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**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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## ABSTRACT

Atmospheric particulate matter, collected over the polluted East Asia/Pacific region in spring 2001 during research flights with the NCAR C-130 aircraft were analyzed for different types of organic compounds using capillary gas chromatography-mass spectrometry. Seventy organic species were detected in the aerosols and grouped into different compound classes based on functional groups, including *n*-alkanes, PAHs, fatty acids, dehydroabietic acid, alkanols, water-soluble sugars (including glucose, sucrose, trehalose and levoglucosan), mono- and dicarboxylic acids, urea, and phthalates. Interestingly, the water-soluble compounds (72–133 ng/m<sup>-3</sup>) 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 estimate their source apportionment. Fossil fuel combustion was recognized as the most significant source for the organic aerosols (contributing 33–80% of TCM, ave. 50%), followed by soil resuspension (5–25%, ave. 19%) and secondary oxidation products (4–15%, ave. 9%). In contrast, the contribution of natural sources such as terrestrial plant wax and marine lipids (fatty acids and alkanols) were relatively small (3.4% and 9.4% on average, respectively). Biomass burning was suggested to contribute only a minor portion to 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 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.

Keywords: Levoglucosan, glucose, sucrose, trehalose, dicarboxylic acids, PAH, hydrocarbons, fatty acids, alkanols, atmospheric aerosols, ACE-Asia

## 1. INTRODUCTION

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, 1996 a,b). 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; Cox et al., 1982; Gagosian et al., 1981, 1982, 1987; 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; Kobayashi et al., 2003; Levine, 1991, 1996; Simoneit, 2002).

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  $2 \times 10^7$  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., greater polarity, which are ideal as cloud condensation nuclei (complementing the inorganic species such as  $\text{SO}_x$ ,  $\text{NO}_x$ ). 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.

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. The Asian continent has been inferred as a source of oceanic dust (e.g., Gagosian et al., 1981) and that was the case during the sampling campaign in April-May 2001 as confirmed by satellite and other observations. The proposed source regions for the Asian dusts are the deserts and loess regions of northern China and Mongolia. 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). 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 (Simoneit et al., 2004a). Here we present the data of the organic tracers in aerosols sampled at different altitudes during research flights with the NCAR C-130 aircraft.

## 2. MATERIALS AND METHODS

### 2.1. Aerosol Samples

Aerosol samples were collected during research flights of the C-130 in April, 2001 over the Korea Strait, Yellow Sea, East China Sea and Sea of Japan (Fig. 1). The altitudes of the flights during sample collection ranged from 20 m to 7000 m. Aerosol samples were collected on a pre-combusted (550°C) quartz fiber filters (25 mm) with a cross section of 5.66 cm<sup>2</sup> and a flow rate of 9.3 liter/min (27 cm/sec) during the aircraft flight patterns. Air was conveyed into the aircraft by a gradually-curved solid diffuser inlet. 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., 2003), was identical to that diagrammed in Eatough et al. (1999) and Lewtas et al. (2001), except that it had only a single filter pack behind the denuder.

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 m<sup>3</sup> (STP), respectively. An aliquot of the filter, representing typically 1 m<sup>3</sup> 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). The filter samples were analyzed for organic carbon (OC) and elemental carbon (EC) by a Sunset Lab’s thermal/optical analyzer (Bertram 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 (Bertram et al., 2003; Kawamura et al., 2003).

### 2.2. Extraction and Fractionation

The samples of filter aliquots were sonicated three times for 10 min each with dichloromethane:methanol (2:1; v/v). The solvent extract was filtered through quartz fiber wool packed in a Pasteur pipette, 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 h at 70°C. This procedure derivatizes COOH and OH groups to the corresponding trimethylsilyl (TMS) esters and ethers, respectively.

### 2.3. Gas chromatography-mass spectrometry

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 (30m x 0.25 mm i.d., 0.25µm film thickness). The GC operating conditions were as follows: temperature hold at 65°C for 2 min, increase from 65 to 300°C at a rate of 6°C min<sup>-1</sup> 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. Recoveries of levoglucosan, glucose, sucrose, palmitic acid and stearic acid were better than 80%.

## 3. RESULTS AND DISCUSSION

### 3.1. Mobile Station: C-130 Flights

The samples for this study, flight paths, altitudes and organic 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. RF 5, 12, 13 and 14 are generally high altitude samples and RF 10 and 16 are taken at low altitudes. The total organic carbon (OC) contents vary from 1.5 to 9.3 µg m<sup>-3</sup>, 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<sup>-3</sup>. The concentrations of the organic compounds in the samples considered here are listed and categorized as classes in Table 2.

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., Fig. 3a, Table 2).

### 3.1.1. Water-soluble sugars and carboxylic acids

The saccharides are mainly the series of polyols (saccharide alditols, i.e., reduced sugars) comprised of sorbitol (D-glucitol, I), xylitol (II), and glycerol (III) (chemical structures are given in Appendix I), with minor levoglucosan (1,6-anhydro- $\beta$ -D-glucopyranose (IV), and mannosan (V), and primary saccharides consisting of  $\alpha$ - and  $\beta$ -glucose (VI), sucrose (VII), and mycose (trehalose) (VIII). Levoglucosan (IV), with 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 and thus are stable during long-range transport (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, b). 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 resuspends soil dust into the passing aerosols. 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 is under further study.

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 ( $C_3$ - $C_6$ ) 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., 2003) due to 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 nor in filter blanks (Simoneit et al., 2004a). Urea may be a secondary product from atmospheric reactions or a primary emission from anthropogenic sources. 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

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 (Fig. 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 depleted mainly the  $n$ -alkanes  $<C_{21}$  and therefore the petroleum tracers (i.e., pristane and phytane) for urban traffic emissions are not detectable. 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 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.

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., Fig. 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, 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).

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 (e.g., Fig. 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,b). Minor amounts of these alkanols may also be injected into the atmosphere by smoke from biomass burning (Oros and Simoneit, 2001a, b; Simoneit, 2002), but  $\beta$ -sitosterol (XI, R= $\beta$ C<sub>2</sub>H<sub>5</sub>), 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 (e.g., Rogge et al., 1991; Simoneit et al., 1983), is not detectable. The vascular plant wax composition (*n*-alkanes >C<sub>23</sub>, *n*-alkanoic acids >C<sub>22</sub> and *n*-alkanol >C<sub>22</sub>) of these samples is consistent with an origin from the northwestern deserts of China when compared to sand and aerosol samples from those areas (Simoneit et al., 2001).

### 3.1.3. Combustion tracers

Biomass burning injects numerous organic compounds from the fuels directly into the atmosphere and based on the major tracers found both the process and the types of fuels can be assessed (Simoneit, 2002). However, in these samples the combustion tracers are quite dilute and often not detectable. The major markers for burning of anything that contains 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).

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.

Polycyclic aromatic hydrocarbons (PAHs) are detectable in only one sample (Table 2). The PAHs range from benzantracene to benzopyrene 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. PAHs are of health concerns because of their carcinogenicity, genotoxicity and endocrine disrupting potential. The PAHs are detectable in samples taken on airplane flights, especially if concentrated extracts are analyzed.

The minor contribution of long chain alkanes C<sub>34</sub> to >C<sub>37</sub>, especially for samples from Sapporo which was interpreted to be from paraffin wax (C<sub>max</sub> 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) may indicate fugitive emissions from burning of urban refuse (Didyk et al., 2000) or contamination during sampling. Source tests from refuse burning sources have not yet been conducted.

### 3.2. Emission Sources

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 long range transport, which produces secondary products. These source groups are summarized in Table 3 and plotted for selected samples in Figure 4. 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 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 probably reflect the influences of urban emissions and sampling contamination. However, the dominant contaminants for these samples taken on the research flights are silicone lubricants (e.g., Fig. 3a).

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 comprise 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 which 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.

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. The remote marine atmosphere has 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). The terrestrial compounds have been subjected to thorough oxidation during transport which is also supported by low concentrations of especially C<sub>18</sub> unsaturated fatty acids.

### 3.3. Water-soluble organic compounds

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). Here we add the following groups of organic compounds to the list of water-soluble organic matter based on their 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 5 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., Mochida et al., 2003; Kawamura 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. (2003) utilized the dibutyl ester derivatization technique and reported that oxalic and malonic acids are one order of magnitude more abundant than succinic acid with a total C<sub>2</sub>-C<sub>5</sub> diacids concentration range of 44–870 ng m<sup>-3</sup>. We report 12–38 ng m<sup>-3</sup> of dicarboxylic acids (C<sub>3</sub>-C<sub>6</sub>) 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 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 (Simoneit and Elias, 2000).

## 4. CONCLUSIONS

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).

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 Pacific (e.g., Gagosian et al., 1981). The major external sources recognized in these samples are: (1) the plant wax composition which fits with an origin from the deserts of northwestern China; (2) high levels of tracers from biomass burning smoke and from agricultural soil resuspension; and (3) high levels of PAHs mainly from coal combustion. The water-soluble fraction of the total organic matter is high and consists of the saccharides, anhydrosaccharides and the secondary dicarboxylic acids.

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**Table 1. Samples and carbon contents of atmospheric particulate matter analyzed from research flights of the NCAR C-130 during the ACE-Asia campaign.**

Sample	Date acquired	Flight path during sampling	Altitude (m) <sup>a</sup>	Ave. Altitude (m)	OC ( $\mu\text{g m}^{-3}$ )	EC ( $\mu\text{g m}^{-3}$ )
RF5	8 April 2001	Sea of Japan	29–4034	2798	4.9	0.7
RF10	18 April 2001	Jeju Is. vicinity	43–1158	464	2.7	1.4
RF12	23 April 2001	Pacific, 139°E 32°N	20–4006	1579	1.5	0.4
RF13	24 April 2001	Yellow Sea	30–5487	1191	9.3	1.7
RF14	25 April 2001	Yellow Sea	37–5553	2144	6.1	1.9
RF16	30 April 2001	East China Sea	26–1605	267	1.7	1.7

<sup>a</sup>Minimum to maximum altitude flown.

**Table 2. Organic compounds identified in samples of aerosol particulate matter acquired during research flights of the NOAA C-130 in the ACE-Asia campaign (ng m<sup>-3</sup>).**

Compound <sup>a</sup>	Composition	M.W.	C-130 Flights					
			RF5	RF10	RF12	RF13	RF14	RF16
<b><i>n</i>-Alkanes</b>								
Eicosane	C <sub>20</sub> H <sub>42</sub>	282	0.1		0.7	0.2	0.1	
Heneicosane	C <sub>21</sub> H <sub>44</sub>	296	0.3		1.0	0.4	0.3	0.5
Docosane	C <sub>22</sub> H <sub>46</sub>	310	0.4	0.5	1.3	0.6	0.5	0.8
Tricosane	C <sub>23</sub> H <sub>48</sub>	324	0.9	1.0	1.6	1.1	0.8	1.0
Tetracosane	C <sub>24</sub> H <sub>50</sub>	338	1.2	1.3	3.0	1.3	0.9	1.5
Pentacosane	C <sub>25</sub> H <sub>52</sub>	352	1.4	1.4	4.3	1.8	1.4	1.8
Hexacosane	C <sub>26</sub> H <sub>54</sub>	366	1.5	2.1	5.6	1.4	1.1	2.0
Heptacosane	C <sub>27</sub> H <sub>56</sub>	380	2.0	2.1	5.9	3.0	1.9	1.9
Octacosane	C <sub>28</sub> H <sub>58</sub>	394	1.9	2.2	6.2	1.5	1.4	1.8
Nonacosane	C <sub>29</sub> H <sub>60</sub>	408	2.5	2.7	8.2	4.1	2.2	2.4
triacontane	C <sub>30</sub> H <sub>62</sub>	422	2.0	2.1	7.9	1.5	1.5	1.8
Hentriacontane	C <sub>31</sub> H <sub>64</sub>	436	2.2	2.3	8.2	4.3	2.3	1.9
Dotriacontane	C <sub>32</sub> H <sub>66</sub>	450	1.6	1.6	7.2	1.0	1.1	1.3
Tritriacontane	C <sub>33</sub> H <sub>68</sub>	464	1.0	1.1	5.2	1.4	0.9	1.2
Pentatriacontane	C <sub>35</sub> H <sub>72</sub>	492	0.6		3.0	0.3	0.5	
<b>Total</b>			<b>19.6</b>	<b>20.4</b>	<b>69.3</b>	<b>26.6</b>	<b>16.8</b>	<b>20.8</b>
CPI (21-35)			1.25	1.08	1.08	2.1	1.4	1.06
<b>Wax alkanes</b>								
Tricosane	C <sub>23</sub> H <sub>48</sub>	324	0.15	0.2		0.2	0.1	
Pentacosane	C <sub>25</sub> H <sub>52</sub>	352	0.05			0.4	0.4	0.1
Heptacosane	C <sub>27</sub> H <sub>56</sub>	380	0.35		0.3	1.5	0.7	
Nonacosane	C <sub>29</sub> H <sub>60</sub>	408	0.5	0.6	1.2	2.6	0.7	0.6
Hentriacontane	C <sub>31</sub> H <sub>64</sub>	436	0.4	0.5	0.7	2.0	1.0	0.35
Tritriacontane	C <sub>33</sub> H <sub>68</sub>	464				0.4		0.1
<b>Total</b>			<b>1.35</b>	<b>1.3</b>	<b>2.2</b>	<b>7.1</b>	<b>2.9</b>	<b>1.2</b>
<b>PAH</b>								
Benz[a]anthracene	C <sub>18</sub> H <sub>12</sub>	228				0.04		
Benzo[k/b]fluoranthene	C <sub>20</sub> H <sub>12</sub>	252				0.09		

Benzo[e]pyrene	C <sub>20</sub> H <sub>12</sub>	252				0.06		
Benzo[a]pyrene	C <sub>20</sub> H <sub>12</sub>	252				0.01		
Indeno[1,2,3-cd]pyrene	C <sub>22</sub> H <sub>12</sub>	276				0.03		
Benzo[ghi]perylene	C <sub>22</sub> H <sub>12</sub>	276				0.04		
<b><i>n</i>-Alkanoic acids</b>								
Dodecanoic acid	C <sub>12</sub> H <sub>24</sub> O <sub>2</sub>	200	4	2.4	2.2	1.7	1.9	2.2
Tetradecanoic acid	C <sub>14</sub> H <sub>28</sub> O <sub>2</sub>	228	4.5	3.0	3.4	2.4	4.6	3.1
Pentadecanoic acid	C <sub>15</sub> H <sub>30</sub> O <sub>2</sub>	242	0.5	1.2	0.7	0.7	2.1	0.5
Hexadecanoic acid	C <sub>16</sub> H <sub>32</sub> O <sub>2</sub>	256	12	12.4	13.3	12.4	12.5	11.2
Heptadecanoic acid	C <sub>17</sub> H <sub>34</sub> O <sub>2</sub>	270	0.3	0.4	0.5	0.5	0.5	0.3
Octadecanoic acid	C <sub>18</sub> H <sub>36</sub> O <sub>2</sub>	284	5.5	9.5	5.1	9.0	3.7	3.5
Nonadecanoic acid	C <sub>19</sub> H <sub>38</sub> O <sub>2</sub>	298				0.1	0.05	
Eicosanoic acid	C <sub>20</sub> H <sub>40</sub> O <sub>2</sub>	312		0.9		0.6	0.3	
Heneicosanoic acid	C <sub>21</sub> H <sub>42</sub> O <sub>2</sub>	326				0.2	0.1	
Docosanoic acid	C <sub>22</sub> H <sub>44</sub> O <sub>2</sub>	340				1.0	0.7	
Tricosanoic acid	C <sub>23</sub> H <sub>46</sub> O <sub>2</sub>	354				0.3	0.2	
Tetracosanoic acid	C <sub>24</sub> H <sub>48</sub> O <sub>2</sub>	368				1.2	1.0	
Pentacosanoic acid	C <sub>25</sub> H <sub>50</sub> O <sub>2</sub>	382				0.2	0.14	
Hexacosanoic acid	C <sub>26</sub> H <sub>52</sub> O <sub>2</sub>	396				0.9	0.5	
Heptacosanoic acid	C <sub>27</sub> H <sub>54</sub> O <sub>2</sub>	410				0.1	0.1	
Octacosanoic acid	C <sub>28</sub> H <sub>56</sub> O <sub>2</sub>	424				0.9	0.5	
Nonacosanoic acid	C <sub>29</sub> H <sub>58</sub> O <sub>2</sub>	438				0.1	0.05	
Triacontanoic acid	C <sub>30</sub> H <sub>60</sub> O <sub>2</sub>	452				0.7	0.2	
Dotriacontanoic acid	C <sub>32</sub> H <sub>64</sub> O <sub>2</sub>	480				0.2	0.05	
CPI (20-32)			-	-	-	5.9	5.0	-
<b><i>n</i>-Alkenoic acids</b>								
Hexadecenoic acid	C <sub>16</sub> H <sub>30</sub> O <sub>2</sub>	254	1.0	0.9	1.0		1.2	0.8
Octadecenoic acid	C <sub>18</sub> H <sub>34</sub> O <sub>2</sub>	282	0.4	0.6	0.5	0.2	0.65	0.5
<b>Total aliphatic acids</b>			<b>28.2</b>	<b>31.3</b>	<b>26.7</b>	<b>33.2</b>	<b>30.84</b>	<b>22.1</b>
Dehydroabiestic acid	C <sub>20</sub> H <sub>28</sub> O <sub>2</sub>	300				0.01		
Benzoic acid	C <sub>7</sub> H <sub>6</sub> O <sub>2</sub>	122	22	5	16	4.2	6	4
Glyceric acid	C <sub>3</sub> H <sub>6</sub> O <sub>4</sub>	106	0.28	2.0	0.69	1.8	10	2.2

**Dicarboxylic acids**

Succinic acid	C <sub>4</sub> H <sub>6</sub> O <sub>4</sub>	118	0.90	2.1	2.3	3.5	9.6	17.3
Glutaric acid	C <sub>5</sub> H <sub>8</sub> O <sub>4</sub>	132	0.04	0.2	1.07	0.3	12	3.1
Adipic acid	C <sub>6</sub> H <sub>10</sub> O <sub>4</sub>	146	0.11	1.6		0.2	1.2	4.1
Malic acid	C <sub>4</sub> H <sub>6</sub> O <sub>5</sub>	134	0.10	0.8		0.7	5	2.4
1,2-Benzenedicarboxylic acid	C <sub>8</sub> H <sub>6</sub> O <sub>4</sub>	166	0.01	0.13	0.08	0.4	1.5	0.8
1,3-Benzenedicarboxylic acid	C <sub>8</sub> H <sub>6</sub> O <sub>4</sub>	166	0.02	0.20	0.31	0.2	2	0.3
1,4-Benzenedicarboxylic acid	C <sub>8</sub> H <sub>6</sub> O <sub>4</sub>	166	0.01	0.12	0.42	0.2	1.5	0.2
<b>Total</b>			<b>23.19</b>	<b>10.15</b>	<b>20.18</b>	<b>9.5</b>	<b>27.8</b>	<b>32.2</b>

***n*-Alkanols**

Docosanol	C <sub>22</sub> H <sub>46</sub> O	326				0.5	0.4	
Tetracosanol	C <sub>24</sub> H <sub>50</sub> O	354				0.8	0.6	
Hexacosanol	C <sub>26</sub> H <sub>54</sub> O	382				1.8	1.2	
Octacosanol	C <sub>28</sub> H <sub>58</sub> O	410				4.8	1.3	
Triacontanol	C <sub>30</sub> H <sub>62</sub> O	438				2.5	0.7	
Dotriacontanol	C <sub>32</sub> H <sub>66</sub> O	466				0.5	0.3	
Nonacosan-10-ol	C <sub>29</sub> H <sub>60</sub> O	424				1.8	1	
<b>Total</b>						<b>12.7</b>	<b>6.5</b>	

**Anhydrosaccharides**

Levoglucosan	C <sub>6</sub> H <sub>10</sub> O <sub>5</sub>	162	4	5	1.3	2.2	6	2
Mannosan	C <sub>6</sub> H <sub>10</sub> O <sub>5</sub>	162				0.2	1.1	

**Saccharides**

Sucrose	C <sub>12</sub> H <sub>22</sub> O <sub>11</sub>	342	0.9	0.7	1.2	0.3	0.8	0.9
Mycose	C <sub>12</sub> H <sub>22</sub> O <sub>11</sub>	342	0.7	0.9	1.8	2.5	0.5	0.1
Xylitol	C <sub>5</sub> H <sub>12</sub> O <sub>5</sub>	152				9.6	1.5	
Sorbitol	C <sub>6</sub> H <sub>14</sub> O <sub>6</sub>	182		10.4		7.1	5.3	2
α-Glucose	C <sub>6</sub> H <sub>12</sub> O <sub>6</sub>	180	3.5	3		1.3	2	0.5
β-Glucose	C <sub>6</sub> H <sub>12</sub> O <sub>6</sub>	180	5	3		1.7	2.4	0.6
Glycerol	C <sub>3</sub> H <sub>8</sub> O <sub>3</sub>	92	52	41	22	40	44	31
<b>Total</b>			<b>66.1</b>	<b>64</b>	<b>26.3</b>	<b>64.9</b>	<b>63.6</b>	<b>37.1</b>

Urea	CH <sub>4</sub> N <sub>2</sub> O	60			40	25	32	38
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**Phthalates**

Dibutyl phthalate	$C_{16}H_{22}O_4$	278	3	8	3	4.5	6.5	3
Diethyl phthalate	$C_{24}H_{38}O_4$	390	16	20	20	14.3	21.5	9
<b>Total</b>			<b>19</b>	<b>28</b>	<b>23</b>	<b>18.8</b>	<b>28</b>	<b>12</b>

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<sup>a</sup>Some samples contained high concentrations of silicones.

**Table 3. Total compound group yields (ng m<sup>-3</sup>).**

Compound Groups	C-130 Flights					
	RF5	RF10	RF12	RF13	RF14	RF16
<i>Plant Wax</i>						
Alkanes <sup>a</sup>	1.35	1.3	2.2	7.1	2.9	1.2
Alkanoic acids <sup>b</sup>	-	-	-	8.3	10.6	-
Alkanols	-	-	-	12.7	6.5	-
Total	1.35	1.3	2.2	28.1	20.0	1.2
<i>Fossil Fuel</i>						
Alkanes <sup>a</sup>	18.25	19.1	67.1	19.5	13.9	19.6
UCM <sup>c</sup>	92.5	96	336	98	70	98
Total	11	115	403	118	84	118
<i>Marine Alkanoic Acids</i> <sup>b</sup>	28.2	31.3	26.7	24.9	20.2	22.1
<i>Combustion</i>						
PAH	-	-	-	0.27	-	-
Levogluconan	4	5	1.3	2.4	7.1	2
<i>Soil</i>						
Saccharides	62	59	25	62.5	56.5	35.1
<i>Secondary Oxidation</i>						
Dicarboxylic acids + glyceric acid	23.5	12.2	20.9	11.5	37.8	34.4
<i>Others</i>						
Urea	-	-	40	25	32	38
Phthalates	19	28	23	18.8	28	12
<i>Water-soluble compounds</i> <sup>d</sup>	89.5	76.2	87.2	101.4	133.4	72.4
<i>Lipid compounds</i>	159.6	175.6	455	165.2	132	153.3

<sup>a</sup>Plant wax alkanes are calculated as the excess odd homolog-adjacent even homologs average (Simoneit et al., 1991b,c).

<sup>b</sup>Plant wax alkanolic acids are apportioned as the sum of all homologs >C<sub>20</sub> and the equal increment of C<sub>22</sub> or C<sub>24</sub> equivalent to C<sub>18</sub> for those <C<sub>20</sub>.

<sup>c</sup>UCM is estimated based on the U:R of 5 for emissions from vehicle traffic in a Los Angeles tunnel (Fraser et al., 1998).

<sup>d</sup>Includes: Levogluconan, saccharides, dicarboxylic acids, glyceric acid and urea.

**Table 4. Sources and water-soluble compound groups in the total compound mass (TCM) of the extractable organic matter from the airborne aerosols (relative%).**

C-130 Sample	Natural			Urban				Secondary Oxidation Products	Total Water-soluble Compounds <sup>a</sup>
	Plant wax Lipids	Marine Lipids		Fossil Fuel	Plastics	Biomass Burning	Soil Resuspension		
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

<sup>a</sup>Includes urea.

## Figure Legends

Figure 1. Map showing the sampling flight paths.

Figure 2. Flight tracks of the NCAR C-130 aircraft during aerosol sample acquisitions: (a) research flight (RF) 5, (b) RF 10, (c) RF 12, (d) RF 13, (e) RF 14, and (f) RF 16 (red tracks are organic matter filtration).

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, P = phthalates, D = dehydroabiatic acid, nol = nonacosan-10-ol).

Figure 4. Pie diagrams showing the source strengths of organic matter in the C-130 aerosol samples (T=terrestrial and M=marine derived natural background lipids).

Figure 5. Bar plot showing the relative proportions of lipid versus water-soluble compounds in the C-130 aerosol samples.

Appendix I – Chemical Structures Cited.

Fig. 1

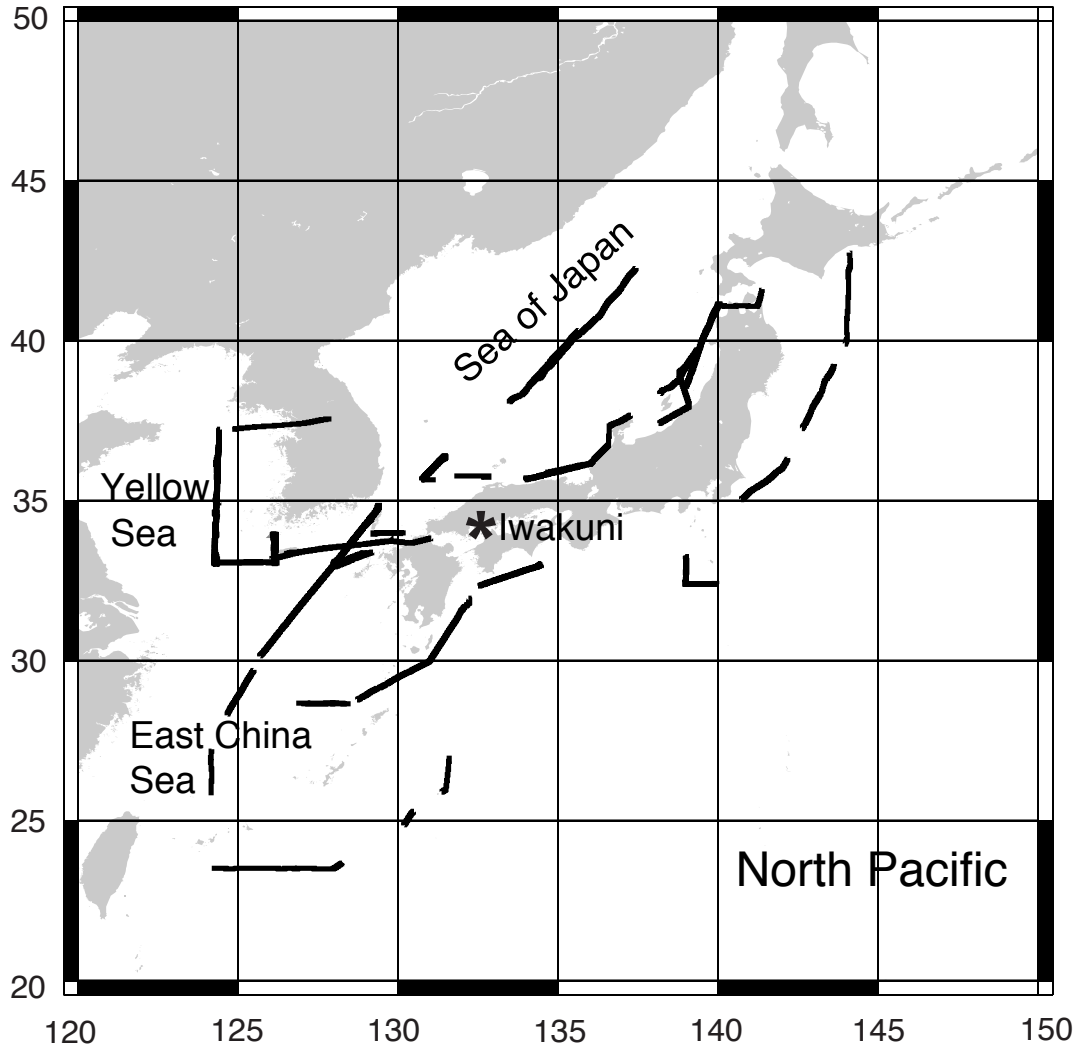


Fig. 2

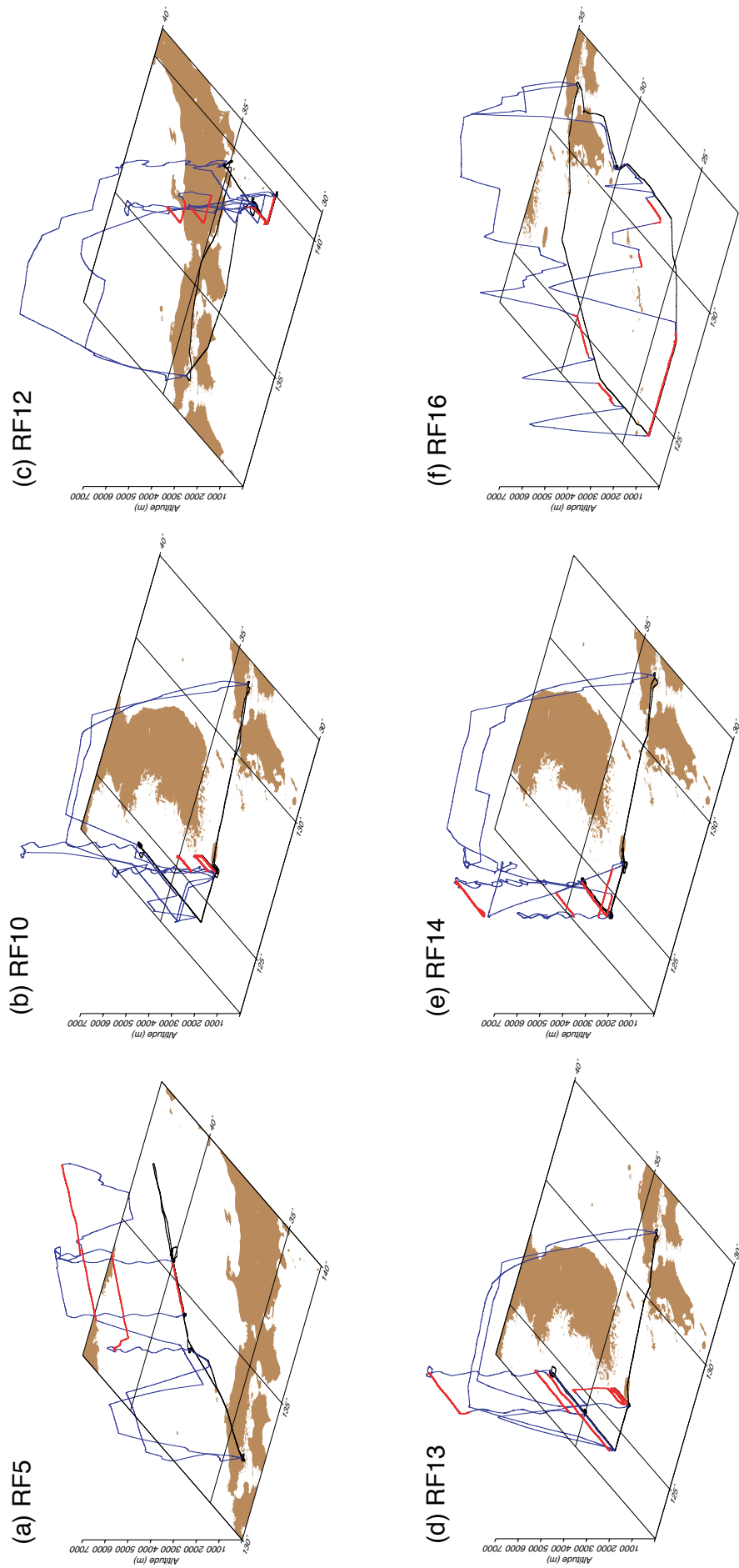
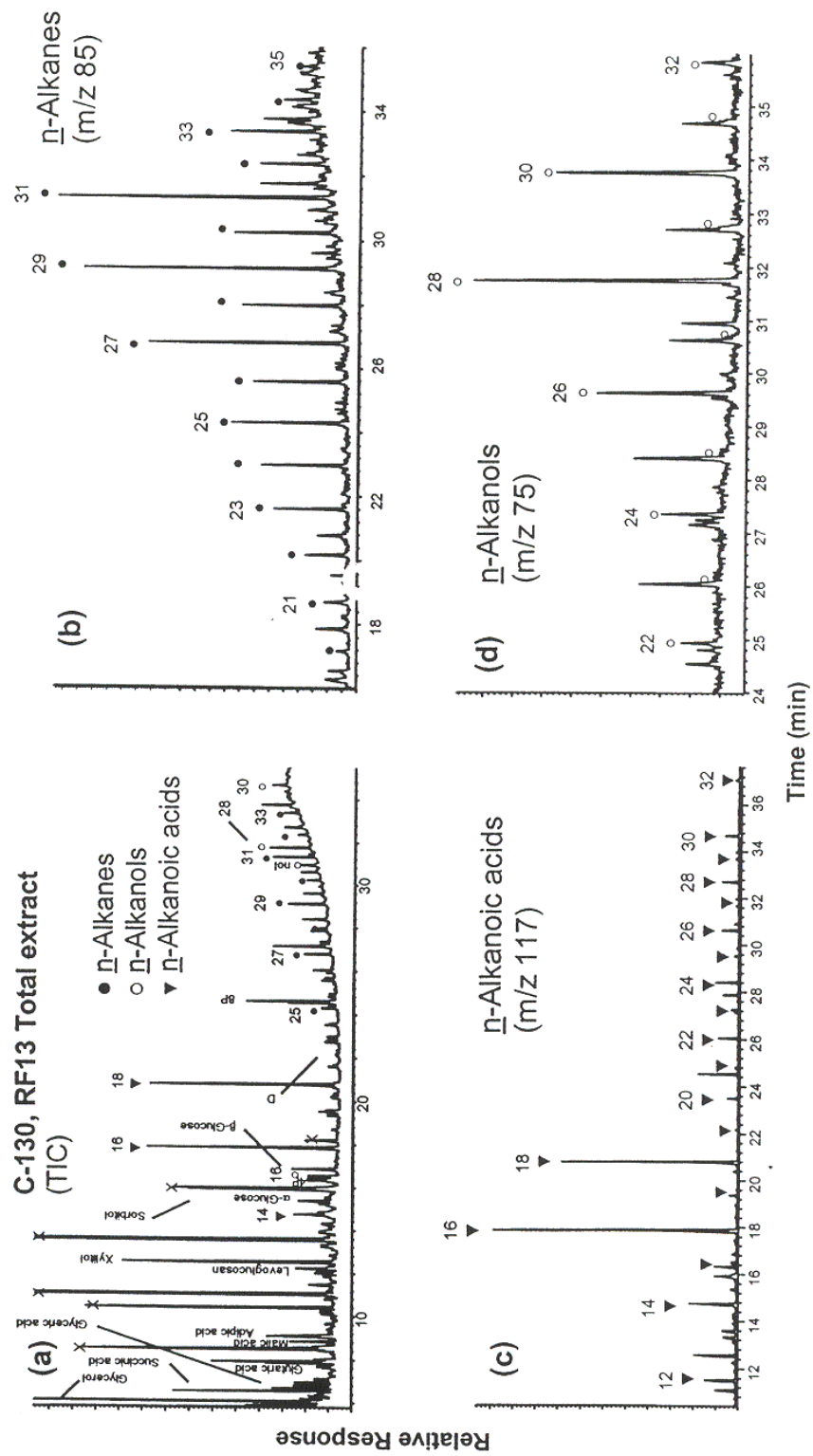


Fig. 3



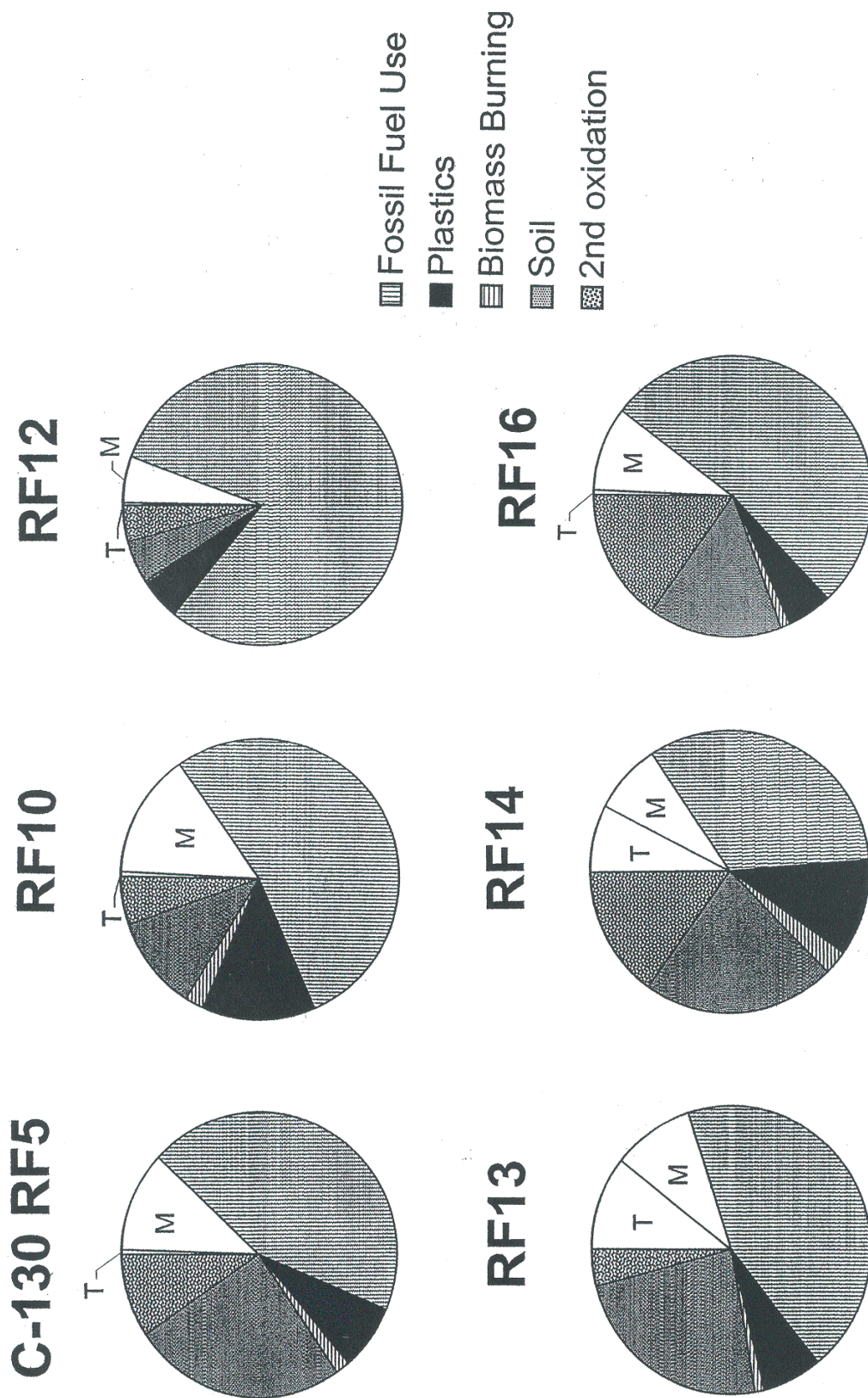
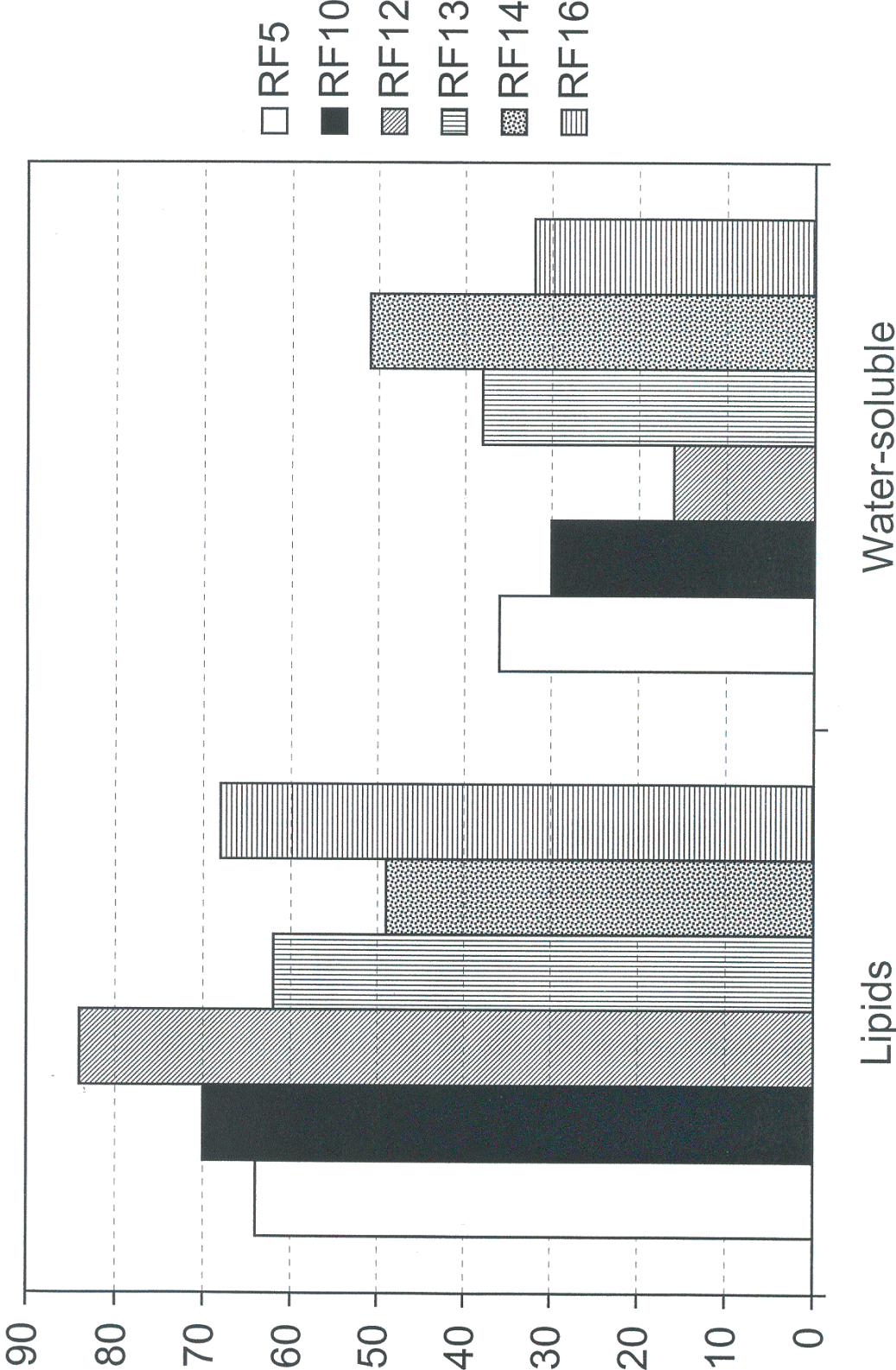


Fig. 5



## Appendix I - Chemical Structures Cited

