

Aerosol particles collected on aircraft flights over the northwestern Pacific region during the ACE-Asia campaign: Composition and major sources of the organic compounds

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[1] Atmospheric particulate matter, collected over the polluted east Asia/Pacific region in spring 2001 during research flights with the National Center for Atmospheric Research (NCAR) C-130 aircraft, was analyzed for different types of organic compounds using capillary gas chromatography–mass spectrometry. More than 70 organic species were detected in the aerosols and grouped into different compound classes on the basis of functional groups, including *n*-alkanes, polycyclic aromatic hydrocarbons, fatty acids, dehydroabietic acid, alkanols, water-soluble sugars (including glucose, sucrose, mycose, and levoglucosan), monocarboxylic and dicarboxylic acids, urea, and phthalates. Interestingly, the water-soluble compounds ($72\text{--}133\text{ ng m}^{-3}$) were found to account for 16–50% (average 34%) of the total identified compound mass (TCM). Organic compounds were further categorized into several groups to suggest their sources. Fossil fuel combustion was recognized as the most significant source for the TCM (contributing 33–80% of TCM, average 50%), followed by soil resuspension (5–25%, average 19%) and secondary oxidation products (4–15%, average 9%). In contrast, the contribution of natural sources such as terrestrial plant wax and marine lipids (fatty acids and alkanols) was relatively small (3.4% and 9.4% on average, respectively). Biomass burning was suggested to contribute only a minor portion to the TCM of the Asian aerosols during the spring season (1.4% on average based on levoglucosan). However, levoglucosan may have been hydrolyzed and/or oxidized in part during long-range transport, and therefore this value represents a lower limit. The organic compound compositions of these samples are very different from those reported for aerosol particles of the Atlantic Ocean and from the earlier data for the mid-Pacific in terms of the abundant presence of water-soluble compounds consisting of saccharides, anhydrosaccharides, and the secondary dicarboxylic acids. This study demonstrated that the organic tracer approach can be carried out on small samples acquired on aircraft and is useful to better understand the sources of organic aerosols over the Asia/Pacific region.

INDEX TERMS: 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0315 Atmospheric Composition and Structure: Biosphere/atmosphere interactions; 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 1055 Geochemistry: Organic geochemistry; 9320 Information Related to Geographic Region: Asia; *KEYWORDS:* levoglucosan, glucose, sucrose, mycose, dicarboxylic acids, PAH

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1. Introduction

[2] The long-range transport of atmospheric particulate matter from continental regions is an active area of research [e.g., Andreae, 1996; Griffin *et al.*, 2002; Prospero, 1996a,

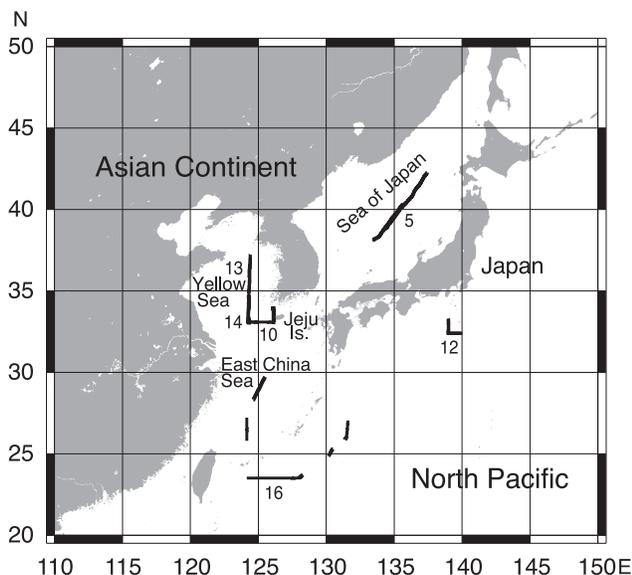


Figure 1. Map of the Asian Pacific with the flight paths for organic aerosol sampling. Solid lines show the flight paths. Numbers refer to research flight (RF). See Table 1 for the flight dates and altitudes.

1996b]. Aerosol particles have relevance for radiative forcing of climate and pollution transport [Seinfeld and Pandis, 1998; Wilkening et al., 2000]. Particles of source emissions from biological organic matter contribute significantly to the total particle burden in the atmosphere, either directly [e.g., Arpino et al., 1972; Conte and Weber, 2002; Cox et al., 1982; Fang et al., 2002; Gagosian et al., 1981, 1982, 1987; Kawamura et al., 2003a; Mazurek and Simoneit, 1997; Simoneit and Mazurek, 1982] or indirectly by burning of biomass and soil resuspension [e.g., Abas et al., 1995; Crutzen and Andreae, 1990; Graham et al., 2002; Kawamura and Gagosian, 1990; Kawamura et al., 1996; Kobayashi et al., 2002; Levine, 1991, 1996; Matsumoto et al., 2001; Simoneit, 2002].

[3] The atmospheric input of terrigenous organic carbon to the world ocean is about equivalent to the organic carbon washed out by the rivers and is estimated to be 2×10^7 ton/year [Buat-Ménard et al., 1989]. This carbon is significantly of a contemporary biological origin and is associated with transport of mineral dust by major wind systems seasonally active from the continents to the oceans [e.g., Kawamura and Gagosian, 1990; Kawamura and Usukura, 1993; Prospero, 1996b, 2001; Simoneit, 1977; Simoneit et al., 1988]. The increasing atmospheric burden of urban particulate matter intermingles with the natural and anthropogenic emissions in the continental rural areas and the total mixture is eventually transported to the oceanic atmosphere. Major oxidative-photochemical reactions alter the organic matter composition during transport, forming derivative products with higher oxygen contents, i.e., more polar, which are ideal as cloud condensation nuclei (complementing the inorganic species such as sulfate and nitrate). Thus molecular characterization and provision of mass balance closure of the initial and downwind aerosols continues to be of interest. Generally, these types of chemical data have been determined on aerosol particles collected near ground level.

[4] The Indian and northwestern Pacific Oceans are currently areas for research of continental dust transport under the auspices of the Aerosol Characterization Experiments (ACE) projects. One aspect of the ACE-Asia project in the northwestern Pacific region is to determine the organic compounds and their sources in the aerosol particles [Huebert et al., 2003]. The Asian continent has been inferred as a source of oceanic dust [e.g., Gagosian et al., 1981; Gagosian and Peltzer, 1986] and that was the case during the sampling campaign in April-May 2001 as confirmed by satellite and other observations [Huebert et al., 2003; Kawamura et al., 2004; Wilkening et al., 2000]. The proposed source regions for the Asian dusts are the deserts and loess plateaus of northern China and Mongolia [Gao et al., 1992; Littmann, 1991; Merrill et al., 1989; Middleton, 1991]. Preliminary organic compound compositions of samples of desert sands from northern China have been presented [Simoneit et al., 2001] and one organic tracer analysis has been reported for a sample from Beijing taken on a 290 m tower, downwind from the sources [Simoneit et al., 1991a]. In the companion paper [Simoneit et al., 2004a], we presented a general assessment of the organic tracer composition of aerosols from Gosan (also Kosan) on Jeju (also Cheju) Island, Korea, and in Sapporo on Hokkaido Island, Japan, as well as on the R/V *Ronald H. Brown* during the ACE-Asia project. Here we present the data of the organic tracers in aerosols sampled at different altitudes during research flights with the National Center for Atmospheric Research (NCAR) C-130 aircraft during the ACE-Asia project. These results should be compared and considered in the same context as those from the ground stations [Simoneit et al., 2004a].

2. Materials and Methods

2.1. Aerosol Samples

[5] Aerosol samples were collected under a variety of conditions (including heavy dust events on research flights RF05 and RF13) from the C-130 in April 2001 over the Korea Strait, Yellow Sea, East China Sea and Sea of Japan (Figure 1). Details of these flights are given by Huebert et al. [2003] and the Technical Appendix to that paper. Constant-altitude level legs were used for the collection of organic aerosols. The altitudes of the flights during sample collection ranged from 35 m to 5500 m. RF5, 12, 13 and 14 are generally high-altitude samples (average altitude: ca. 1200–2800 m) and RF10 and 16 are taken at low altitudes (average ca. 270–460 m). Table 1 summarizes the information of RF samples for this study, flight paths, and flight altitudes. Aerosol samples were collected on a precombusted (550°C) quartz fiber filters with an unmasked area of 5.66 cm² and a flow rate of 9.3 liter/min (27 cm/s) during the aircraft flight patterns. Air was conveyed into the aircraft by a gradually curved solid diffuser inlet and then through a section of black, conductive silicone tubing. Aerosol samples were collected using a side channel of a Particle Concentrator-Brigham Young University Organic Sampling System (PC-BOSS). This PC-Boss sampler, whose schematic diagram has been given elsewhere [Kawamura et al., 2003b], was identical to that diagrammed by Eatough et al. [1999] and Lewtas et al. [2001], except that it had only a single filter pack behind the denuder.

Table 1. Samples and Carbon Contents of Atmospheric Particulate Matter Analyzed From the C-130 Research Flights During the ACE-Asia Campaign in 2001

C-130 Sample	Date Acquired	Flight Path During Sampling	Altitude, ^a m	Altitude for Organic Aerosol Sampling, ^b m	Average Altitude, ^c m	Average OC, ^d $\mu\text{g m}^{-3}$	Average EC, ^d $\mu\text{g m}^{-3}$	Average OC/EC Ratio	nss-SO ₄ , ^e $\mu\text{g m}^{-3}$	Soluble Ca, ^e $\mu\text{g m}^{-3}$
RF5	8 April	Sea of Japan	29–4034	2686, 43, 4006, 3869	2798	4.9	0.7	7.0	14	21
RF10	18 April	Jeju Island vicinity	43–1158	59, 323, 1065	464	2.7	1.4	1.9	7.3	4.1
RF12	23 April	Pacific, 139°E 32°N	20–4006	36, 35, 2825, 3844	1579	1.5	0.4	3.8	3.2	3.3
RF13	24 April	Yellow Sea	30–5487	5450, 673, 248, 322	1191	9.3	1.7	5.5	5.2	13.8
RF14	25 April	Yellow Sea	37–5553	184, 634, 182, 4703, 633, 63, 1615	2144	6.1	1.9	3.2	4.3	2.2
RF16	30 April	East China Sea, Pacific	26–1605	487, 1479, 144, 247, 245	267	1.7	1.7	1.0	6.8	0.4

^aMinimum to maximum altitude flown.

^bAltitudes of horizontal flight conducted for organic aerosol sampling with different sectors.

^cAveraged altitudes for the period of organic aerosol sampling.

^dOC and EC data are from Huebert *et al.* [2004].

^eNon-sea-salt sulfate and soluble Ca data are from Kline *et al.* [2004].

[6] A quartz filter that sampled undenuded air upstream on the side channel PC-BOSS system is referred to as the “sideall” filter. One sideall filter sample was collected during each flight. Total sideall sampling times and volumes were roughly 1.5–4 hours and 1.1–2.8 m³ (STP), respectively. An aliquot of the filter, representing typically 1 m³ of air sampled, was used in this study for lipid class compounds and water-soluble organic compounds including dicarboxylic acids and sugars. Concurrent with each flight’s sideall sample, 3 to 5 sequential aerosol samples were collected using baked quartz filters (47 mm) in the main stream of the PC-BOSS sampler, in which a denuder removed volatile organic compounds [Lewtas *et al.*, 2001]. These filter samples were analyzed for organic carbon (OC) and elemental carbon (EC) by a Sunset Lab’s thermal/optical analyzer [Huebert *et al.*, 2004]. The averaged concentrations of OC were used for TOC estimation in this study. Results of organic carbon analyses and low-molecular-weight dicarboxylic acid analyses are presented elsewhere [Huebert *et al.*, 2004; Kawamura *et al.*, 2003b].

2.2. Extraction and Fractionation

[7] The filter aliquots of the samples and blank were sonicated three times for 10 min each with dichloromethane:methanol (2:1; v/v). The solvent extracts were filtered through quartz fiber wool packed in Pasteur pipettes, and concentrated by use of a rotary evaporator and then under blow-down with dry nitrogen gas. Aliquots of the total extracts were converted to the trimethylsilyl derivatives by reaction with N, O-bis-(trimethylsilyl)trifluoroacetamide (BSTFA) with 1% trimethylsilyl chloride and pyridine for 3 hours at 70°C. This procedure derivatizes COOH and OH groups to the corresponding trimethylsilyl (TMS) esters and ethers, respectively [e.g., Schauer *et al.*, 1996; Simoneit *et al.*, 2004a]. Unexposed portions of the filters were used as surrogate field blanks for background analysis [Kawamura *et al.*, 2003b]. Quartz fiber filters were also used to monitor the laboratory blanks.

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 a Hewlett-Packard model 5973 MSD. Separation was achieved on a fused silica capillary column coated with

DB5 (30 m × 0.25 mm i.d., 0.25 μm film thickness). The GC operating conditions were as follows: temperature hold at 50°C for 2 min, increase from 50 to 300°C at a rate of 6°C min⁻¹ with final isothermal hold at 300°C for 20 min. Helium was used as carrier gas. The sample was injected splitless with the injector temperature at 300°C. The silylated extracts were diluted (to 1:1) with *n*-hexane prior to injection. The mass spectrometer was operated in the electron impact mode (EI) at 70 eV and scanned from 50 to 650 dalton. Data were acquired and processed with the Chemstation software. Individual compounds were identified by comparison of mass spectra with literature and library data, comparison with authentic standards and interpretation of mass spectrometric fragmentation patterns. GC-MS response factors were determined using authentic standards [Simoneit *et al.*, 2004a]. Recoveries of levoglucosan, glucose, sucrose, palmitic acid and stearic acid that were spiked to quartz filter were better than 80%.

3. Results and Discussion

3.1. Mobile Station: C-130 Flights

[9] Organic and elemental carbon contents are summarized in Table 1 and the general sampling locations are shown in Figures 1 and 2. The sampling periods covered the ACE-Asia dust event in April 2001: the “perfect dust storm” (RF05) and an unexpected storm originating in northeast China (RF13). Table 1 also shows the average total soluble calcium (a proxy for dust) on these six flights. The total organic carbon (OC) contents vary from 1.5 to 9.3 μg m⁻³, with the samples from the Yellow Sea exhibiting the highest levels. The total elemental carbon (EC, also called black carbon) contents are less abundant than OC and more uniform at 0.4 to 1.9 μg m⁻³. These values are comparable to those (3.9–8.6, average 7.0 μg m⁻³) obtained at Gosan station on Jeju Island [Simoneit *et al.*, 2004a] and the results reported by other group [Lim *et al.*, 2003] for the same period. OC/EC ratios are fairly high (>3.2), except for two samples (1.0–1.9, RF10 and RF16) (Table 1). The concentrations of the organic compounds in the samples considered here are listed and categorized as classes in Table 2 in the same manner as for the more extensive database for the ground stations [Simoneit *et al.*, 2004a]. The average concentration data for the Gosan station are also listed as a footnote in Table 2. Because of the stable atmosphere over the cold Yellow Sea, inorganic

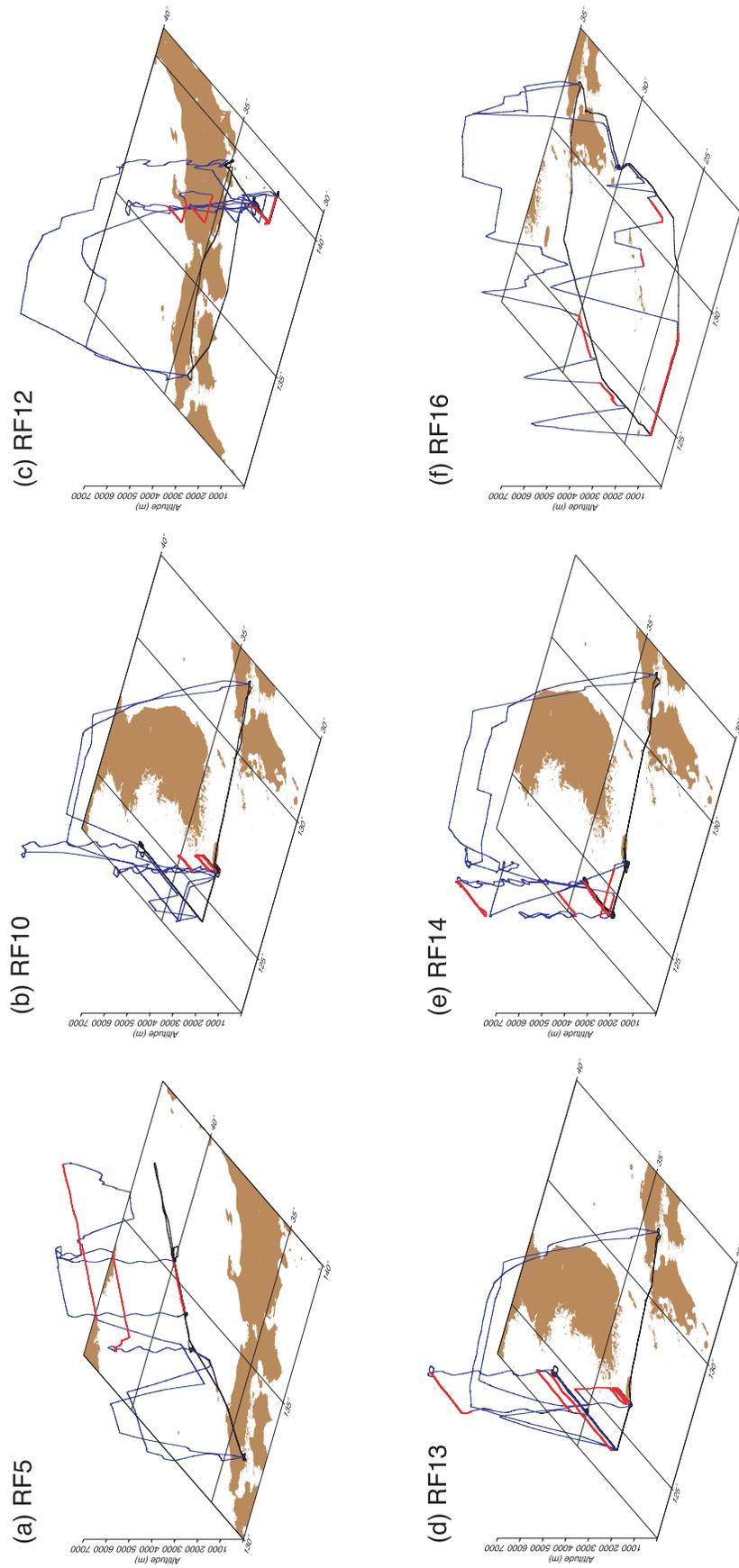


Figure 2. Flight tracks of the NCAR C-130 aircraft during aerosol sample acquisitions: (a) research flight (RF) 5, (b) RF10, (c) RF12, (d) RF13, (e) RF14, and (f) RF16 (red tracks mean the paths for organic matter filtration).

Table 2. Organic Compounds Identified in Samples of Aerosol Particulate Matter Acquired During Research Flights of the NCAR C-130 in the ACE-Asia Campaign^a

Compound ^b	Composition	M.W.	C-130 Flights					
			RF5	RF10	RF12	RF13	RF14	RF16
<i>n</i> -Alkanes								
Eicosane	C ₂₀ H ₄₂	282	0.1		0.7	0.2	0.1	
Heneicosane	C ₂₁ H ₄₄	296	0.3		1.0	0.4	0.3	0.5
Docosane	C ₂₂ H ₄₆	310	0.4	0.5	1.3	0.6	0.5	0.8
Tricosane	C ₂₃ H ₄₈	324	0.9	1.0	1.6	1.1	0.8	1.0
Tetracosane	C ₂₄ H ₅₀	338	1.2	1.3	3.0	1.3	0.9	1.5
Pentacosane	C ₂₅ H ₅₂	352	1.4	1.4	4.3	1.8	1.4	1.8
Hexacosane	C ₂₆ H ₅₄	366	1.5	2.1	5.6	1.4	1.1	2.0
Heptacosane	C ₂₇ H ₅₆	380	2.0	2.1	5.9	3.0	1.9	1.9
Octacosane	C ₂₈ H ₅₈	394	1.9	2.2	6.2	1.5	1.4	1.8
Nonacosane	C ₂₉ H ₆₀	408	2.5	2.7	8.2	4.1	2.2	2.4
Triacontane	C ₃₀ H ₆₂	422	2.0	2.1	7.9	1.5	1.5	1.8
Hentriacontane	C ₃₁ H ₆₄	436	2.2	2.3	8.2	4.3	2.3	1.9
Dotriacontane	C ₃₂ H ₆₆	450	1.6	1.6	7.2	1.0	1.1	1.3
Tritriacontane	C ₃₃ H ₆₈	464	1.0	1.1	5.2	1.4	0.9	1.2
Pentatriacontane	C ₃₅ H ₇₂	492	0.6		3.0	0.3	0.5	
Total			19.6	20.4	69.3	26.6	16.8	20.8
CPI (21–35) ^c			1.25	1.08	1.08	2.1	1.4	1.06
Wax alkanes								
Tricosane	C ₂₃ H ₄₈	324	0.15	0.2		0.2	0.1	
Pentacosane	C ₂₅ H ₅₂	352	0.05			0.4	0.4	0.1
Heptacosane	C ₂₇ H ₅₆	380	0.35		0.3	1.5	0.7	
Nonacosane	C ₂₉ H ₆₀	408	0.5	0.6	1.2	2.6	0.7	0.6
Hentriacontane	C ₃₁ H ₆₄	436	0.4	0.5	0.7	2.0	1.0	0.35
Tritriacontane	C ₃₃ H ₆₈	464				0.4		0.1
Total			1.35	1.3	2.2	7.1	2.9	1.2
PAH								
Benz[a]anthracene	C ₁₈ H ₁₂	228				0.04		
Benzo[k/b]fluoranthene	C ₂₀ H ₁₂	252				0.09		
Benzo[e]pyrene	C ₂₀ H ₁₂	252				0.06		
Benzo[a]pyrene	C ₂₀ H ₁₂	252				0.01		
Indeno[1,2,3-cd]pyrene	C ₂₂ H ₁₂	276				0.03		
Benzo[ghi]perylene	C ₂₂ H ₁₂	276				0.04		
<i>n</i> -Alkanoic acids								
Dodecanoic acid	C ₁₂ H ₂₄ O ₂	200	4.0	2.4	2.2	1.7	1.9	2.2
Tetradecanoic acid	C ₁₄ H ₂₈ O ₂	228	4.5	3.0	3.4	2.4	4.6	3.1
Pentadecanoic acid	C ₁₅ H ₃₀ O ₂	242	0.5	1.2	0.7	0.7	2.1	0.5
Hexadecanoic acid	C ₁₆ H ₃₂ O ₂	256	12	12.4	13.3	12.4	12.5	11.2
Heptadecanoic acid	C ₁₇ H ₃₄ O ₂	270	0.3	0.4	0.5	0.5	0.5	0.3
Octadecanoic acid	C ₁₈ H ₃₆ O ₂	284	5.5	9.5	5.1	9.0	3.7	3.5
Nonadecanoic acid	C ₁₉ H ₃₈ O ₂	298				0.1	0.05	
Eicosanoic acid	C ₂₀ H ₄₀ O ₂	312		0.9		0.6	0.3	
Heneicosanoic acid	C ₂₁ H ₄₂ O ₂	326				0.2	0.1	
Docosanoic acid	C ₂₂ H ₄₄ O ₂	340				1.0	0.7	
Tricosanoic acid	C ₂₃ H ₄₆ O ₂	354				0.3	0.2	
Tetracosanoic acid	C ₂₄ H ₄₈ O ₂	368				1.2	1.0	
Pentacosanoic acid	C ₂₅ H ₅₀ O ₂	382				0.2	0.14	
Hexacosanoic acid	C ₂₆ H ₅₂ O ₂	396				0.9	0.5	
Heptacosanoic acid	C ₂₇ H ₅₄ O ₂	410				0.1	0.1	
Octacosanoic acid	C ₂₈ H ₅₆ O ₂	424				0.9	0.5	
Nonacosanoic acid	C ₂₉ H ₅₈ O ₂	438				0.1	0.05	
Triacontanoic acid	C ₃₀ H ₆₀ O ₂	452				0.7	0.2	
Dotriacontanoic acid	C ₃₂ H ₆₄ O ₂	480				0.2	0.05	
CPI (20-32) ^c			-	-	-	5.9	5.0	-
<i>n</i> -Alkenoic acids								
Hexadecenoic acid	C ₁₆ H ₃₀ O ₂	254	1.0	0.9	1.0		1.2	0.8
Octadecenoic acid	C ₁₈ H ₃₄ O ₂	282	0.4	0.6	0.5	0.2	0.65	0.5
Total aliphatic acids			28.2	31.3	26.7	33.2	30.8	22.1
Dehydroabietic acid	C ₂₀ H ₂₈ O ₂	300				0.01		
Benzoic acid	C ₇ H ₆ O ₂	122	22	5	16	4.2	6	4
Glyceric acid	C ₃ H ₆ O ₄	106	0.28	2.0	0.69	1.8	10	2.2
Dicarboxylic acids								
Succinic acid	C ₄ H ₆ O ₄	118	0.90	2.1	2.3	3.5	9.6	17.3

Table 2. (continued)

Compound ^b	Composition	M.W.	C-130 Flights					
			RF5	RF10	RF12	RF13	RF14	RF16
Glutaric acid	C ₅ H ₈ O ₄	132	0.04	0.2	1.07	0.3	12	3.1
Adipic acid	C ₆ H ₁₀ O ₄	146	0.11	1.6		0.2	1.2	4.1
Malic acid	C ₄ H ₆ O ₅	134	0.10	0.8		0.7	5	2.4
1,2-Benzenedicarboxylic acid	C ₈ H ₆ O ₄	166	0.01	0.13	0.08	0.4	1.5	0.8
1,3-Benzenedicarboxylic acid	C ₈ H ₆ O ₄	166	0.02	0.20	0.31	0.2	2	0.3
1,4-Benzenedicarboxylic acid	C ₈ H ₆ O ₄	166	0.01	0.12	0.42	0.2	1.5	0.2
Total			23.2	10.1	20.2	9.5	27.8	32.2
<i>n</i> -Alkanols								
Docosanol	C ₂₂ H ₄₆ O	326				0.5	0.4	
Tetracosanol	C ₂₄ H ₅₀ O	354				0.8	0.6	
Hexacosanol	C ₂₆ H ₅₄ O	382				1.8	1.2	
Octacosanol	C ₂₈ H ₅₈ O	410				4.8	1.3	
Triacosanol	C ₃₀ H ₆₂ O	438				2.5	0.7	
Dotriacontanol	C ₃₂ H ₆₆ O	466				0.5	0.3	
Nonacosan-10-ol	C ₂₉ H ₆₀ O	424				1.8	1.0	
Total						12.7	6.5	
Anhydrosaccharides								
Levoglucosan	C ₆ H ₁₀ O ₅	162	4.0	5.0	1.3	2.2	6.0	2.0
Mannosan	C ₆ H ₁₀ O ₅	162				0.2	1.1	
Saccharides								
Sucrose	C ₁₂ H ₂₂ O ₁₁	342	0.9	0.7	1.2	0.3	0.8	0.9
Mycose	C ₁₂ H ₂₂ O ₁₁	342	0.7	0.9	1.8	2.5	0.5	0.1
Xylitol	C ₅ H ₁₂ O ₅	152				9.6	1.5	
Sorbitol	C ₆ H ₁₄ O ₆	182		10.4		7.1	5.3	2.0
α-Glucose	C ₆ H ₁₂ O ₆	180	3.5	3.0		1.3	2.0	0.5
β-Glucose	C ₆ H ₁₂ O ₆	180	5.0	3.0		1.7	2.4	0.6
Glycerol	C ₃ H ₈ O ₃	92	52	41	22	40	44	31
Total			66.1	64	26.3	64.9	63.6	37.1
Urea	CH ₄ N ₂ O	60			40	25	32	38
Phthalates								
Dibutyl phthalate	C ₁₆ H ₂₂ O ₄	278	3.0	8.0	3.0	4.5	6.5	3.0
Diocetyl phthalate	C ₂₄ H ₃₈ O ₄	390	16	20	20	14.3	21.5	9.0
Total			19	28	23	18.8	28	12

^aSome samples contained high concentrations of silicones. Values are given in ng m⁻³. M.W., molecular weight.

^bAveraged concentrations (ng m⁻³) for major compound classes observed at the Gosan ground station (G1-G20) during the same period [Simoneit *et al.*, 2004a]: *n*-alkanes, 47.8; PAH, 4.6; *n*-alkanoic acids, 53; *n*-alkanols, 49; anhydrosaccharides, 47.7; saccharides, 224; and phthalates, 8.1.

^cCPI, carbon preference index: (C₂₁ + C₂₃ + C₂₅ + C₂₇ + C₂₉ + C₃₁ + C₃₃ + C₃₅ + C₃₇)/(C₂₂ + C₂₄ + C₂₆ + C₂₈ + C₃₀ + C₃₂ + C₃₄ + C₃₆ + C₃₈) for *n*-alkanes, (C₂₀ + C₂₂ + C₂₄ + C₂₆ + C₂₈ + C₃₀ + C₃₂)/(C₂₁ + C₂₃ + C₂₅ + C₂₇ + C₂₉ + C₃₁ + C₃₃) for *n*-alkanoic acids.

species were often more concentrated aloft than at the Gosan site [Kline *et al.*, 2004].

[10] Typical examples of organic compositions of aerosol particles are discussed and shown in the following text and figures. The major resolved components of the total aerosol extracts are water-soluble saccharides, dihydroxy carboxylic acid and dicarboxylic acids with minor hydrophobic lipids from higher plant waxes and anthropogenic sources (*n*-alkanes, *n*-alkanols, *n*-alkanoic acids) (e.g., Figure 3a and Table 2). The extracts from the filter blanks contained traces of methyl alkanoates, *n*-alkanes, and dioctyl adipate and the amounts were subtracted from the concentrations of these compounds in the samples if present. The following discussion is in the same format as that for the companion study [Simoneit *et al.*, 2004a].

3.1.1. Water-Soluble Sugars and Carboxylic Acids

[11] The saccharides are mainly the series of polyols (saccharide alditols, i.e., reduced sugars) composed of sorbitol (D-glucitol, I), xylitol (II), and glycerol (III) (chemical structures are given in Figure 4), with minor levoglucosan (1,6-anhydro-β-D-glucopyranose (IV), and mannosan (V),

and primary saccharides consisting of α- and β-glucose (VI), sucrose (VII), and mycose (trehalose) (VIII). Levoglucosan (IV) and mannosan (V) are the key tracers for smoke particulate matter from burning of biomass (i.e., thermal alteration products from cellulose, Simoneit *et al.* [1999]). These compounds have been found in aerosols over the ocean [Simoneit and Elias, 2000; Fraser and Lakshmanan, 2000]. They were more concentrated than the primary saccharides and polyols in the aerosols from the ground-based stations [Simoneit *et al.*, 2004a], indicating that smoke from biomass burning is not advected significantly to the higher altitudes of these research flights. The primary saccharides (e.g., glucose, sucrose, etc.) and the saccharide polyols (e.g., sorbitol, xylitol, etc.) are candidate tracers for surface soil dust [Simoneit *et al.*, 2004a, 2004b]. Saccharides are a major fraction of soil organic matter [Gleixner *et al.*, 2002] and have been characterized in riverine particulate matter [da Cunha *et al.*, 2002]. These compounds are present at relatively low atmospheric concentrations during the early spring and become dominant into the summer season, paralleling the agricultural tilling practices on the Asian continent, which resuspend soil dust into the passing aero-

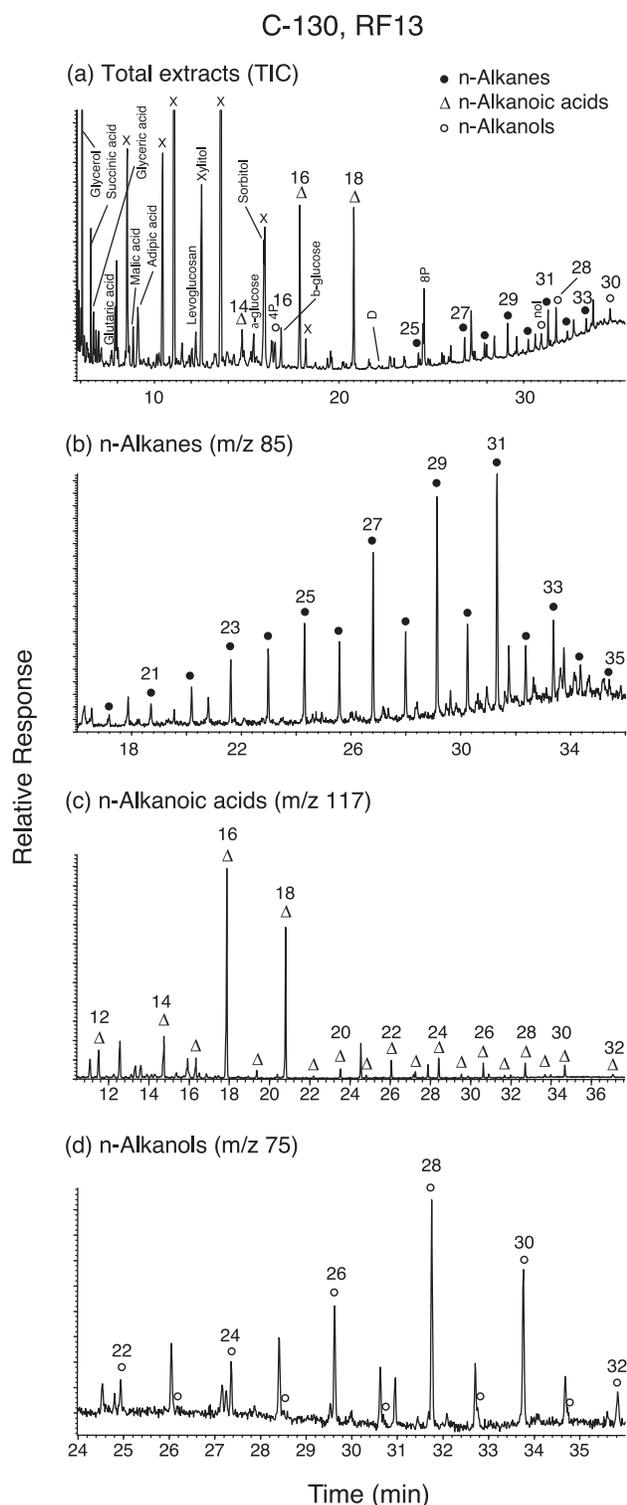


Figure 3. Salient features of the GC-MS data for a total extract (TMS derivatized) of aerosol particles from RF13: (a) total ion current trace, (b) m/z 85 key ion plot for n -alkanes, (c) m/z 117 key ion plot for n -alkanoic acids (TMS esters), and (d) m/z 75 key ion plot for n -alkanols (TMS ethers). Numbers refer to carbon chain length of homologous series; x, silicone contaminants; 4P, dibutyl phthalates; 8P, dioctyl phthalate; D, dehydroabietic acid; nol, nonacosan-10-ol.

sols. The enhanced concentration of the saccharide polyols over that of the primary and dehydrosaccharides indicates advection of soil dust to higher altitudes with concomitant potentially more severe degradation to lower molecular weight products. The characterization, atmospheric chemistry, and implications of these sugars are under further study.

[12] Secondary oxidation products of organic compounds are found mainly as short-chain dicarboxylic acids ranging from oxalic acid to adipic acid and minor aromatic acids (e.g., benzoic, benzenedicarboxylic acids) [Kawamura and Sakaguchi, 1999; Mochida *et al.*, 2003; Simoneit *et al.*, 2004a]. The dicarboxylic acids detected in these total extracts are succinic, glutaric and adipic acids (C_4 - C_6) and the aromatic acids (e.g., 1,2-, 1,3- and 1,4-benzenedicarboxylic acids). Succinic acid is somewhat depleted compared to the data from the direct analyses [e.g., Kawamura *et al.*, 2003b] because of the volatility of the trimethylsilyl esters used in this method, although succinic acid was quantified in another study without evaporative loss using same procedure [Graham *et al.*, 2002]. Some aerosol samples also contain hydroxy carboxylic acids, mainly glyceric (2,3-dihydroxypropionic, C_3) and malic (hydroxysuccinic, C_4) acids, which may be secondary oxidation products from the saccharides. Some of these samples contain high concentrations of urea (IX, carbamide, Table 2), which was not detected in aerosols taken near ground level or in filter blanks [Simoneit *et al.*, 2004a]. This is consistent with the finding of Cornell *et al.* [2001] that urea was a significant fraction of the organic nitrogen found in Hawaii, although it is not found over the Atlantic. Urea may be a secondary product from atmospheric reactions or a primary emission from anthropogenic sources. It was not detected in the ground-based samples. Thus the saccharides, dihydroxy carboxylic acid, dicarboxylic acids and urea comprise the major water-soluble organic compounds of these aerosols.

3.1.2. Aliphatic Lipids

[13] The lipid components, secondary in concentration in these aerosols (Table 2), are derived from terrestrial plant waxes, marine microbial detritus and emissions from fossil fuel use. This is illustrated by the key ion plots for the total extract GC-MS data as the m/z 85 plot for n -alkanes, m/z 117 plot for n -alkanoic acids as the TMS esters, and m/z 75 for the n -alkanols as the TMS ethers (Figure 3). The n -alkanes typically range from C_{20} to C_{35} , some with a strong odd carbon number predominance (CPI ranges from 1.05 to 2.0) and carbon number maxima (C_{max}) at C_{29} or C_{31} . Volatile compound blow-off from the filters over the acquisition time has probably depleted mainly the n -alkanes $<C_{21}$ and other petroleum tracers (i.e., pristane and phytane) for urban traffic emissions, which were not detected. The heavier components from petroleum product utilization (i.e., urban or air traffic) are present as n -alkanes $>C_{21}$ with no carbon number predominance but the supporting biomarkers (hopanes and steranes) are not detectable [Simoneit, 1984, 1985]. Leaf surface (epicuticular) wax input to aerosols is recognized by the n -alkanes from C_{21} - C_{35} with a strong odd carbon number predominance, and n -alkanoic acids from C_{20} - C_{32} and n -alkanols from C_{22} - C_{30} , both with strong even carbon number predominances [Simoneit, 1989]. The contribution of epicuticular wax to the total n -alkanes has been

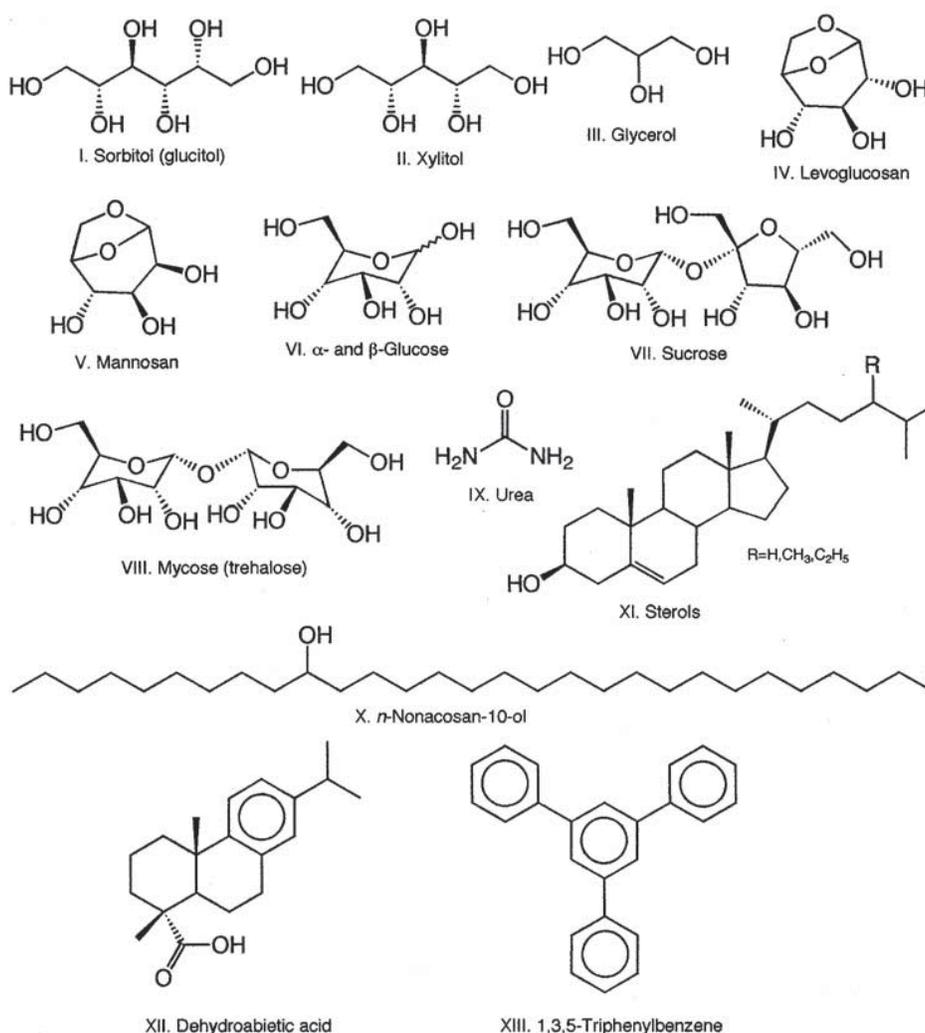


Figure 4. Chemical structures cited.

estimated by the subtraction of the smooth alkane envelope attributed to fossil fuel emissions from the total alkanes [Simoneit *et al.*, 1991b] and the wax alkane concentrations are listed in Table 2, confirming the C_{\max} at 29 or 31.

[14] The *n*-alkanoic acids (fatty acids) range mainly from C_{12} to C_{32} , with C_{\max} at 16, 18 and minor C_{\max} at 22 or 24 and strong even carbon number predominances (CPI = 5 and 5.9 for C_{20} - C_{32} , Table 2) (e.g., Figure 3c). There are only traces of unsaturated alkenoic acids (e.g., $C_{16:1} > C_{18:1}$) indicating that the organic matter of these aerosols is aged, has a significant input of marine-derived lipids (based on $C_{16:1}$ content), and has been transported over significant distances. The alkenoic acids are rapidly oxidized once in the atmosphere [e.g., Kawamura and Gagosian, 1987]. The *n*-alkanoic acids $<C_{20}$ have multiple sources and thus indicate mainly a biogenic input. However, the alkenoic acids $<C_{20}$ are present at much higher levels than the homologs $>C_{20}$ further supporting a marine lipid input. The *n*-alkanoic acids from C_{20} to C_{32} in some samples are interpreted to derive from higher plant waxes

and they have very similar distributions of $C_{22} \approx C_{24} > C_{26} < C_{28}$ as reported for the ground station samples during the same time interval [Simoneit *et al.*, 2004a].

[15] The *n*-alkanols (fatty alcohols) range from C_{20} to C_{32} , with strong even carbon number predominances (essentially no odd numbered alkanols, CPI all >4 , Table 2) and C_{\max} at C_{28} during the dust event of RF13 (e.g., Figure 3d). Some aerosol samples contain *n*-nonacosan-10-ol (X) at significant levels (Table 2). The normal alkanols indicate an input of plant waxes and nonacosan-10-ol indicates an input of waxes from forests consisting of both soft and hardwoods [Oros and Simoneit, 2001a, 2001b]. Minor amounts of these alkanols may also be injected into the atmosphere by smoke from biomass burning [Oros and Simoneit, 2001a, 2001b; Simoneit, 2002], but β -sitosterol (XI, $R = \beta C_2H_5$), a supporting tracer is not detectable. Cholesterol (XI, $R = H$), found in most of the ground-based samples [Simoneit *et al.*, 2004a] and indicating an input from marine (algal) sources and possibly from emissions from cooking in urban areas

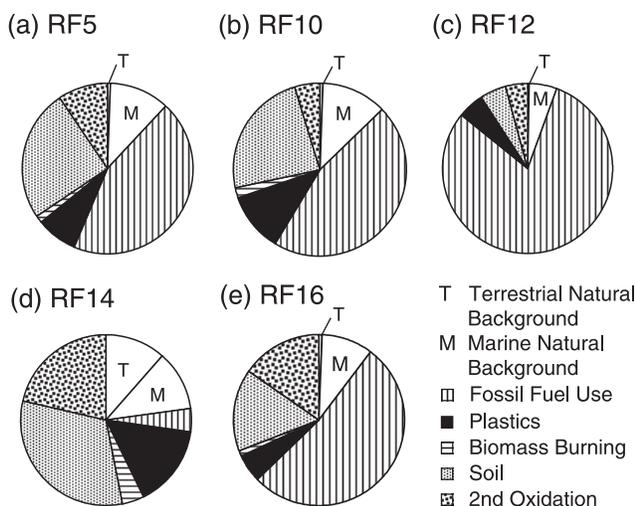


Figure 5. (a–e) Pie diagrams showing the source strengths of organic matter in the C-130 aerosol samples. See Table 1 for information on the RF samples.

[e.g., Rogge *et al.*, 1991; Simoneit *et al.*, 1983], is not detectable. The vascular plant wax composition (n -alkanes $>C_{23}$, n -alkanoic acids $>C_{22}$ and n -alkanols $>C_{22}$) of these samples is consistent with an origin from continental areas [Simoneit *et al.*, 2001].

3.1.3. Combustion Tracers

[16] Biomass burning injects numerous organic compounds from the fuels directly into the atmosphere, and on the basis of the tracer compositions both the combustion process and the fuel types burned can be assessed [Simoneit, 2002]. However, in these samples the subordinate combustion tracers are too dilute and thus not detectable in most samples. The major markers for burning of fuel containing cellulose are levoglucosan (IV) with lesser amounts of the other anhydrosaccharides (e.g., V). Levoglucosan is a relatively minor component in most aerosol samples analyzed here (Table 2).

[17] A secondary tracer for biomass burning is dehydroabietic acid (XII), the key marker for smoke from burning of conifer wood [Standley and Simoneit, 1994; Rogge *et al.*, 1998; Simoneit, 1999]. It is found as a minor component in one of these samples (Table 2). Lignin is the other major biopolymer of wood and upon burning yields phenolic tracers in the smoke characteristic of the fuel type [Simoneit *et al.*, 1993]. These tracers are not found in the aerosol samples analyzed here. This is probably due to the relative low concentration of these compounds in the total extracts.

[18] Polycyclic aromatic hydrocarbons (PAHs) are detectable in only one sample (Table 2). The PAHs range from benzantracene to benzoperylene and their distributions are approximately similar as those for the ground-based stations [Simoneit *et al.*, 2004a]. 2,9-Dimethylpicene, the tracer for coal burning, or 1,3,5-triphenylbenzene (XIII), is not detectable.

[19] The minor contribution of long-chain alkanes C_{34} to $>C_{37}$, especially for samples from Sapporo which was interpreted to be from paraffin wax (C_{max} at 28), a petroleum product used for example on food containers was not obvious in these samples. However, the enhanced contents

of phthalates in these aerosol samples (Table 2, the ground station average is 8 ng m^{-3}) may indicate fugitive emissions from burning of urban refuse [Didyk *et al.*, 2000] or contamination during sampling from our silicone tubing. Source tests from refuse burning sources have not yet been conducted.

3.2. Emission Sources

[20] The organic compound groups characterized and quantitated for these aerosols are similar to those reported for the ground-based stations [Simoneit *et al.*, 2004a] and can be apportioned to six emission sources and to significant oxidation reactions during longer-range transport, which produces secondary products. These source groups are summarized in Table 3 and plotted for selected samples in Figure 5. Although this apportionment approach is useful to roughly estimate the relative contribution from major emission sources to the organic aerosols, it does not necessarily exclude a potential overlapping problem; that is, some substances may come from more than one source. The terrestrial natural background compounds are minor and consist of vascular plant wax lipids ranging from 0.5 to 10.6% of the total identified compound mass (TCM) for the research flights (Table 4). The emissions from fossil fuel utilization range from 33–80% of TCM, but petroleum markers cannot be distinguished from coal-derived hydrocarbons. These values are in general greater than those reported in the ground-based aerosols from Gosan, Sapporo and Chichi-jima island from the western North Pacific, as

Table 3. Total Compound Group Yields^a

Compound Groups	C-130 Flights					
	RF5	RF10	RF12	RF13	RF14	RF16
Plant wax						
Alkanes ^b	1.3	1.3	2.2	7.1	2.9	1.2
Alkanoic acids ^c	-	-	-	8.3	10.6	-
Alkanols	-	-	-	12.7	6.5	-
Total	1.3	1.3	2.2	28.1	20.0	1.2
Fossil fuel						
Alkanes ^b	18.2	19.1	67.1	19.5	13.9	19.6
UCM ^d	92.5	96	336	98	70	98
Total	11	115	403	118	84	118
Marine alkanolic acids ^c	28.2	31.3	26.7	24.9	20.2	22.1
Combustion						
PAH	-	-	-	0.27	-	-
Levoglucosan	4	5	1.3	2.4	7.1	2
Soil						
Saccharides	62	59	25	62.5	56.5	35.1
Secondary oxidation						
Dicarboxylic acids + glyceric acid	23.5	12.2	20.9	11.5	37.8	34.4
Others						
Urea	-	-	40	25	32	38
Phthalates	19	28	23	19	28	12
Water-soluble compounds ^e	89.5	76.2	87.2	101	133.4	72.4
Lipid compounds	159.6	175.6	455	165.2	132	153.3

^aValues are given in ng m^{-3} .

^bPlant wax alkanes are calculated as the excess odd homolog-adjacent even homologs average [Simoneit *et al.*, 1991b, 1991c].

^cPlant wax alkanolic acids are apportioned as the sum of all homologs $>C_{20}$ and the equal increment of C_{22} or C_{24} equivalent to C_{18} for those $<C_{20}$.

^dUCM is estimated on the basis of the U:R (unresolved: resolved compounds) of 5 for emissions from vehicle traffic in a Los Angeles tunnel [Fraser *et al.*, 1998].

^eIncludes levoglucosan, saccharides, dicarboxylic acids, glyceric acid, and urea.

Table 4. Sources and Water-Soluble Compound Groups in the Total Compound Mass of the Extractable Organic Matter From the Airborne Aerosols^a

C-130 Sample	Natural ^b		Urban ^b		Biomass Burning ^c	Soil Resuspension ^d	Secondary Oxidation Products ^e	Total Water-Soluble Compounds ^f
	Plant Wax Lipids	Marine Lipids	Fossil Fuel	Plastics				
RF5	0.5	11.3	44.6	7.6	1.6	24.9	9.4	36
RF10	0.5	12.4	45.7	11.1	2	23.4	4.8	30.3
RF12	0.4	5.3	80.3	4.6	0.3	5	4.2	16
RF13	10.6	9.4	44.3	7.1	0.9	23.5	4.3	38
RF14	7.9	8	3.1	11	2.8	22.3	14.9	50.3
RF16	0.5	9.8	52.5	5.3	0.9	15.6	15.3	32
Average	3.4	9.4	50	7.8	1.4	19.1	8.8	33.8

^aValues are given as relative percent.

^bTotals from Table 4 divided by total of all compounds + UCM.

^c(Total anhydrosaccharides + resin acids)/(total for biomass burning).

^d(Total saccharides)/(total for soil resuspension).

^e(Total dicarboxylic, glyceric, and benzoic acids)/(total for secondary oxidation).

^fWater-soluble compounds: sum of biomass burning, soil resuspension, urea, and secondary oxidation products.

well as mobile station R/V *Ron H. Brown* [Simoneit et al., 2004a]. These fossil-fuel-derived hydrocarbons are interpreted to originate from numerous sources. Emissions from possible burning of refuse range from 5 to 11% of TCM as phthalates and possibly reflect the influences of urban emissions and sampling contamination. The dominant emittants for these samples taken on the research flights are silicone lubricants (e.g., Figure 3a), which could be from our sample tubing.

[21] Biomass burning, as reflected by wood smoke markers (levoglucosan and dehydroabietic acid), is very low and accounts for 0.3 to 2.8% of TCM of the compound mass during the research flights. The saccharides compose from 5 to 24.9% of TCM. These compounds are interpreted to represent viable biomass as in soil and possibly in marine particulate matter [Simoneit et al., 2004a]. Saccharides have been characterized in urban aerosols that contain entrained soil dust [e.g., Didyk et al., 2000; Simoneit et al., 2004b], and thus they are used here as tracers for soil resuspension by agricultural tilling or wind erosion. The marine lipid input to these aerosols is reflected mainly by the apportioned fatty acid content, which ranges from 5.3 to 12.4% of TCM. This marine component is in all cases greater than the continental plant waxes transported over longer distances.

[22] The secondary products derived from oxidative reactions of the organic compounds during transport consist mainly of dicarboxylic acids, hydroxy carboxylic acids and aromatic acids. This compound group varies from 4.2 to 15.3% of TCM for the aerosols of the research flights. It is of interest to note that relative abundance of secondary oxidation products in TCM become the largest when OC content is highest (in the dust from a northeast China source in RF13, see Tables 1 and 4). This may suggest that water-soluble carboxylic acids largely contribute to OC in the atmospheric aerosols by gas-to-particle conversion. The remote marine atmospheric samples collected on board R/V *Ron H. Brown* over the North Pacific showed a higher proportion of secondary oxidation products than the atmosphere downwind from Asia at Gosan and Sapporo [Simoneit et al., 2004a]. This has also been demonstrated earlier on prior cruises in the western Pacific [e.g., Kawamura and Sakaguchi, 1999; Sempéré and Kawamura,

2003]. The terrestrial compounds have been subjected to thorough oxidation during transport which is also supported by low concentrations of especially C₁₈ unsaturated fatty acids.

3.3. Water-Soluble Organic Compounds

[23] The water solubility of the carbonaceous organic fraction of aerosols is a major open question in climate models [Facchini et al., 1999]. Oxalic acid, with the other short-chain dicarboxylic acids, has been documented as the dominant water-soluble components of organic aerosol matter [e.g., Kawamura and Usukura, 1993; Kawamura and Sakaguchi, 1999; Sempéré and Kawamura, 2003]. Here we add the following groups of organic compounds to the list of water-soluble organic matter on the basis of their

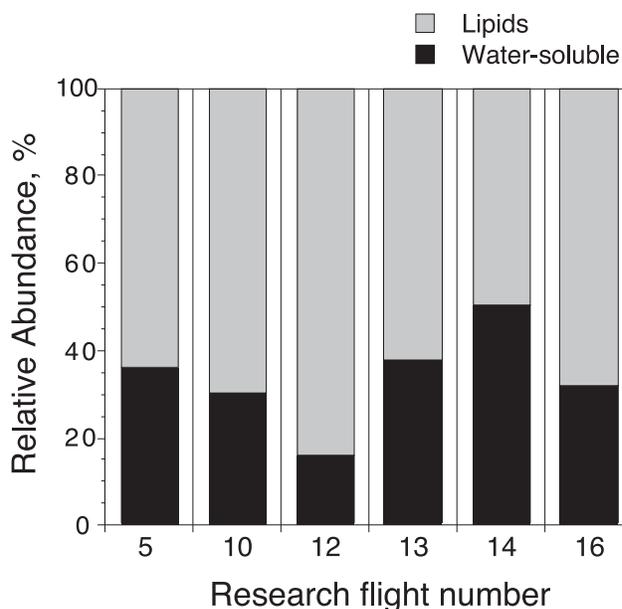


Figure 6. Bar plot showing the relative proportions of lipid (organic solvent extractable) versus water-soluble compounds in the C-130 aerosol samples. Numbers under the bars refer to sample ID (see Table 1).

complete aqueous solubility properties: saccharides and anhydrosaccharides, as well as the secondary oxidation products such as dicarboxylic, hydroxy dicarboxylic, dihydroxy monocarboxylic and aromatic acids. The sum of these water-soluble compounds ranges from 16 to 50% of TCM (Figure 6 and Table 4) for all the samples analyzed here. These water-soluble fractions are unusually high and when coupled with the oxalic and malonic acid contents determined separately [e.g., Kawamura *et al.*, 2003b, 2004; Mochida *et al.*, 2003] make a major portion of the total compound mass soluble in water. For the filter samples collected on the C-130 flights, Kawamura *et al.* [2003b] utilized the dibutyl ester derivatization technique and reported that oxalic and malonic acids are 1 order of magnitude more abundant than succinic acid with a total C₂-C₅ dicarboxylic acid concentration range of 44–870 ng m⁻³. We report 12–38 ng m⁻³ of dicarboxylic acids (C₄-C₆) in the total extracts of aerosol particles (Table 3). This is quite different from the urban aerosols such as Los Angeles or Santiago, where the bulk of the organic compounds are more hydrophobic [e.g., Schauer *et al.*, 1996; Didyk *et al.*, 2000]. It is also different when compared to the Saharan dust aerosols from Africa over the North Atlantic, which have low contents of these water-soluble compounds (0.02–0.04 ng m⁻³, 3–9% of TCM, a lower limit due to less efficient sample acquisition) [Simoneit and Elias, 2000].

4. Conclusions

[24] Ultra small samples of aerosol particles taken on aircraft can be analyzed for organic compound tracers by the simple total extract derivatization GC-MS method. The primary inputs of organic compounds to ACE-Asia aerosols sampled aloft by airplane are: (1) secondary products consisting of dicarboxylic acids and aromatic acids, which are derived from the oxidation of atmospheric organic matter during long-range transport (2) natural emissions of terrestrial plant wax and marine lipids based on the alkanolic acid, alkanol and alkane distributions; (3) smoke from biomass burning using levoglucosan as the tracer; (4) soil resuspension due to spring agricultural activity as inferred from major primary and reduced sugars (e.g., sucrose, glucose, sorbitol); and (5) urban/industrial emissions from fossil fuel use and other urban emissions based on the alkanes and plasticizers. The general compound distributions and sources are analogous and comparable as reported for the ground stations during the same time period [Simoneit *et al.*, 2004a].

[25] The organic compound compositions of these samples are very different from those reported for aerosol particles of the Atlantic Ocean [Simoneit and Elias, 2000] and from the data reported earlier for the mid-Pacific [e.g., Gagosian *et al.*, 1981]. The major external sources recognized in these samples are as follows: (1) the plant wax composition which fits with an origin from the deserts of northwestern China; (2) tracers from biomass-burning smoke and from agricultural soil resuspension; and (3) PAHs from fossil fuel combustion. The water-soluble fraction of the total organic matter is high and consists of saccharides, anhydrosaccharides and secondary dicarboxylic acids.

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