	13.3
C_{27}	0.98	41.3	13.4	10.3	2.89	64.8	16.8	17.1
C_{28}	0.59	31.9	10.5	8.76	1.84	53.4	13.7	15.4
C_{29}	0.94	40.2	15.7	10.1	3.02	64.3	18.3	18.1
C_{30}	0.24	25.6	8.93	7.09	1.48	49.8	12.2	14.3
C_{31}	1.03	34.2	13.1	8.40	2.24	63.7	16.4	18.1
C_{32}	0.22	22.6	7.68	6.37	1.10	49.5	11.1	13.8
C_{33}	0.20	27.1	10.0	7.11	1.42	52.2	14.0	15.8
C_{34}	0.05	17.6	5.13	4.79	0.70	37.3	8.58	11.0
C_{35}	0.15	14.2	4.80	3.88	0.71	36.5	7.54	10.1
C_{36}	n.d.[b]	12.4	4.02	3.56	0.18	32.8	6.90	9.15
C_{37}	n.d.	16.6	3.23	3.80	0.25	43.5	7.39	12.3
C_{38}	n.d.	8.23	2.06	2.38	n.d.	23.0	3.91	6.32
C_{39}	n.d.	7.32	1.81	2.35	n.d.	10.4	1.46	2.71
C_{40}	n.d.	4.01	0.68	1.30	n.d.	5.56	0.65	1.36
Subtotal	15.2	437	141	110	30.9	727	187	200
CPI (C_{21}–C_{40})[c]	1.17	2.34	1.56	0.30	1.20	1.97	1.52	0.23
Plant wax-alkanes[d]								
C_{23}	0	0.62	0.12	0.15	0	0.76	0.28	0.28
C_{25}	0	7.38	1.79	1.58	0	7.67	2.42	2.00
C_{27}	0.32	6.77	2.59	1.68	0.79	14.7	3.40	3.08
C_{29}	0.52	11.4	6.02	2.56	1.33	12.7	5.36	3.31
C_{31}	0.80	10.1	4.79	2.02	0.95	23.3	4.81	4.86
C_{33}	0.06	7.50	3.59	2.06	0.52	14.8	4.23	3.67
C_{35}	0	3.04	0.42	0.67	0	1.49	0.20	0.40
C_{37}	0	6.84	0.50	1.31	0	15.6	2.09	4.83
Subtotal	2.26	49.6	19.8	9.67	4.85	76.3	22.8	19.8
II. Fatty acids								
$C_{8:0}$	0.54	2.40	1.02	0.45	0.86	2.43	1.58	0.41
$C_{9:0}$	0.19	2.05	0.74	0.38	0.86	12.1	3.40	3.39
$C_{10:0}$	0.11	0.79	0.46	0.14	0.47	1.84	0.97	0.45
$C_{11:0}$	n.d.	0.90	0.34	0.25	0.01	1.99	0.67	0.50
$C_{12:0}$	1.64	9.14	3.63	1.90	3.26	19.6	9.08	5.81

Table 1. Continued.

Compounds	Winter (*n*=29)				Summer (*n*=20)			
	Min	Max	Average	SD	Min	Max	Average	SD
$C_{13:0}$	0.18	1.35	0.66	0.31	0.56	2.18	1.15	0.43
$C_{14:0}$	1.85	21.4	8.00	4.40	7.80	86.4	24.6	18.6
$C_{15:0}$	0.38	4.67	2.38	0.99	3.29	14.7	6.53	3.25
$C_{16:0}$	6.45	217	90.9	63.4	77.7	780	275	218
$C_{17:0}$	0.29	5.27	2.54	1.51	1.27	14.6	5.19	4.03
$C_{18:0}$	2.04	110	43.7	33.2	14.4	287	85.4	83.9
$C_{19:0}$	0.01	3.31	1.46	0.92	0.49	6.53	2.22	1.85
$C_{20:0}$	0.45	14.6	6.49	4.44	1.27	24.5	7.31	7.26
$C_{21:0}$	0.18	4.18	1.98	1.00	0.45	6.70	2.34	1.90
$C_{22:0}$	0.72	29.6	12.6	7.75	1.84	28.7	9.38	8.64
$C_{23:0}$	0.45	10.7	6.08	2.54	0.87	14.4	5.09	4.21
$C_{24:0}$	1.51	49.5	23.3	11.8	3.02	36.5	13.9	9.61
$C_{25:0}$	0.60	9.25	5.41	1.93	0.75	12.2	3.93	3.26
$C_{26:0}$	1.56	32.6	19.8	7.87	2.11	27.6	10.4	6.77
$C_{27:0}$	0.58	6.87	4.14	1.39	0.47	9.49	2.89	2.48
$C_{28:0}$	1.31	25.4	15.4	5.31	2.21	22.9	10.1	5.40
$C_{29:0}$	0.42	6.08	3.79	1.26	0.50	6.26	2.55	1.79
$C_{30:0}$	1.52	19.1	11.4	3.43	1.61	13.2	6.82	3.19
$C_{31:0}$	0.06	3.12	1.63	0.74	0.14	4.55	1.34	1.16
$C_{32:0}$	0.15	8.74	5.36	1.83	0.74	6.34	2.88	1.34
$C_{33:0}$	n.d.	1.13	0.47	0.36	n.d.	1.29	0.47	0.33
$C_{34:0}$	n.d.	2.67	1.30	0.76	0.42	2.54	1.27	0.58
$C_{16:1}$	n.d.	2.03	0.25	0.45	n.d.	1.85	0.12	0.42
$C_{18:1}$	0.13	116	22.4	35.2	n.d.	74.8	7.19	16.8
$C_{18:2}$	n.d.	17.0	3.29	5.30	n.d.	2.67	0.17	0.61
$C_{20:1}$	n.d.	3.01	0.63	1.08	n.d.	n.d.	n.d.	n.d.
$C_{22:1}$	n.d.	3.34	0.45	0.93	n.d.	n.d.	n.d.	n.d.
Subtotal	40.5	657	302	179	141	1448	504	400
CPI ($C_{20:0}$–$C_{34:0}$)[c]	3.11	6.40	3.99	0.84	2.80	5.09	3.61	0.56
III. Fatty alcohols								
C_{14}	n.d.	0.95	0.27	0.23	n.d.	6.96	1.51	1.67
C_{15}	n.d.	0.64	0.19	0.15	0.03	1.32	0.33	0.29
C_{16}	0.10	4.18	1.72	1.05	0.50	6.86	2.58	1.53
C_{17}	0.03	0.67	0.24	0.16	0.25	1.35	0.72	0.31
C_{18}	0.24	3.04	1.10	0.67	0.98	13.9	3.58	3.03
C_{19}	0.01	3.22	1.15	1.04	0.31	13.1	3.21	3.57
C_{20}	n.d.	3.20	1.12	0.67	0.38	4.34	1.46	1.06
C_{21}	0.01	1.79	0.58	0.43	0.02	3.40	0.87	0.81
C_{22}	0.04	3.30	1.62	0.93	0.07	4.36	1.67	1.34
C_{23}	0.24	1.92	0.95	0.48	0.02	4.55	1.15	1.24
C_{24}	0.26	4.79	2.65	1.31	0.14	10.8	2.68	2.60
C_{25}	0.05	2.22	0.93	0.68	0.08	7.97	1.55	1.98
C_{26}	0.78	10.6	6.24	2.47	0.36	10.2	3.85	2.29
C_{27}	0.07	3.44	1.26	0.86	0.03	5.90	1.49	1.62
C_{28}	3.82	32.5	18.9	7.82	4.21	30.1	13.4	6.59

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Table 1. Continued.

Compounds	Winter (n=29)				Summer (n=20)			
	Min	Max	Average	SD	Min	Max	Average	SD
C_{29}	0.24	3.20	1.37	0.79	0.23	8.88	1.43	1.92
C_{30}	1.73	30.7	17.9	7.42	4.69	49.7	20.3	11.1
C_{31}	n.d.	1.26	0.57	0.29	0.04	0.80	0.40	0.18
C_{32}	n.d.	14.8	7.81	3.17	2.53	15.2	7.21	3.37
C_{33}	n.d.	1.41	0.35	0.36	n.d.	1.04	0.28	0.30
C_{34}	n.d.	3.57	0.92	0.97	n.d.	2.13	0.67	0.51
Subtotal	11.0	116	67.9	26.6	21.0	155	70.4	34.2
CPI (C_{20}–C_{34})^c	4.68	16.8	9.75	2.94	3.34	27.7	10.9	6.77
IV. Anhydrosugars								
Galactosan	0.35	23.7	8.07	6.22	2.71	10.8	5.84	2.12
Mannosan	0.26	42.6	13.4	12.0	3.75	20.3	9.90	4.52
Levoglucosan	4.30	361	112	95.4	50.7	213	111	45.2
Subtotal	4.99	427	134	112	59.6	244	127	51.6
V. Sugars/sugar alcohols								
Glycerol	1.13	7.34	2.59	1.38	2.83	18.7	5.49	3.47
Erythritol	0.28	6.99	2.74	1.88	1.40	5.20	3.20	0.94
Xylose	0.32	9.03	3.13	2.13	1.80	6.81	3.91	1.49
Arabitol	0.16	2.49	1.05	0.67	0.78	4.06	1.51	0.70
Fructose	0.16	4.98	1.51	1.14	0.69	2.72	1.53	0.61
Glucose	0.49	20.6	3.86	4.00	2.61	14.8	6.82	3.24
Mannitol	0.06	2.38	0.54	0.52	0.82	5.56	2.20	1.11
Inositol	0.03	1.37	0.48	0.30	0.30	0.87	0.53	0.15
Sucrose	0.29	13.8	2.06	2.68	0.83	7.65	2.23	1.54
Maltose	0.01	7.27	0.81	1.40	0.44	3.14	1.08	0.58
Trehalose	0.06	2.66	1.22	0.81	0.34	1.63	1.00	0.33
Subtotal	4.39	54.2	20.0	11.8	19.9	48.8	29.5	7.83
VI. Lignin products								
3-hydroxybenzoic acid	0.14	2.24	0.86	0.43	0.46	3.33	1.11	0.85
4-hydroxybenzoic acid	0.49	29.5	9.93	6.58	2.46	9.60	5.29	1.93
3,4-dihydroxybenzoic acid	0.11	7.80	3.58	2.46	0.86	9.04	3.85	2.33
Vanillin	0.10	3.97	1.35	0.90	0.23	1.15	0.56	0.19
Vanillic acid	0.04	2.28	1.00	0.62	0.33	1.08	0.79	0.20
Syringic acid	0.03	4.21	2.07	1.27	0.24	1.13	0.62	0.25
Shonanin	0.02	1.26	0.54	0.38	n.d.	0.27	0.09	0.08
Subtotal	1.18	45.4	19.3	11.2	5.85	18.6	12.3	3.49
VII. Terpenoid biomarkers								
Pimaric acid	n.d.	0.59	0.15	0.18	0.01	0.13	0.05	0.03
Isopimaric acid	n.d.	1.24	0.26	0.34	n.d.	0.19	0.05	0.04
Dehydroabietic acid	0.58	14.2	4.30	3.16	0.76	5.53	2.56	1.35
Abietic acid	n.d.	0.64	0.13	0.18	0.01	0.05	0.02	0.01
7-oxodehydroabietic acid	0.15	1.16	0.38	0.28	0.09	0.59	0.19	0.10
$\alpha+\beta$-amyrin	0.07	5.96	1.93	1.50	0.34	3.60	1.22	0.86
Subtotal	0.98	23.3	7.16	5.37	1.32	9.64	4.09	2.24
VIII. Sterols								
Cholesterol	0.30	17.5	5.30	5.31	0.11	29.1	5.06	7.89

Table 1. Continued.

Compounds	Winter (n=29)				Summer (n=20)			
	Min	Max	Average	SD	Min	Max	Average	SD
Stigmasterol	0.34	26.0	7.34	8.25	n.d.	12.4	2.48	3.57
β-sitosterol	0.42	156	42.3	50.1	1.70	77.8	17.3	18.6
Subtotal	1.55	195	55.0	63.2	2.14	119	24.8	29.3
IX. Aromatic acids								
Benzoic acid	0.54	2.16	1.25	0.39	0.95	2.56	1.44	0.40
o-toluic acid	0.00	0.30	0.16	0.07	0.04	0.40	0.15	0.09
m-toluic acid	0.07	0.38	0.22	0.08	0.13	0.35	0.22	0.07
p-toluic acid	0.06	0.53	0.24	0.10	0.08	0.34	0.22	0.08
Phthalic acid	0.42	31.2	5.79	5.87	1.92	11.5	5.82	2.81
Isophthalic acid	0.11	2.47	0.75	0.51	0.18	0.78	0.50	0.17
Terephthalic acid	1.61	111	28.1	21.0	11.8	117	43.4	34.8
Subtotal	3.90	118	36.5	21.9	15.9	124	51.8	33.9
X. Plasticizers/antioxidants								
Dimethyl phthalate (DMP)	n.d.	0.12	0.04	0.04	0.04	0.16	0.09	0.03
Diethyl phthalate (DEP)	0.32	23.0	4.00	5.05	0.26	15.8	5.03	4.54
Diisobutyl phthalate (DiBP)	13.5	373	104	66.2	7.44	294	127	68.7
Di-*n*-butyl phthalate(DnBP)	6.80	103	24.9	17.8	10.5	211	42.4	44.7
Di-(2-ethylhexyl) phthalate (DEHP)	96.2	270	171	43.5	177	579	379	118
Subtotal of phthalate esters	175	598	303	89.0	295	857	553	140
Tris(2,4-*di*-*tert*-butyl-phenyl)phosphate[e]	n.d.	1.46	0.33	0.30	n.d.	1.54	0.42	0.48
XI. Hopanes								
$C_{27}\alpha$	n.d.	1.36	0.39	0.36	n.d.	0.32	0.08	0.08
$C_{27}\beta$	n.d.	0.18	0.05	0.06	n.d.	0.11	0.01	0.03
$C_{29}\alpha\beta$	0.41	8.22	2.97	1.76	0.33	1.45	0.92	0.30
$C_{29}\beta\alpha$	n.d.	1.04	0.27	0.24	n.d.	0.22	0.08	0.07
$C_{30}\alpha\beta$	0.26	5.38	1.93	1.13	0.31	1.13	0.65	0.22
$C_{30}\beta\alpha$	n.d.	0.59	0.11	0.13	n.d.	0.28	0.05	0.06
$C_{31}\alpha\beta$S	n.d.	3.95	1.52	0.89	0.21	1.86	0.64	0.43
$C_{31}\alpha\beta$R	n.d.	4.14	1.32	0.88	0.17	1.29	0.47	0.27
$C_{31}\beta\alpha$	n.d.	0.94	0.24	0.24	n.d.	0.39	0.06	0.10
$C_{32}\alpha\beta$S	n.d.	3.20	1.25	0.77	0.17	1.06	0.50	0.26
$C_{32}\alpha\beta$R	n.d.	2.95	0.97	0.67	0.15	0.58	0.34	0.13
$C_{33}\alpha\beta$S	n.d.	2.50	0.80	0.56	0.10	0.67	0.29	0.13
$C_{33}\alpha\beta$R	n.d.	1.78	0.52	0.37	0.09	0.41	0.23	0.09
$C_{34}\alpha\beta$S	n.d.	2.28	0.67	0.50	n.d.	0.48	0.25	0.13
$C_{34}\alpha\beta$R	n.d.	2.05	0.52	0.43	n.d.	0.27	0.16	0.09
$C_{35}\alpha\beta$S	n.d.	1.95	0.51	0.42	n.d.	0.55	0.14	0.16
$C_{35}\alpha\beta$R	n.d.	1.56	0.38	0.32	n.d.	0.27	0.09	0.08
Subtotal	0.75	43.2	14.4	9.09	2.47	8.88	4.97	1.91
XII. PAHs								
1,2,4-triphenylbenzene	n.d.	0.68	0.20	0.18	0.08	1.24	0.31	0.33
1,3,5-triphenylbenzene	0.08	15.4	4.24	3.54	0.78	38.7	9.22	11.3
Phenanthrene	0.01	0.60	0.20	0.14	0.01	0.15	0.05	0.03
Anthracene	n.d.	0.16	0.05	0.04	n.d.	0.06	0.02	0.01
Fluoranthene	0.19	1.57	0.72	0.32	0.12	0.24	0.18	0.03

Table 1. Continued.

Compounds	Winter (n=29)				Summer (n=20)			
	Min	Max	Average	SD	Min	Max	Average	SD
Pyrene	0.18	1.69	0.81	0.35	0.19	0.38	0.27	0.06
Benzo(b)fluorine	0.01	0.59	0.20	0.13	n.d.	0.15	0.06	0.05
Benz(a)anthracene	0.07	2.51	0.97	0.58	0.12	0.33	0.20	0.05
Chrysene/triphenylene	0.20	2.83	1.16	0.64	0.20	0.52	0.31	0.09
Benzo(b)fluoranthene	0.39	12.7	6.36	3.28	0.73	2.20	1.34	0.41
Benzo(k)fluoranthene	0.20	3.78	1.98	0.94	0.28	0.50	0.40	0.07
Benzo(e)pyrene	0.20	4.66	2.24	1.13	0.30	0.93	0.56	0.18
Benzo(a)pyrene	0.17	6.85	3.04	1.79	0.31	0.86	0.54	0.14
Perylene	0.01	1.70	0.74	0.45	0.11	0.24	0.16	0.04
Indeno(1,2,3-cd)pyrene	0.13	7.89	2.86	2.18	0.47	1.25	0.92	0.23
Dibenz(a,h)anthracene	n.d.	10.8	0.90	1.93	0.09	0.29	0.17	0.05
Benzo(ghi)perylene	0.19	5.69	2.36	1.67	0.50	1.24	0.89	0.23
Anthanthrene	n.d.	6.94	1.67	1.77	0.08	0.30	0.18	0.06
Coronene	0.05	19.0	4.23	4.60	0.30	0.89	0.60	0.16
Dibenzo(a,e)pyrene	n.d.	1.78	0.74	0.41	0.05	0.30	0.18	0.08
Subtotal	2.23	83.4	35.7	18.7	5.50	47.3	16.5	12.3
Total organics (ng m^{-3})	*362*	*2381*	*1136*	*462*	*611*	*3268*	*1586*	*766*
OC (µg m^{-3})	3.20	15.6	9.12	3.71	4.15	17.6	9.69	3.65
WSOC (µg m^{-3})	1.71	6.95	3.76	1.28	2.15	6.03	4.10	0.90

[a] SD: standard deviation.

[b] n.d., not detected.

[c] CPI, carbon preference index: $(C_{21}+C_{23}+C_{25}+C_{27}+C_{29}+C_{31}+C_{33}+C_{35}+C_{37}+C_{39})/(C_{22}+C_{24}+C_{26}+C_{28}+C_{30}+C_{32}+C_{34}+C_{36}+C_{38}+C_{40})$ for *n*-alkanes; $(C_{20}+C_{22}+C_{24}+C_{26}+C_{28}+C_{30}+C_{32}+C_{34})/(C_{21}+C_{23}+C_{25}+C_{27}+C_{29}+C_{31}+C_{33})$ for fatty acids and fatty alcohols.

[d] Plant wax *n*-alkanes are calculated as the excess odd homologues – adjacent homologues average, and the difference from the total *n*-alkanes is the petroleum-derived amount. Negative values of plant wax *n*-alkanes were taken as zero.

[e] Tris(2,4-*di*-*tert*-butyl-phenyl)phosphate (TBPP): due to a lack of the authentic standard, TBPP was quantified using 1,3,5-triphenylbenzene.

Table 2. Results of principal component analysis with Varimax rotation for the dataset of sugar compounds (including anhydrosugars) in the urban aerosols from Chennai, tropical India.

Sugars	Winter			Summer		
	Component 1	Component 2	Component 3	Component 1	Component 2	Component 3
Galactosan	*0.87*	0.10	0.34	*0.92*	–0.23	–0.02
Mannosan	*0.98*	0.02	0.12	*0.94*	–0.23	–0.13
Levoglucosan	*0.98*	0.07	0.07	*0.98*	–0.11	–0.11
Glycerol	0.29	0.21	*0.89*	–0.06	–0.07	*0.78*
Erythritol	*0.92*	0.33	0.11	*0.91*	0.02	0.25
Xylose	*0.93*	–0.02	0.28	*0.93*	–0.03	–0.20
Arabitol	*0.62*	*0.65*	0.31	0.01	*0.97*	0.07
Fructose	*0.55*	*0.63*	–0.12	0.19	0.30	*0.84*
Glucose	0.20	*0.56*	0	0.10	*0.86*	–0.36
Mannitol	0.12	*0.91*	0.22	–0.09	*0.92*	–0.01
Inositol	*0.87*	0.11	0.37	*0.82*	0.29	0.14
Sucrose	–0.03	*0.98*	0.02	–0.21	*0.88*	0.30
Trehalose	–0.12	*0.96*	0.12	–0.01	*0.92*	0.25
Maltose	*0.88*	0.20	–0.28	*0.74*	0.08	0.23
Variance (%)	48.1	29.1	10.0	40.8	31.8	12.9

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Table 3. Contribution of individual organic compound classes to OC and WSOC in the aerosols from Chennai, tropical India (%)[a].

Compound class	Winter (n=29)				Summer (n=20)			
	Min	Max	Average	SD[b]	Min	Max	Average	SD
			To OC					
n-Alkanes	0.25	2.39	1.18	0.55	0.50	3.72	1.37	0.91
Fatty acids	0.58	3.51	2.37	0.72	1.65	6.56	3.54	1.44
Fatty alcohols	0.17	0.90	0.62	0.14	0.36	0.85	0.57	0.14
Anhydrosugars	0.04	1.50	0.59	0.40	0.36	0.88	0.59	0.16
Sugars/sugar alcohols	0.02	0.34	0.10	0.07	0.05	0.28	0.14	0.05
Lignin products	0.01	0.23	0.12	0.05	0.05	0.11	0.08	0.02
Terpenoid biomarkers	0.02	0.13	0.06	0.03	0.02	0.05	0.03	0.01
Sterols	0.04	2.07	0.57	0.56	0.03	0.62	0.19	0.14
Aromatic acids	0.05	0.55	0.26	0.13	0.20	0.50	0.32	0.10
Plasticizers/antioxidants	0.99	7.95	2.97	1.73	2.20	8.39	4.50	1.53
Hopanes	0.01	0.27	0.14	0.06	0.03	0.06	0.05	0.01
PAHs	0.04	0.68	0.37	0.16	0.07	0.27	0.15	0.06
Total quantified	*6.04*	*13.8*	*9.35*	*1.77*	*7.70*	*15.0*	*11.5*	*1.93*
			To WSOC					
Sugars/sugar alcohols	0.08	0.65	0.21	0.12	0.21	0.58	0.30	0.09
Levoglucosan	0.07	3.49	1.17	0.84	0.70	1.97	1.20	0.39
Galactosan+mannosan	0.01	0.64	0.23	0.16	0.09	0.29	0.17	0.06
Benzoic acid	0.01	0.05	0.02	0.01	0.01	0.04	0.02	0.01
Phthalic acid	0.01	0.59	0.10	0.12	0.04	0.22	0.09	0.04
Vanillic acid[c]	0.001	0.031	0.015	0.009	0.007	0.017	0.011	0.003
4-Hydroxybenzoic acid[c]	0.01	0.35	0.16	0.09	0.04	0.12	0.08	0.03
Total soluble	*0.22*	*4.91*	*1.90*	*1.10*	*1.19*	*2.78*	*1.87*	*0.46*

[a] OC, organic carbon; WSOC, water-soluble organic carbon. All the quantified organic compounds were converted to carbon contents to calculate the OC and WSOC ratios.

[b] SD, standard deviation.

[c] Slightly soluble in water.

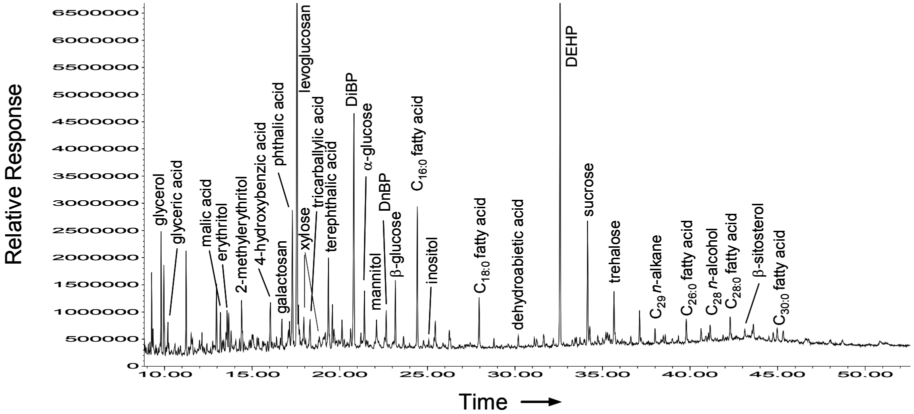

Fig. 1. A typical GC/MS trace (total ion current: TIC) for a total extract (TMS derivatized) of aerosol particles from tropical India. For the abbreviation, see Table 1.

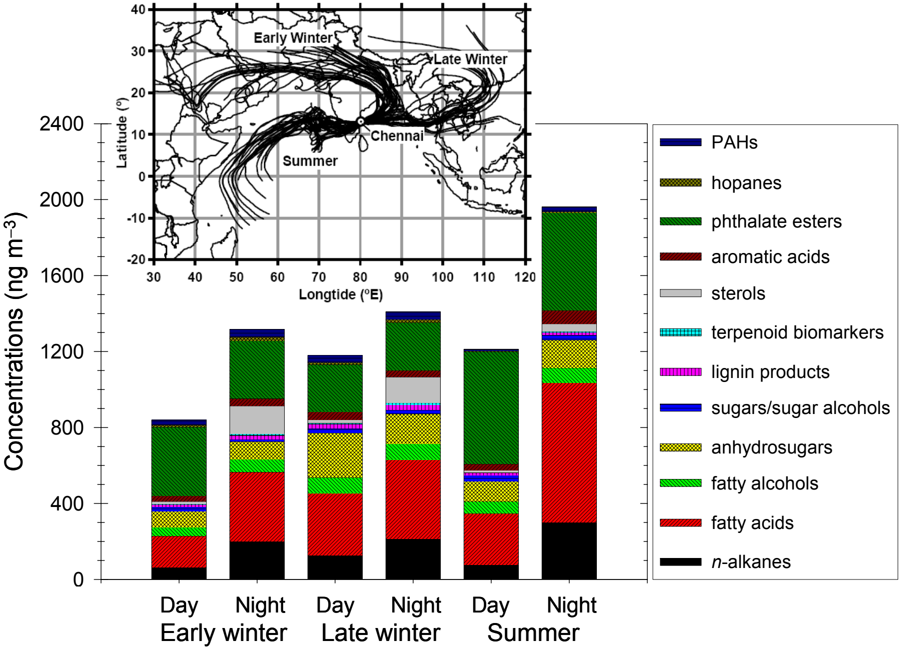

Fig. 2. Chemical composition of organic compounds detected in the atmospheric aerosols from Chennai, tropical India. Ten-day back trajectories of air masses arriving in Chennai during the sampling periods were also plotted here. Detailed information for air mass back trajectory analysis is described by Pavuluri et al. (2009a).

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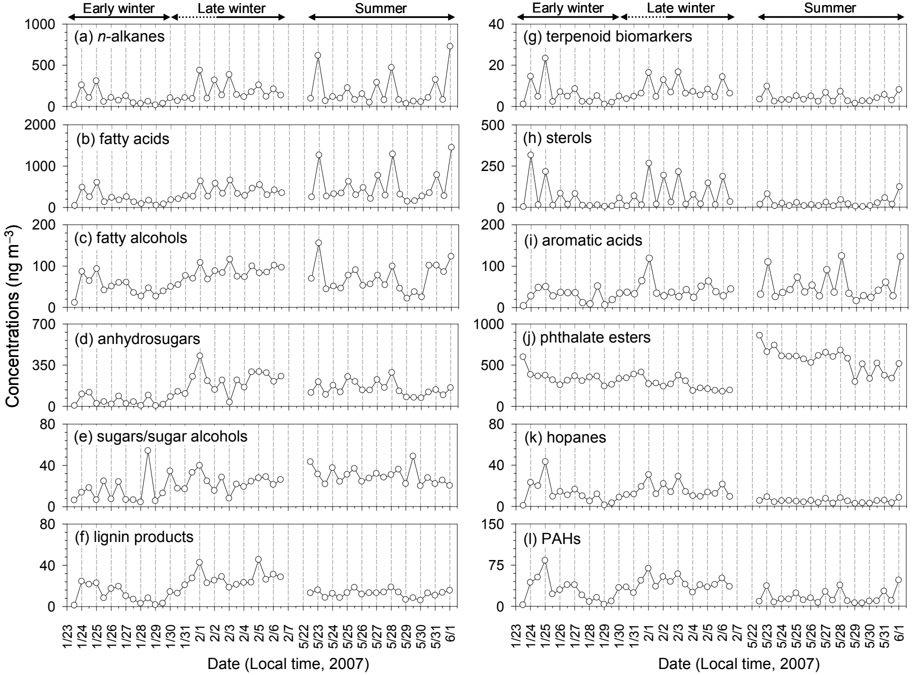

Fig. 3. Temporal variations in the concentrations of aliphatic lipids and other organic compound classes detected in the tropical Indian aerosols.

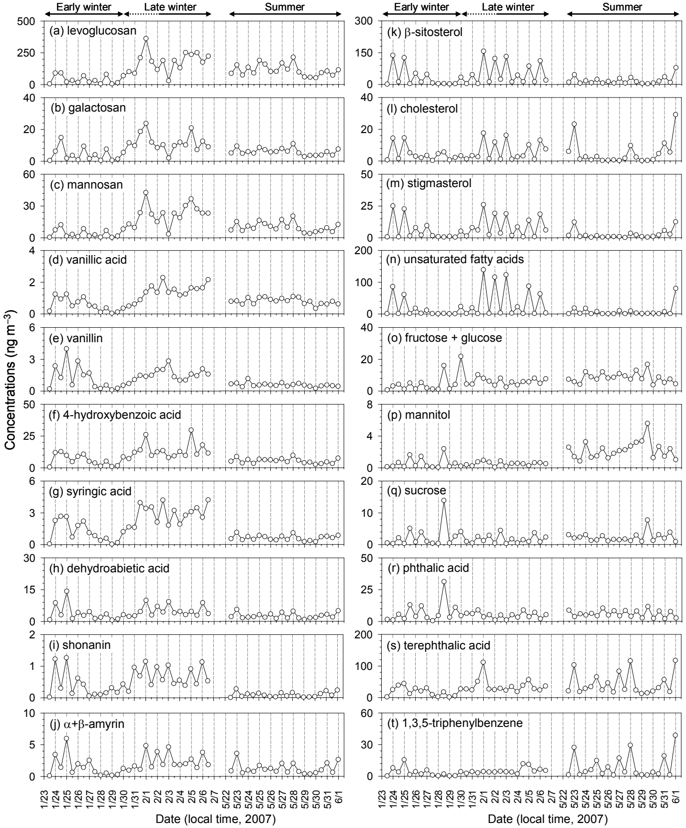

Fig. 4. Temporal variations in the concentrations of biomass-burning tracers and other individual organic compounds detected in the tropical Indian aerosols.

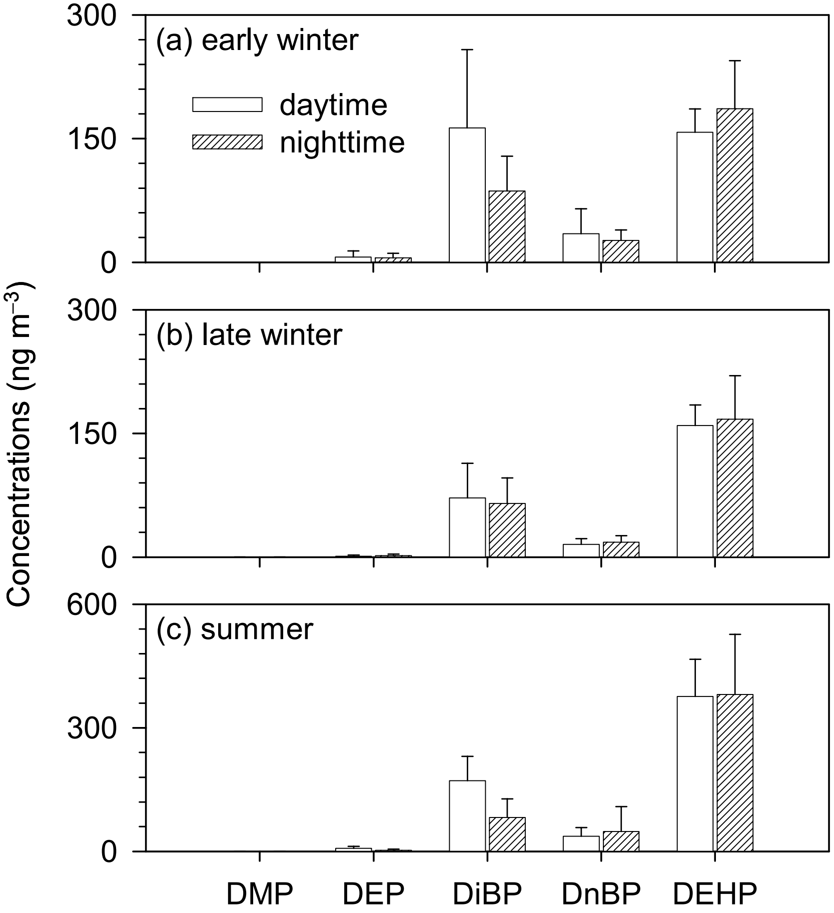

Fig. 5. Molecular distributions of phthalate esters in PM_{10} aerosols collected in Chennai, tropical India.

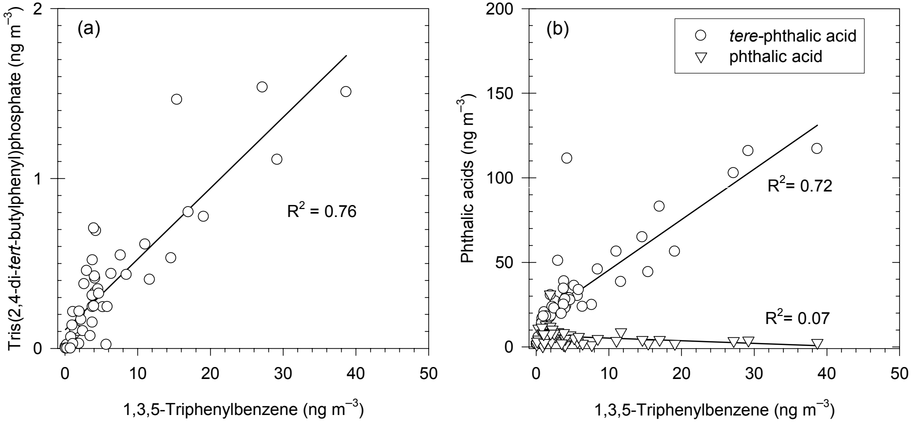

Fig. 6. Correlations between the concentration of 1,3,5-triphenylbenzene (a tracer for plastic burning) and **(a)** tris(2,4-*di*-*tert*-butyl-phenyl)phosphate, and **(b)** phthalic acids.

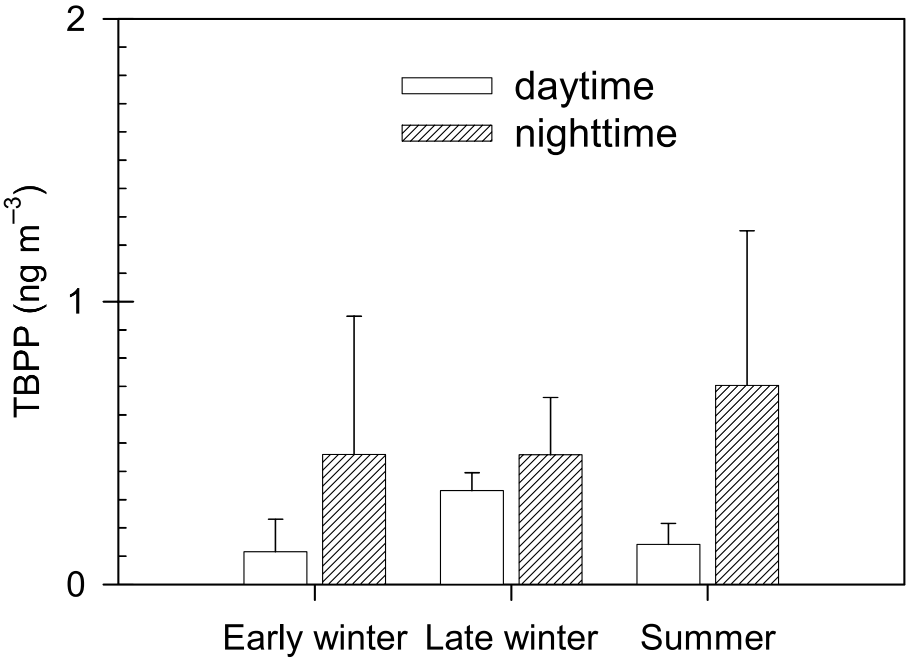

Fig. 7. Concentrations of tris(2,4-*di*-*tert*-butyl-phenyl)phosphate (TBPP) in PM_{10} aerosols collected in Chennai, tropical India.

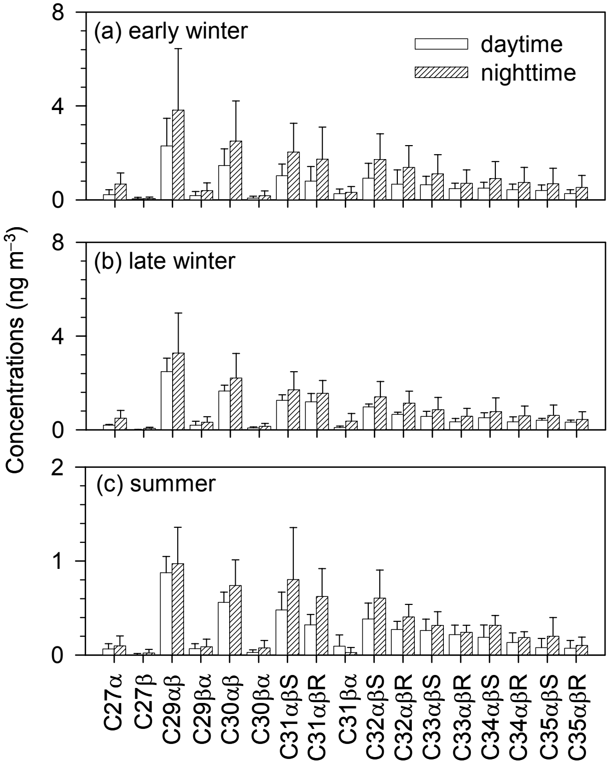

Fig. 8. Molecular distributions of hopanes in PM_{10} aerosols collected in Chennai, tropical India.

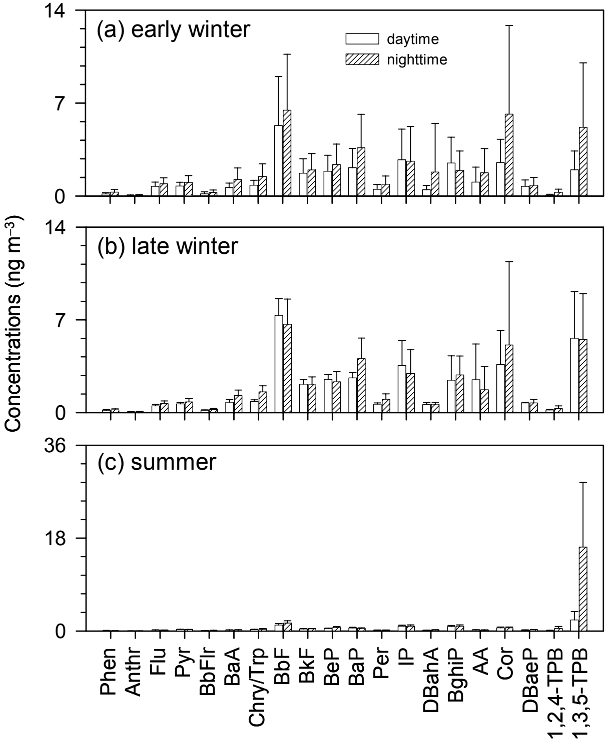

Fig. 9. Molecular distributions of PAHs in PM_{10} aerosols collected in Chennai, tropical India.

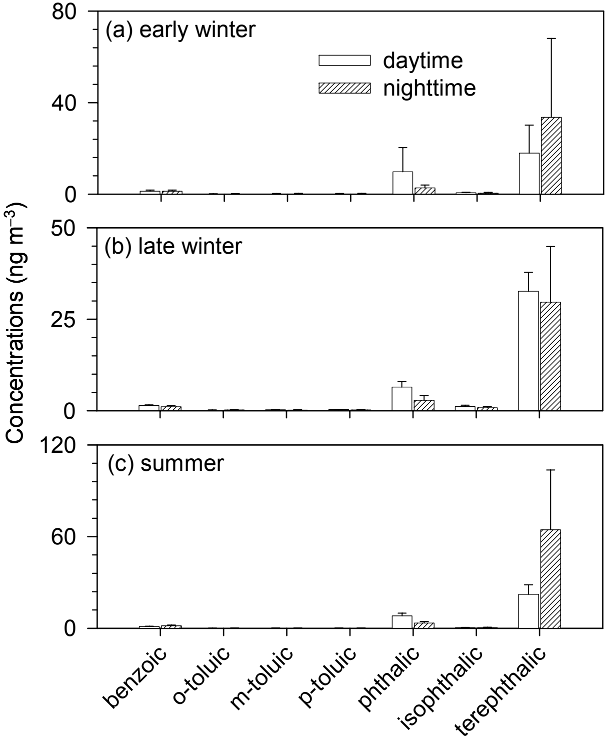

Fig. 10. Molecular distributions of aromatic acids in PM_{10} aerosols collected in Chennai, tropical India.

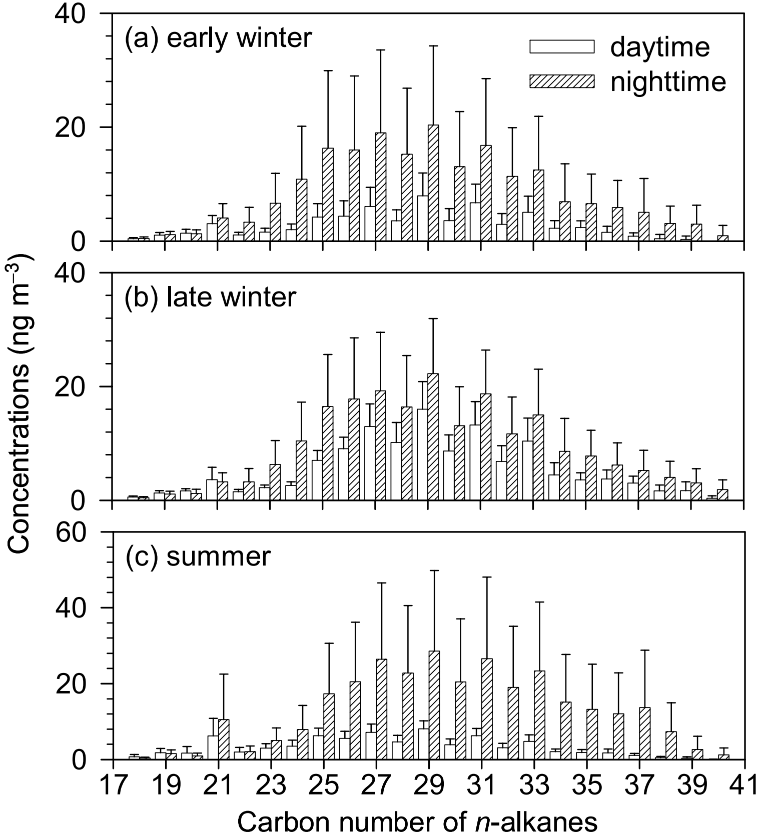

Fig. 11. Molecular distributions of *n*-alkanes in PM_{10} aerosols collected in Chennai, tropical India.

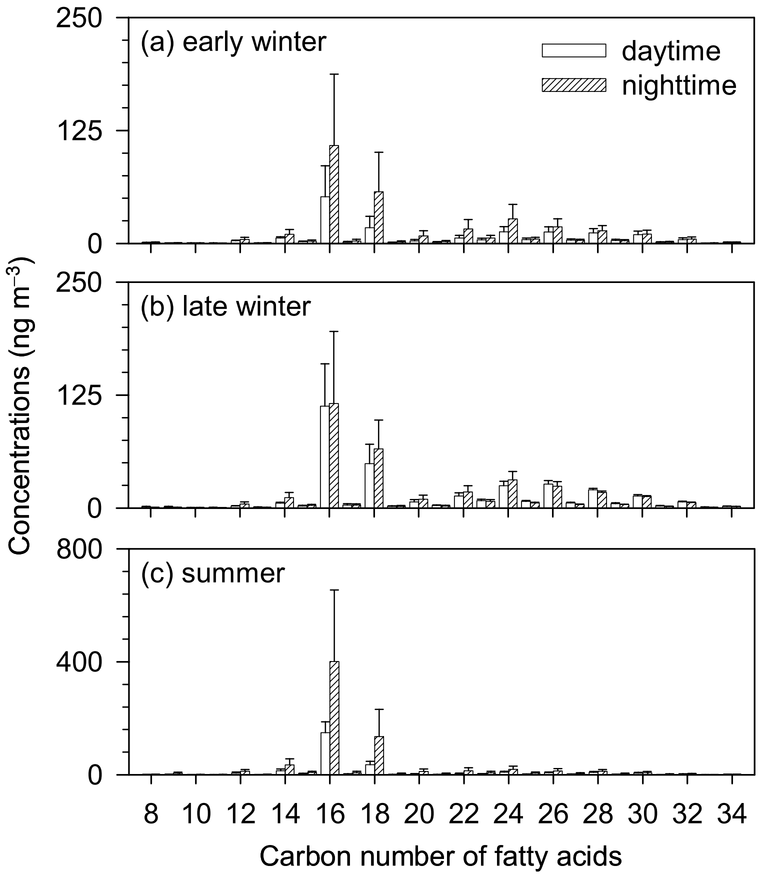

Fig. 12. Molecular distributions of fatty acids in PM_{10} aerosols collected in Chennai, tropical India.

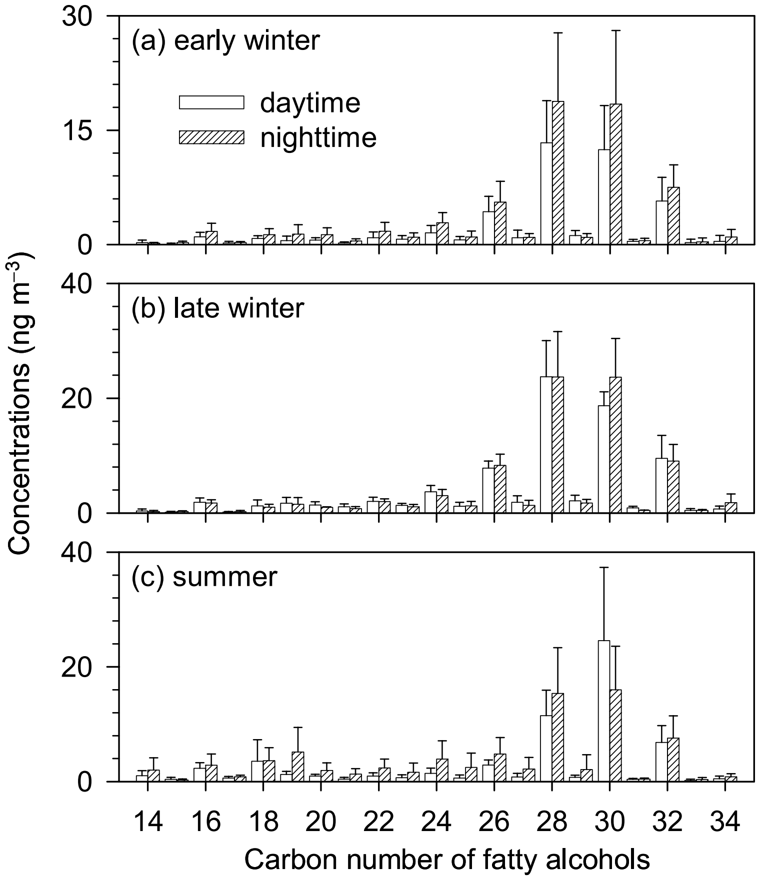

Fig. 13. Molecular distributions of fatty alcohols in PM_{10} aerosols collected in Chennai, tropical India.

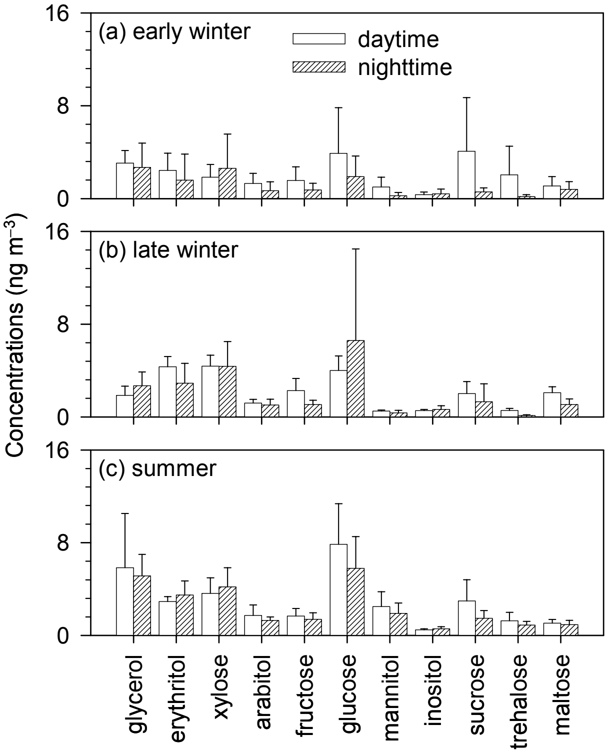

Fig. 14. Molecular distributions of sugar compounds in PM_{10} aerosols collected in Chennai, tropical India.