

See discussions, stats, and author profiles for this publication at: <https://www.researchgate.net/publication/228705725>

Organic molecular compositions and temporal variations of summertime mountain aerosols over Mt. Tai, North China Plain

Article in *Journal of Geophysical Research Atmospheres* · October 2008

DOI: 10.1029/2008JD009900

CITATIONS

207

READS

280

8 authors, including:



Pingqing Fu
Tianjin University

559 PUBLICATIONS 18,991 CITATIONS

[SEE PROFILE](#)



Kimitaka Kawamura
Chubu University

658 PUBLICATIONS 32,586 CITATIONS

[SEE PROFILE](#)



Shankar G. Aggarwal
National Physical Laboratory - India

130 PUBLICATIONS 2,915 CITATIONS

[SEE PROFILE](#)



Gehui Wang
East China Normal University

231 PUBLICATIONS 13,336 CITATIONS

[SEE PROFILE](#)



Organic molecular compositions and temporal variations of summertime mountain aerosols over Mt. Tai, North China Plain

Pingqing Fu,^{1,2} Kimitaka Kawamura,¹ Kazuhiro Okuzawa,¹ Shankar Gopala Aggarwal,¹ Gehui Wang,^{1,3} Yugo Kanaya,⁴ and Zifa Wang⁵

Received 1 February 2008; revised 3 May 2008; accepted 17 July 2008; published 8 October 2008.

[1] Total suspended particles (TSP) were collected at the summit of Mt. Tai (1534 m above sea level) on a daytime and nighttime basis during a summertime campaign (May–June 2006) and were characterized for organic molecular compositions using solvent extraction/derivatization and gas chromatography/mass spectrometry technique. The *n*-Alkanes, fatty acids, fatty alcohols, sugars, glycerol and polyacids, and phthalate esters were found as major organic compound classes, whereas lignin and resin products, sterols, aromatic acids, hopanes, and polycyclic aromatic hydrocarbons (PAHs) were detected as minor classes. Sugars (49.8–2115 ng m⁻³, average 640 ng m⁻³ in daytime; 18.1–4348 ng m⁻³, 799 ng m⁻³ in nighttime) were found to be the dominant compound class. Levoglucosan, a specific cellulose pyrolysis product, was detected as the most abundant single compound, followed by C₂₈ fatty alcohol, diisobutyl and di-*n*-butyl phthalates, C₂₉ *n*-alkane, C₁₆ and C₂₈ fatty acids, and malic acid. By grouping organic compounds based on their sources, we found that emission of terrestrial plant waxes was the most significant source (30–34%) of the TSP, followed by biomass burning products (25–27%) (e.g., levoglucosan and lignin and resin products), soil resuspension (15–18%) due to agricultural activities, secondary oxidation products (8–10%), plastic emission (3–10%), marine/microbial sources (6%), and urban/industrial emissions from fossil fuel use (4%). However, low molecular weight dicarboxylic acids (such as oxalic acid) of photochemical origin were not included in this study. Malic acid was found to be much higher than those reported in the ground level, suggesting an enhanced photochemical production in the free troposphere over mountain areas. Temporal variations of biomass burning tracers (e.g., levoglucosan, galactosan, mannosan) and some higher plant wax derived compound classes suggested that there were two major (E1 and E2) and one minor (E3) biomass-burning events during this campaign. Most of the compound classes showed higher concentrations in nighttime samples when organic aerosols can be long-range transported from different source regions to the summit of Mt. Tai above the planetary boundary layer (PBL). This study also demonstrates that the free troposphere over Mt. Tai is heavily influenced by field burning of agricultural wastes such as wheat straws in the North China Plain during the harvest season in early summer.

Citation: Fu, P., K. Kawamura, K. Okuzawa, S. G. Aggarwal, G. Wang, Y. Kanaya, and Z. Wang (2008), Organic molecular compositions and temporal variations of summertime mountain aerosols over Mt. Tai, North China Plain, *J. Geophys. Res.*, *113*, D19107, doi:10.1029/2008JD009900.

1. Introduction

[2] Both natural processes and anthropogenic activities produce tropospheric aerosol particles. They play significant roles in regional and global changes of climate, chemistry of the atmosphere, biogeochemical carbon cycle, pollution transport, and human health [Andreae and Crutzen, 1997; Charlson *et al.*, 1992; Jacobson *et al.*, 2000; Jacobson, 2001; Kaufman *et al.*, 2002; Ramanathan *et al.*, 2001; Rudich, 2003; Seinfeld and Pandis, 1998; Wilkening *et al.*, 2000]. All these features are strongly influenced by the chemical composition and concentrations of the aerosol particles [Pöschl, 2005]. Their chemical constituents can

¹Institute of Low Temperature Science, Hokkaido University, Sapporo, Japan.

²Institute of Geochemistry, Chinese Academy of Sciences, Guiyang, China.

³School of the Environment, Nanjing University, Nanjing, China.

⁴Frontier Research Center for Global Change, Japan Agency for Marine-Earth Science and Technology, Kanagawa, Japan.

⁵Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, China.

be divided into inorganic and organic groups. Organic constituents are recently highlighted because they account for a substantial portion of atmospheric particles, up to 50% in mass [Seinfeld and Pandis, 1998], potentially influence the hygroscopic properties [Hemming and Seinfeld, 2001; Kanakidou et al., 2005], contribute to a large portion of terrestrial input of organic carbon over the remote oceans [Gagosian and Peltzer, 1986; Kawamura and Gagosian, 1990], and cause human health problems (e.g., PAHs and phthalate esters) [Pöschl, 2005; Sidhu et al., 2005].

[3] China has been facing a serious air pollution problem due to the increased usage of fossil fuels, combined with soil dust emissions. One fourth of primary anthropogenic organic aerosols on the globe is generated in China, roughly 70% of which comes from coal burning [Cooke et al., 1999; Streets et al., 2004]. Atmospheric emissions of aerosols and their precursors from China have been recognized to have a significant impact on atmospheric composition at regional and global scales via long-range atmospheric transport [Feng et al., 2007; Gao and Anderson, 2001; Heald et al., 2006; Simoneit et al., 2004c]. Moreover, field burning of agriculture residues (e.g., wheat straws) in countryside sometimes causes serious air pollution even in urban areas (e.g., Beijing), especially in the North China Plain [Li et al., 2007]. Some studies have reported high abundances of organic aerosols collected at ground sites from different Chinese urban areas [Simoneit et al., 1991; Wang and Kawamura, 2005; Wang et al., 2006a; Zheng et al., 2005] and Changdao Island in the East China Sea [Feng et al., 2007]. Aircraft measurements of organic aerosols over China also demonstrated high loadings of organic matter in the troposphere above the planetary boundary layer (PBL) [Wang et al., 2007].

[4] The large usage of coal and biofuel, increased number of automobiles, rapid industrialization/urbanization, high density of population, and agricultural activities during specific seasons all make Chinese air pollution very complicated and different from those of other countries [Wang et al., 2006a]. To better understand the air quality in China and the outflow of Asian aerosols to the Pacific Ocean, studies on the organic compounds in aerosols from rural areas including high mountains are also needed. High mountain areas provide a unique situation for atmospheric chemistry because they sometimes exist in the free troposphere due to the upward and downward movement of the PBL in daytime and nighttime, respectively. Thus, significant difference may exist in the chemical composition of aerosols in high mountains from those of urban and rural aerosols, although the mountain atmosphere receives aerosols and their precursors mostly from the flat ground levels in urban areas.

[5] In this study, we collected day/night aerosol samples using quartz filters and high volume air sampler at the summit of Mt. Tai during a summertime field campaign in 2006 when agricultural burning was active in the region. Here we report the results of organic chemical analyses of the aerosols, that is, compositions, concentrations, and temporal variations of molecular markers. Using molecular tracers, we also discuss source apportionments of organic aerosols, including fossil fuel combustion, biomass burning,

marine/microbial emissions, soil dusts, industrial and residential emissions.

2. Sample and Method

2.1. Aerosol Sampling

[6] Mt. Tai (36.25°N and 117.10°E, 1534 m above sea level) is located in Shandong Province of China, which is on the transport path of the Asian continent outflow (Figure 1). It is a tilted fault block mountain with height increasing from the north to the south, facing to the East China Sea, Korean Peninsula, and Japanese Islands. It lies in the zone of deciduous forest; about 80% of its area is covered with vegetation. Almost 1000 species are known to grow in this area. However, vegetations are limited to bushes and the ground surfaces are mostly covered with rocks at the mountaintop. Daytime/nighttime and 3-h aerosol sampling were performed from 28 May to 28 June 2006 at the balcony of the second floor of observatory (~10 m above ground level) on the top of Mt. Tai using high-volume air sampler. All the samples were collected onto precombusted (450°C for 6 h) quartz fiber filters (20 × 25 cm, Pallflex 2500QAT-UP) with a flow rate of 1.0 m³ min⁻¹. Before sampling, filter was placed in a precombusted (450°C for 6 h) clean glass jar with a Teflonlined screw cap during the transport and storage. After the sampling, the filter was recovered into the glass jar, transported to the laboratory and stored at -20°C prior to analysis. Eighty-one suspended particulate (TSP) samples (including 5 whole-day, 19 daytime (0600–1800, local time), 20 nighttime (1800–0600), and 37 three-hour samples) were collected during this campaign. Among them the results of daytime/nighttime samples (n = 39) were presented here.

2.2. Extraction and Derivatization

[7] A filter aliquot (approximately 10 cm²) was cut into pieces and extracted three times with dichloromethane/methanol (2:1, v/v) under ultrasonication for 10 min. The solvent extracts were filtered through quartz wool packed in a Pasteur pipette, concentrated by the use of a rotary evaporator, and then blown down to dryness with pure nitrogen gas. The extracts were reacted 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 to derive trimethylsilyl derivatives of carboxylic acids, alcohols and sugars, etc. During this procedure, COOH and OH groups are derivatized to the corresponding trimethylsilyl (TMS) esters and ethers, respectively [Schauer et al., 1996; Simoneit et al., 2004b; Simoneit et al., 2004c; Wang and Kawamura, 2005]. After the reaction, derivatives were diluted with 140 μl of *n*-hexane containing the internal standard (C₁₃ *n*-alkane, 1.43 ng μl⁻¹). Field and laboratory blank filters were treated as the real samples for quality assurance.

2.3. Gas Chromatography-Mass Spectrometry

[8] Gas chromatography-mass spectrometry (GC-MS) analyses of the derivatized total extracts were performed on a Hewlett-Packard model 6890 GC coupled to Hewlett-Packard model 5973 mass-selective detector (MSD). The GC separation was achieved on a DB-5MS fused silica capillary column (30 m × 0.25 mm i.d., 0.5 μm film thickness) with a GC oven temperature program: tempera-

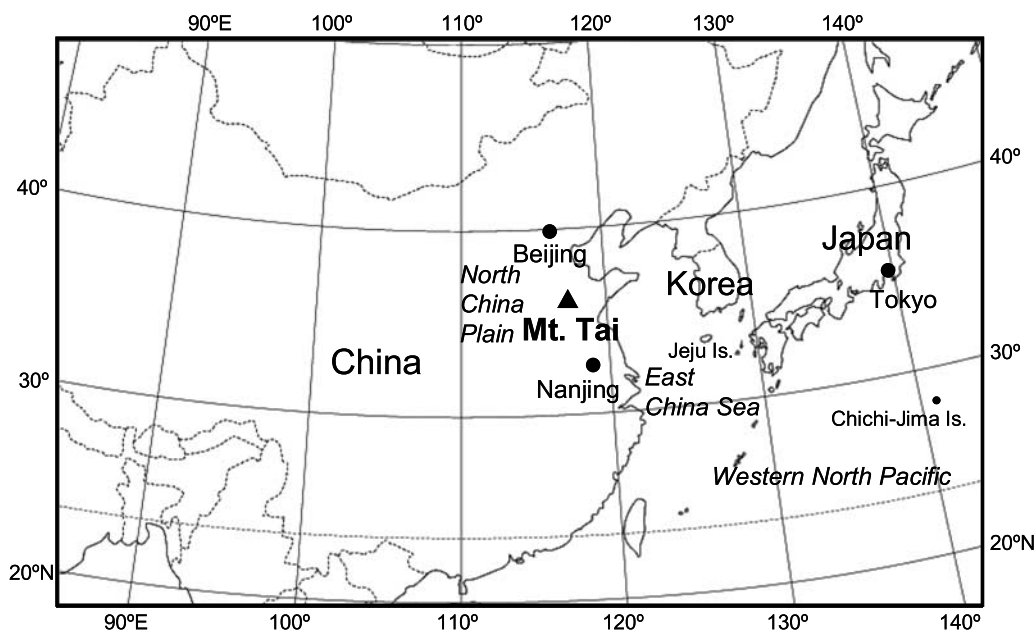


Figure 1. A map of east Asia with the sampling site of Mt. Tai in the North China Plain.

ture hold at 50°C for 2 min, increase from 50 to 120°C at a rate of 30°C min⁻¹, then further increase from 120 to 300°C at a rate of 6°C min⁻¹ with a final isotherm hold at 300°C for 16 min. The sample was injected on a splitless mode with the injector temperature at 280°C. The mass spectrometer was operated on the electron impact (EI) mode at 70 eV and scanned from 50 to 650 Da. Mass spectral data were acquired and processed with the Chemstation software. Individual compounds were identified by comparing mass spectra with those of literature and library data and authentic standards and by interpretation of mass fragmentation patterns. GC-MS response factors of individual compound were determined using authentic standards.

[9] Values of 100–200 ng of each standard were spiked to 10 cm² of blank filter and treated as a real sample. This recovery experiment was repeated three times. The results showed that average recoveries of the 66 standards, including *n*-alkanes, fatty alcohols, fatty acids, sugars, lignin and resin products, phthalate esters, aromatic acids, and sterols, were better than 80% except for polyacids (malic acid, tartaric acid, and citric acid), whose recovery was less than 60%. The field and the laboratory blank filters were also analyzed by the procedure described earlier in this section; their results showed no significant contamination except for bis(2-ethylhexyl) phthalate, whose field blank levels sometimes were high compared to real samples and thus were not used in this study. The data reported here were corrected for the field blanks but not corrected for the recoveries.

3. Results and Discussion

[10] Homologs of eleven compound classes, i.e., *n*-alkanes, fatty acids, fatty alcohols, sugars, glycerol and polyacids, lignin and resin products, sterols, aromatic acids, phthalate esters, hopanes, and polycyclic aromatic hydrocarbons (PAHs), were detected in the aerosol samples from Mt. Tai with total concentration ranges of 339–4265 ng m⁻³ (average 1731 ng m⁻³) in daytime and 221–8760 ng m⁻³

(1970 ng m⁻³) in nighttime. Figure 2 shows the major resolved organic components of total aerosol extracts. Table 1 presents the concentrations of all the identified organic compounds. Among them, *n*-alkanes, fatty acids, fatty alcohols, sugars, glycerol and polyacids, and phthalate esters are the major compound classes, while the others are relatively minor (Figure 3). Levoglucosan (on average) was the most abundant single compound, followed by C₂₈ fatty alcohol, diisobutyl and di-*n*-butyl phthalates, C₂₉ *n*-alkane, C₁₆ and C₂₈ fatty acids, and malic acid (Table 1).

3.1. Major Organic Compound Classes

3.1.1. Aliphatic Lipids

[11] The lipid components (*n*-alkanes, fatty acids, and *n*-alcohols), abundant in the Mt. Tai aerosols, are derived from terrestrial plant waxes, marine/microbial detritus, and emissions from fossil fuel use. The *n*-alkanes were detected in a range of C₂₀–C₃₆. Their concentrations were higher in nighttime (7.68–619 ng m⁻³, average 194 ng m⁻³) than daytime (16.9–445 ng m⁻³, 161 ng m⁻³) (Table 1). These values are in good agreement with those reported from urban cities in China (10–328 ng m⁻³, 143 ng m⁻³ in summer) [Wang *et al.*, 2006a] and Tokyo (31–323 ng m⁻³) [Kawamura *et al.*, 1995] but 1–2 orders of magnitude higher than those reported from mountain forest in Japan (14–34 ng m⁻³) [Kawamura *et al.*, 1995] and Chichi-Jima Island in the western North Pacific (0.11–14.1 ng m⁻³) [Kawamura *et al.*, 2003].

[12] Their molecular distributions are characterized by an odd-carbon-numbered predominance with a maximum at C₂₉ (Figure 2b). Such a molecular distribution suggests that *n*-alkanes over Mt. Tai were mainly derived from higher plant waxes, in which C₂₇, C₂₉, C₃₁, and C₃₃ are dominant species. Lower molecular weight *n*-alkanes (LMW *n*-alkanes) such as C₂₀–C₂₆ show the weaker odd/even predominance, suggesting that they are derived from the incomplete combustion of fossil fuels and petroleum residue. The carbon preference

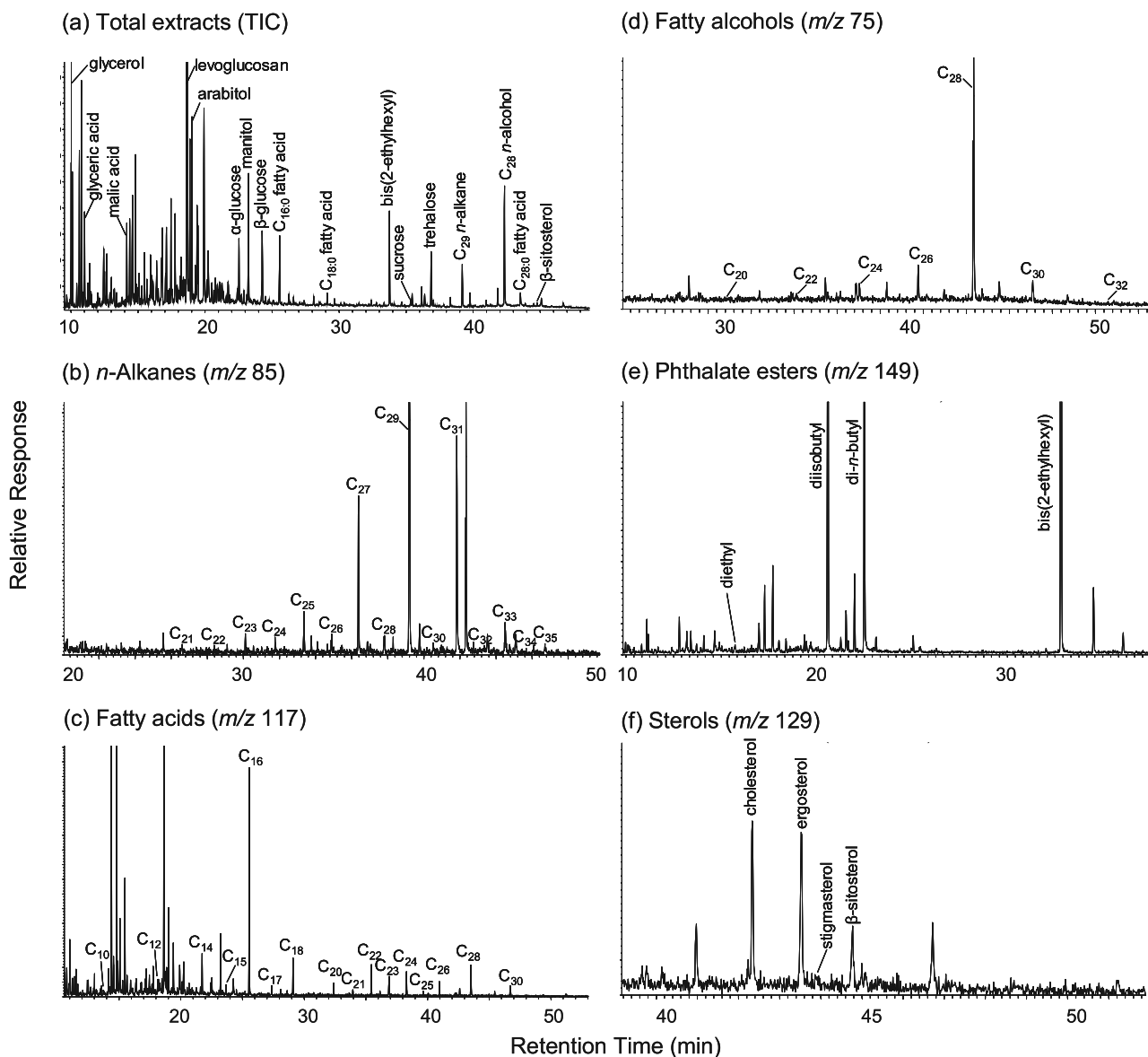


Figure 2. Salient features of the gas chromatography-mass spectrometry data for total extracts of aerosol sample from Mt. Tai. (a) Total ion current traces (TIC), (b) m/z 85 plots for n -alkanes, (c) m/z 117 plots for fatty acids (trimethylsilyl (TMS) esters), (d) m/z 75 plots for fatty alcohols (TMS ethers), (e) m/z 149 plots for phthalate esters, and (f) m/z 129 plots for sterols (TMS ethers).

indexes (CPI) for C_{20} – C_{36} are 1.12–8.03 (average 4.42) in daytime and 1.45–7.31 (4.63) in nighttime. They are much higher than those reported in urban aerosols from Chinese cities (1.16 ± 0.12) [Wang *et al.*, 2006a] and Tokyo (1.1–2.8, average 1.5) [Kawamura *et al.*, 1995]. However, they are similar to those (CPI > 5) reported in the aerosols from the mountain area in Japan [Kawamura *et al.*, 1995] and those (1.8–14.6, average 4.5) reported from Chichi-Jima Island [Kawamura *et al.*, 2003]. These results suggest that n -alkanes in the aerosols over Mt. Tai are significantly influenced by higher plant wax emissions rather than fossil fuel combustion. However, the higher concentrations in nighttime cannot be explained by local emissions of plant waxes. They should be long-range transported from different source regions via the atmosphere above the PBL. During this field campaign, the heights of PBL were generally approximately 1800–

4200 m in daytime (M. Takigawa *et al.*, unpublished data, 2007) when more organic aerosols can be transported upward from local sources by the vertical mixing. In contrast, the summit of Mt. Tai was present in the free troposphere in nighttime when the heights of PBL were decreased to 200–800 m (M. Takigawa *et al.*, unpublished data, 2007).

[13] A homologous series of straight chain $C_{12:0}$ – $C_{34:0}$ fatty acids, including unsaturated ($C_{18:1}$ and $C_{18:2}$) acids were detected in the aerosol samples. Their distributions are characterized by a strong even carbon number predominance with two maximum at $C_{16:0}$ and $C_{28:0}$ (Table 1 and Figure 2c). A similar bimodal distribution has been reported in continental [Simoneit and Mazurek, 1982; Wang and Kawamura, 2005; Wang *et al.*, 2006a] and marine aerosols [Gagosian *et al.*, 1981; Kawamura *et al.*, 2003; Mochida *et al.*, 2002]. Concentration ranges of total fatty acids were

Table 1. Concentrations of Organic Compounds in the Aerosol Samples (TSP) Collected at the Summit of Mt. Tai, North China Plain (May to June 2006)^a

Compounds	Daytime (n = 19)				Nighttime (n = 20)			
	Minimum	Maximum	Mean	SD	Minimum	Maximum	Mean	SD
<i>n-Alkanes</i>								
C ₂₀	0.13	1.78	0.62	0.40	0.03	2.18	0.51	0.48
C ₂₁	0.37	5.85	2.52	1.72	0.06	10.9	2.72	2.61
C ₂₂	0.22	3.98	1.62	1.09	0.18	5.82	1.73	1.56
C ₂₃	0.87	11.7	5.24	2.94	0.14	16.0	5.21	3.98
C ₂₄	0.57	6.19	3.09	1.68	0.19	12.0	3.57	2.75
C ₂₅	0.84	27.3	9.88	6.28	0.24	31.0	10.4	8.04
C ₂₆	1.08	13.9	5.79	3.43	0.21	19.3	5.91	4.51
C ₂₇	1.66	69.7	21.6	15.1	0.56	84.5	26.0	22.8
C ₂₈	1.37	19.2	8.72	4.94	0.36	27.5	9.77	7.13
C ₂₉	3.15	180	61.2	48.9	1.80	230	76.6	68.0
C ₃₀	0.83	9.54	4.53	2.61	0.41	15.6	5.17	3.61
C ₃₁	2.43	86.1	25.7	20.5	1.64	123	33.4	34.2
C ₃₂	0.38	6.33	2.28	1.63	0.45	7.36	2.40	1.62
C ₃₃	0.40	14.0	5.60	4.07	0.38	30.6	7.73	8.70
C ₃₄	0.11	3.02	1.02	0.84	n.d.	10.1	1.34	2.20
C ₃₅	n.d.	2.74	0.64	0.79	n.d.	3.46	0.64	0.80
C ₃₆	n.d.	2.96	0.64	0.86	n.d.	3.48	0.60	0.90
Subtotal	16.9	445	161	105	7.68	619	194	167
CPI ^b	1.12	8.03	4.42	1.79	1.45	7.31	4.63	1.68
<i>Fatty Acids</i>								
C _{10:0}	0.29	2.17	1.10	0.53	0.20	5.57	1.39	1.32
C _{11:0}	0.04	1.45	0.36	0.39	0.01	2.23	0.59	0.66
C _{12:0}	1.15	9.36	3.82	1.74	0.41	10.3	3.06	2.09
C _{13:0}	0.16	5.85	1.87	1.39	0.04	3.51	1.34	0.99
C _{14:0}	2.57	32.0	13.0	6.39	0.31	53.2	17.0	13.3
C _{15:0}	0.74	7.07	4.22	1.90	0.17	12.6	3.75	2.82
C _{16:0}	16.9	175	55.9	32.5	4.02	269	62.8	60.6
C _{17:0}	0.39	4.86	2.18	1.18	0.18	6.53	1.97	1.79
C _{18:0}	6.99	70.4	19.9	12.8	1.87	50.1	18.0	11.9
C _{19:0}	0.17	3.57	1.09	0.83	0.02	5.26	1.06	1.26
C _{20:0}	1.40	21.4	5.63	4.37	0.31	24.5	5.68	5.77
C _{21:0}	0.23	9.24	2.05	2.26	0.07	10.9	2.55	3.25
C _{22:0}	3.02	50.7	15.1	11.9	0.86	62.1	16.1	16.5
C _{23:0}	0.69	32.8	8.41	8.48	0.23	30.4	8.73	9.28
C _{24:0}	3.86	56.9	19.8	13.6	1.05	65.0	20.5	19.4
C _{25:0}	0.33	17.9	5.44	5.16	0.17	18.1	5.33	5.45
C _{26:0}	2.34	57.4	22.5	16.6	1.17	61.8	23.4	20.1
C _{27:0}	0.12	20.1	6.05	5.64	0.33	27.7	6.67	7.28
C _{28:0}	4.12	133	43.5	37.6	1.85	164	49.9	51.4
C _{29:0}	0.08	12.5	3.37	3.60	0.02	18.8	4.04	4.64
C _{30:0}	1.92	74.1	21.4	18.3	0.70	97.7	26.1	24.8
C _{31:0}	n.d.	2.10	0.57	0.69	n.d.	4.04	0.80	1.01
C _{32:0}	n.d.	9.02	3.19	3.34	0.06	13.9	3.39	4.31
C _{18:1}	0.81	12.1	2.77	2.47	0.61	7.90	3.21	2.05
C _{18:2}	n.d.	5.20	0.73	1.19	n.d.	11.8	1.73	2.76
Subtotal	63.0	783	262	172	15.6	892	287	250
CPI	3.31	11.9	6.04	2.00	2.99	9.50	5.53	1.42
<i>Fatty Alcohols</i>								
C ₁₈	0.51	4.27	1.72	1.06	0.13	6.16	2.20	1.65
C ₁₉	0.05	0.83	0.36	0.22	0.02	3.40	0.67	0.81
C ₂₀	0.27	3.39	1.09	0.86	0.14	5.23	1.47	1.41
C ₂₁	0.09	3.01	0.70	0.74	0.03	1.78	0.77	0.53
C ₂₂	1.12	8.86	4.29	2.31	0.13	20.5	4.73	5.31
C ₂₃	0.16	2.99	0.83	0.73	0.02	4.18	0.92	0.93
C ₂₄	0.61	32.9	10.8	7.33	0.91	40.0	12.4	11.8
C ₂₅	0.07	2.74	1.00	0.77	0.06	6.38	1.41	1.43
C ₂₆	1.69	92.6	32.4	24.7	0.20	129	36.8	33.7
C ₂₇	0.16	8.19	2.41	2.56	0.03	21.8	4.43	5.64
C ₂₈	6.17	597	161	176	3.51	1573	244	374
C ₂₉	0.25	9.15	2.37	2.20	0.19	18.2	3.26	4.38
C ₃₀	1.76	100	25.5	22.2	0.78	141	33.9	35.5
C ₃₁	0.07	4.46	1.25	1.04	n.d.	3.76	1.30	1.09
C ₃₂	0.25	11.6	2.96	2.77	n.d.	9.61	2.99	2.42
C ₃₄	n.d.	2.56	0.45	0.74	n.d.	2.24	0.57	0.70
Subtotal	15.1	866	249	230	11.1	1930	352	466
CPI	7.95	66.5	25.6	15.2	12.2	53.4	22.8	9.25

Table 1. (continued)

Compounds	Daytime (n = 19)				Nighttime (n = 20)			
	Minimum	Maximum	Mean	SD	Minimum	Maximum	Mean	SD
<i>Sugars</i>								
Galactosan	0.44	46.6	8.42	11.7	0.02	103	12.8	24.1
Mannosan	0.60	43.1	9.71	11.1	0.10	61.6	9.97	15.2
Levoglucofan	18.5	1733	391	454	0.37	3432	459	805
Arabitol	2.01	182	52.5	46.2	2.83	216	77.8	71.5
Fructose	4.30	40.9	16.7	9.85	0.54	70.3	21.3	18.8
Glucose	16.7	147	49.5	32.9	3.30	348	74.7	76.9
Mannitol	3.01	142	56.4	38.2	3.76	276	83.9	75.4
Inositol	0.58	11.8	3.56	2.46	0.09	13.2	4.40	3.69
Sucrose	0.28	95.4	27.6	26.1	0.25	120	25.4	32.6
Trehalose	0.36	59.9	24.5	16.9	0.43	93.1	29.8	29.3
Subtotal	49.8	2115	640	542	18.1	4348	799	1030
<i>Phthalate Esters^c</i>								
Dimethyl (DMP)	n.d.	1.41	0.48	0.39	0.04	0.94	0.39	0.24
Diethyl (DEP)	n.d.	4.07	1.11	1.04	n.d.	8.30	1.19	1.78
Diisobutyl (DiBP)	3.92	646	105	152	2.76	96.1	33.4	30.0
Di- <i>n</i> -butyl (DnBP)	5.38	335	63.3	78.5	5.53	81.2	27.0	20.9
Subtotal	9.83	985	170	230	9.61	160	62.0	49.1
<i>Glycerol and Polyacids</i>								
Glycerol	2.18	93.4	19.9	22.4	0.78	200	34.4	45.4
Glyceric acid	3.71	173	42.3	40.9	2.63	180	48.7	50.7
Malic acid	10.2	236	68.2	52.6	2.97	219	63.4	55.3
Tartaric acid	0.65	25.9	5.07	5.63	0.28	11.3	3.50	3.40
Citric acid	0.19	12.5	4.00	3.01	n.d.	12.7	3.06	2.97
Subtotal	26.9	536	139	118	7.10	487	153	144
<i>Aromatic Acids</i>								
Benzoic acid	0.98	27.1	9.01	7.16	0.59	39.2	10.9	10.7
Phthalic acid	7.08	57.5	26.3	13.3	0.95	60.8	24.0	16.7
Isophthalic acid	0.64	5.87	2.61	1.70	0.33	8.15	2.42	2.10
Terephthalic acid	2.44	53.7	12.2	11.8	1.07	20.4	7.82	4.87
Subtotal	19.6	105	50.1	25.7	14.8	113	45.2	31.4
<i>Lignin and Resin Products</i>								
4-Hydroxybenzoic acid	0.98	37.4	8.91	9.82	0.13	52.7	8.12	12.0
Vanillic acid	0.21	5.81	2.15	1.75	0.03	25.2	2.91	5.51
Syringic acid	0.02	4.23	1.36	1.26	0.02	8.77	1.35	1.99
Dehydroabietic acid	0.21	42.3	3.88	9.15	0.18	3.14	1.19	0.89
Subtotal	1.78	57.5	16.3	15.9	0.84	89.8	13.6	19.9
<i>Sterols</i>								
Cholesterol	0.91	23.7	7.16	6.69	0.44	32.5	8.39	8.00
β -Cholesterol	n.d.	31.9	4.10	7.67	n.d.	10.7	1.99	3.26
Ergosterol	n.d.	10.0	1.73	2.69	n.d.	45.9	4.37	10.0
Stigmasterol	n.d.	3.01	1.10	1.12	n.d.	17.9	2.63	4.18
β -Sitosterol	n.d.	43.2	11.2	10.9	0.67	200	29.5	49.8
Subtotal	1.41	64.5	25.3	17.3	2.45	250	46.9	65.5
<i>Hopanes</i>								
C ₂₇ α	0.04	0.62	0.17	0.13	n.d.	0.78	0.26	0.23
C ₂₇ β	0.02	0.41	0.14	0.11	n.d.	0.37	0.14	0.11
C ₂₉ $\alpha\beta$	0.02	1.12	0.33	0.26	0.02	1.45	0.33	0.31
C ₂₉ $\beta\alpha$	n.d.	1.26	0.29	0.31	0.04	1.23	0.28	0.28
C ₃₀ $\alpha\beta$	0.09	1.02	0.47	0.27	0.05	1.67	0.48	0.39
C ₃₀ $\beta\alpha$	n.d.	0.76	0.17	0.17	0.01	0.58	0.16	0.14
C ₃₁ $\alpha\beta$ S	n.d.	1.85	0.46	0.49	0.02	1.48	0.37	0.34
C ₃₁ $\alpha\beta$ R	0.02	0.86	0.28	0.22	n.d.	0.78	0.31	0.27
C ₃₁ $\beta\alpha$	n.d.	1.45	0.48	0.41	n.d.	2.42	0.65	0.69
C ₃₂ $\alpha\beta$ S	n.d.	0.95	0.31	0.24	n.d.	0.83	0.33	0.23
C ₃₂ $\alpha\beta$ R	n.d.	0.82	0.32	0.26	n.d.	0.91	0.31	0.24
Subtotal	0.82	7.34	3.41	1.96	0.75	11.1	3.63	2.58
<i>PAHs</i>								
Phenanthrene (Phen)	0.04	2.75	0.68	0.58	0.13	2.28	0.57	0.47
Anthracene (Anthr)	n.d.	0.18	0.09	0.05	n.d.	0.42	0.11	0.10
Fluoranthene (Flu)	0.72	2.58	1.52	0.48	0.29	3.18	1.54	0.76
Pyrene (Pyr)	0.51	2.36	1.35	0.48	0.20	2.71	1.39	0.71
Benzo(b)fluorine (BbFlr)	n.d.	0.35	0.14	0.11	n.d.	0.38	0.16	0.14
Benz(a)anthracene (BaA)	0.02	0.82	0.31	0.20	0.02	1.58	0.48	0.41

Table 1. (continued)

Compounds	Daytime (n = 19)				Nighttime (n = 20)			
	Minimum	Maximum	Mean	SD	Minimum	Maximum	Mean	SD
Chrysenetriphenylene(Chry/Trp)	0.27	2.57	1.17	0.64	0.21	4.01	1.62	0.96
Benzo(b)fluoranthene (BbF)	0.59	3.62	2.15	0.75	0.33	5.32	2.32	1.16
Benzo(k)fluoranthene (BkF)	0.47	2.80	1.26	0.67	0.31	2.94	1.41	0.86
Benzo(e)pyrene (BeP)	0.31	2.20	1.25	0.50	0.16	2.76	1.24	0.63
Benzo(a)pyrene (BaP)	0.22	1.92	0.92	0.42	0.10	1.56	0.85	0.41
Perylene (Per)	0.03	0.67	0.27	0.16	0.01	0.89	0.24	0.19
Indeno(1,2,3-cd)pyrene (IP)	0.22	1.85	1.03	0.44	0.16	2.17	1.06	0.59
Dibenz(a,h)anthracene (DbahA)	n.d.	0.32	0.12	0.12	n.d.	0.22	0.09	0.07
Benzo(ghi)perylene (BghiP)	0.23	2.35	1.30	0.52	0.19	2.50	1.26	0.65
Anthanthrene (AA)	n.d.	0.56	0.21	0.13	n.d.	0.48	0.17	0.14
Coronene (Cor)	0.11	1.93	0.81	0.48	0.11	1.83	0.70	0.52
Subtotal	5.18	25.2	14.6	5.24	2.32	31.4	15.2	7.60

^aConcentration is measured in ng m^{-3} . SD is standard deviation and n.d. is not detected.

^bCPI: carbon preference index: $(C_{21}+C_{23}+C_{25}+C_{27}+C_{29}+C_{31}+C_{33}+C_{35})/(C_{20}+C_{22}+C_{24}+C_{26}+C_{28}+C_{30}+C_{32}+C_{34}+C_{36})$ for *n*-alkanes; $(C_{20}+C_{22}+C_{24}+C_{26}+C_{28}+C_{30}+C_{32})/(C_{21}+C_{23}+C_{25}+C_{27}+C_{29}+C_{31})$ for fatty acids; $(C_{18}+C_{20}+C_{22}+C_{24}+C_{26}+C_{28}+C_{30}+C_{32}+C_{34})/(C_{19}+C_{21}+C_{23}+C_{25}+C_{27}+C_{29}+C_{31}+C_{33})$ for fatty alcohols.

^cBis(2-ethylhexyl) phthalate was neglected owing to the contamination from the sampling campaign.

63–783 ng m^{-3} (average 262 ng m^{-3}) in daytime and 15.6–892 ng m^{-3} (287 ng m^{-3}) in nighttime. Although the difference in the total concentrations was not significant between daytime and nighttime (Figure 3), concentrations of long-chain fatty acids (C_{26} – C_{32}) are much higher in nighttime (Table 1). This is consistent with plant wax derived C_{27} – C_{33} *n*-alkanes whose concentrations are higher in nighttime. The fatty acid concentrations over Mt. Tai are about half of those from ground surface in Chinese megacities (155–877 ng m^{-3} , average 473 ng m^{-3} in summer) [Wang *et al.*, 2006a] but are several times higher than those from the aircraft samples collected over coastal China (26–160 ng m^{-3} , 68 ng m^{-3}) [Wang *et al.*, 2007] and 1–2 orders of magnitude higher than those of Chichi-Jima aerosols (2.46–60.2 ng m^{-3} , 13.8 ng m^{-3}) [Kawamura *et al.*, 2003]. Higher molecular weight fatty acids ($\geq C_{20}$) (HFAs) are derived from terrestrial higher plant wax, while lower molecular weight fatty acids ($\leq C_{19}$) (LFAs) have multiple sources such as vascular plants, microbes, and marine phytoplankton as well as kitchen emissions [Kawamura *et al.*, 2003; Kolattukudy, 1976; Simoneit and Mazurek, 1982]. As for the summertime Mt. Tai samples, LFAs/HFAs ratios varied with samples (average 1.02 ± 0.80). The CPI values (even/odd) of the fatty acids are 6.04 in daytime and 5.53 in nighttime.

[14] Oleic ($C_{18:1}$) and linoleic ($C_{18:2}$) acids were detected in the Mt. Tai aerosols (Table 1). They were directly emitted from many sources, including leaf surfaces of plants with $C_{18:2} > C_{18:1}$ [Rogge *et al.*, 1993b], meat charbroiling with $C_{18:2} > C_{18:1}$ [Schauer *et al.*, 1999], wood combustion with $C_{18:2} > C_{18:1}$ [Fine *et al.*, 2001]. However, only $C_{18:1}$ was reported in automobile source [Rogge *et al.*, 1993a]. The concentration range of $C_{18:1}$ was 0.81–12.1 ng m^{-3} (average 2.77 ng m^{-3}) in daytime and 0.61–7.90 ng m^{-3} (3.21 ng m^{-3}) in nighttime, while $C_{18:2}$ was n.d.–5.20 ng m^{-3} (0.73 ng m^{-3}) in daytime and n.d.–11.8 ng m^{-3} (1.73 ng m^{-3}) in nighttime. The difference in their concentrations indicates a fast destruction of these compounds via photochemical oxidation of double bonds [Kawamura and Gagosian, 1987]. $C_{18:2}$ acid was often nondetectable in daytime, indicating that $C_{18:2}$ degrades more quickly than $C_{18:1}$ under the solar radiation

due to two double bonds in $C_{18:2}$. Polyunsaturated fatty acids are more reactive toward OH, NO_3 radicals, and ozone. The $C_{18:0}/C_{18:1}$ ratios obtained during this study were 9.4 ± 5.0 in daytime versus 6.5 ± 4.1 in nighttime, suggesting an enhanced photochemical degradation of unsaturated fatty acids during daytime.

[15] Molecular distributions of C_{18} – C_{34} fatty alcohols are characterized by strong even carbon number predominance with C_{max} at C_{28} in both day and night samples (Table 1 and Figure 2d). Concentrations of total *n*-alcohols (C_{18} – C_{34}) were 15.1–866 ng m^{-3} (average 249 ng m^{-3}) in daytime and 11.1–1930 ng m^{-3} (352 ng m^{-3}) in nighttime. Again we found higher concentrations of *n*-alcohols in nighttime, especially for C_{24} – C_{30} species, being consistent with plant wax derived *n*-alkanes and fatty acids. Their concentrations were within the range of 8–1200 ng m^{-3} reported for the aerosols from Chinese cities [Simoneit *et al.*, 1991; Wang *et al.*, 2006a]. Long-chain fatty alcohols are abundant in higher plant waxes and loess deposits [Simoneit *et al.*, 1991]. They may also be emitted to the air by biomass burning process [Kawamura *et al.*, 2003; Simoneit, 2002].

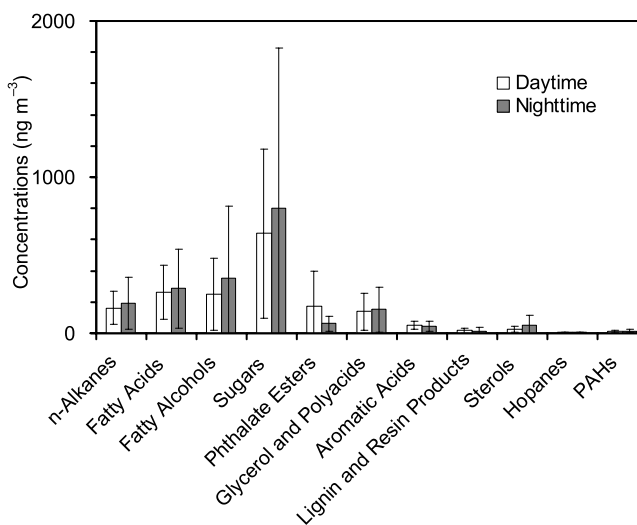


Figure 3. Concentrations of organic compound classes detected in the aerosols over Mt. Tai.

Table 2. Concentrations of Levoglucosan and Its Two Diastereomers (Galactosan and Mannosan) in Ambient Aerosols^a

Sample	Galactosan	Mannosan	Levoglucosan	Reference
Santiago, Chile (TSP)	1.7–127 (45)	1–233 (81)	12–2452 (839)	[Simoneit et al., 2004a]
Kuala Lumpur, Malaysia (TSP)	84–2410 (1014)	154–4430 (1422)	1162–33,400 (14,460)	[Simoneit et al., 2004a]
Sapporo, Japan (TSP)	0.6–2.4	0.2–15	6–56	[Simoneit et al., 2004a]
Gent, Belgium (PM10, summer)	0.67–1.17 (1.02)	2.0–3.9 (3.0)	9.1–27 (19.1)	[Pashynska et al., 2002]
Gent, Belgium (PM10, winter)	4.9–115 (25)	10–290 (61)	96–1900 (420)	[Pashynska et al., 2002]
Rondônia, Brazil (PM10, dry season)	7.6–61.5 (31.2)	21–259 (116)	446–4106 (2006)	[Zdrahal et al., 2002]
Rondônia, Brazil (PM10, wet season)	n.d.–1.19 (0.63)	n.d.–2.05 (0.98)	0.40–13.2 (4.40)	[Zdrahal et al., 2002]
Rondônia (Pasture), Brazil (PM2.5)	2.3–148 (55.4)	6.0–371 (126)	1182–6900 (2460)	[Graham et al., 2002]
Rondônia (Forest), Brazil (PM2.5)	1.6–44.6 (22.7)	1.7–127 (49.5)	39.9–2660 (1180)	[Graham et al., 2002]
Bakersfield, Canada (PM3)	96	171	2390	[Nolte et al., 2002]
Fresno, Canada (PM3)	144	322	2980	[Nolte et al., 2002]
Howland Forest, Maine (TSP)	n.d.–2.6	n.d.–10.4	1.0–55.1	[Medeiros et al., 2006]
Mount Lulin, Taiwan (PM2.5)	0.06–0.75	n.d.–0.49	1.6–132	[Hsu et al., 2007]
Gosan, Jeju Island (TSP)	n.d.–3.8	0.2–4.2	8–74	[Simoneit et al., 2004c]
Yellow Sea (TSP, aircraft samples)	-	0.2–1.1	2.2–6.0	[Simoneit et al., 2004b]
Datong, China (TSP)	106	108	1350	[Simoneit et al., 2004a]
Mt. Tai, China (TSP, daytime)	0.44–46.6 (8.42)	0.60–43.1 (9.71)	18.5–1733 (391)	this study
Mt. Tai, China (TSP, nighttime)	0.02–103 (12.8)	0.1–61.6 (9.97)	0.37–3432 (459)	this study

^aConcentration is measured in ng m^{-3} . Numbers in parentheses are the mean.

CPIs of fatty alcohols were 25.6 ± 15.2 in daytime versus 22.8 ± 9.25 in nighttime. The CPI values together with the molecular distributions (Figure 2d) suggested that *n*-alcohols in the troposphere over Mt. Tai are most likely derived from waxes of higher plant origin. However, the molecular composition and concentrations of lipid components do not differentiate two emission pathways of plant waxes, that is, wind-driven sloughing/ablation or biomass burning-related evaporation.

3.1.2. Sugar Compounds

[16] Sugars are water-soluble and thus contribute to water-soluble organic carbon (WSOC) in aerosols and can be used as tracers for primary biological aerosol particles [Elbert et al., 2007; Graham et al., 2003; Medeiros et al., 2006; Simoneit and Mazurek, 1982; Yttri et al., 2007]. They are believed to influence the hygroscopicity of aerosols [Mochida and Kawamura, 2004] and thus potentially play an important role in the regulation of climate [Kanakidou et al., 2005]. Ten sugars (levoglucosan, galactosan, mannosan, arabinol, fructose, glucose, mannitol, inositol, sucrose, and trehalose) were detected in the aerosols over Mt. Tai (Table 1). The concentrations of sugars were $49.8\text{--}2115 \text{ ng m}^{-3}$ (average 640 ng m^{-3}) in daytime versus $18.1\text{--}4348 \text{ ng m}^{-3}$ (799 ng m^{-3}) in nighttime. The higher nighttime concentrations are consistent with those of wax components such as *n*-alkanes, fatty acids, and *n*-alcohols as described in section 3.1.1. Interestingly, their concentrations are several times higher than those reported in Chinese coastal urban aerosols (average 262 ng m^{-3} in summer) [Wang et al., 2006a], and an order of magnitude higher than those reported in the aircraft samples over China ($31 \pm 25 \text{ ng m}^{-3}$ in summer) and in the northwestern Pacific ($4.1\text{--}22.5 \text{ ng m}^{-3}$) [Simoneit et al., 2004b].

[17] Levoglucosan (1,6-anhydro- β -D-glucopyranose), specifically formed by a pyrolysis of cellulose, has been recognized as an excellent tracer of biomass-burning [Simoneit, 2002; Simoneit et al., 1999]. It is often reported in biomass burning plumes [Graham et al., 2002; Hays et al., 2005; Inuma et al., 2007; Sheesley et al., 2003], urban areas [Chowdhury et al., 2007; Oliveira et al., 2007; Pashynska et al., 2002; Robinson et al., 2006; Wang and

Kawamura, 2005; Wang et al., 2006a; Zheng et al., 2005] and urban dust reference materials [Larsen et al., 2006], and oceans [Simoneit and Elias, 2000; Simoneit et al., 2004c]. In the aerosols over Mt. Tai, levoglucosan was found to be the most abundant among the individually identified compounds (Table 1), being similar to previous studies [Graham et al., 2002; Hoffer et al., 2006]. The percentage of levoglucosan to WSOC ($1.27\text{--}37.2 \text{ } \mu\text{g m}^{-3}$) ranged from 0.27 to 7.18% (average 1.50%) in daytime versus 0.01–4.52% (1.13%) in nighttime, while the percentage of levoglucosan to TC ($3.45\text{--}87.4 \text{ } \mu\text{g m}^{-3}$) was 0.10–3.16% (0.64%) in daytime and 0.01–1.75% (0.43%) in nighttime. In the urban aerosols from Chinese megacities, levoglucosan could contribute up to 90% of total identified sugars [Wang et al., 2006a]. In the Mt. Tai aerosols, levoglucosan contributed $46.1 \pm 20.4\%$ of total sugars in daytime versus $39.0 \pm 18.0\%$ in nighttime. This suggests that the tropospheric aerosols over Mt. Tai contain sugar compounds derived from different sources than biomass burning, a point to be discussed later.

[18] In addition to levoglucosan, galactosan and mannosan were identified in all the samples studied. The concentration range of galactosan was $0.44\text{--}46.6 \text{ ng m}^{-3}$ (8.42 ng m^{-3}) in daytime and $0.02\text{--}103 \text{ ng m}^{-3}$ (12.8 ng m^{-3}) in nighttime, while mannosan was $0.60\text{--}43.1 \text{ ng m}^{-3}$ (9.71 ng m^{-3}) in daytime and $0.10\text{--}61.6 \text{ ng m}^{-3}$ (9.97 ng m^{-3}) in nighttime (Table 1). These anhydrosugars are also formed through the pyrolysis of cellulose/hemicelluloses present in biomass [Simoneit, 2002; Simoneit et al., 1999]. They have been often identified in smoke particulate matter from woods [Graham et al., 2002; Inuma et al., 2007; Oros and Simoneit, 2001a; Schmidl et al., 2008; Simoneit et al., 1999], grasses [Inuma et al., 2007; Oros et al., 2006], and agricultural wastes such as rice straws [Sheesley et al., 2003]. Reports on the concentrations of galactosan, mannosan, and levoglucosan in ambient aerosols are summarized in Table 2. In the aerosols over Mt. Tai, the level of galactosan was slightly lower than mannosan in daytime. However, galactosan became more abundant than mannosan in nighttime. This is in agreement with some studies on smoke particles derived from rice straws, biomass briquettes and grasses, in which the levels of galactosan are higher than mannosan [Oros et al., 2006;

Table 3. Results of Principal Component Analysis With Varimax Rotation for the Data Set of Sugars in the Daytime/Nighttime Mt. Tai Aerosols

Sugars	Daytime			Nighttime	
	Component 1	Component 2	Component 3	Component 1	Component 2
Galactosan	0.96	0.14	0.02	0.90	0.28
Mannosan	0.91	0.15	-0.05	0.92	0.30
Levoglucozan	0.95	0.10	0.01	0.90	0.28
Arabitol	0.27	0.91	0.17	0.72	0.61
Fructose	0.75	0.50	0.16	0.93	0.30
Glucose	0.91	-0.02	0.01	0.86	-0.15
Mannitol	0.09	0.96	0.12	0.51	0.57
Inositol	0.29	0.18	0.90	0.53	0.76
Sucrose	-0.29	0.29	0.85	-0.21	0.88
Trehalose	0.04	0.89	0.24	0.33	0.79
Variance (%)	42.8	29.6	16.6	52.9	30.4

Sheesley *et al.*, 2003]. Thus, the higher abundance of galactosan over mannosan in nighttime again suggests that more biomass burning products from agricultural wastes in the North China Plain could be transported over Mt. Tai above the PBL.

[19] Primary saccharides consist of glucose, fructose, inositol, sucrose, and trehalose, as well as some sugar polyols (reduced sugars) including arabitol and mannitol. They have been proposed as tracers for resuspension of surface soil and unpaved road dust [Simoneit *et al.*, 2004a], which contain biological materials including fungi and bacteria. Their concentrations are relatively high in the atmospheric particles during summer season, paralleling the agricultural tilling practices on the Asian continent that resuspend soil dust into the passing aerosols [Simoneit *et al.*, 2004c]. Previous study mentioned that field burning of wheat straws in the North China Plain during May–June 2006 caused serious air pollution even in urban areas such as Beijing [Li *et al.*, 2007]. These agricultural activities played a significant role in controlling the molecular composition and concentrations of sugar compounds in the lower troposphere on a local and regional scale. In order to get further insight to the sources of sugars in the tropospheric aerosols over Mt. Tai, principal component analysis (PCA) was performed using the concentrations of various sugars as variables.

[20] PCA is a useful approach for verifying the sources of aerosols [Hopke, 1985]. It has been successfully used for a series of dicarboxylic acids (DCAs) and other atmospheric trace species [Kawamura and Sakaguchi, 1999; Mochida *et al.*, 2003; Wolff and Korsog, 1985]. The data sets of sugars for daytime ($n = 19$) and nighttime ($n = 20$) aerosol samples from Mt. Tai 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 3. Three and two components were set for daytime and nighttime samples, respectively, by the scree tests.

[21] For the daytime data set, three components were found to account for 89.0% of the total variance, with the first component corresponding to 42.8%. Levoglucozan, galactosan, mannosan, and glucose showed loadings of >0.90 in component 1, which is mainly associated with the emissions from biomass burning. Medeiros *et al.* [2006] reported that wildfires strongly enhanced emissions of uncombusted saccharides including glucose. Fructose also

showed a loading of 0.75 in component 1. In contrast, arabitol, mannitol, and trehalose showed loadings of >0.89 in component 2, suggesting a common biological origin. These sugar polyols are produced in large amounts 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]. They are also used to trace the resuspension of surface soil and unpaved road dust [Simoneit *et al.*, 2004c; Wang and Kawamura, 2005]. Inositol and sucrose showed loadings of >0.85 in component 3 (16.6%).

[22] Pashynska *et al.* [2002] showed that the atmospheric levels of inositol and sucrose and their contributions to organic carbon (OC) were highest at the beginning of summer (June) owing to the developing leaves. Sucrose is the predominant sugar in the phloem of plants and is important in developing flower buds [Bieleski, 1995]. The present campaign was conducted during the early summer (May/June). Thus, the third component may be associated with sugar compounds emitted from the developing leaves and pollen in addition to suspended soil dust. Jaenicke [2005] showed that the contributions of primary bioaerosols (including plant fragments, pollen, etc.) to total atmospheric particles ($>0.2 \mu\text{m}$) from Lake Baikal (Russia) and Mainz (Germany) ranged from 20% to 30%. 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 “giant” bioaerosols. However, the importance of bioaerosols in the atmospheric aerosol content is still controversial [Hoffmann and Warnke, 2007].

[23] During nighttime, two components were found to account for 83.3% of the total variance, with the first component corresponding to 52.9% (Table 3). Levoglucozan, galactosan, mannosan, fructose, and glucose, which are all derived from biomass burning, showed loadings of >0.86 in the first component. Arabitol, mannitol, and inositol also showed loadings of >0.51 in component 1, suggesting that they are influenced by biomass burning as well. These results suggest that biomass burning is the primary source of sugars in nighttime aerosols. Biomass burning plumes may be relatively more transported over Mt. Tai than biogenic sugars by long-range atmospheric transport under the less active conditions of vegetation in nighttime. How-

ever, concentrations of inositol, sucrose, and trehalose did not show high loadings with component 1, but have large loadings >0.76 in component 2. Meanwhile, arabitol and mannitol showed loadings of 0.61 and 0.57 in component 2. As mentioned earlier in this section, these primary saccharides and sugar polyols could be derived from suspended soil dust.

[24] In the troposphere over Mt. Tai, the average concentration of primary saccharides (fructose, glucose, inositol, sucrose, and trehalose) was $122 \pm 54 \text{ ng m}^{-3}$ in daytime, being higher than sugar polyols (arabitol and mannitol) ($109 \pm 83 \text{ ng m}^{-3}$). In contrast, the nighttime concentration of primary saccharides was $156 \pm 110 \text{ ng m}^{-3}$, being lower than sugar polyols ($162 \pm 144 \text{ ng m}^{-3}$). These results suggest that sugar compounds in the lower troposphere over Mt. Tai mainly originate from biomass burning and suspended soil dusts. However, the direct release of bioaerosols (e.g., pollen) and emission from developing leaves in the Mt. Tai area is also an important source of sugars and sugar polyols in the atmosphere, especially in daytime when these materials can be transported toward the summit of Mt. Tai as a result of the expansion of mixing layer and the uplift of PBL.

3.1.3. Glycerol and Polyacids

[25] Glycerol and polyacids (glyceric, malic, tartaric, and citric acids) were detected in the aerosol samples. They also contribute to WSOC in aerosols. Their concentrations were $26.9\text{--}536 \text{ ng m}^{-3}$ in daytime (average 139 ng m^{-3}) versus $7.10\text{--}487 \text{ ng m}^{-3}$ (153 ng m^{-3}) in nighttime (Table 1). The contributions of glycerol and polyacids to WSOC ranged from 0.15 to 0.79% (0.45%) in daytime versus 0.19–0.81% (0.46%) in nighttime.

[26] Malic acid is the most abundant species in this group (Table 1), accounting for about 40% of glycerol and polyacids. These values are much higher than those from ground surface aerosols in Nanjing (about 15%), but are lower than those reported in the aircraft samples over China, where malic acid accounted for more than 60% [Wang *et al.*, 2007]. Glycerol is a metabolic product in soil organisms [Simoneit *et al.*, 2004a], whereas polyacids are secondary photooxidation products of atmospheric organic precursors. For example, malic acid can be produced by the photochemical oxidation of succinic acid in the atmosphere [Kawamura and Ikushima, 1993; Kawamura and Sakaguchi, 1999; Simoneit *et al.*, 2004b]. The photooxidation of isoprene also results in secondary organic aerosol (SOA) formation such as malic acid and tartaric acid [Claeys *et al.*, 2004]. The increased percentage of malic acid in the Mt. Tai aerosols compared to the urban aerosols as discussed earlier, strongly suggests an enhanced photochemical production of polyacids in the troposphere over high mountains.

3.1.4. Phthalate Esters

[27] Phthalate esters are manufactured worldwide and are used as plasticizers in synthetic polymers and a softener in polyvinylchloride. Phthalate esters 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 phthalate esters due to their potential carcinogenic and endocrine disrupting properties [Sidhu *et al.*, 2005]. Four phthalate esters were detected in this study, i.e., dimethyl (DMP), diethyl (DEP), diisobutyl (DiBP), and di-*n*-butyl (DnBP) phthalates (Figure 2e). Summertime concentrations

of phthalates were $9.83\text{--}985 \text{ ng m}^{-3}$ (average 170 ng m^{-3}) in daytime and $9.61\text{--}160 \text{ ng m}^{-3}$ (62 ng m^{-3}) in nighttime (Table 1). Their higher concentrations in daytime are opposite to the trends of higher plant derived lipids and sugar compounds. The higher daytime concentrations may be caused by enhanced emission of phthalates from plastics due to the higher ambient temperature. In fact, higher concentrations of phthalates in summertime than wintertime were reported in Chinese megacities [Wang *et al.*, 2006a]. The enhanced daytime concentrations can also be explained by the upward transport of phthalates following the uplift of PBL from the near surface of the North China Plain to the height over Mt. Tai.

[28] DiBP was in general a dominant species in this group, followed by DnBP, DEP, and DMP (Table 1). Among them, DiBP and DnBP showed a strong linear correlation in both daytime ($r^2 = 0.99$, $p < 0.05$, $n = 19$) and nighttime ($r^2 = 0.81$, $p < 0.05$, $n = 20$) (Figures 4a and 4d), suggesting that they are commonly used plasticizers in China and emitted into the atmosphere in a similar way. However, the correlation coefficient in daytime is much higher than in nighttime, suggesting that DiBP and DnBP may have different processes and lifetime in the atmosphere because the summit of Mt. Tai locates in the free troposphere at night and thus the nighttime aerosols are more aged than daytime aerosols.

3.2. Minor Organic Compound Classes

3.2.1. Lignin and Resin Products and Sterols

[29] Three lignin products (4-hydroxybenzoic acid, vanillic and syringic acids) and one resin product (dehydroabietic acid), as well as sterols (cholesterol, ergosterol, stigmasterol, and β -sitosterol) were detected in the Mt. Tai samples. Total concentrations of lignin and resin products were $1.78\text{--}57.5 \text{ ng m}^{-3}$ (average 16.3 ng m^{-3}) in daytime and $0.84\text{--}89.8 \text{ ng m}^{-3}$ (13.6 ng m^{-3}) in nighttime. These values are similar to those reported in Chinese urban aerosols ($1.3\text{--}53 \text{ ng m}^{-3}$ in summer) [Wang *et al.*, 2006a]. The 4-Hydroxybenzoic acid was the dominant species in daytime, followed by dehydroabietic acid, vanillic acid, and syringic acid. Dehydroabietic acid is produced by pyrolytic dehydration of abietic acid that is abundant in pine resin. Thus, this compound can be used as more specific marker of biomass burning of pine trees [Simoneit *et al.*, 1993]. In contrast, vanillic acid became more abundant than dehydroabietic acid in nighttime (Table 1). These lignin- and resin-derived organic acids have been reported in the continental aerosols and in the smoke particles from biomass burning [Simoneit, 2002].

[30] Total concentrations of sterols were $1.41\text{--}64.5 \text{ ng m}^{-3}$ (average 25.3 ng m^{-3}) in daytime and $2.45\text{--}250 \text{ ng m}^{-3}$ (46.9 ng m^{-3}) in nighttime. All the sterols, except for β -sitosterol, showed higher concentrations in nighttime, being consistent with the results of higher plant waxes (*n*-alkanes, fatty acids, and *n*-alcohols). β -Sitosterol was in general a dominant species, followed by cholesterol and other sterols. β -Sitosterol and stigmasterol are present in terrestrial higher plant leaves and emitted to the air via their biomass burning processes [Kawamura *et al.*, 2003; Simoneit, 2002]. Cholesterol is a tracer for smoke particles generated from meat cooking [Rogge *et al.*, 1991] and has been proposed as a source marker of marine organisms

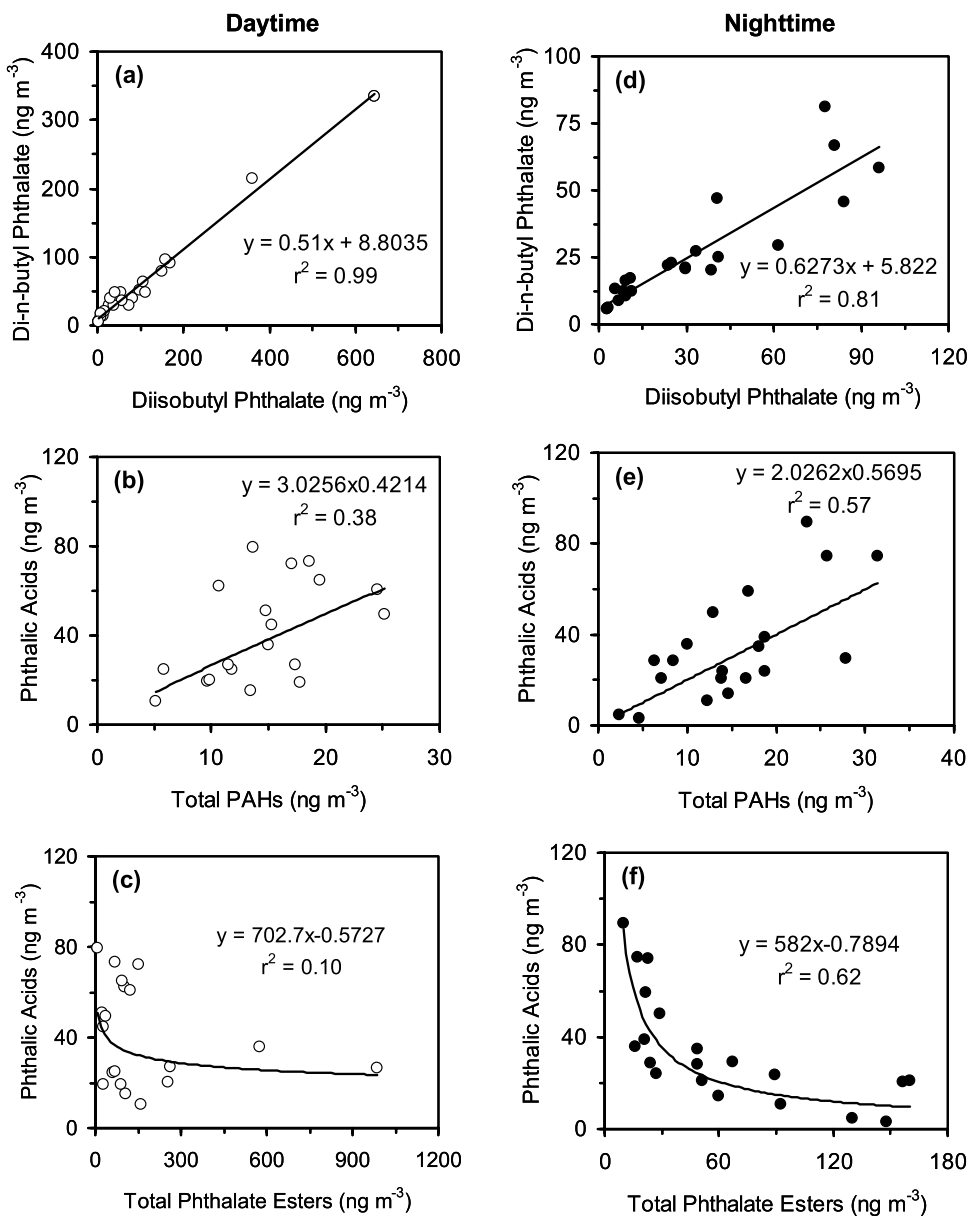


Figure 4. Relationships between the concentrations of individual compounds or compound classes in the Mt. Tai aerosols. (a, b, and c) Daytime samples and (d, e, and f) nighttime samples.

[Kawamura *et al.*, 2003]. β -Sitosterol can also be produced by cooking because they are present in plant oils [Hoffmann and Warnke, 2007]. The concentration ranges of ergosterol, a biomarker for fungal biomass in atmospheric aerosols [Lau *et al.*, 2006], were n.d.–10.0 ng m⁻³ (average 1.73 ng m⁻³) in daytime and n.d.–45.9 ng m⁻³ (4.37 ng m⁻³) in nighttime, which was comparable to those reported in urban samples from Chongqing, Xi'an, Tianjing and Xiamen [Wang *et al.*, 2006a] but much higher than those (0.03–0.41 ng m⁻³) reported from Hong Kong [Lau *et al.*, 2006]. The higher level of ergosterol found in this study is probably related to the higher fungal activities associated with soil or decomposition of large vegetation in the mountain area.

[31] The abundance of lignin and resin products in the troposphere over Mt. Tai, as well as levoglucosan as

mentioned in section 3.1.2, indicates that biomass burning is an important source to control organic aerosol composition and concentration in this region. In China, especially the rural areas, biomass is still a major domestic energy source for cooking and heating, and field burning of agriculture residues is a common way to eliminate the wastes after harvesting crops [Li *et al.*, 2007]. For example, annual rate of biomass burning in Shandong Province of China, where Mt. Tai locates, is up to 7.4 Tg of crop residue and 31 Tg of biofuels [Streets *et al.*, 2003]. These values account for more than 1% of the biomass burned in the world [Fernandes *et al.*, 2007].

3.2.2. Aromatic Acids

[32] Four aromatic acids, i.e., benzoic acid and three phthalic acids (including *iso* and *tere* isomers), were detected in the aerosol samples. The concentration ranges of benzoic

acid were 0.98–27.1 ng m⁻³ (average 9.01 ng m⁻³) in daytime and 0.59–39.2 ng m⁻³ (10.9 ng m⁻³) in nighttime (Table 1). Benzoic acid has been proposed as a primary pollutant in the exhaust of motor vehicles [Kawamura *et al.*, 1985, 2000; Rogge *et al.*, 1993a] and a secondary product from photochemical degradation of aromatic hydrocarbons such as toluene emitted by automobiles [Suh *et al.*, 2003].

[33] The concentration ranges of total phthalic acids were 10.3–79.5 ng m⁻³ (41.1 ng m⁻³) in daytime and 3.29–89.3 ng m⁻³ (34.2 ng m⁻³) in nighttime. Their day/night trends are different from those of plant wax-derived components and sugars. Their concentrations are higher than those reported from aircraft aerosols over China (17 ± 13 ng m⁻³ in summer) [Wang *et al.*, 2007] and over the northwestern Pacific (average 1.5 ng m⁻³) [Simoneit *et al.*, 2004b]. The isomer distribution of phthalic acids was characterized by a predominance of phthalic acid (Table 1) followed by terephthalic acid, being consistent with those reported in the aircraft aerosols over China [Wang *et al.*, 2007] and East China Sea [Simoneit *et al.*, 2004b]. Phthalic acids have been proposed as SOA products that can be used as a surrogate for the contributions of SOA to an ambient sample [Oliveira *et al.*, 2007]. A possible source of phthalic acids is the oxidation of naphthalene or other PAHs [Fine *et al.*, 2004].

[34] In this study, a positive correlation was found between the concentrations of phthalic acids and PAHs in nighttime ($r^2 = 0.57$, see Figure 4e). Similar positive but weaker correlation was also found in daytime ($r^2 = 0.38$, Figure 4b). Interestingly, a strong negative correlation between phthalic acids and phthalate esters was found in nighttime ($r^2 = 0.62$, Figure 4f), while no correlation was found in daytime samples ($r^2 = 0.10$, Figure 4c). The intrinsic relationship between phthalic acids and phthalate esters in the atmosphere generally suggests that they are linked with similar anthropogenic sources; however, their behaviors in the atmosphere are different. Phthalic acids may be produced by the photochemical oxidation of aromatic hydrocarbons in the atmosphere over the North China Plain. In contrast, phthalate esters are directly emitted from plastics and synthetic polymers used in urban and other areas, as discussed in section 3.1.4.

3.2.3. Hopanes (Hopanoid Hydrocarbons)

[35] Hopanes are specific biomarkers of petroleum and coal. They are enriched in motor oils [Kawamura *et al.*, 1995] and are emitted to the atmosphere from internal combustion engines and the usage of coal [Rogge *et al.*, 1993a; Simoneit *et al.*, 1991]. A series of hopanes (C₂₇–C₃₂ except for C₂₈) were detected in the aerosols (see Table 1), with average concentrations of 3.41 ± 1.96 ng m⁻³ in daytime and 3.63 ± 2.58 ng m⁻³ in nighttime. There is no significant difference in the daytime and nighttime concentrations. Their concentrations are similar to those reported in Chinese megacities (3.1 ± 4.6 ng m⁻³) [Wang *et al.*, 2006a], Tokyo (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]. However, they are generally not detectable in the remote marine aerosols from the Pacific [Simoneit *et al.*, 2004c]. Our results suggest that the upward transport is probably an important pathway for hopanes detected over Mt. Tai in nighttime.

3.2.4. PAHs

[36] PAHs are of health concerns due to their carcinogenicity, genotoxicity, and potential endocrine disruptiveness. Their sources include coal combustion, automobile emissions, biomass burning, and natural gas combustion. In this study, 17 PAHs (3- to 7-rings) were detected in the Mt. Tai samples ranging from phenanthrene to coronene. Their total concentrations were 5.18–25.2 ng m⁻³ (average 14.6 ng m⁻³) in daytime and 2.32–31.4 ng m⁻³ (15.2 ng m⁻³) in nighttime (Table 1). There is no significant change between the averaged daytime and nighttime samples, being consistent with fossil fuel-derived hopanes. Their concentrations are lower than those observed in the ground surface aerosols from Chinese megacities (1.7–168 ng m⁻³, average 28 ng m⁻³ in summer) [Wang *et al.*, 2006a] but much higher than those reported in the marine aerosol samples from the East China Sea (average 0.44 ng m⁻³) [Kaneyasu and Takada, 2004] and those reported in the aircraft samples (0–0.27 ng m⁻³) over the East China Sea and Sea of Japan [Simoneit *et al.*, 2004b]. Among the PAHs detected, benzo(b)fluoranthene (BbF) was the dominant species (Table 1), being consistent with the results of ground surface aerosols from Chinese cities [Wang *et al.*, 2006a] and the aircraft aerosols over China [Wang *et al.*, 2007] and the East China Sea [Simoneit *et al.*, 2004b]. BbF is the most abundant PAH detected in Chinese megacities, which is predominantly produced by the burning of Chinese coals [Wang *et al.*, 2006a].

3.3. Temporal Variations

[37] To investigate the general sources of organic aerosols, PCA was applied for the daytime/nighttime data sets of 11 compound classes (Table 4). In both data sets, most of the organic compound classes (lipid components, sugars, glycerol and polyacids, aromatic acids, lignin and resin products) showed loadings of >0.80 in component 1, which is most likely associated with the biogenic inputs, mostly with the field burning of wheat straws. Phthalate esters showed large loadings of >0.77 in component 2, indicating an anthropogenic inputs. Hopanes showed loadings of >0.70 in component 2, as well as loadings of >0.5 in component 1 in both daytime and nighttime, suggesting a mixed source of hopanes in the aerosols. PAHs showed daytime loadings of 0.43 and 0.60 in components 1 and 2, respectively. While in nighttime, PAHs showed higher loading of 0.86 in component 1. This probably results from the fact that PAHs may also be derived from biomass burning and transported over the PBL to the summit of Mt. Tai in nighttime together with other organic components.

[38] Figure 5 presents the overall temporal variations of organic compound classes in the aerosol samples collected during summertime in 2006. The *n*-Alkanes, fatty alcohols, and sugars showed two major peaks, one during 5–7 June (Event 1, E1) and the other during 12–14 June (Event 2, E2). We also found a minor peak on 27 June (Event 3, E3). Concentrations of the organic compounds including more water-soluble species generally increased from the beginning of the campaign to E1, then decreased rapidly during 8–11 June. Although a rain event occurred in the evening of 7 June, the precipitation was rather small (0.8 mm). Instead, clean air may have arrived over Mt. Tai from the north as suggested by air mass trajectory (Figure 7c). On the basis of the air mass

Table 4. Results of Principal Component Analysis with Varimax Rotation for the Data Set of the Organic Compound Classes Detected in the Daytime/Nighttime Mt. Tai Aerosols

Compound Classes	Daytime		Nighttime	
	Component 1	Component 2	Component 1	Component 2
<i>n</i> -Alkanes	0.92	0.17	0.98	0.06
Fatty acids	0.92	0.28	0.98	0.06
Fatty alcohols	0.91	0.25	0.96	0.20
Sugars	0.88	0.35	0.96	0.18
Phthalate esters	-0.04	0.77	0.01	0.99
Glycerol and polyacids	0.78	0.37	0.86	0.12
Aromatic acids	0.88	0.28	0.77	0.27
Lignin and resin products	0.83	0.30	0.93	0.25
Sterols	0.87	-0.03	0.90	0.16
Hopanes	0.50	0.70	0.65	0.71
PAHs	0.43	0.60	0.86	0.31
Variance (%)	59.4	18.5	71.9	16.6

trajectory and low CO concentrations (P. Pochanart et al., unpublished data, 2007), we consider that a clean air mass may have intruded from the north over Mt. Tai during 8–11 June. During E2, the highest concentrations for these organic compounds were observed. In late June, concentrations stayed at relatively low levels with a small increase during E3 (Figure 5).

[39] Similar variations were observed for biomass burning tracers such as levoglucosan, galactosan, mannosan, β -sitosterol, 4-hydroxybenzoic acid, vanillic acid, and syringic acid (Figures 6a–6g). Thus, the observed E1 and E2 should indicate major episodes of field burning of agricultural residues such as wheat straws in the North China Plain. This is further supported by the covariation of CO with above tracers during 1–15 June covering E1 and E2 (P. Pochanart et al., unpublished data, 2007). Positive correlations between higher plant-derived biomarkers (*n*-alkanes, fatty acids, and fatty alcohols) and levoglucosan were found ($r^2 = 0.60, 0.54, \text{ and } 0.84$, respectively), indicating that these compounds in the atmospheric aerosols over Mt. Tai during early summer are mainly derived from the open burning of agricultural residues in the North China Plain. On the other hand, dehydroabietic acid, the smoke marker of coniferous trees, showed different trend (Figure 6h) with a major peak during E3, suggesting that E3 may be associated with the biomass burning source different from E1 and E2.

[40] Similar concentration peaks, as well as some additional peaks, were observed for glycerol and polyacids and aromatic acids (Figures 5f and 5i). For example, malic acid that can be produced by photochemical oxidation of succinic acid showed the highest concentrations during E1, followed by E2 and some other concentration peaks during 1–2 June, 19–22 June, and E3 (Figure 6l). Benzoic acid, which has been proposed to be a primary pollutant in the exhaust of motor vehicles [Kawamura et al., 1985; Rogge et al., 1993a] and a secondary product from photochemical degradation of aromatic hydrocarbons [Suh et al., 2003] also showed high ambient concentrations during E1 and E2. This suggests that benzoic acid is derived from local emission sources or rapid atmospheric reactions associated with biomass burning, which can also emit their organic precursors in significant quantities.

[41] Temporal variations of oleic ($C_{18:1}$) and linoleic ($C_{18:2}$) acids showed four concentration peaks during 1–

2 June, E1, E2, and 25–26 June (Figure 6j). Their enhanced concentrations during E1 and E2 should be associated with biomass burning episodes. This is reasonable because these unsaturated fatty acids are abundant in plant leaves. Unsaturated fatty acids are also emitted to the atmosphere from many other sources such as meat charbroiling [Rogge et al., 1991] and automobile [Rogge et al., 1993a]. The temporal variations of total phthalate esters (Figure 5e) showed different pattern from other compound classes. Phthalates are emitted from plastics, and the emission rates are increased with an increase in ambient temperature.

[42] Throughout the samples studied, hopanes showed several concentration peaks during E2, 16–18 June, 22–23 June, and E3 (Figure 5j). As mentioned in section 3.2.3, hopanes are emitted to the atmosphere from internal combustion engines and the usage of coal; thus their peaks at E2 and E3 may suggest a contribution from automobile exhausts from urban regions. This may be possible because PAHs also showed their concentration maximum at E2, followed by E1 and several other peaks (Figure 5k). PAHs have been reported in biomass smoke in addition to motor exhausts; however, their emission from biomass burning is generally less significant compared to cellulose/hemicellulose or lignin decomposition products [Fine et al., 2001; Hays et al., 2005; Inuma et al., 2007; Oros et al., 2006; Oros and Simoneit, 2001a; Oros and Simoneit, 2001b; Rogge et al., 1998; Simoneit, 2002]. We consider that PAHs in the aerosols over Mt. Tai were mainly derived from coal combustion as discussed in section 3.2.4. A relatively weak but positive correlation was found between PAHs and levoglucosan ($r^2 = 0.30$), indicating that biomass burning has contributed, in part, to the PAHs in the tropospheric aerosols over Mt. Tai. Hays et al. [2005] reported that the enrichment of PAHs (≥ 178 amu) in wheat straw combustion-derived aerosols is considerable.

[43] Air mass back trajectory analyses (CGER/METEX) were conducted for the Mt. Tai aerosol samples from 29 May to 28 June 2006. Figure 7 shows six typical examples for the events of relative high concentrations (7 June during E1 and 13 June during E2) and low concentrations (1 June, 8 June, 19 June, and 27 June during E3) of levoglucosan. According to the 10-day back trajectory analysis, the source areas of the air masses were different during the field campaign. In the beginning of the campaign, air masses came mainly from mid-Siberia on the way to the East China

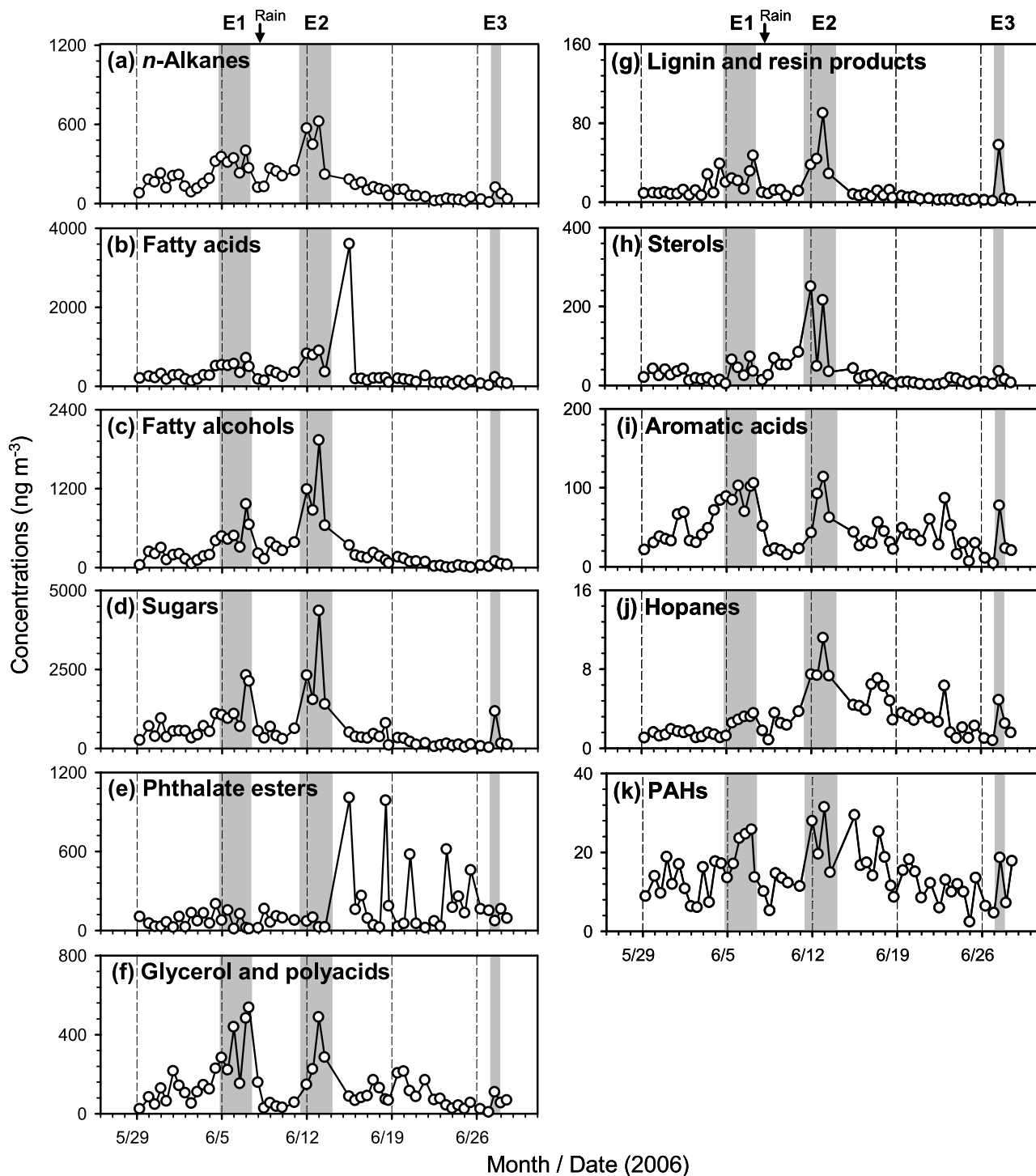


Figure 5. Temporal variations in the concentrations of lipid and various compound classes detected in the tropospheric aerosols over Mt. Tai, North China Plain (the daytime/nighttime data during 2–5 June, 23 June, and 25 June in Figure 5 and Figure 6 were obtained by the calculation of the unpublished data on 3-h samples).

Sea (Figure 7a). The heights of trajectories arriving at Mt. Tai were higher than 1500 m (Figure 7a). For E1 and E2, air masses mostly came from the northeast or north direction on the way to the south part of the North China Plain and then turned back to Mt. Tai (Figures 7b and 7d), when the agricultural waste burning peaked after wheat harvest in

the provinces of Anhui, Jiangsu, Shandong, Henan, and Hebei in the North China Plain as observed by the remote sensing meteorological satellite (<http://www.sepa.gov.cn/hjjc/stjc/index.htm>). Figures 7c and 7e showed that the air masses came straightly from the north when very low levels of organic compound classes were recorded. However,

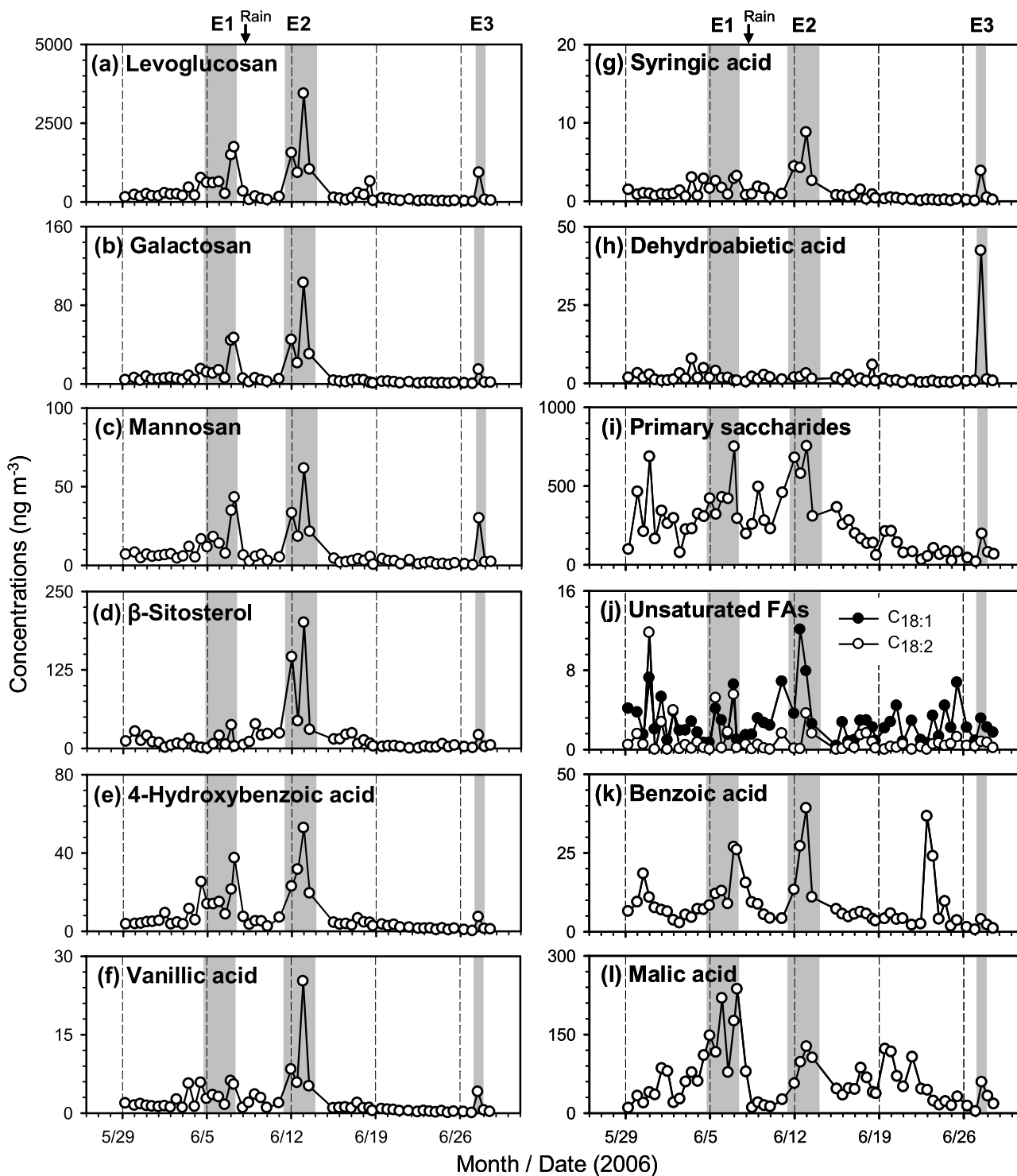


Figure 6. Temporal variations in the concentrations of biomass-burning tracers and other individual organic compounds detected in the tropospheric aerosols over Mt. Tai, North China Plain.

Figure 7f showed that during E3 most of the air masses came from the South China, when the harvesting areas were in the North China and the field burning of wheat straws was rather weak. This is consistent with our results that the biomass burning sources during E3 were different from those of E1 and E2.

3.4. Long-Range Atmospheric Transport of Higher Plant Waxes

[44] Long chain *n*-alkanes, fatty acids, and fatty alcohols were found to be more abundant in nighttime than in daytime as mentioned in section 3.1.1. This suggests that these biomarkers are abundantly present in the free troposphere over the North China Plain. Relative abundances

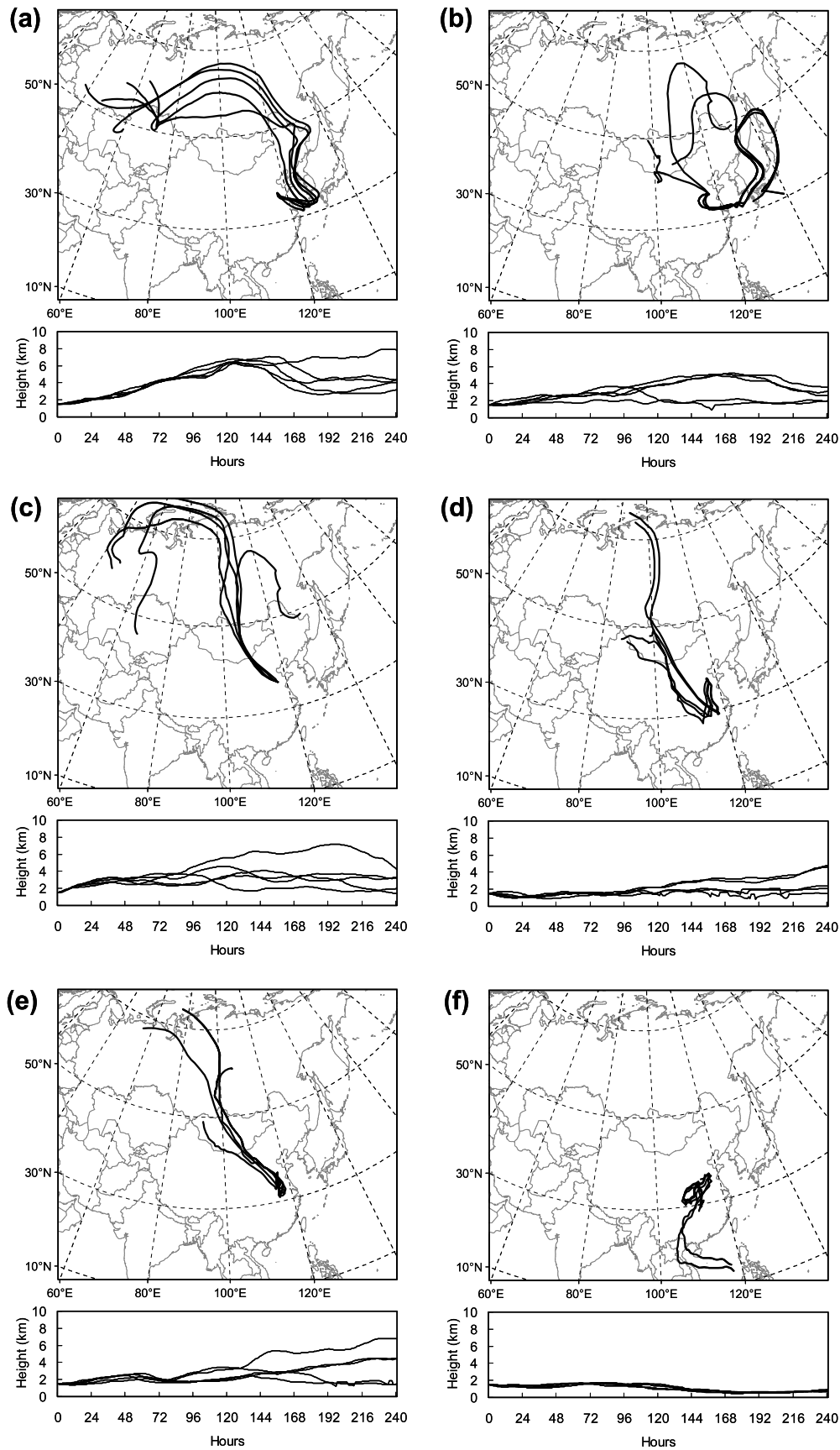


Figure 7. Ten-day air mass back trajectories arriving at 1500 m (above sea level) above Mt. Tai on: (a) 1 June, (b) 7 June, (c) 8 June, (d) 13 June, (e) 19 June, and (f) 27 June.

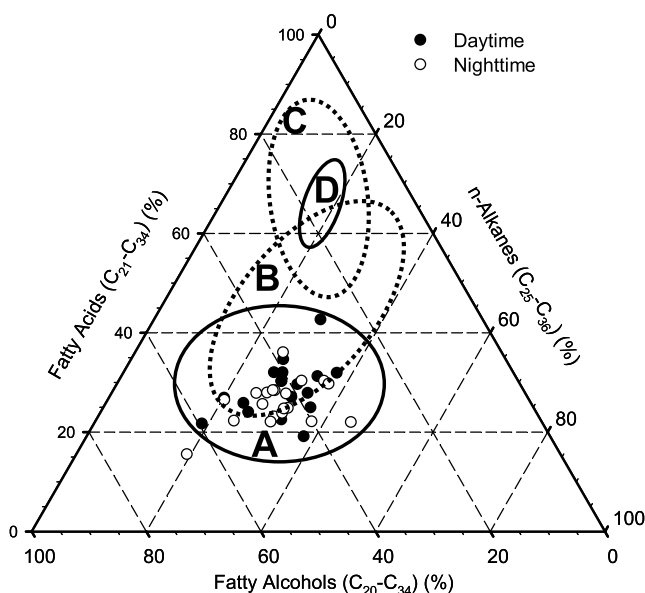


Figure 8. Triangular plots of terrestrial biomarkers detected in the Mt. Tai aerosols. The *n*-Alkanes (C_{25} – C_{36}), fatty alcohols (C_{20} – C_{34}), and fatty acids (C_{21} – C_{34}) in the Mt. Tai samples fall in category A (daytime is shown with filled circles and nighttime is shown with open circles). Marine aerosol samples collected from Chichi-Jima Island, the western North Pacific, fall in category B (during winter/spring seasons: November to June) and category C (during summer/autumn seasons: July to October) [Kawamura *et al.*, 2003]. Triangular plots of terrestrial biomarkers in the marine sediment samples collected from the central Pacific (15°N – 48°N , 175°E) fall in category D [Ohkouchi *et al.*, 1997].

(given in percent) of three terrestrial biomarker classes in the Mt. Tai aerosols are plotted in a triangular diagram (see Figure 8). The concentrations of higher plant-derived biomarkers (*n*-alkanes, fatty acids, and fatty alcohols) generally covaried in the Mt. Tai aerosol samples (strong linear correlations were found among three classes, $r^2 > 0.83$, $n = 39$), suggesting that the different compound classes were transported to the summit of Mt. Tai from similar sources. It is interesting to note that both daytime and nighttime samples are generally plotted in the lower part of the triangle, falling in category A (Figure 8).

[45] Kawamura *et al.* [2003] analyzed terrestrial biomarkers (*n*-alkanes, fatty acids, and fatty alcohols) in the marine aerosol samples collected in 1990–1993 from Chichi-Jima Island in the western North Pacific (For the location, see Figure 1). The *n*-alkanes were reported in a range of C_{20} – C_{43} with a C_{max} at C_{29} or C_{31} . The CPI ratios ranged from 1.8 to 14.6 (average 4.5). Their data were plotted as relative abundances in the same diagram. Those samples were grouped in category B and C, where samples were collected under the meteorological conditions of westerly and trade wind regimes, respectively. Asian continent is considered as a major source region for the terrestrial organic matter transported over the western North Pacific in winter to spring [Kawamura *et al.*, 2003].

[46] The present results of Mt. Tai show that the group A is much closer to B rather than C (Figure 8), suggesting that

the Asian aerosols are one of the important sources of terrestrial biomarkers detected in the western North Pacific. Moreover, Ohkouchi *et al.* [1997] analyzed terrestrial biomarkers (long-chain *n*-alkanes, *n*-alcohols, and *n*-fatty acids) in the pelagic sediments collected at 175°E transect (15°S – 48°N) in the Pacific, whose relative abundances are also plotted in the same triangular figure. As shown in Figure 8, data from sediment samples collected in the middle to high latitudes (15°N – 48°N) are plotted in the upper part of the triangle as category D, which is included in category C and overlapped with category B, the latter means westerly regime.

[47] The plots in Figure 8 indicate that long-chain fatty acids in Mt. Tai aerosols are less abundant than *n*-alkanes and fatty alcohols compared to the marine aerosols from Chichi-Jima Island (categories B and C). This may suggest that lipid compositions of biomass burning products in the North China Plain are different from those of leaf waxes in the terrestrial plants. Long-chain fatty acids are important

Table 5. Concentrations of Various Compound Classes and Groups^a

Compound Groups	Daytime	Nighttime
<i>Plant Wax</i>		
<i>n</i> -Alkanes ^b	104	131
HMW fatty acids ^c	157	173
Fatty alcohols	249	349
Subtotal	510	656
<i>Marine/Microbial Source^d</i>		
LMW fatty acids ^c	106	114
Ergosterol	1.73	4.37
Subtotal	108	118
<i>Biomass Burning</i>		
Levoglucosan	391	459
Galactosan	8.43	12.8
Mannosan	9.71	9.97
Lignin and resin products	16.3	13.6
β -Sitosterol	11.2	29.5
Subtotal	437	525
<i>Fossil Fuel (Coal + Petroleum) Combustion</i>		
<i>n</i> -Alkanes ^b	57	63
Hopanes	3.41	3.63
PAHs	14.6	15.2
Subtotal	74.7	81.8
<i>Soil Resuspension</i>		
Primary saccharides	231	317
Glycerol	19.9	34.4
Subtotal	251	351
<i>Plastics</i>		
Phthalate esters	170	62
<i>Secondary Oxidation</i>		
Aromatic acids	50	45
Polyacids	120	119
Subtotal	170	164
Total	1720	1958

^aConcentration is measured in ng m^{-3} .

^bPlant wax *n*-alkanes are calculated as the excess odd homologous-adjacent even homologous average, and the difference from the total *n*-alkanes is the petroleum-derived amount [Simoneit *et al.*, 1991].

^cHigh molecular weight fatty acids ($C_{20:0}$ – $C_{34:0}$); low molecular weight fatty acids ($C_{10:0}$ – $C_{19:0}$), including $C_{18:1}$ and $C_{18:2}$.

^dHoffmann and Warnke [2007], Simoneit *et al.* [2004c], Wang and Kawamura [2005].

Table 6. Source Strengths of Organic Matter in the Tropospheric Aerosols Over Mt. Tai

Source	Source Strengths (%)	
	Daytime	Nighttime
Plant emission	30	34
Marine/microbial source	6	6
Biomass burning	25	27
Fossil fuel combustion	4	4
Soil resuspension	15	18
Plastics	10	3
Secondary oxidation	10	8

components of leaf waxes, but may not be so abundant in the cellulose-oriented wheat straws and ears of wheat. This may be the reason why the Mt. Tai aerosols, affected by field burning of wheat straws, are depleted with long-chain fatty acids compared to *n*-alkanes and fatty alcohols (Figure 8). Although there are some differences in the relative abundances of these compound classes, the comparisons of terrestrial biomarkers in the aerosols from Mt. Tai, Chichi-Jima Island in the western North Pacific, and deep-sea sediments from the central Pacific indicate that terrestrial biomarkers can be transported long distances from Asian continent over the Pacific Ocean under the control of westerly wind. This study indicates the importance of Asian mountain aerosols as a source of organic matter in the remote marine atmosphere and deep-sea sediment, further signifying a detailed analysis and study.

3.5. Source Apportionment of Organic Matter in Aerosols Over Mt. Tai

[48] On the basis of the above discussion and other studies on the extensive emission profiles of organic compounds and their compositions [Alves *et al.*, 2007; Schauer *et al.*, 1996; Simoneit, 2002; Simoneit *et al.*, 2004a, 2004c; Wang and Kawamura, 2005], organic compound classes quantified in Mt. Tai aerosols can roughly be apportioned to six emission sources and one secondary oxidation products as follows (Table 5): (1) plant emission characterized by higher plant wax *n*-alkanes, HMW fatty alcohols, and fatty acids; (2) marine or microbial lipids characterized mainly by LMW fatty acids (<C₂₀, including unsaturated fatty acids C_{18:1} and C_{18:2}) [Simoneit *et al.*, 2004c; Wang and Kawamura, 2005]; (3) biomass burning characterized by levoglucosan, β -sitosterol, lignin, and resin products; (4) fossil fuel combustion characterized by petroleum-derived *n*-alkanes, hopanes, and PAHs; (5) soil resuspension characterized by primary saccharides and glycerol; (6) plasticizers (phthalate esters); and (7) secondary oxidation products characterized by aromatic acids and polyacids.

[49] On the average concentrations of the compound classes, the source strengths in terms of their organic tracers are apportioned as shown in Table 6. It should be noted that low molecular weight dicarboxylic acids (such as oxalic acid) that are also of photochemical origin and abundant in atmospheric aerosols [Kawamura and Ikushima, 1993] were not measured in this study. It should also be noted that bioaerosols (e.g., pollen) might contribute to the sugars in daytime as mentioned in section 3.1.2. As seen from Table 6, the most abundant compound group in summer was higher plant waxes (average 30% in daytime and 34% in nighttime), followed by biomass burning emissions (25% in daytime and 27% in nighttime)

and soil resuspension (15% in daytime and 18% in nighttime). There was no significant difference in the source strength of secondary oxidation products between daytime (10%) and nighttime (8%). During nighttime, secondary oxidation products can also be transported over Mt. Tai (above the PBL). Phthalate esters accounted for 10% in daytime and only 3% in nighttime. This suggests that the ambient temperature and height of PBL seriously influence the atmospheric concentrations of plasticizer-derived phthalate esters.

4. Summary and Conclusions

[50] During the summer campaign at Mt. Tai in the North China Plain, two major biomass-burning episodes were identified based on the organic chemical analyses of mountain aerosols that were often collected in the free troposphere. Height of PBL is an important factor to control the concentration of organic aerosols over Mt. Tai. Furthermore, this study demonstrated that the tropospheric aerosols over Mt. Tai are contributed from various sources including emissions of terrestrial plant waxes, biomass burning, soil resuspension, coal and petroleum combustion, as well as significant secondary oxidation products in the atmosphere.

[51] Most of the organic compounds identified in Mt. Tai aerosols are less abundant than those reported from ground surface sites in Chinese urban areas but much more abundant than those found in the East China Sea or northwestern Pacific. Under the condition of westerly winds, the continent-derived organic compounds detected over Mt. Tai can be transported further to the western North Pacific through atmospheric long-range transport, affecting the organic composition of aerosol particles in the remote marine atmosphere. Polyacids were observed as a major compound class in the troposphere over Mt. Tai, being different from those in ground surface sites in urban area where polyacids were minor. Such a trend was also observed in aircraft samples over China [Wang *et al.*, 2007]. This study indicates an enhanced photochemical production of secondary organic aerosols in the lower troposphere over mountaintop in the North China Plain, although small dicarboxylic acids such as oxalic acid were not studied here. Our results provided useful information to better understand the air qualities of Northeast China and the downstream sites with the Asian continental outflow.

[52] **Acknowledgments.** This study is supported by the Japanese Ministry of Education, Culture, Sports, Science and Technology (MEXT) through grants-in-aid 17340166 and 19204055. P.F. appreciates MEXT for the Monbukagakusho Scholarship. We also acknowledge the financial support by the Global Environment Research Fund (B-051) of the Ministry of the Environment, Japan for the shipping of the instruments to Mt. Tai.

References

- Alves, C., T. Oliveira, C. Pio, A. J. D. Silvestre, P. Fialho, F. Barata, and M. Legrand (2007), Characterisation of carbonaceous aerosols from the Azorean Island of Terceira, *Atmos. Environ.*, *41*(7), 1359–1373, doi:10.1016/j.atmosenv.2006.10.022.
- Andreae, M. O., and P. J. Crutzen (1997), Atmospheric aerosols: Biogeochemical sources and role in atmospheric chemistry, *Science*, *276*, 1052–1058, doi:10.1126/science.276.5315.1052.
- Bielecki, R. L. (1995), Onset of phloem export from senescent petals of daylily, *Plant Physiol.*, *109*, 557–565.
- Charlson, R. J., S. E. Schwartz, J. M. Hales, R. D. Cess, J. A. Coakley, J. E. Hansen, and D. J. Hofmann (1992), Climate forcing by anthropogenic aerosols, *Science*, *255*(5043), 423–430, doi:10.1126/science.255.5043.423.

- Chowdhury, Z., M. Zheng, J. J. Schauer, R. J. Sheesley, L. Salmon, G. R. Cass, and A. Russell (2007), Speciation of ambient fine organic carbon particles and source apportionment of PM_{2.5} in Indian cities, *J. Geophys. Res.*, *112*, D15303, doi:10.1029/2007JD008386.
- Claeys, M., et al. (2004), Formation of secondary organic aerosols through photooxidation of isoprene, *Science*, *303*(5661), 1173–1176, doi:10.1126/science.1092805.
- Cooke, W. F., C. Liousse, H. Cachier, and J. Feichter (1999), Construction of a 1 degrees × 1 degrees fossil fuel emission data set for carbonaceous aerosol and implementation and radiative impact in the ECHAM4 model, *J. Geophys. Res.*, *104*(D18), 22,137–22,162, doi:10.1029/1999JD900187.
- Elbert, W., P. E. Taylor, M. O. Andreae, and U. Pöschl (2007), Contribution of fungi to primary biogenic aerosols in the atmosphere: Wet and dry discharged spores, carbohydrates, and inorganic ions, *Atmos. Chem. Phys.*, *7*, 4569–4588.
- Feng, J., Z. Guo, C. K. Chan, and M. Fang (2007), Properties of organic matter in PM_{2.5} at Changdao Island, China-A rural site in the transport path of the Asian continental outflow, *Atmos. Environ.*, *41*(9), 1924–1935, doi:10.1016/j.atmosenv.2006.10.064.
- Fernandes, S. D., N. M. Trautmann, D. G. Streets, C. A. Roden, and G. C. Bond (2007), Global biofuel use, 1980–2000, *Global Biogeochem. Cycles*, *21*, GB2019, doi:10.1029/2006GB002836.
- Fine, P. M., G. R. Cass, and B. R. T. Simoneit (2001), Chemical characterization of fine particle emissions from fireplace combustion of woods grown in the northeastern United States, *Environ. Sci. Technol.*, *35*(13), 2665–2675, doi:10.1021/es001466k.
- Fine, P. M., B. Chakrabarti, M. Krudysz, J. J. Schauer, and C. Sioutas (2004), Diurnal variations of individual organic compound constituents of ultrafine and accumulation mode particulate matter in the Los Angeles basin, *Environ. Sci. Technol.*, *38*(5), 1296–1304, doi:10.1021/es0348389.
- Gagosian, R. B., and E. T. Peltzer (1986), The importance of atmospheric input of terrestrial organic material to deep sea sediments, *Org. Geochem.*, *10*, 661–669, doi:10.1016/S0146-6380(86)80002-X.
- Gagosian, R. B., E. T. Peltzer, and O. C. Zafrioui (1981), Atmospheric transport of continentally derived lipids to the tropical North Pacific, *Nature*, *291*, 312–314, doi:10.1038/291312a0.
- Gao, Y., and J. R. Anderson (2001), Characteristics of Chinese aerosols determined by individual-particle analysis, *J. Geophys. Res.*, *106*(D16), 18,037–18,045, doi:10.1029/2000JD900725.
- Graham, B., O. L. Mayol-Bracero, P. Guyon, G. C. Robert, S. Decesari, M. C. Facchini, P. Artaxo, W. Maenhaut, P. Köll, and M. O. Andreae (2002), Water-soluble organic compounds in biomass burning aerosols over Amazonia: 1. Characterization by NMR and GC-MS, *J. Geophys. Res.*, *107*(D20), 8047, doi:10.1029/2001JD000336.
- Graham, B., P. Guyon, P. E. Taylor, P. Artaxo, W. Maenhaut, M. M. Glovsky, R. C. Flagan, and M. O. Andreae (2003), Organic compounds present in the natural Amazonian aerosol: Characterization by gas chromatography-mass spectrometry, *J. Geophys. Res.*, *108*(D24), 4766, doi:10.1029/2003JD003990.
- Hays, M. D., P. M. Fine, C. D. Geron, M. J. Kleeman, and B. K. Gullett (2005), Open burning of agricultural biomass: Physical and chemical properties of particle-phase emissions, *Atmos. Environ.*, *39*(36), 6747–6764, doi:10.1016/j.atmosenv.2005.07.072.
- Heald, C. L., D. J. Jacob, R. J. Park, B. Alexander, T. D. Fairlie, R. M. Yantosca, and D. A. Chu (2006), Transpacific transport of Asian anthropogenic aerosols and its impact on surface air quality in the United States, *J. Geophys. Res.*, *111*, D14310, doi:10.1029/2005JD006847.
- Hemming, B. L., and J. H. Seinfeld (2001), On the hygroscopic behavior of atmospheric organic aerosols, *Ind. Eng. Chem. Res.*, *40*(20), 4162–4171, doi:10.1021/ie000790l.
- Hoffer, A., A. Gelencsér, M. Blazsó, P. Guyon, P. Artaxo, and M. O. Andreae (2006), Diel and seasonal variations in the chemical composition of biomass burning aerosol, *Atmos. Chem. Phys.*, *6*, 3505–3515.
- Hoffmann, T., and J. Warnke (2007), Organic aerosols, in *Volatile Organic Compounds in the Atmosphere*, edited by R. Koppmann, pp. 342–387, Blackwell, Oxford, U.K.
- Hopke, P. K. (1985), *Receptor Modeling in Environmental Chemistry*, John Wiley, New York.
- Hsu, C.-L., C.-Y. Cheng, C.-T. Lee, and W.-H. Ding (2007), Derivatization procedures and determination of levoglucosan and related monosaccharide anhydrides in atmospheric aerosols by gas chromatography-mass spectrometry, *Talanta*, *72*, 199–205, doi:10.1016/j.talanta.2006.10.018.
- Iinuma, Y., E. Brüggemann, T. Gnauk, K. Müller, M. O. Andreae, G. Helas, R. Parmar, and H. Herrmann (2007), Source characterization of biomass burning particles: The combustion of selected European conifer, African hardwood, savanna grass, and German and Indonesian peat, *J. Geophys. Res.*, *112*, D08209, doi:10.1029/2006JD007120.
- Jacobson, M. Z. (2001), Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols, *Nature*, *409*(6821), 695–697, doi:10.1038/35055518.
- Jacobson, M. C., H. C. Hansson, K. J. Noone, and R. J. Charlson (2000), Organic atmospheric aerosols: Review and state of science, *Rev. Geophys.*, *38*(2), 267–294, doi:10.1029/1998RG000045.
- Jaenicke, R. (2005), Abundance of cellular material and proteins in the atmosphere, *Science*, *308*, 73, doi:10.1126/science.1106335.
- Kanakidou, M., et al. (2005), Organic aerosol and global climate modelling: A review, *Atmos. Chem. Phys.*, *5*, 1053–1123.
- Kaneyasu, N., and H. Takada (2004), Seasonal variations of sulfate, carbonaceous species (black carbon and polycyclic aromatic hydrocarbons), and trace elements in fine atmospheric aerosols collected at subtropical islands in the East China Sea, *J. Geophys. Res.*, *109*, D06211, doi:10.1029/2003JD004137.
- Kaufman, Y. J., D. Tanré, and O. Boucher (2002), A satellite view of aerosols in the climate system, *Nature*, *419*, 215–223, doi:10.1038/nature01091.
- Kawamura, K., and R. B. Gagosian (1987), Implications of ω -oxocarboxylic acids in the remote marine atmosphere for photo-oxidation of unsaturated fatty acids, *Nature*, *325*, 330–332, doi:10.1038/325330a0.
- Kawamura, K., and R. B. Gagosian (1990), Atmospheric transport of soil-derived dicarboxylic acids over the North Pacific Ocean, *Naturwissenschaften*, *77*, 25–27, doi:10.1007/BF01131790.
- Kawamura, K., and K. Ikushima (1993), Seasonal changes in the distribution of dicarboxylic acids in the urban atmosphere, *Environ. Sci. Technol.*, *27*(10), 2227–2235, doi:10.1021/es00047a033.
- Kawamura, K., and F. Sakaguchi (1999), Molecular distributions of water soluble dicarboxylic acids in marine aerosols over the Pacific Ocean including tropics, *J. Geophys. Res.*, *104*(D3), 3501–3509, doi:10.1029/1998JD100041.
- Kawamura, K., L.-L. Ng, and I. R. Kaplan (1985), Determination of organic acids (C₁-C₁₀) in the atmosphere, motor exhausts, and engine oils, *Environ. Sci. Technol.*, *19*(11), 1082–1086, doi:10.1021/es00141a010.
- Kawamura, K., M. Kosaka, and R. Sempéré (1995), Distributions and seasonal changes of hydrocarbons in urban aerosols and rainwaters, *Geochemistry*, *29*, 1–15.
- Kawamura, K., S. Steinberg, and I. R. Kaplan (2000), Homologous series of C₁-C₁₀ monocarboxylic acids and C₁-C₆ carbonyls in Los Angeles air and motor vehicle exhausts, *Atmos. Environ.*, *34*(24), 4175–4191, doi:10.1016/S1352-2310(00)00212-0.
- Kawamura, K., Y. Ishimura, and K. Yamazaki (2003), Four years' observations of terrestrial lipid class compounds in marine aerosols from the western North Pacific, *Global Biogeochem. Cycles*, *17*(1), 1003, doi:10.1029/2001GB001810.
- Kolattukudy, P. E. (1976), *Chemistry and Biochemistry of Natural Waxes*, Elsevier, New York.
- Larsen, R. K., M. M. Schantz, and S. A. Wise (2006), Determination of levoglucosan in particulate matter reference material, *Aerosol Sci. Technol.*, *40*, 781–787, doi:10.1080/02786820600596909.
- Lau, A. P. S., A. K. Y. Lee, C. K. Chan, and M. Fang (2006), Ergosterol as a biomarker for the quantification of the fungal biomass in atmospheric aerosols, *Atmos. Environ.*, *40*(2), 249–259, doi:10.1016/j.atmosenv.2005.09.048.
- Li, X., S. Wang, L. Duan, J. Hao, C. Li, Y. Chen, and L. Yang (2007), Particulate and trace gas emissions from open burning of wheat straw and corn stover in China, *Environ. Sci. Technol.*, *41*(17), 6052–6058, doi:10.1021/es0705137.
- Medeiros, P. M., M. H. Conte, J. C. Weber, and B. R. T. Simoneit (2006), Sugars as source indicators of biogenic organic carbon in aerosols collected above the Howland Experimental Forest, Maine, *Atmos. Environ.*, *40*(9), 1694–1705, doi:10.1016/j.atmosenv.2005.11.001.
- Mochida, M., and K. Kawamura (2004), Hygroscopic properties of levoglucosan and related organic compounds characteristic to biomass burning aerosol particles, *J. Geophys. Res.*, *109*, D21202, doi:10.1029/2004JD004962.
- Mochida, M., Y. Kitamori, K. Kawamura, Y. Nojiri, and K. Suzuki (2002), Fatty acids in the marine atmosphere: Factors governing their concentrations and evaluation of organic films on sea-salt particles, *J. Geophys. Res.*, *107*(D17), 4325, doi:10.1029/2001JD001278.
- Mochida, M., A. Kawabata, K. Kawamura, H. Hatsushika, and K. Yamazaki (2003), Seasonal variation and origins of dicarboxylic acids in the marine atmosphere over the western North Pacific, *J. Geophys. Res.*, *108*(D6), 4193, doi:10.1029/2002JD002355.
- Nolte, C. G., J. J. Schauer, G. R. Cass, and B. R. T. Simoneit (2002), Trimethylsilyl derivatives of organic compounds in source samples and in atmospheric fine particulate matter, *Environ. Sci. Technol.*, *36*(20), 4273–4281, doi:10.1021/es020518y.
- Ohkouchi, N., K. Kawamura, H. Kawahata, and A. Taira (1997), Latitudinal distributions of terrestrial biomarkers in the sediments from the Central Pacific, *Geochim. Cosmochim. Acta*, *61*(9), 1911–1918, doi:10.1016/S0016-7037(97)00040-9.
- Oliveira, C., et al. (2007), Seasonal distribution of polar organic compounds in the urban atmosphere of two large cities from the North and South of

- Europe, *Atmos. Environ.*, *41*(27), 5555–5570, doi:10.1016/j.atmosenv.2007.03.001.
- Oros, D. R., and B. R. T. Simoneit (2001a), Identification and emission factors of molecular tracers in organic aerosols from biomass burning Part 1. Temperate climate conifers, *Appl. Geochem.*, *16*, 1513–1544, doi:10.1016/S0883-2927(01)00021-X.
- Oros, D. R., and B. R. T. Simoneit (2001b), Identification and emission factors of molecular tracers in organic aerosols from biomass burning Part 2. Deciduous trees, *Appl. Geochem.*, *16*, 1545–1565, doi:10.1016/S0883-2927(01)00022-1.
- Oros, D. R., M. R. B. Abas, N. Y. M. J. Omar, N. A. Rahman, and B. R. T. Simoneit (2006), Identification and emission factors of molecular tracers in organic aerosols from biomass burning: Part 3. Grasses, *Appl. Geochem.*, *21*, 919–940, doi:10.1016/j.apgeochem.2006.01.008.
- Pashynska, V., R. Vermeylen, G. Vas, W. Maenhaut, and M. Claeys (2002), Development of a gas chromatographic/ion trap mass spectrometric method for the determination of levoglucosan and saccharidic compounds in atmospheric aerosols, Application to urban aerosol, *J. Mass Spectrom.*, *37*, 1249–1257, doi:10.1002/jms.391.
- Pöschl, U. (2005), Atmospheric aerosols: Composition, transformation, climate and health effects, *Angew. Chem. Int. Ed.*, *44*, 7520–7540, doi:10.1002/anie.200501122.
- Ramanathan, V., P. J. Crutzen, J. T. Kiehl, and D. Rosenfeld (2001), Aerosols, climate, and the hydrological cycle, *Science*, *294*, 2119–2124, doi:10.1126/science.1064034.
- Robinson, A. L., R. Subramanian, N. M. Donahue, A. Bernardo-Bricker, and W. F. Rogge (2006), Source apportionment of molecular markers and organic aerosol. 2. Biomass smoke, *Environ. Sci. Technol.*, *40*(24), 7811–7819, doi:10.1021/es060782h.
- Rogge, W. F., L. M. Hildemann, M. A. Mazurek, G. R. Cass, and B. R. T. Simoneit (1991), Source of fine organic aerosols: 1. Charbroilers and meat cooking operations, *Environ. Sci. Technol.*, *25*(6), 1112–1125, doi:10.1021/es00018a015.
- Rogge, W. F., L. M. Hildemann, M. A. Mazurek, G. R. Cass, and B. R. T. Simoneit (1993a), Sources of fine organic aerosol. 2. Noncatalyst and catalyst-equipped automobiles and heavy-duty diesel trucks, *Environ. Sci. Technol.*, *27*(4), 636–651, doi:10.1021/es00041a007.
- Rogge, W. F., L. M. Hildemann, M. A. Mazurek, G. R. Cass, and B. R. T. Simoneit (1993b), Sources of fine organic aerosol. 4. Particulate abrasion products from leaf surfaces of urban plants, *Environ. Sci. Technol.*, *27*(13), 2700–2711, doi:10.1021/es00049a008.
- Rogge, W. F., L. M. Hildemann, M. A. Mazurek, G. R. Cass, and B. R. T. Simoneit (1998), Sources of fine organic aerosol. 9. Pine, oak and synthetic log combustion in residential fireplaces, *Environ. Sci. Technol.*, *32*(1), 13–22, doi:10.1021/es960930b.
- Rudich, Y. (2003), Laboratory perspectives on the chemical transformations of organic matter in atmospheric particles, *Chem. Rev.*, *103*, 5097–5124, doi:10.1021/cr020508f.
- Schauer, J. J., W. F. Rogge, L. M. Hildemann, M. A. Mazurek, and G. R. Cass (1996), Source apportionment of airborne particulate matter using organic compounds as tracers, *Atmos. Environ.*, *30*(22), 3837–3855, doi:10.1016/1352-2310(96)00085-4.
- Schauer, J. J., M. J. Kleeman, G. R. Cass, and B. R. T. Simoneit (1999), Measurement of emissions from air pollution sources. 1. C₁ through C₂₉ organic compounds from meat charbroiling, *Environ. Sci. Technol.*, *33*(10), 1566–1577, doi:10.1021/es980076j.
- Schmidl, C., I. L. Marr, A. Caseiro, P. Kotianová, A. Berner, H. Bauer, A. Kasper-Giebl, and H. Puxbaum (2008), Chemical characterisation of fine particle emissions from wood stove combustion of common woods growing in mid-European Alpine regions, *Atmos. Environ.*, *42*(1), 126–141, doi:10.1016/j.atmosenv.2007.09.028.
- Seinfeld, J. H., and S. N. Pandis (1998), *Atmospheric Chemistry and Physics*, John Wiley, New York.
- Sheesley, R. J., J. J. Schauer, Z. Chowdhury, G. R. Cass, and B. R. T. Simoneit (2003), Characterization of organic aerosols emitted from the combustion of biomass indigenous to South Asia, *J. Geophys. Res.*, *108*(D9), 4285, doi:10.1029/2002JD002981.
- Sidhu, S., B. Gullett, R. Striebich, J. Klosterman, J. Contreras, and M. DeVito (2005), Endocrine disrupting chemical emissions from combustion sources: Diesel particulate emissions and domestic waste open burn emissions, *Atmos. Environ.*, *39*(5), 801–811, doi:10.1016/j.atmosenv.2004.10.040.
- Simoneit, B. R. T. (2002), Biomass burning—a review of organic tracers for smoke from incomplete combustion, *Appl. Geochem.*, *17*, 129–162, doi:10.1016/S0883-2927(01)00061-0.
- Simoneit, B. R. T., and V. O. Elias (2000), Organic tracers from biomass burning in atmospheric particulate matter over the ocean, *Mar. Chem.*, *69*(3–4), 301–312, doi:10.1016/S0304-4203(00)00008-6.
- Simoneit, B. R. T., and M. A. Mazurek (1982), Organic matter of the troposphere-II. Natural background of biogenic lipid matter in aerosols over the rural western United States, *Atmos. Environ.*, *16*(9), 2139–2159, doi:10.1016/0004-6981(82)90284-0.
- Simoneit, B. R. T., G. Y. Sheng, X. J. Chen, J. M. Fu, J. Zhang, and Y. P. Xu (1991), Molecular marker study of extractable organic-matter in aerosols from urban areas of China, *Atmos. Environ. Part A*, *25*(10), 2111–2129, doi:10.1016/0960-1686(91)90088-O.
- Simoneit, B. R. T., W. F. Rogge, M. A. Mazurek, L. J. Standley, L. M. Hildemann, and G. R. Cass (1993), Lignin pyrolysis products, lignans, and resin acids as specific tracers of plant classes in emissions from biomass combustion, *Environ. Sci. Technol.*, *27*(12), 2533–2541, doi:10.1021/es00048a034.
- Simoneit, B. R. T., J. J. Schauer, C. G. Nolte, D. R. Oros, V. O. Elias, M. P. Fraser, W. F. Rogge, and G. R. Cass (1999), Levoglucosan, a tracer for cellulose in biomass burning and atmospheric particles, *Atmos. Environ.*, *33*(2), 173–182, doi:10.1016/S1352-2310(98)00145-9.
- Simoneit, B. R. T., V. O. Elias, M. Kobayashi, K. Kawamura, A. I. Rushdi, P. M. Medeiros, W. F. Rogge, and B. M. Didyk (2004a), Sugars-dominant water-soluble organic compounds in soils and characterization as tracers in atmospheric particulate matter, *Environ. Sci. Technol.*, *38*(22), 5939–5949, doi:10.1021/es0403099.
- Simoneit, B. R. T., M. Kobayashi, M. Mochida, K. Kawamura, and B. J. Huebert (2004b), Aerosol particles collected on aircraft flights over the northwestern Pacific region during the ACE-Asia campaign: Composition and major sources of the organic compounds, *J. Geophys. Res.*, *109*, D19S09, doi:10.1029/2004JD004565.
- Simoneit, B. R. T., M. Kobayashi, M. Mochida, K. Kawamura, M. Lee, H. J. Lim, B. J. Turpin, and Y. Komazaki (2004c), Composition and major sources of organic compounds of aerosol particulate matter sampled during the ACE-Asia campaign, *J. Geophys. Res.*, *109*, D19S10, doi:10.1029/2004JD004598.
- Streets, D. G., K. F. Yarber, J. H. Woo, and G. R. Carmichael (2003), Biomass burning in Asia: Annual and seasonal estimates and atmospheric emissions, *Global Biogeochem. Cycles*, *17*(4), 1099, doi:10.1029/2003GB002040.
- Streets, D. G., T. C. Bond, T. Lee, and C. Jang (2004), On the future of carbonaceous aerosol emissions, *J. Geophys. Res.*, *109*, D24212, doi:10.1029/2004JD004902.
- Suh, I., R. Zhang, L. T. Molina, and M. J. Molina (2003), Oxidation mechanism of aromatic peroxy and bicyclic radicals from OH-toluene reactions, *J. Am. Chem. Soc.*, *125*, 12,655–12,665, doi:10.1021/ja0350280.
- Wang, G., and K. Kawamura (2005), Molecular characteristics of urban organic aerosols from Nanjing: A case study of a mega-city in China, *Environ. Sci. Technol.*, *39*(19), 7430–7438, doi:10.1021/es051055+.
- Wang, G., K. Kawamura, S. Lee, K. F. Ho, and J. J. Cao (2006a), Molecular, seasonal, and spatial distributions of organic aerosols from fourteen Chinese cities, *Environ. Sci. Technol.*, *40*(15), 4619–4625, doi:10.1021/es060291x.
- Wang, G., K. Kawamura, S. Hatakeyama, A. Takami, H. Li, and W. Wang (2007), Aircraft measurement of organic aerosols over China, *Environ. Sci. Technol.*, *41*(9), 3115–3120, doi:10.1021/es062601h.
- Wang, H. B., K. Kawamura, and D. Shooter (2006b), Wintertime organic aerosols in Christchurch and Auckland, New Zealand: Contributions of residential wood and coal burning and petroleum utilization, *Environ. Sci. Technol.*, *40*(17), 5257–5262, doi:10.1021/es052523i.
- Wilkening, K. E., L. A. Barrie, and M. Engle (2000), Trans-Pacific air pollution, *Science*, *290*, 65–66, doi:10.1126/science.290.5489.65.
- Wolff, G. T., and P. E. Korsog (1985), Estimates of the contributions of sources to inhalable particulate concentrations in Detroit, *Atmos. Environ.*, *19*(9), 1399–1409, doi:10.1016/0004-6981(85)90277-X.
- Yttri, K. E., C. Dye, and G. Kiss (2007), Ambient aerosol concentrations of sugars and sugar-alcohols at four different sites in Norway, *Atmos. Chem. Phys.*, *7*, 4267–4279.
- Zdrahal, Z., J. Oliveira, R. Vermeylen, M. Claeys, and W. Maenhaut (2002), Improved method for quantifying levoglucosan and related monosaccharide anhydrides in atmospheric aerosols and application to samples from urban and tropical locations, *Environ. Sci. Technol.*, *36*(4), 747–753, doi:10.1021/es015619v.
- Zheng, M., L. G. Salmon, J. J. Schauer, L. Zeng, C. S. Kiang, Y. Zhang, and G. R. Cass (2005), Seasonal trends in PM_{2.5} source contributions in Beijing, China, *Atmos. Environ.*, *39*(22), 3967–3976, doi:10.1016/j.atmosenv.2005.03.036.

S. G. Aggarwal, P. Fu, K. Kawamura, K. Okuzawa, and G. Wang, Institute of Low Temperature Science, Hokkaido University, Sapporo 060-0819, Japan. (kawamura@lowtem.hokudai.ac.jp)

Y. Kanaya, Frontier Research Center for Global Change, Japan Agency for Marine-Earth Science and Technology, Kanagawa 236-0001, Japan.

Z. Wang, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China.