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Composition and major sources of organic compounds of aerosol particulate matter sampled during the ACE-Asia campaign

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ABSTRACT

The organic compound tracers of atmospheric particulate matter, as well as organic carbon (OC) and elemental carbon (EC), have been characterized for samples acquired during the ACE-Asia campaign from Gosan, Jeju Island, Korea; Sapporo, Japan, and Chichi-jima Island in the western North Pacific, as well as on the NOAA *R/V Ronald H. Brown*. Total extracts were analyzed by GC-MS to determine both polar and aliphatic compounds. Total particles, organic matter and lipid and saccharide compounds were high during the Asian dust episode (early April 2001) compared to levels at other times. The organic matter can be apportioned to seven emission sources and to significant oxidation producing secondary products during long-range transport. Terrestrial natural background compounds are vascular plant wax lipids derived from direct emission and as part of desert sand dust. Fossil fuel utilization is obvious and derives from petroleum product and coal combustion emissions. Saccharides are a major polar (water-soluble) carbonaceous fraction derived from soil resuspension (agricultural activities). Biomass burning smoke is evident in all samples and seasons. It contributes up to 13% of the total compound mass as water-soluble constituents. Burning of refuse is another source of organic particles. Varying levels of marine-derived lipids are superimposed during aerosol transport over the ocean. Secondary oxidation products increase with increasing transport distance and time. The ACE-Asia aerosols are comprised not only of desert dust, but also of soil dust, smoke from biomass and refuse burning, and emissions from fossil fuels use in urban areas.

Keywords: Levoglucosan, Glucose, Sucrose, Mycose, Saccharides, Atmospheric aerosols, Dicarboxylic acids, PAHs, Hydrocarbons, Fatty acids, Alkanols, ACE-Asia

1. INTRODUCTION

Long-range atmospheric transport of continentally derived particulate matter is an active area of research [e.g., Andreae, 1996; Griffin et al., 2002; Prospero, 1996a,b]. Aerosol particles have relevance for radiative forcing of climate and pollution transport [Wilkening et al., 2000]. The direct radiative effect of aerosols is strongly influenced by particle size and composition. Radiative effects of anthropogenic aerosols are relatively large compared to their mass contribution because they are in the size range which is radiatively most active [Seinfeld and Pandis, 1998]. Particles of source emissions from biological organic matter also 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 [e.g., Abas et al., 1995; Crutzen and Andreae, 1990; Levine, 1991, 1996; Simoneit, 2002].

Dust fallout and haze were first described for the North Atlantic Ocean off west Africa [Darwin, 1846; Ehrenberg, 1847] and has been studied most extensively in terms of long-range transport. Initial research was concerned primarily with the inorganic chemical composition and mineralogy of the atmospheric particles [e.g., Aston et al., 1973; Chester et al., 1972; Delany et al., 1967; Glaccum and Prospero, 1980; Parkin et al., 1972; Prospero et al., 1970]. The effects of long-range transport of aerosol particles on direct radiative forcing, on ultimate receptor areas, and on oceanic nutrient deposition are continuing to be investigated [e.g., Artaxo et al., 1994; Charlson et al., 1992; Husar et al., 1997; IPCC, 1994; Li et al., 1996; Moulin et al., 1997; Penner, 1995; Penner et al., 1994; Prospero et al., 1996; Sokolik and Toon, 1996; Swap et al., 1992; Tegen et al., 1996]. Another major concern is the potential detrimental human health effects of fine aerosol particulate matter with the associated microbes, spores, and toxic organic compounds [e.g., PAHs, Abelson, 1998; Hannigan et al., 1998; Prospero, 2001]. The primary study area has been the North Atlantic Ocean and the export of Saharan dust over the ocean has been estimated to be about 10^9 ton/year [D'Almeida, 1986]. But the Indian and northwestern Pacific Oceans are currently also active research areas under the auspices of the Aerosol Characterization Experiments (ACE) projects.

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 $2x10^7$ ton/year [Buat-Ménard et al., 1989]. Most of this carbon is 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., Gagosian and Peltzer, 1986; Kawamura and Gagosian, 1990; Kawamura and Usukura, 1993; Kawamura et al., 2004; Prospero, 1996 b, 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 SO_x, NO_x). Thus, molecular characterization and provision of mass balance closure of the initial and downwind aerosols continues to be of interest.

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] 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 290m tower, downwind from the sources [Simoneit et al., 1991b]. Here we present a general assessment of the organic tracer composition of aerosols from ground stations at Gosan (also Kosan) on Jeju (also Cheju) Island, Korea, in Sapporo on Hokkaido Island, Japan, and in Chichi-jima Island in the western North pacific, as well as from the North Pacific on the *R/V Ronald H. Brown* during the ACE-Asia project. One-year observation at Gosan site (Kawamura et al., 2004) and 4-year observation at Chichi-jima (Mochida et al., 2003) for water-soluble dicarboxylic acids (C_2-C_{10}) in aerosols have been reported elsewhere.

2. MATERIALS AND METHODS

2.1. Samples

Atmospheric particles were collected on quartz fiber filters using high-volume air samplers during the ACE-Asia campaign in 2001 spring to autumn at island sites: Gosan on Cheju (Jeju) Island (Korea), and Sapporo on Hokkaido Island (Figure 1). Aerosol samples from Chichi-jima (Japan) were collected in 1990 to 1991. Marine aerosol samples were also collected over the western North Pacific, the East China Sea and the Sea of Japan during the cruise of the *R/V Ronald H. Brown* in the same time period (Figure 1). Aliquots of filters were prepared for high molecular weight organic compound characterization, organic carbon analyses, and dicarboxylic acid analyses.

2.2. Extraction and Derivatization

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 wool packed in a Pasteur pipette, and concentrated by use of a rotary evaporator and then under blowdown with dry nitrogen gas. Aliquots of the total extracts were reacted with N, O-bis-(trimethylsilyl)trifluoroacetamide (BSTFA) with 1% trimethylsilyl chloride and pyridine for 3 h at 70°C to derive the trimethylsilyl derivatives of carboxylic acids, alcohols and sugars. 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 DB-5 (30m x 0.25 mm i.d., 0.25μm 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. Recoveries of levoglucosan, glucose, sucrose, palmitic acid, and stearic acid were better than 80%.

2.4. OC and EC measurements

The amounts of organic carbon (OC) and elemental carbon (EC) on the filter punches (2 cm^2) were determined using a thermal-optical OC/EC analyzer from Sunset Laboratory (Tigard, OR). OC was evolved under a stream of ultrahigh purity He while heating the sample in four temperature steps to 870°C. To evolve EC and pyrolyzed OC, the sample was first cooled to 550°C and then heated under a mixture of 2 % O_2 + 98 % He in six temperature steps to a final temperature of 900°C. The transmittance through the filter was monitored with a He-Ne laser throughout the analysis to correct for the pyrolytic conversion of OC to EC.

3. RESULTS AND DISCUSSION

The samples for this study, locations, and ambient conditions are summarized in Table 1 and the general locations are shown in Figure 1. Ground stations were located in Gosan, Sapporo and Chiji-jima. Marine aerosols over the North Pacific (see locations 1-10 in Figure 1) were sampled during the cruise of the *R/V Ronald H. Brown.* The sampling periods covered the ACE-Asia dust event in April 2001 and extended to fall of 2001 and winter of 2002 for the stations in Sapporo. The concentrations of the organic compounds in the samples considered here are listed and categorized as classes in Table 2 for the ground stations and in Table 3 for the samples onboard the *R/V Ronald H. Brown*. Typical examples of organic compositions of aerosol particles are discussed and shown in the following text and figures.

3.1. Ground Stations: Gosan (Kosan) and Sapporo

3.1.1. General results

The total suspended particles (TSP) in the atmosphere during the dust period at the land stations ranged from 142–440 μg m^{-3}, compared to 31–120 μg m^{-3} during other times (Table 1). The TSP for the shipboard samples varied from 23–270 μg m^{-3} with low values far from continental outflow (Table 1). The Chichi-jima samples had high TSP during April and September. The organic carbon (OC) contents were high during the dust episode (3.9–14 μg m^{-3} for the land stations and 5–12 μg m^{-3} for the shipboard samples, Table 1) and lower during other times. Elemental carbon (EC or black C) increased only slightly during the dust event and ranged from 0.65–3.2 μg m^{-3} for the land stations, from 0.53–2.6 μg m^{-3} for samples taken on the mobile platform and 0.01–0.10 μg m^{-3} for background marine samples (Table 1). OC is several times more abundant than EC (OC/EC ratios: 2-9), except for few samples collected in urban Sapporo and Chichi-jima (ratios: 0.7-0.8). The OC/EC ratios obtained at Gosan site (av. 6.0) are higher than those of Sapporo (av. 3.5). In contrast, the ratios obtained onboard *R/V Ron H. Brown* (av. 4.9) are consistent with those (av. 3.7) reported for offshore of Japan (Lim et al., 2003).

The major resolved components of the total aerosol extracts are water-soluble saccharides and dicarboxylic acids with minor hydrophobic lipids from higher plant waxes and fossil fuel sources (*n*-alkanes, *n*-alkanols, *n*-alkanoic acids) (e.g., Figures 2a; 3a; 4a).

3.1.2. Water-soluble compounds

The saccharides are comprised of levoglucosan (1,6-anhydro-β-D-glucopyranose, I, chemical structures are given in Appendix I), with minor mannosan (II), galactosan (III), and 1,6-anhydro-β-D-glucofuranose (levoglucosan isomer, IV), and primary saccharides consisting of α -and β-glucose (V), inositols (VI), sucrose (VII) and mycose (trehalose VIII). In addition there is the series of saccharide polyols (alditols, i.e., reduced sugars) ranging from sorbitol (D-glucitol, IX), xylitol (X), to glycerol (XI). Minor amounts of other monosaccharides (e.g., arabinose, α- and β-fructose, galactofuranose) are found in some samples. Levoglucosan, with mannosan, galactosan, and 1,6-anhydro-β-D-glucofuranose (I-IV) are the key tracers for smoke particulate matter from burning of biomass [i.e., thermal alteration products from cellulose, Simoneit et al., 1999] and have concentrations from 7 to 78 ng m^{-3} (Table 2). 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]. The primary saccharides (e.g., glucose, sucrose, trehalose, etc.) and the saccharide polyols (e.g., sorbitol, xylitol, etc.) have concentrations ranging from 14 to 574 ng m^{-3} (Table 2) and are candidate tracers for surface soil dust. 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 aerosols. 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 aromatic acids (e.g., benzoic, benzenedicarboxylic acids) [Kawamura and Sakaguchi, 1999; Mochida et al., 2003]. The dicarboxylic acids detected in these total extracts are malonic, succinic, glutaric, and adipic acids (C_3-C_6) and the aromatic acids (e.g., 1,2-, 1,3- and 1,4-benzenedicarboxylic acids), with concentration ranges from 8 to 302 ng m^{-3} (Table 2). Malonic and succinic acids are somewhat depleted due to the volatility of the trimethylsilyl esters used in this method compared to the data from the direct analyses, where the C_2-C_6 dicarboxylic acids were determined by separate specific analyses [e.g., Mochida et al., 2003; Kawamura et al., 2004]. Some aerosol samples also contain hydroxydicarboxylic acids, mainly malic, and hydroxyglutaric acids (C_4-C_5), which may be secondary oxidation products from the saccharides. Thus, the saccharides and dicarboxylic acids comprise the major water-soluble organic compounds of these aerosols.

3.1.3. Aliphatic lipids

The lipid components, secondary in concentration in these aerosols (Table 2), are derived from terrestrial plant waxes. 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 (Figures 2–3). The *n*-alkanes typically range from C_{19} to C_{35} and in some cases (especially in Sapporo) to C_{39+}, with a strong odd carbon number predominance (carbon preference index, CPI, ranges from 1.2 to 6.0) and carbon number maxima (C_{max}) at C_{29} or C_{31}. Volatile compound blow-off from the filters over the 1–2 day 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 traffic) are present as *n*-alkanes $>C_{21}$ with no carbon number predominance and traces of biomarkers (hopanes and steranes) [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 from the total alkanes [Simoneit et al., 1991a] and the wax alkane concentrations are listed in Table 2 and plotted in a separate figure as discussed below.

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 range 1.9 to 2.6 for C_{20}-C_{32}, Table 2) (Figures 2b,f and 3b). There are only traces of alkenoic acids (e.g., $C_{18:1}$) indicating that the organic matter of these aerosols is aged and has been transported over significant distances. There is also no palmitoleic acid ($C_{16:1}$) detectable as it is found in the marine aerosols discussed below. 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 only indicate a biogenic input. The *n*-alkanoic acids from C_{20} to C_{32} are interpreted to derive from higher plant waxes and they have very similar distributions of $C_{22} \approx C_{24} > C_{26} < C_{28}$ for most of the ground station samples during the same time interval (cf. Figures 2b,f and 3b). This distribution changes later in the season (e.g., Figure 2).

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 and at C_{26} afterwards (Figures 2d,h and 3d). All aerosol samples contain *n*-nonacosan-10-ol (XII) at unusually high levels (Table 2). The normal alkanols indicate an input of plant waxes and noncosan-10-ol indicates an input of waxes from forests consisting of both soft and hardwoods [Oros and Simoneit, 2001a,b]. Some of these alkanols may also be injected into the atmosphere by smoke from biomas burning [Oros and Simoneit, 2001a,b; Simoneit, 2002], which is supported by the presence of β-sitosterol (XIII) and in some samples lupadienol (XIV) (Table 2). The latter lupadienol is an altered natural product and its precursor and β-sitosterol occur as internal lipids, which can become airborne by volatilization during burning [Simoneit, 2002]. Cholesterol (XV) is found in most of these samples (Table 2) and indicates an input from marine (algal) sources and possibly from emissions from cooking in urban areas [e.g., Rogge et al., 1991; Schauer et al., 1999; Simoneit et al., 1983].

3.1.4. Biomass burning tracers

Biomass burning injects numerous organic compounds from the fuels directly into the atmosphere and based on the major tracers detected both the process and the types of fuels can be assessed [Simoneit, 2002]. The major markers for burning of anything that contains cellulose are levoglucosan (I) with lesser amounts of the other anhydrosaccharides (II-IV) mentioned above. Levoglucosan is found in all aerosol samples analyzed here (range 0.2–74 ng m^{-3}, Table 2), often as the major organic compound, and indicates that biomass burning is impacting the whole Western Pacific region.

The secondary tracers for biomass burning are various natural product compounds or their derivatives [Simoneit, 2002]. β-Sitosterol (XIII) is a directly emitted phytosterol present in all vegetation and is thus not fuel specific but indicates biomass burning. High concentrations of β-sitosterol (XIII) were reported in the marine aerosols from the western North Pacific during 1991 El Ninõ forest fire event occurred in Indonesia and other Southeast Asian countries (Kawamura et al., 2003). Lupadienol (XIV) is indicative for burning of hardwood. Another secondary tracer from biomass burning emissions is dehydroabietic acid (XVI), the key marker for smoke from burning of conifer wood [Standley and Simoneit, 1994; Oros and Simoneit, 2001a; Rogge et al., 1998; Simoneit, 1999]. Dehydroabietic acid is present as a minor component in these samples (Table 2), indicating significant softwood burning during winter-spring in the aerosol source regions. Lignin is the other major biopolymer of wood and upon burning yields phenolic tracers in the smoke characteristic of the fuel type [Hawthorne et al., 1988; Simoneit et al., 1993, 1996]. The major tracers for lignin burning products in oxidized aerosols are 4-hydroxybenzoic acid (XVII) indicative of grasses and other non-woody vegetation, vanillic acid (XVIII) indicative of mainly softwood but also hardwood, and syringic acid (XIX) indicative of hardwood [Simoneit et al., 1993]. These three phenolic compounds are found in most of the aerosol samples analyzed here (Table 2) and the concentrations are similar for Gosan and Sapporo indicating an upwind source on the Asian continent. These compounds are also present in the marine aerosol samples taken near land (Table 3). The predominance of 4-hydroxybenzoic acid in all cases supports the interpretation that the biomass fuels being burned are primarily grasses (e.g., dung, agricultural refuse, bamboo, etc.) [e.g., Sheesley et al., 2003].

3.1.5. PAHs

Examples of typical PAH compositions and confirming biomarkers are shown in Figure 4. The PAHs range from phenanthrene to coronene (Figure 4a, b) and their total concentrations are significant, ranging from 0.005 to 7.8 ng m^{-3} in Gosan and 1.7 to 19.3 ng m^{-3} in Sapporo (Table 2). Their distribution patterns are similar during the dust event, indicating common sources. 1,3,5-Triphenylbenzene (XX) is present at significant concentrations indicating a major input source of PAHs from incineration of refuse (Fu et al., 1997). 1,3,5-Triphenylbenzene has been detected in particles from solid waste incinerators and from burning of plastics (Tong et al., 1984; Karasek and Tong, 1985). It may be a useful urban tracer. Similar PAH distributions have also been reported for various cities in China [Simoneit et al., 1991b; Fu et al., 1997; Okuda et al., 2002] with total PAH concentrations of 8–450 ng m^{-3}. Retene from burning conifer wood is not detectable in any of these samples. PAHs are of health concerns because of their carcinogenicity, genotoxicity, and endocrine disrupting potential. The benzopyrenes and cyclopenta[c,d]pyrene are found at high concentrations in these aerosols and the ratio of benzo[a]pyrene to benzo[e]pyrene (Bap/Bep) ranges from 0.4 – 0.8 which is high (Table 2). The Bap/Bep values for samples from urban areas of China ranged from 0.1 to 1.0 [Simoneit et al., 1991b; Okuda et al., 2002]. Benzo[a]pyrene has a shorter atmospheric lifetime than benzo[e]pyrene due to photochemical degradation [Lane, 1989]. Thus, the high concentrations of benzo[a]pyrene indicate that these PAHs reside in tar particulate matter of coal smoke emissions (small fire type burning) and are thus protected from secondary reactions during transport [Oros and Simoneit, 2000].

The urban aerosols of China also contained equivalent concentrations of oxy-PAHs as the PAHs, both primary emissions (e.g., benzo[a]fluoren-11-one), as well as secondary oxidation products from the PAHs (e.g., 9,10-anthraquinone) [Simoneit et al., 1991b]. Oxy-PAHs are not significant components of these samples, probably reflecting their greater reactivity toward further atmospheric oxidation upon emission to for example the benzenedicarboxylic acids and/or entrapment in coal tar emissions.

3.1.6. Biomarker confirmations

The compositions of the hopane biomarkers characteristic of petroleum utilization are shown for two examples in Figure 4c,d. They are present at trace levels (Table 2) and are thus utilized for confirmation of urban traffic emissions. The hopanes consist of only the 17α(H) series (XXI), ranging from C_{27} to C_{35} (no C_{28}) with the fully mature configuration for the homologs $>C_{30}$. Gammacerane (XXII) is present and these compositions match those reported earlier for some urban areas of China [Simoneit et al., 1991b]. Steranes and carotane detected in urban aerosols of China during the 1980's were below the detection limit in these samples.

Examples of key ion traces (m/z 204 + 361) for GC-MS confirmation of the glucose and sucrose anomers are shown in Figure 4e,f. Although these compounds are dominant resolved peaks in most of the total ion current traces (e.g., Figures 2a,e and 3a), such key ion plots allow confirmation of trace levels of an anomer. The interconversion of sucrose to its suspected aldol anomer during atmospheric transport is still under investigation.

3.1.7. Wax alkanes

The *n*-alkane distributions attributable to vascular plant wax in these samples have been calculated as described [Table 2, Simoneit et al., 1991a] and plotted (Figure 5). None of these distributions are the same and even the air parcel over Gosan moving to Sapporo over a two-day period shows slightly different *n*-alkane distributions (e.g., Figure 5a vs. 5e or 5c vs. 5f). This variation may reflect source attributes or additional local input. Nevertheless, the C_{max} at 29 or 31 reflects a more tropical source region [Simoneit, 1977, 1979], confirming the long-range transport of these aerosols from southeast Asia to the sampling locations. A minor contribution of *n*-alkanes from C_{34} to $>C_{37}$, especially for samples from Sapporo (Figure 3c, Table 2), should be pointed out. These long chain alkanes are interpreted to be the high molecular weight distribution of paraffin wax (C_{max} at 28), a petroleum product used for example to coat food containers such as paper milk and juice cartons. The enhanced contents of phthalates in the Sapporo aerosol samples (24–86 ng m^{-3}) compared to those from Gosan (2–23 ng m^{-3}, Table 2) and the possible presence of paraffin wax indicate fugitive emissions from burning of urban refuse [Didyk et al., 2000]. Source tests from such sources have not yet been conducted.

3.2. Ground Station: Chichi-jima

Chichi-jima is located to the southeast of the major dust outflow trajectories from Asia, but is still impacted (Figure 1). The organic compositions of typical aerosol samples taken on the island in 1990 and 1991 during dust episodes are given in Table 2 and an example is shown in Figure 6.

3.2.1. Water-soluble compounds

The dominant compounds are water-soluble saccharides and their derivatives and dicarboxylic acids with minor amounts of plant wax lipids, phthalates, and dehydroabietic acid. The saccharides and derivatives consist of glycerol (XI), glyceric acid, xylitol (X), sorbitol (IX), α- and β- glucose (V), sucrose (VII), and sucrose aldol (VIII), with a lesser amount of levoglucosan (I) (Table 2). Levoglucosan and dehydroabietic acid indicate an input from biomass burning with a significant amount of softwood [Simoneit, 2002]. The saccharides are interpreted to derive from soil dust. The dicarboxylic acids, ranging from succinic to adipic acids (C_4-C_6), and the benzenedicarboxylic acids (Table 2) are oxidation products from other organic compounds, reflecting the long-range transport of the aerosols.

3.2.2. Lipid compounds

The lipid compounds of the example aerosol taken in April 1990 are shown in the key ion plots (Figure 6b–d). The dominant aliphatic homologs are derived from terrestrial plant wax as indicated by the *n*-alkanes, ranging from C_{21} to C_{35} with C_{max} at 29 and CPI=4.8, the *n*-alkanoic acids $>C_{20}$ with C_{max} at 24 and CPI=3.2, and the *n*-alkanols, ranging from C_{22} to C_{30} with C_{max} at 28. Nonacosan-10-ol is also derived from terrestrial plant wax. The wax alkanes have been replotted after subtraction of the even carbon numbered homologs (Figure 5h) and clearly show the C_{max} at 29. The source region for this plant wax is interpreted to be tropical areas based on the high C_{max} of these lipids. Marine derived lipids are minor and consist mainly of *n*-alkanoic acids $>C_{20}$ for this example (Figure 6b). However, during June the terrestrial wax component is small and the marine lipids are major, with a dominance of saccharides (Table 2). In that case the saccharides, especially the alditols (polyols), may also have a marine origin. This is consistent with the seasonal trend of lipid distributions observed for Chichi-jima aerosols (Kawamura et al., 2003), where concentrations of terrestrial lipids are generally high in winter/spring and low in summer/autumn. Petroleum and coal hydrocarbons, PAH and biomarkers, from continental urban areas or marine traffic are not detectable in these aerosol samples. The minor amounts of phthalates may indicate an input from urban areas as for example smoke from garbage burning or contaminants.

3.3. Mobile Station: *R/V Ronald H. Brown*

The organic compound signatures of the aerosols collected during the cruise of the *R/V Ronald H. Brown* from Hawaii to the East Asian region (Figure 1) changed from solely marine to dominantly terrestrial sources. The data are summarized in Table 3 and typical examples of GC-MS data are illustrated in Figures 7.

3.3.1. Marine aerosol composition

Samples RB1-3 have mainly marine derived lipid components (2–37 ng m^{-3}) and secondary oxidation products (e.g., dicarboxylic acids) become superimposed for samples closer to land (e.g., samples 5-6). The marine lipids are comprised of *n*-alkanoic acids $<C_{20}$, significant amounts of monounsaturated fatty acids (i.e., hexadecenoic and octadecenoic acids, Table 3), cholesterol (XVI), and glycerol (XI). The high amount of glycerol may reflect hydrolysis of fatty acid glycerides from the marine lipid pool in the sea surface slick, which can become part of the aerosol with the other lipid markers by wind activity [Simoneit et al., 1991c]. The dicarboxylic acids (1–134 ng m^{-3}, Table 3, e.g., Figure 7a) become more prevalent in the samples nearer land and consist of succinic to adipic acids and the benzenedicarboxylic acids.

3.3.2. Terrestrial tracers

Levoglucosan (I) is detectable in all samples (0.2–40 ng m^{-3}), indicating biomass burning smoke is spread over the western North Pacific. Dehydroabietic acid (XI), the secondary biomass burning tracer specific for conifer wood, is not detectable. The terrestrial lipids are found as minor components in aerosols taken near land (mainly samples 6–10, 3–27 ng m^{-3}). They consist of *n*-alkanes ranging from C_{21} to C_{33}, with C_{max} at 29 or 31 and CPI values from 1.1 to 2.1, *n*-alkanoic acids >C_{20} (CPI from 4.5-11.0) and C_{max} at 24, and *n*-alkanols from C_{22} to C_{30}, with C_{max} at 26, as well as nonacosan-10-ol (XIII) (Table 3, Figure 7). Minor amounts of sterols and triterpenoids are also found in some samples (Table 3). The plant wax alkanes are replotted after subtraction of the even carbon numbered homologs for two examples (Figure 5i,j), thus showing the C_{max} more clearly. These homolog distributions and C_{max} confirm the terrestrial plant wax origin and a semitropical to temperate source region of the minor aliphatic components, with their transport from Asia to the ocean. At sea level these continental aerosols are admixed and even overwhelmed with marine derived organic matter from the sea slick. The *n*-alkane distributions of the marine samples (e.g., Figure 7c) reflect traces of petroleum-derived hydrocarbons with no carbon number preference (CPI=1.1) and C_{max} at 26. Petroleum biomarkers were not detectable but the alkane distribution can be interpreted to be derived from diesel exhaust particulate matter (i.e., marine ship traffic) which had become resuspended from the sea surface by wind action and had the volatiles removed by evaporation or once on the filter by blow-off [Simoneit et al., 1991c]. The *n*-alkanoic acids derived from marine emissions are mainly composed of the homologs <C_{20} (C_{max}=16) with high contents of alkenoic acids, especially palmitoleic acid ($C_{16:1}$) common in phytoplankton (cf. Figure 7b). The saccharides and phthalates (Table 3) are found in most samples, but PAHs or other terrestrial tracers from urban anthropogenic activity are not detectable. The sources of the sometimes dominant saccharides are not clear at this time, they may have both a terrigenous and marine origin.

3.4. Emission Sources

The organic compound groups characterized and quantitated for these aerosols can be apportioned to seven emission sources and to significant oxidation reactions during long-range transport, which produces secondary products. This apportionment is based on the extensive emission profile studies of organic compound compositions carried out for major stationary and fugitive sources in Los Angeles, CA and other regions (e.g., Rogge et al., 1991, 1998; Fraser et al., 1998; Oros and Simoneit, 2000, 2001a, b; Simoneit, 1989, 1999, 2002). The source strengths in terms of organic compound tracers can thus be modeled and apportioned (Schauer et al., 1996). These source groups are summarized in Table 4 and plotted for selected samples in Figure 8. The terrestrial natural background compounds are the vascular plant wax lipids and they range from 4.6 to 24% of the total identified compound mass (TCM) during the dust season (Table 5, especially G5, S2033, and C181 in Figure 8b-d). The emissions from fossil fuel utilization range from 5–29% at Gosan and 19–58% of TCM at Sapporo for petroleum markers and 0.0–2.6% of TCM at Gosan and 0.3-2.9% at Sapporo for coal derived PAHs (both coal and petroleum are summed in Figure 8). Thus, there is an additional input of petroleum hydrocarbons from the

Sapporo urban area to the organic matter transported over long range from Asia (Figure 8c). The high PAH levels compared to the petroleum derived alkanes and unresolved complex mixture (UCM) of branched and cyclic hydrocarbonds indicate coal as the major source for PAH [Oros and Simoneit, 2000]. Emissions from possible burning of refuse range from about 1 to 24% of TCM and probably reflect the influences of local urban and onboard ship emissions (e.g., G5, S2033 and RB7 in Table 5 and Figure 8b,c,f).

Biomass burning, as reflected by wood smoke markers accounts for 5 to 12% of TCM during the major dust period (G1–G20, S2033, S2039, and RB4-8 in Table 5, and Figure 8a,c,e). The value rises to 13% during the winter in Sapporo and has been reported at much higher levels in other regions [e.g., Schauer et al., 2002]. During summer the wood smoke impact decreases (<3% of TCM). The saccharides comprise from 13 to 26% of TCM and have isolated higher values over the ocean (up to 63%, Table 5, e.g., G1–G30, S2039, S2443, RB1–3 and RB8). These compounds are interpreted to represent viable biomass as in soil and possibly in marine particulate matter. Saccharides have been characterized in urban aerosols, which contain entrained soil dust [e.g., Didyk et al., 2000] and in agricultural soils [Simoneit et al., 2004]. Thus, saccharides 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 ($<C_{20}$, including $C_{16:1}$ and $C_{18:1}$) content, which ranges from 1 to 12% of TCM for the Gosan and Sapporo locations and from 1–45% of TCM for the samples taken onboard the *R/V Ronald H. Brown* (see Table 5 and Figure 8f). Obviously, the marine samples contain more autochthonous components such as phytoplankton lipids, but the long range continental signature is still dominant.

The secondary products derived from oxidative reactions of the organic compounds during transport consist mainly of aliphatic dicarboxylic acids, dihydroxy carboxylic (glyceric) acid and aromatic acids. This compound group varies from 3–40% of TCM for the aerosols from the land stations (e.g., S2033 vs. G1 and G5, Figure 8a,b,c) and from 6–66% of TCM for those taken onboard the ship (e.g., RB2, RB5, and RB6, see Table 5 and Figure 8a,b,e). The remote marine atmosphere has a higher proportion of secondary oxidation products than the atmosphere downwind from Asia at Gosan and Sapporo. This has been demonstrated earlier on prior cruise 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 as also supported by low concentrations of unsaturated fatty acids.

3.5. 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; Kawamura et al., 2004]. 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, anhydrosaccharides, and secondary oxidation products such as aliphatic dicarboxylic, dihydroxy carboxylic, and aromatic acids. The sums of these water-soluble compounds range from 14 to 89% of TCM for all the samples analyzed here and are about 40–70% of TCM (Table 5) during the dust episode at the ground stations. Bar graphs showing the total lipid versus water-soluble compounds are illustrated in Figure 9 for all samples analyzed. 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., 2004] make essentially the total compound mass soluble in water (e.g., G1 and G3 in Figure 9). The Gosan, Chichi-jima, and *R/V Ronald H. Brown* (RB2, RB5, and RB6) have a greater proportion of water-soluble organic matter due to the long range transport of the aerosols. The samples from Sapporo and RB1, RB3, RB4, and RB7 have a higher proportion of lipids due to additional input from urban emissions being superimposed on the aerosol from the Asian continent. This is quite different from other urban aerosols such as Los Angeles, CA or Santiago, Chile 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].

3.6. Contributions of each compound classes to aerosol OC: mass closure for organics

Table 6 presents contribution of each compound class to total OC in aerosols collected from Gosan, Sapporo, Chichi-jima and *R/V Ron H. Brown*. The results for total n-alkanes show that higher relative abundances (> 1 %) of OC were found in the aerosols from ground stations at Gosan and Sapporo, whereas those of Chichi-jima and Ron Brown samples are less than 0.6 %. UCM hydrocarbons of fossil fuel combustion origin are found to comprise 0.4-6.6 % of aerosol OC, where the highest value (6.6%) was obtained in Sapporo and the lowest value (0.4%) was in Chichi-jima. The values from Sapporo are similar with those (1-6%, av. 4%) reported for the aerosols from Tokyo (Kawamura et al., 1995). This indicates that local emissions of UCM hydrocarbons from fossil fuel combustion sources largely contribute to OC in the urban atmosphere. Relatively high UCM/OC ratios were also obtained at Gosan and the offshore of Japanese Islands during the cruise of *R/V Ron Brown*. This indicates that anthropogenic combustion sources also affect on the organic composition of remote aerosols. Other class of compounds that importantly contribute to OC includes biogenic fatty acids (0.2-1.6 %), photochemically-derived diacids (0.003-1.7%), n-alcohols (0.04-1.5 %), anhydrosaccharides

(0.003-0.78 %), saccharides (0.1-2.5 %) and phthalates (0.02-4.1 %). However, sum of these organic compounds determined account for only less than 16 % of OC.

Although oxalic acid that is the most abundant diacid species was not determined in this study and malonic acid is possibly underestimated as stated above, optimal contribution of these diacids to OC should be less than 10 % [Kawamura and Ikushima, 1993; Kawamura and Sakaguchi, 1999]. With the consideration of oxalic and malonic acids, the maximum total amounts of identifiable organic compounds are at most 26 % of OC. This indicates that major portion (three quarters) of organic aerosols studied in the Asian Pacific region is unknown in terms of chemical structures. They may include amino acids, peptides, amines, and fulvic and humic acids, etc. Our organic analytical study demonstrates that soil-derived organic materials are sometimes detected as major species in aerosols (see Figure 8a,d), suggesting that humic substances may substantially be present in atmospheric particles and significantly contribute to the aerosol OC. Humic and fulvic acids are one of the most abundant organic fractions of soil organic matter as well as humin (kerogen) [e.g., Schnitzer and Khan, 1972; Stevenson, 1982]. Thus, humic materials may be emitted to the atmosphere during strong winds, followed by uplift process of air mass and be subjected to long-range atmospheric transport. It is of interest to note that organic carbon is enriched in fine aerosol fraction when soil particles are emitted to the air under a strong wind condition [Zenchelsky et al., 1976].

Matsumoto et al. [2001] isolated fatty acids collected in June 1999 from Sapporo and conducted compound-specific radiocarbon analysis of individual fatty acids using accelerator mass spectrometry (AMS). They found that ^{14}C ages of C_{24} plus C_{26} fatty acids to be 5860 (±200) years, suggesting that these terrestrial fatty acids had been stored in geochemical reservoirs such as soils at least for hundreds to thousands of years and then emitted to the atmosphere followed by long-range atmospheric transport from the Asian continent. They also analyzed the humic-like substances for ^{14}C, which were left in the acidified fraction of the CH_2Cl_2/methanol extracts of the aerosols after the fatty acids were removed. The humic-like substances are insoluble in acidified solution and non-extractable with CH_2Cl_2. They were isolated, converted to CO_2, and then subjected to AMS measurement. The humic-like substances showed an age of ca. 5000 years, again suggesting that aerosol particles contain fairly old carbons probably derived from aged soil organic matter transported long distances from the Asian continent.

4. CONCLUSIONS

The primary inputs of organic compounds to ACE-Asia aerosols are: (1) natural emissions of terrestrial plant wax and marine lipids based on the alkanoic acid, alkanol, and alkane distributions; (2) smoke from biomass burning (mainly non-conifer fuels) using levoglucosan and dehydroabietic acid as the tracers; (3) soil resuspension due to spring agricultural activity as inferred from major primary and reduced sugars (e.g., sucrose, glucose, sorbitol); and (4) urban/industrial emissions from fossil fuel use based on the biomarkers in this case from coal and petroleum. Significant secondary products derive from the oxidation of atmospheric organic matter during long-range transport and consist of dicarboxylic acids, aromatic acids, and hydroxy carboxylic acids (e.g., glyceric acid).

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. The organic source apportionment needs to be coupled with the inorganic/mineralogical composition data to provide the appropriate weighting of desert dust versus soil dust.

Comparison of each compound classes to OC further supported the source strength and source apportionment obtained by the molecular signature and concentrations. Although measured compound classes in the aerosols explained up to 16% of OC, a mass closure approach for organic compounds demonstrated that we still have a significant fraction (>80% of OC) whose chemical structures are unknown. Our molecular analyses of organic aerosols suggest that this fraction may contain soil-derived organic geopolymers such as humic and fulvic acids that are abundantly present in soil reservoirs and can be emitted to the atmosphere by wind motions.

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Figure Legends

Figure 1. Map showing the sampling locations and major deserts of China. See Table 1 for the sampling dates.

Figure 2. Salient features of the GC-MS data for total extracts of aerosol samples from Gosan: (a-d) G3 and (e-h) G20 as the trimethylsilyl derivatives. (a,e) total ion current traces, (b,f) m/z 117 key ion plots for *n*-alkanoic acids, (c,g) m/z 85 key ion plots for *n*-alkanes, and (d,h) m/z 75 key ion plots for *n*-alkanols (numbers refer to carbon chain length of homologous compounds, •=*n*-alkanes, o=*n*-alkanols, Δ=*n*-alkanoic acids, G=galactosan, M=mannosan, Lf=1,6-anhydro-β-D-glucofuranose, FA=alkanoic acid, OH Glutaric=hydroxyglutaric acid, DHA=dehydroabietic acid, 8P=dioctyl phthalate, C=cholesterol, S=β-sitosterol).

Figure 3. Salient features of the GC-MS data for total extracts of aerosol sample (S2033) from Sapporo. (a) Total ion current traces, (b) m/z 117 plots for *n*-alkanoic acids, (c) m/z 85 plots for *n*-alkanes, and (d) m/z 75 plots for *n*-alkanols (numbers and symbols as in Figure 2).

Figure 4. Examples of key ion plots for PAHs and biomarkers: (a,c,e) G4 and (b,d,f) S2033. (a,b) Sums of m/z 178, 202, 226, 228, 252, 276, 300 and 306 for P=phenanthrene, Fl=fluoranthene, Py=pyrene, Bzf=benzo(ghi)fluoranthene, Cppy=cyclopenta(cd)pyrene, BzA=benz(a)anthracene, Chry=chrysene, Bzfl=benzofluoranthenes, Bep=benzo(e)pyrene, Bap=benzo(a)pyrene, Per=perylene, Ph_3B_2=1,3,5,-triphenylbenzene, Infl=indeno(1,2,3-cd)fluoranthene, Inpy=indeno(1,2,3-cd)pyrene, Bzper=benzo(ghi)perylene, and Cor=coronene; (c,d) m/z 191 fragmentogram for hopanes (numbers refer to carbon skeleton, G=gammacerane, S and R=configuration at C-22); and (e,f) m/z 204 and 361 fragmentogram for glucose and sucrose isomers.

Figure 5. Distribution plots of the odd carbon number alkanes attributable to plant wax in the aerosols (see Table 1 for the information of samples).

Figure 6. Salient features of the GC-MS data for the total extract of an aerosol sample taken on Chiji-jima in 1990. (a) total ion current trace, (b) m/z 117 plot for *n*-alkanoic acids, (c)

m/z 85 plot for *n*-alkanes, and (d) m/z 75 plot for *n*-alkanols (numbers and symbols as in Figure 2).

Figure 7. Salient features of the GC-MS data for total extracts of marine aerosol samples (RB7) taken onboard the *R/V Ronald H. Brown* from the North Pacific. (a) total ion current trace, (b) m/z 117 plot for *n*-alkanoic acids, (c) m/z 85 plot for *n*-alkanes, and (d) m/z 75 plot for *n*-alkanols (numbers and symbols as in Figure 2).

Figure 8. Pie diagrams showing the source strengths of organic matter in aerosols from (a,b) Gosan, (c) Sapporo, (d) Chichi-jima and (e,f) onboard the *R/V Ronald H. Brown* (see Table 1 for the information of samples).

Figure 9. Bar graphs showing the lipid vs. water-soluble compounds in the aerosols from: (a) Gosan, (b) Sapporo, (c) Chichi-jima island, and (d) onboard the *R/V Ronald H. Brown*. Numbers under the bars mean sample ID (see Table 1).

Appendix I. Chemical Structures Cited.

Table 1. Atmospheric particulate matter samples, dates, locations, total suspended particles, organic carbon, and elemental carbon during the ACE-Asia campaign.

Sample	**Date acquired**	**Location**	**TSP** (μg m^{-3})	**OC** (μg m^{-3})	**EC** (μg m^{-3})	**OC/EC** ratios	**OC** (%)
G1	Apr. 8-9, 2001	Gosan, Jeju Island	110	7.2	0.81	8.9	90
G3	Apr. 10-11, 2001	Gosan, Jeju Island	440	8.6	2.0	4.3	81
G4	Apr. 11-12, 2001	Gosan, Jeju Island	270	8.1	1.4	5.8	85
G5	Apr. 12-13, 2001	Gosan, Jeju Island	170	3.9	1.3	3.0	75
G14	Apr. 21-22, 2001	Gosan, Jeju Island	140	7.5	1.4	5.4	84
G20	Apr. 27-28, 2001	Gosan, Jeju Island	260	6.7	0.8	8.4	89
G30	May 14-17, 2001	Gosan, Jeju Island	120	n.d.	n.d.		
S2033	Apr. 8-10, 2001	Sapporo	230	14	3.2	4.4	81
S2039	Apr. 14-16, 2001	Sapporo	66	6.7	1.7	3.9	80
S2443	June 28-July 5, 2001	Sapporo	40	4	0.65	6.2	86
S2447	Sept. 11-12, 2001	Sapporo	31	1.5	1.9	0.8	44
S2504	Jan. 10-12, 2002	Sapporo	40	4.5	2.1	2.1	68
C181	Apr. 9-12, 1990	Chichi-jima Island	62	1.4	0.52	2.7	73
C188	June 3-6, 1990	Chichi-jima Island	16	0.28	0.41	0.7	41
C221	Sept. 17-20, 1991	Chichi-jima Island	280	0.76	0.04	19.0	95
RB1(2329)[a]	Mar 16-17, 2001	*R/V Ronald H. Brown*	23	0.00	0.01		
RB2(2332)[a]	Mar 18-19, 2001	*R/V Ronald H. Brown*	24	0.49	0.10	4.9	83
RB3(2333)[a]	Mar. 19, 2001	*R/V Ronald H. Brown*	n.d.	n.d.	n.d.		
RB4(2350)[a]	Apr. 2-3, 2001	*R/V Ronald H. Brown*	130	1.4	0.53	2.6	73
RB5(2357)[a]	Apr. 6, 2001	*R/V Ronald H. Brown*	65	3.9	0.56	7.0	87
RB6(2360)[a]	Apr. 7-8, 2001	*R/V Ronald H. Brown*	120	5.2	0.93	5.6	85
RB7(2376)[a]	Apr. 14-15, 2001	*R/V Ronald H. Brown*	60	2.9	0.62	4.7	82
RB8(2384)[a]	Apr. 19-20, 2001	*R/V Ronald H. Brown*	270	12	2.6	4.6	82

[a] Approximate location shown in Figure 1.

TSP was weighed in the lab with air conditioning.

but relative humidity was not controlled.

n.d.= not determined.

Table 2. Organic compounds (ng m^{-3}) identified in aerosol particulate matter sampled at the ground stations during the ACE-Asia campaign and at other times.

Compound	Composition	M.W.	Gosan							Sapporo					Chichi-jima		
			G1	G3	G4	G5	G14	G20	G30	S2033	S2039	S2443	S2447	S2504	C181	C188	C221
n **-Alkanes (total)**																	
Octadecane	$C_{18}H_{38}$	254				0.03			0.1	0.5				0.4			
Nonadecane	$C_{19}H_{40}$	268	0.17	0.38	0.48	0.06			0.2	0.7	0.3			0.7	0.06		
Eicosane	$C_{20}H_{42}$	282	0.19	0.25	0.60	0.12		0.1	0.3	1.1	0.5		0.4	3.5	0.06		
Heneicosane	$C_{21}H_{44}$	296	0.18	0.63	0.96	0.23	1.5	0.6	0.2	2.4	1.2	0.3	0.3	11.3	0.13	0.008	
Docosane	$C_{22}H_{46}$	310	0.13	0.75	1.20	0.29	1.8	0.6	0.2	2.7	1.2	0.2	1.0	14.1	0.1	0.013	0.01
Tricosane	$C_{23}H_{48}$	324	0.35	1.50	2.88	0.87	4.7	1.6	0.8	3.8	2.4	0.6	2.4	11.7	0.26	0.023	0.04
Tetracosane	$C_{24}H_{50}$	338	0.44	1.63	3.00	0.76	6.9	1.4	1	4.4	3.3	0.5	2.8	8.2	0.19	0.025	0.07
Pentacosane	$C_{25}H_{52}$	352	0.79	3.50	4.92	2.04	11.8	3.2	4.2	5.8	4.0	4.0	3.6	6.5	0.64	0.026	0.15
Hexacosane	$C_{26}H_{54}$	366	0.83	2.38	2.88	0.87	10.0	1.9	2.9	5.5	3.0	0.6	2.6	3.5	0.32	0.02	0.22
Heptacosane	$C_{27}H_{56}$	380	1.20	5.00	6.60	4.08	20.6	4.4	7.7	8.2	4.9	10.0	4.0	4.8	1.31	0.031	0.46
Octacosane	$C_{28}H_{58}$	394	0.79	1.88	2.40	0.87	11.8	1.9	2.3	5.6	2.4	1.3	1.8	2.1	0.29	0.02	0.42
Nonacosane	$C_{29}H_{60}$	408	1.50	6.00	7.20	6.70	29.4	5	6.6	8.2	5.0	7.0	3.6	4.2	1.6	0.05	1.1
Triacontane	$C_{30}H_{62}$	422	0.58	1.38	1.68	0.58	4.4	1.3	1.6	5.6	2.0	0.8	1.2	1.6	0.16	0.014	0.37
Hentriacontane	$C_{31}H_{64}$	436	1.20	6.25	6.84	2.04	25.0	3.3	3.1	8.8	4.1	3.7	2.8	3.1	1.38	0.028	1.01
Dotriacontane	$C_{32}H_{66}$	450	0.40	0.75	0.96	0.30	7.1	0.4	0.9	5.1	1.3	0.6	0.8	0.8	0.06	0.013	0.2
Tritriacontane	$C_{33}H_{68}$	464	0.44	1.88	1.92	0.60	10.3	0.9	0.8	10.9	1.5	1.2	1.2	0.7	0.38	0.014	0.31
Tetratriacontane	$C_{34}H_{70}$	478	0.18	0.25	0.29		2.1	0.1	0.4	4.2	0.2	0.2				0.005	0.04
Pentatriacontane	$C_{35}H_{72}$	492	0.17	0.25	0.36		2.9	0.1	0.1	1.8	0.2	0.3			0.03	0.006	0.08
Hexatriacontane	$C_{36}H_{74}$	506	0.07	0.13					0.1	1.3	0.1	0.1					
Heptatriacontane	$C_{37}H_{76}$	520								0.7							
Total			**9.61**	**34.8**	**45.2**	**20.4**	**150**	**25.8**	**33.5**	**87.3**	**37.6**	**31.45**	**28.52**	**77.2**	**6.97**	**0.3**	**4.48**
CPI (21-37)[a]			1.60	2.70	2.40	4.40	2.40	2.50	2.50	1.40	1.70	6.00	1.70	1.20	4.80	1.80	2.30
Plant Wax Alkanes[b]																	
Tricosane	$C_{23}H_{48}$	324		0.31	0.84	0.35	0.3	0.6	0.2	0.25	0.15	0.3	0.52	0.6	0.35	0.004	
Pentacosane	$C_{25}H_{52}$	352	0.16	1.50	2.04	1.22	33.5	1.6	2.7	0.85	0.9	3.5	0.92	0.7	1.2	0.003	0.08
Heptacosane	$C_{27}H_{56}$	380	0.39	2.87	3.96	3.21	9.7	2.5	5.1	2.7	2.2	9.0	1.80	2.0	3.2	0.011	0.14
Nonacosane	$C_{29}H_{60}$	408	0.82	4.37	5.16	5.98	21.3	3.4	4.6	2.6	2.8	5.8	2.12	2.4	4.3	0.033	0.71
Hentriacontane	$C_{31}H_{64}$	436	0.71	5.19	5.52	1.60	19.3	2.5	1.8	3.4	2.5	2.8	1.80	2.0	3.9	0.014	0.73
Tritriacontane	$C_{33}H_{68}$	464	0.15	1.38	1.32	0.30	5.7	0.6	0.1	6.3	0.75	0.6	0.60	0.3	1.1	0.005	0.19
Pentatriacontane	$C_{35}H_{72}$	492	0.05	0.06			1.8				0.05	0.1				0.001	0.04
Total			**2.28**	**15.7**	**18.8**	**12.7**	**91.6**	**11.1**	**14.5**	**16.1**	**9.35**	**22.1**	**7.76**	**8**	**14.05**	**0.071**	**1.89**
PAHs																	
Phenanthrene	$C_{14}H_{10}$	178	0.09	0.30	0.69	0.60	0.80		0.04	0.8	0.10	0.7	0.05	0.22			
Fluoranthene	$C_{16}H_{10}$	202	0.04	0.80	1.10	1.20	1.30	0	0.10	1.9	0.20	1.3	0.11	0.6	0.016		
Pyrene	$C_{16}H_{10}$	202	0.03	0.60	1.00	1.40	1.00	0	0.10	2.0	0.19	1.4	0.1	0.52	0.013		
Benzo(ghi)fluoranthene	$C_{18}H_{10}$	226	0.01	0.20	0.22	0.30	0.20		0.02	1.3	0.08	0.5	0.09	0.38	0.003		
Cyclopenta(c,d)pyrene	$C_{18}H_{10}$	226	0	0.05	0.07	0.05	0.06		0.006	0.5	0.04	0.3	0.01	0.21	0.001		
Benz(a)anthracene	$C_{18}H_{12}$	228	0.02	0.12	0.14	0.15	0.40		0.04	0.9	0.07	0.5	0.07	0.33	0.002		
Chrysene	$C_{18}H_{12}$	228	0.02	0.35	0.69	0.70	0.52		0.08	1.3	0.11	0.7	0.12	0.56	0.007		
Benzo(k/b)fluoranthenes	$C_{20}H_{12}$	252	0.04	0.55	1.24	1.40	1.60	0	0.12	2.8	0.27	2.2	0.22	0.9	0.01		
Benzo(e)pyrene	$C_{20}H_{12}$	252	0.2	0.30	0.62	0.70	0.40		0.05	1.2	0.13	1.1	0.11	0.35	0.007		
Benzo(a)pyrene	$C_{20}H_{12}$	252	0.01	0.18	0.28	0.30	0.25		0.02	0.9	0.09	0.5	0.05	0.16	0.003		
Perylene	$C_{20}H_{12}$	252		0.01	0.03	0.03	0.07			0.1	0.01	0.15	0.02	0.03			
Indeno(1,2,3-cd)pyrene	$C_{22}H_{12}$	276	0.01	0.12	0.41	0.40	0.24		0.03	1.2	0.11	0.8	0.03	0.26	0.003		
Indeno(1,2,3-cd)fluoranthene	$C_{22}H_{12}$	276		0.04	0.14	0.15	0.07		0.01	0.3	0.04	0.4	0.07	0.08			
Benzo(ghi)perylene	$C_{22}H_{12}$	276	0.02	0.14	0.41	0.35	0.41		0.03	1.6	0.17	1.3	0.12	0.35	0.007		
Coronene	$C_{24}H_{12}$	300		0.01	0.14	0.05	0.12		0.01	0.5	0.05	0.2	0.02	0.06			
1,3,5- Triphenylbenzene	$C_{24}H_{18}$	306	0.11	0.18	0.28	0.03	0.24	0	0.09	2.0	0.05		0.06	0.06	0.003		
Total			**0.6**	**3.95**	**7.46**	**7.78**	**7.66**	**0.05**	**0.75**	**19.3**	**1.71**	**12.05**	**1.24**	**5.07**	**0.078**	**0**	**0**
Bap/Bep			0.8	0.6	0.45	0.43	0.63	-	0.4	0.8	0.7	0.45	0.45	0.5	0.43	-	-
n-Alkanoic acids																	
Tetradecanoic acid	$C_{14}H_{28}O_2$	228		4.0	3.0	2.0	3.0	2.0		9.0	2.0	0.6	2.0	2.0	0.23	0.3	
Pentadecanoic acid	$C_{15}H_{30}O_2$	242			1.4	1.0	1.3	0.5		3.0	1.5	0.3		0.8	0.05		
Hexadecanoic acid	$C_{16}H_{32}O_2$	256	13.0	20.2	21.0	11.0	22.0	6.0	6.1	29.8	21.0	5.5	11.5	8.0	1.40	1.6	1.4
Heptadecanoic acid	$C_{17}H_{34}O_2$	270	0.8	1.2	1.0	0.5	1.2	0.3	0.6	2.0	1.4	0.4	0.4	0.4	0.11	0.01	0.05
Octadecanoic acid	$C_{18}H_{36}O_2$	284	8.0	11.4	9.0	6.0	12.0	3.0	3.0	25.0	17.0	6.1	4.0	1.9	0.65	0.52	0.44
Nonadecanoic acid	$C_{19}H_{38}O_2$	298	0.4	0.5	0.4	0.3	0.7	0.4	0.7	1.5	0.4	0.06		0.2	0.08		0.01
Eicosanoic acid	$C_{20}H_{40}O_2$	312	1.6	2.9	3.0	1.5	11.5	1.9	2.3	6.0	1.9	0.8	0.3	0.4	0.25	0.02	0.05
Heneicosanoic acid	$C_{21}H_{42}O_2$	326	0.6	0.8	1.0	0.5	1.7	0.9	1.2	2.0	0.6	0.08		0.1	0.13		0.02
Docosanoic acid	$C_{22}H_{44}O_2$	340	3.2	4.9	5.2	3.0	11.6	4.1	2.5	9.0	3.0	1.1	0.3	0.5	0.50	0.03	0.19
Tricosanoic acid	$C_{23}H_{46}O_2$	354	1.8	1.7	1.5	1.0	3.5	1.6	1.2	3.0	1.2	0.1		0.1	0.23		0.11
Tetracosanoic acid	$C_{24}H_{48}O_2$	368	2.8	6.2	3.0	3.0	6.6	2.0	2.5	9.0	3.3	1.7	0.4	0.6	0.54	0.06	0.51
Pentacosanoic acid	$C_{25}H_{50}O_2$	382	0.8	0.5	0.7	0.4	1.5	0.5	0.7	2.5	0.6	0.3			0.14		0.11
Hexacosanoic acid	$C_{26}H_{52}O_2$	396	1.8	1.6	3.0	1.4	6.0	1.6	1.0	14.0	1.8	2.5	0.3	0.3	0.45	0.02	0.49
Heptacosanoic acid	$C_{27}H_{54}O_2$	410	0.4	0.2	0.5		0.7	0.2	0.3	0.6	0.3	0.1			0.11		0.05
Octacosanoic acid	$C_{28}H_{56}O_2$	424	1.6	1.3	3.4	1.4	6.0	1.1	0.6	14.6	1.6	1.4	0.1	0.2	0.44	0.01	0.33
Nonacosanoic acid	$C_{29}H_{58}O_2$	438	0.2	0.1	0.2		0.4	0.1	0.1	0.6	0.1	0.3			0.05		0.01
Triacontanoic acid	$C_{30}H_{60}O_2$	452	1.2	0.4	1.6	0.4	2.2	0.5	0.3	0.9	0.7	0.3	0.1	0.1	0.16		0.09
Hentriacontanoic acid	$C_{31}H_{62}O_2$	466													0.01		
Dotriacontanoic acid	$C_{32}H_{64}O_2$	480	0.4		0.2		0.5	0.1		0.3	0.1				0.02		
CPI (20-32)[a]			3.0	4.2	5.3	4.9	5.1	3.3	1.9	3.4	3.8	11.7	26.0	5.3	3.2	-	6.2
n **-Alkenoic acids**																	

Hexadecenoic acid	$C_{16}H_{30}O_2$	254		0.9	0.6											0.01	
Octadecenoic acids	$C_{18}H_{34}O_2$	282	0.4	1.2	1.5	0.8	3.5	0.1	0.2	2.0	0.5	0.3	1.0	0.3	0.05	3.2	0.7
Total aliphatic acids			**39.8**	**60.0**	**61.2**	**34.2**	**95.9**	**26.8**	**23.3**	**143.0**	**59.0**	**21.7**	**16.0**	**16.1**	**5.6**	**3.2**	**7.6**
Dehydroabietic acid	$C_{20}H_{28}O_2$	300	0.5	0.2	0.2	0.1	3	0.2	0.4	10.5	1.8	2	0.5	0.6	0.5	0.2	0.5
Benzoic acid	$C_7H_6O_2$	122	2	38	9	28	8	2	6	0.9	1.5	0.3	0.2	0.5	0.1	0.05	0.005
Glyceric acid	$C_3H_6O_4$	106	45														
Dicarboxylic acids																	
Malonic acid	$C_3H_4O_4$	104	20	35	15	16	6	52	2	0.8	2.0	3.2	0.2	0.6	0.3	1.2	0.002
Succinic acid	$C_4H_6O_4$	118	114	70	43	29	46	95	43	5.8	14.4	13.5	6.0	6.0	9.0	2.4	0.02
Glutaric acid	$C_5H_8O_4$	132	21	15	11	7	10	18	8	2.0	2.6	2.9	0.6	1.0	2.0	0.2	0.005
Adipic acid	$C_6H_{10}O_4$	146	6	6	10	2	8		4	1.3		0.6	0.2	0.2	1.9	1.0	0.002
Malic acid	$C_4H_6O_5$	134	72		10												
Hydroxyglutaric acid	$C_5H_8O_5$	148	27														
1,2-Benzenedicarboxylic acid	$C_8H_6O_4$	166	14	15	16	11	10	4	3	2.7	3.1	1.6	0.4	-	0.5	0.06	0.02
1,3-Benzenedicarboxylic acid	$C_8H_6O_4$	166	10	8	6	4	6	1	1								
1,4-Benzenedicarboxylic acid	$C_8H_6O_4$	166	16	12	11	7	9	2	6	6.3	1.1	0.6	0.2	-	0.2	0.04	0.004
Total			**302**	**197**	**131**	**104**	**103**	**176**	**83**	**19.8**	**24.7**	**22.7**	**7.9**	**8.3**	**14**	**4.95**	**0.05**
***n* -Alkanols**																	
Eicosanol	$C_{20}H_{42}O$	312		0.8	1.4	0.1	0.8	1.5	0.8	1.2	0.6	0.2		0.4			0.02
Docosanol	$C_{22}H_{46}O$	326	0.7	2.2	2.3	1.7	4.1	3.0	1.7	2.4	1.5	2.2	0.10	0.5	0.5		0.1
Tetracosanol	$C_{24}H_{50}O$	354	0.8	5.8	5.1	1.6	26.4	5.6	2.8	3.8	2.3	4.6	0.20	0.8	1		0.4
Hexacosanol	$C_{26}H_{54}O$	382	1.2	9.2	9.9	4.2	46.0	6.0	5.2	4.2	4.0	18.0	0.30	1.0	2.9		0.6
Octacosanol	$C_{28}H_{58}O$	410	0.8	17.0	29.0	13.0	36.5	3.8	3.1	8.8	2.6	5.0	0.10	0.5	13.6		0.9
Triacontanol	$C_{30}H_{62}O$	438	0.8	4.2	3.7	1.1	5.4	1.5	0.8	5.9	1.2	1.8	0.05	0.1	0.9		0.6
Dotriacontanol	$C_{32}H_{66}O$	466	0.2	1.0	0.3	0.1	1.0		0.1	1.2	0.2	0.2			0.2		0.4
Nonacosan-10-ol	$C_{29}H_{60}O$	424	0.5	8.0	7.0	1.3	14.0	3.5	1.0	5.5	3.8	4.0	0.05	5.0	2.8		1.6
Nonacosan-5,10-diol	$C_{29}H_{60}O_2$	440										2.0					
Total			**5.0**	**48.3**	**58.7**	**23.1**	**134.2**	**24.9**	**15.5**	**33.0**	**16.2**	**38.0**	**0.80**	**8.3**	**21.9**		**4.6**
Steroids																	
Cholesterol	$C_{27}H_{46}O$	386	0.12	0.41	7.4	6.0	4.2	0.5	-	1.6	0.2	0.6	0.01	0.5	0.2	0.02	
β-Sitosterol	$C_{29}H_{50}O$	414	0.14	0.34	0.2	4.8	10.3	0.2	-	2.2	0.6	0.5	0.005	0.2	0.1	0.01	0.01
Lupadienol	$C_{30}H_{48}O$	424			1.4												
Anhydrosaccharides																	
Levoglucosan	$C_6H_{10}O_5$	162	74	54	25	11	64	26	8	54	52	6.4	23	56	1.4	0.5	1.2
Mannosan	$C_6H_{10}O_5$	162	2.5	3.5	1.5	0.6	4.2	1.6	0.2	6	4.6	0.2	2.4	15			
Galactosan	$C_6H_{10}O_5$	162	1.5	2.5	1	0.4	3.8	1		2	2.4		0.6	2			
1,6-Anhydroglucofuranose	$C_6H_{10}O_5$	162	1.3	2.1	0.8	0.3	3	0.8		1.7	2		0.3	1.7			
Total			**79.3**	**62.1**	**28.3**	**12.3**	**75**	**29.4**	**8.2**	**63.7**	**61**	**6.6**	**26.3**	**74.7**	**1.4**	**0.5**	**1.2**
Saccharides																	
Sucrose	$C_{12}H_{22}O_{11}$	342	72	21	10	6	444	25	18	9.2	2.6	4.4	3.5	0.4	2.7	0.5	0.2
Mycose	$C_{12}H_{22}O_{11}$	342	2.5	27	30	9	9	5	7	15.5	0.2	7.7	12.0	0.1	9.2	0.1	0.1
Arabitol	$C_5H_{12}O_5$	152														3.8	
Xylitol	$C_5H_{12}O_5$	152			3	6		8	22	2.0	5.0	22.0	19.5		1.5	2.1	8.1
Sorbitol (glucitol)	$C_6H_{14}O_6$	182			4	4		7	9	3.0	4.0	18.0	26.0		1.4	3.8	7.8
α-Fructose	$C_6H_{12}O_6$	180	54	19					34			7.8				4.9	
β-Fructose	$C_6H_{12}O_6$	180	41	20					42			3.8					
α-Glucose	$C_6H_{12}O_6$	180	40	41	5	6	9	10	48	3.0	5.0	14.5	11.5	0.4	2.4	3.6	0.5
β-Glucose	$C_6H_{12}O_6$	180	50	44	7	5	10	12	63	2.7	7.0	19.0	15.0	0.8	2.1	4.3	1.8
Inositols	$C_6H_{12}O_6$	180	82	20			52	17	8	2.9	104.0						
Glycerol	$C_3H_8O_3$	92	10.5	18	10	4	50	16	40	2.5	24.0	8.9	5.5	12.0	7.4	6.0	5.3
Total			**352**	**210**	**69**	**40**	**574**	**100**	**291**	**41**	**152**	**106**	**93**	**14**	**27**	**29**	**24**
Lignin products																	
4-Hydoxybenzoic acid	$C_7H_6O_3$	138	0.9	6.5	2.8	0.3	3.1	2.2	2.0	2.8	3.0	2.0	0.5	0.5	1.5	2.1	0.4
Vanillic acid	$C_8H_8O_4$	168	0.2	2.5	1.6	0.1	1.1	0.5	0.4	1.5	0.4	0.5	0.2	0.1	0.2	0.3	
Syringic acid	$C_9H_{10}O_5$	198	0.1		0.6	0.1	0.2	0.2	0.1	1.4							
Total			**1.2**	**9.0**	**5.0**	**0.5**	**4.4**	**2.9**	**2.5**	**5.7**	**3.4**	**2.5**	**0.7**	**0.6**	**1.7**	**2.4**	**0.4**
Phthalates																	
Dibutyl phthalates	$C_{16}H_{22}O_4$	278					2			5.4	4	1.7	44	23	3.6	2.7	0.8
Dioctyl phthalate	$C_{24}H_{38}O_4$	390	5	5	6	7.3	21	2	12	34	20	26	42	15	2.6	4	16
Total			**5**	**5**	**6**	**7.3**	**23**	**2**	**12**	**39.4**	**24**	**28**	**86**	**38**	**7.2**	**6.7**	**16.8**

Calculated by subfraction of non-biogenic *n* -alkanes (Simoneit et al., 1991a).

[a] CPI = carbon prefence index = (C_{21} + C_{23} + C_{25} + C_{27} + C_{29} + C_{31} + C_{33} + C_{35} + C_{37})/(C_{22} + C_{24} + C_{26} + C_{28} + C_{30} + C_{32} + C_{34} + C_{36} + C_{38}) 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} + C_{33}) for *n* -alkanoic acids.

Table 3. Organic compounds (ng m^{-3}) identified in aerosol particulate matter sampled onboard the *R/V Ronald H. Brown.*

Compound	**Composition**	**M.W.**	RB1 (2329)	RB2 (2332)	RB3 (2333)	RB4 (2350)	RB5 (2357)	RB6 (2360)	RB7 (2376)	RB8 (2384)
***n* -Alkanes (Total)**										
Nonadecane	$C_{19}H_{40}$	268							0.09	
Eicosane	$C_{20}H_{42}$	282		0.09	0.20				0.08	0.96
Heneicosane	$C_{21}H_{44}$	296	0.03	0.18	0.47	0.14	0.08	0.54	0.08	2.52
Docosane	$C_{22}H_{46}$	310	0.04	0.20	0.46	0.26	0.10	0.56	0.40	3.00
Tricosane	$C_{23}H_{48}$	324	0.06	0.19	0.50	0.30	0.20	1.28	0.72	3.24
Tetracosane	$C_{24}H_{50}$	338	0.11	0.22	0.60	0.62	0.24	1.39	0.64	2.16
Pentacosane	$C_{25}H_{52}$	352	0.18	0.35	0.87	0.86	0.34	2.89	1.04	2.76
Hexacosane	$C_{26}H_{54}$	366	0.19	0.37	0.83	0.84	0.30	1.93	0.64	1.80
Heptacosane	$C_{27}H_{56}$	380	0.19	0.40	0.83	1.02	0.35	3.21	1.28	2.88
Octacosane	$C_{28}H_{58}$	394	0.12	0.28	0.67	0.84	0.26	1.50	0.56	1.68
Nonacosane	$C_{29}H_{60}$	408	0.10	0.30	0.76	1.20	0.50	3.00	1.60	3.60
Triacontane	$C_{30}H_{62}$	422	0.07	0.17	0.67	0.96	0.28	1.07	0.40	1.44
Hentriacontane	$C_{31}H_{64}$	436	0.07	0.23	0.70	1.10	0.33	2.14	1.04	2.16
Dotriacontane	$C_{32}H_{66}$	450	0.04	0.09	0.43	0.52	0.18	0.86	0.16	0.96
Tritriacontane	$C_{33}H_{68}$	464	0.03	0.08	0.40	0.60	0.19	0.86	0.28	0.96
Tetratriacontane	$C_{34}H_{70}$	478			0.17	0.12	0.06	0.43	0.08	0.36
Total			**1.3**	**3.2**	**8.6**	**9.4**	**3.4**	**21.6**	**9.1**	**30.5**
CPI (19-34)^{a}			1.1	1.2	1.2	1.3	1.4	1.7	2.1	1.6
Squalene	$C_{30}H_{50}$	410		2						
Plant Wax Alkanes^{b}										
Tricosane	$C_{23}H_{48}$	324				0.04		0.31		0.66
Pentacosane	$C_{25}H_{52}$	352	0.03	0.06	0.16	0.13	0.05	1.23	0.40	0.80
Heptacosane	$C_{27}H_{56}$	380	0.03	0.08	0.08	0.18	0.05	1.50	0.68	1.14
Nonacosane	$C_{29}H_{60}$	408	0.01	0.08	0.11	0.30	0.23	1.72	1.02	2.09
Hentriacontane	$C_{31}H_{64}$	436	0.02	0.10	0.15	0.36	0.10	1.18	0.76	0.96
Tritriacontane	$C_{33}H_{68}$	464	0.01	0.04	0.10	0.28	0.07	0.22	0.16	0.30
Total			**0.10**	**0.38**	**0.60**	**1.29**	**0.50**	**6.16**	**3.02**	**5.95**
PAH										
Fluoranthene	$C_{16}H_{10}$	202	-	-	-	0.012	-	0.10	0.50	-
Pyrene	$C_{16}H_{10}$	202	-	-	-	0.008	-	0.08	0.40	-
Benzo(ghi)fluoranthene	$C_{18}H_{10}$	226	-	-	-	-	-	0.02	0.15	-
Benz(a)anthracene	$C_{18}H_{12}$	228	-	-	-	0.003	-	-	0.10	-
Chrysene	$C_{18}H_{12}$	228	-	-	-	-	-	0.02	0.25	-
Benzo(b/k)fluoranthene	$C_{20}H_{12}$	252	-	-	-	0.007	-	0.06	0.40	-
Benzo(e)pyrene	$C_{20}H_{12}$	252	-	-	-	-	-	0.04	0.20	-
Benzo(a)pyrene	$C_{20}H_{12}$	252	-	-	-	-	-	-	0.10	-
Indeno(1,2,3-cd)pyrene	$C_{22}H_{12}$	276	-	-	-	-	-	-	0.20	-
Benzo(ghi)perylene	$C_{22}H_{12}$	276	-	-	-	-	-	-	0.20	-
1,3,5-Triphenylbenzene	$C_{24}H_{18}$	306	-	-	-	-	-	0.02	0.05	-
Total						**0.03**		**0.34**	**2.4**	
***n* -Alkanoic acids**										
Dodecanoic acid	$C_{12}H_{24}O_2$	200	0.08	0.9	4.6				8.82	4.05
Tetradecanoic acid	$C_{14}H_{28}O_2$	228	0.21	1.5	5.8	1	1.56	3.20	20.16	4.56
Pentadecanoic acid	$C_{15}H_{30}O_2$	242	0.07	0.6	2.5	0.3	0.63	1.50	8.40	2.03
Hexadecanoic acid	$C_{16}H_{32}O_2$	256	1.2	3.2	15.2	2.4	2.50	11.50	42.00	22.80
Heptadecanoic acid	$C_{17}H_{34}O_2$	270	0.01	0.08	0.4		0.23	0.69	2.10	0.51
Octadecanoic acid	$C_{18}H_{36}O_2$	284	0.13	0.6	3.9	0.7	1.03	4.26	7.14	6.59
Nonadecanoic acid	$C_{19}H_{38}O_2$	298					0.08	0.12	0.02	
Eicosanoic acid	$C_{20}H_{40}O_2$	312		0.03	0.05	0.1	0.27	1.15	0.63	0.76
Heneicosanoic acid	$C_{21}H_{42}O_2$	326					0.13	0.58	0.04	
Docosanoic acid	$C_{22}H_{44}O_2$	340		0.03	0.05	0.1	0.42	1.96	0.08	1.01
Tricosanoic acid	$C_{23}H_{46}O_2$	354					0.30	1.04	0.04	
Tetracosanoic acid	$C_{24}H_{48}O_2$	368		0.05	0.07	l2	0.55	2.30	0.13	1.01
Pentacosanoic acid	$C_{25}H_{50}O_2$	382					0.11	0.46	0.02	
Hexacosanoic acid	$C_{26}H_{52}O_2$	396		0.02	0.02	0.1	0.28	1.04	0.04	0.51
Heptacosanoic acid	$C_{27}H_{54}O_2$	410					0.08	0.35		
Octacosanoic acid	$C_{28}H_{56}O_2$	424					0.22	0.81	0.04	0.25
Nonacosanoic acid	$C_{29}H_{58}O_2$	438						0.12		
Triacontanoic acid	$C_{30}H_{60}O_2$	452						0.46		
CPI (12-30)^{a}			10	9.3	10.1	11	4.5	5.8	7.2	14.2
***n* -Alkenoic acids**										
Hexadecenoic acid	$C_{16}H_{30}O_2$	254	0.04	0.8	2.07	0.2		1.15	12	1.01

Octadecenoic acids	$C_{18}H_{34}O_2$	282	0.02	0.38	1.84	0.2		1.04	9	2.03
Total aliphatic acids			**11.8**	**17.5**	**46.6**	**16.1**	**12.9**	**39.5**	**117.9**	**61.3**
Glyceric acid	$C_3H_6O_4$	106	0.3	2.2	1.7		15	18	5	21
Benzoic acid	$C_7H_6O_2$	122	0.2	0.2	0.3	3.2	2	1.2	0.8	3.9
Dicarboxylic acids										
Malonic acid	$C_3H_4O_4$	104	0.1	1.5	3.8	5.6	21	23	2.5	16
Succinic acid	$C_4H_6O_4$	118	0.6	7	3.3	2.2	70	58	11	50
Glutaric acid	$C_5H_8O_4$	132	0.1	2.2	2.4		14	13	3	11
Adipic acid	$C_6H_{10}O_4$	146					8	22	2.5	10
Malic acid	$C_4H_6O_5$	134		0.2	0.8				3	26
1,2-Benzenedicarboxylic a	$C_8H_6O_4$	166			1.2		11.4	11.6	7	18
1,3-Benzenedicarboxylic a	$C_8H_6O_4$	166			0.1		2.2	2.6	2.1	4.1
1,4-Benzenedicarboxylic a	$C_8H_6O_4$	166			0.1		3.1	4	3.1	9
Total			**0.8**	**10.9**	**11.7**	**7.8**	**129.7**	**134.2**	**34.2**	**124.9**
***n* -Alkanols**										
Docosanol	$C_{22}H_{46}O$	326							0.6	
Tetracosanol	$C_{24}H_{50}O$	354							0.5	
Hexacosanol	$C_{26}H_{54}O$	382							2	
Octacosanol	$C_{28}H_{58}O$	410							1.3	
Triacontanol	$C_{30}H_{62}O$	438							0.5	
Nonacosan-10-ol	$C_{29}H_{60}O$	424							1.3	
Total									**6.2**	
Steroids										
Cholesterol	$C_{27}H_{46}O$	386	0.24	0.8	1.2			2	3.6	0.8
β-Sitosterol	$C_{29}H_{50}O$	414								3.2
Triterpenoids										
Lupa-2,19(22)-dienol	$C_{30}H_{48}O$	424							0.3	0.2
Friedelin	$C_{30}H_{50}O$	426	0.04	0.15						0.1
Anhydrosaccharides										
Levoglucosan	$C_6H_{10}O_5$	162	0.2	1.3	0.4	0.6	28	40	15	27
Mannosan	$C_6H_{10}O_5$	162					1.2	2.8	0.2	0.9
Saccharides										
Sucrose	$C_{12}H_{22}O_{11}$	342			1.8		0.6	1.2	3.7	422
Mycose	$C_{12}H_{22}O_{11}$	342			0.2		0.3	0.8	1.8	16
Sorbitol	$C_6H_{14}O_6$	182								1
α-Fructose	$C_6H_{12}O_6$	180								1
β-Fructose	$C_6H_{12}O_6$	180								3
α-Glucose	$C_6H_{12}O_6$	180			4		1.2	3.1	5	5
β-Glucose	$C_6H_{12}O_6$	180			3		1.4	4	3.5	22
Glycerol	$C_3H_8O_3$	92	6	15	32	-	8.1	7	9	12
Total			**6**	**15**	**41**	**0**	**11.6**	**16.1**	**23**	**482**
Lignin products										
4-Hydroxybenzoic acid	$C_7H_6O_3$	138				3	1.5	3	2	
Vanillic acid	$C_8H_8O_4$	168				0.3		0.4	0.2	
Total						**3.3**	**1.5**	**3.4**	**2.2**	
Phthalates										
Dibutyl phthalates	$C_{16}H_{22}O_4$	278			2.2	0.5				
Dioctyl phthalate	$C_{24}H_{38}O_4$	390	3.8	3.3	7.6	18	9.7	83	4.8	27
Total			**3.8**	**3.3**	**9.8**	**18.5**	**9.7**	**83**	**4.8**	**27**

[a] CPI as defined in Table 2.

[b] Calculated by subfraction of the non-biogenic *n* -alkanes (Simoneit et al., 1991a).

Table 4. Total compound group yields (ng m^{-3}).

Compound Groups	Gosan							Sapporo					Chichi-jima			*R/V Ronald Brown*							
	G1	G3	G4	G5	G14	G20	G30	S2033	S2039	S2443	S2447	S2504	C181	C188	C221	RB1	RB2	RB3	RB4	RB5	RB6	RB7	RB8
Lipids																							
Plant Wax Lipids																							
n -Alkanes[a]	2.3	15.7	18.8	12.7	91.6	11.1	14.5	16.6	9.4	22.1	7.8	8	4.5	0.06	1.9	0.1	0.38	0.6	1.29	0.5	6.16	3.02	5.95
n -Alkanols	5	48.3	58.7	23	134	25	15.5	33	16	38	0.8	8.3	22	0	4.6	0	0	0	0	0	0	6.2	0
n -Alkanoic acids[b]	33	3.7	37.5	24	81.3	26.8	19.4	36	22	13.2	2.7	5.8	5	0.47	6.1	0	0.6	0.7	1.4	5.44	21.1	2.9	13.1
Total	**40.3**	**67.7**	**115**	**59.7**	**307**	**63**	**49.4**	**86**	**47.4**	**73.3**	**11.3**	**24.1**	**29.5**	**0.53**	**12.6**	**0.1**	**1**	**1.3**	**2.7**	**5.9**	**27.2**	**12.1**	**19**
Fossil Fuel (Coal/Petroleum)																							
n -Alkanes[a]	7.3	19.1	26.4	6.7	58.7	14.7	19	64.2	26.2	9.4	20.8	69.2	2.5	0.24	2.6	1.16	2.78	8	8.09	2.9	15.4	6.07	24.5
UCM[c]	37	95.5	132	33.5	294	74	95	321	131	47	104	346	12.5	1.2	13	5.8	13.9	40	40.5	14.5	77	30.5	123
Total	**44.3**	**115**	**158**	**40.2**	**353**	**88.7**	**114**	**385**	**157**	**56.4**	**125**	**415**	**15**	**1.4**	**15**	**7.0**	**16.7**	**48**	**48.6**	**17.4**	**92.4**	**36.6**	**148**
PAHs	0.4	3.95	7.46	7.8	7.7	0	0.75	19.3	1.7	12	1.2	5.1	0.08	0	0	0	0	0	0.03	0	0.34	2.4	0
Marine Lipids																							
Alkanoic Acids[b]	7	2.3	23.7	10.2	14.6	0	2.9	107	37	8.5	13.3	10.3	0.6	2.73	1.5	1.76	6.96	35.8	3.86	2.95	11.6	108	34
Sterols	0.26	0.75	9	10.8	14.5	0.7	0	3.8	0.8	1.1	0.015	0.7	0.3	0.03	0.01	0.24	0.8	1.2	0	0	2	3.6	4
Polar Compounds																							
Saccharides (Soil resuspension)	352	210	69	40	574	100	291	41	152	95	93	14	27	29	24	6	15	41	0	12	16.4	23	482
Anhydrosaccharides (biomass burning)	78	60	28	12	72	29	8	62	59	7	26	73	1.4	0.5	1.2	0.2	1.3	0.4	0.6	29	43	15	28
Dicarboxylic, Glyceric and Benzoic Acids (Secondary oxidation)	347	197	131	104	103	176	83	20	25	23	8	8.3	14	5	0.05	1.3	13.3	13.7	11	147	153	40	169
Plastics																							
Phthalates	5	5	6	7.3	23	2	12	39	24	28	86	38	7	7	17	4	3	10	19	10	83	5	27
Total All Compounds + UCM	**874**	**661**	**547**	**292**	**1469**	**459**	**561**	**667**	**504**	**304**	**364**	**589**	**95**	**46**	**72**	**20.6**	**58**	**151**	**85.8**	**224**	**429**	**246**	**911**

[a] Plant wax alkanes are calculated as the excess odd homolog - adjacent even homologs average [Simoneit et al., 1991a,c] and the difference from the total alkanes is the petroleum derived amount.

[b] Plant wax alkanoic 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}.

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

Table 5. Relative percentages of the various compound source groups in the total compound mass (TCM) of the extractable organic matter from the aerosols.

Sample	Natural[a]		Urban[a]			Biomass Burning[b]	Soil Resuspension[b]	Secondary Oxidation Products[b]	Total Water-soluble Compounds[c]
	Plant Wax Lipids	Marine Lipids	Petroleum	PAHs (Coal)	Plastics				
Gosan									
G1	4.6	0.8	5	0.05	0.6	9	40	40	89
G3	10.2	0.5	17	0.6	0.8	9	32	30	71
G4	21	6	29	1.4	1.1	5	13	24	42
G5	20	7.2	14	2.6	2.5	4	14	36	54
G14	21	2	24	0.5	1.6	5	39	7	51
G20	14	0.2	19	0.00	0.4	6	22	38	66
G30	8.8	0.5	20	0.1	2.1	1.4	52	15	68
Sapporo									
S2033	13	2.2	58	2.9	5.8	9	6	3	19
S2039	9.4	7.5	31	0.3	5	12	30	5	47
S2443	24	3.2	19	0.4	9.2	2.3	31	7.6	41
S2447	3.1	3.7	34	0.3	24	7	26	2	35
S2504	4.2	2	73	0.9	6.7	13	3	2	14
Chichi-jima									
C181	31	0.9	16	0.08	7.4	2	28	15	45
C188	1.2	0.06	3.1	0.0	15	1	63	11	75
C221	18	2.1	22	0.0	24	1.7	33	0.07	35
Mobile-*RV Ronald H. Brown*									
RB1	0.5	10	34	0.0	19	1	29	6	36
RB2	1.7	13.5	29	0.0	5	2	26	23	51
RB3	0.9	24	32	0.0	6.6	0.3	27	9	36
RB4	3.1	4.5	57	0.04	22	0.7	0	13	14
RB5	2.6	1.3	8	0.0	5	13	5	66	84
RB6	6.3	2.9	22	0.08	19	10	4	36	50
RB7	4.9	45	15	1.0	2	6	9	16	31
RB8	2	4.2	16	0.0	3	3	53	19	75

[a] Totals from Table 4 divided by total of all compounds + UCM.

[b] (Total anhydrosaccharides + resin acids) / (total for biomass burning), (total saccharides) / (total for siol resuspension), (total dicarboxylic glyceric and benzoic acids) / (total for secondary oxidation) (from Table 4).

[c] Water-soluble compounds; Sum of biomass burning, soil resuspension and secondary oxidation products.

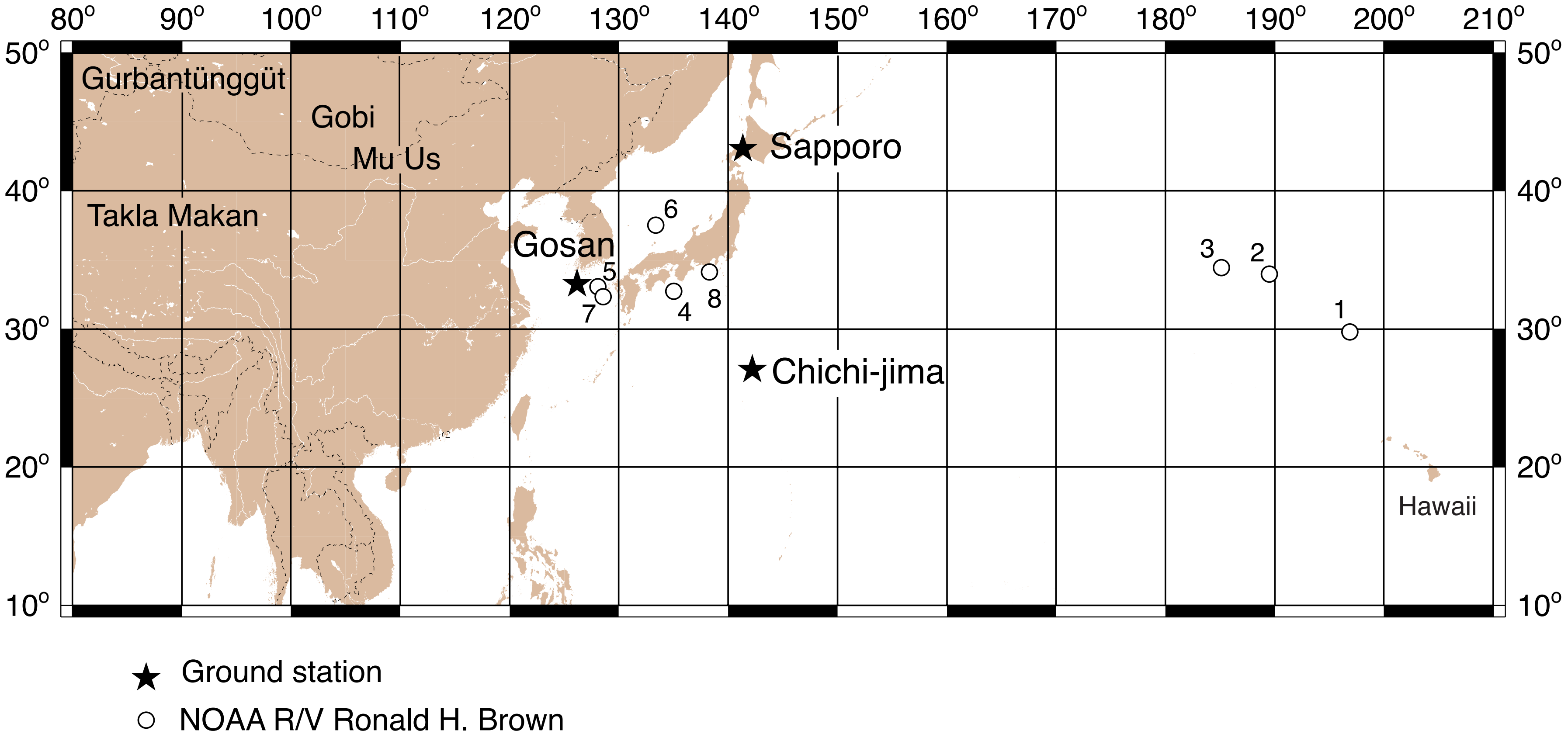
Table 6. Contribution (%) of each organic compound classes to total organic carbon (OC) in aerosols.

Compound class [a]	Gosan		Sapporo		Chichi-jima		R/V Ron H. Brown	
	range	average	range	average	range	average	range	average
Total n-alkanes	0.1-1.7	0.57	0.53-1.6	1.0	0.09-0.50	0.34	0.07-0.57	0.34
UCM hydrocarbons (fossil fuel) [b]	0.44-3.3	1.3	1.0-6.6	3.4	0.37-1.5	0.86	0.87-2.5	1.4
Wax alkanes (higher plant)	0.03-1.0	0.31	0.10-0.47	0.26	0.02-0.86	0.36	0.01-0.10	0.06
PAHs (combustion)	0.00-0.20	0.07	0.02-0.29	0.13	0.005	0.005	0.002-0.09	0.03
Fatty acids (biogenic)	0.31-0.98	0.58	0.27-1.0	0.62	0.31-1.6	0.78	0.16-1.1	0.82
Diacids (C3-C8)	0.60-1.7	0.96	0.07-0.24	0.15	0.003-0.43	0.39	0.20-1.1	0.79
n-Alcohols (higher plant)	0.06-1.5	0.56	0.04-0.77	0.27	0.04-1.3	0.89	0.34	0.34
Anhydrosaccharides (biomass burning)	0.14-0.49	0.29	0.07-0.78	0.44	0.04-0.08	0.06	0.003-0.37	0.18
Other saccharides	0.3-2.0	1.2	0.12-2.5	0.94	0.80-1.2	2.1	0.1-1.7	0.69
Lignin products	0.01-0.06	0.03	0.01-0.04	0.03	0.03-0.52	0.21	0.02-0.14	0.06
Phthalates (plasticizer)	0.02-0.23	0.09	0.21-4.1	1.1	0.31-1.7	1.2	0.12-1.2	0.52
Total compounds	3.8-12	6.0	4.3-16	8.1	4.5-9.3	6.6	2.8-5.7	4.9

[a] Compound concentrations (ngC m^{-3}) were calculated for each species and then divided by OC (µgC m^{-3}).

[b] UCM hydrocarbon-carbon was calculated assuming that UCM carbon/mass ratio is equivalent to that of C28 alkane. See Table 4 for UCM data.
Results of minor species such as steroidal alcohols, benzoic acid, glyceric acid, and dehydroabietic acid are not presented here.
But, they are all included in Total compounds as shown above.

Fig. 1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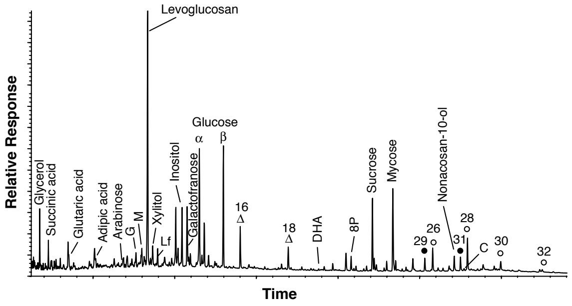
120°
130°
140°
150°
160°
170°
180°
190°
200°
210°
50°
40°
30°
20°
10°
Gurbantünggüt
Gobi
Mu Us
Takla Makan
Gosan
Sapporo
Chichi-jima
Hawaii
1
2
3
4
5
6
7
8
★ Ground station
○ NOAA R/V Ronald H. Brown

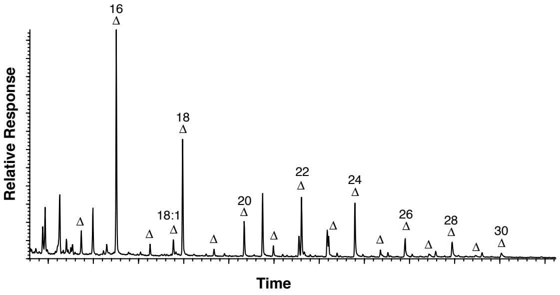
Fig. 2

Gosan 3
April 10-11, 2001

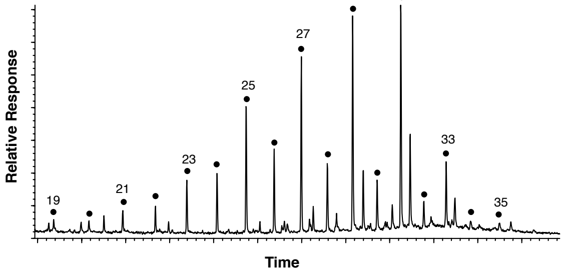
(a) Total extracts (TIC)



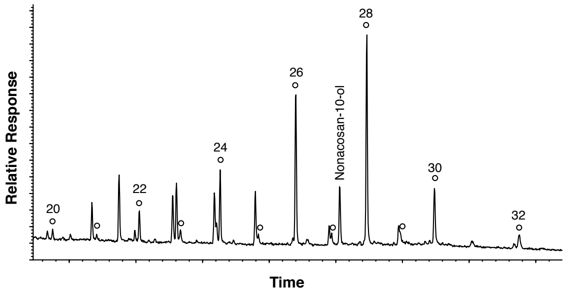
(b) n-Alkanoic acids (m/z 117)



(c) n-Alkanes (m/z 85)

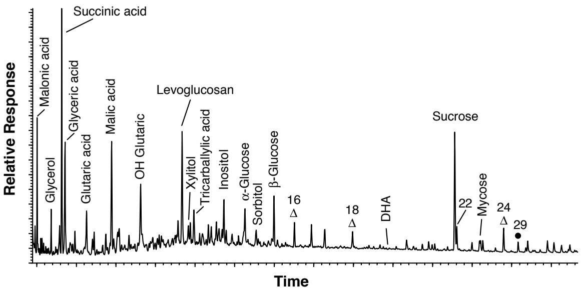


(d) n-Alkanols (m/z 75)

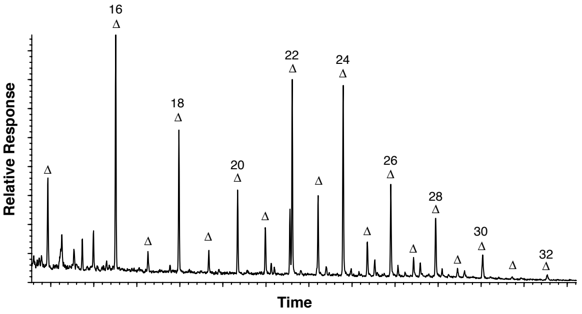


Gosan 20
April 27-28, 2001

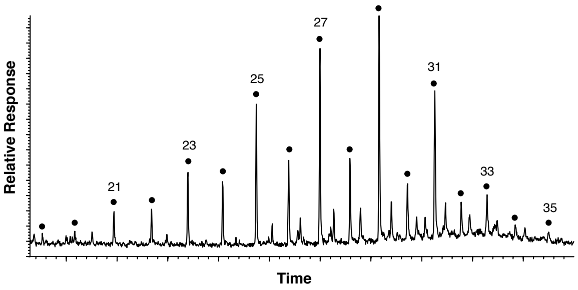
(e) Total extracts (TIC)



(f) n-Alkanoic acids (m/z 117)



(g) n-alkanes (m/z 85)



(h) n-alkanols (m/z 75)

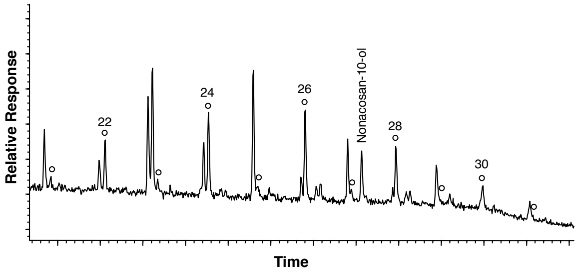
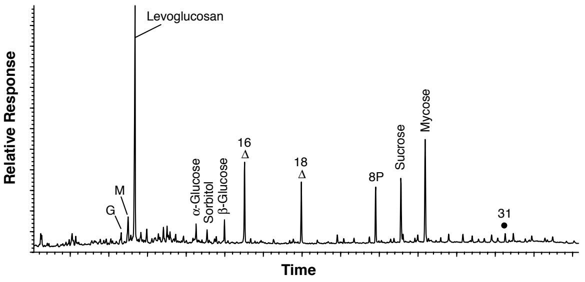


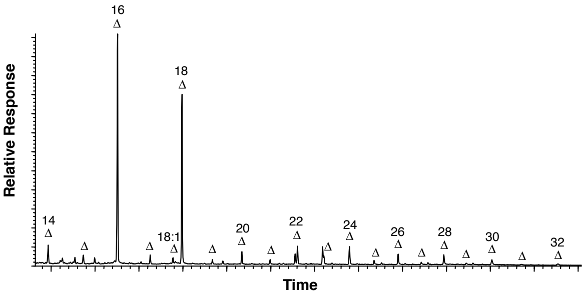
Fig. 3

Sapporo 2033A
April 8-10, 2001

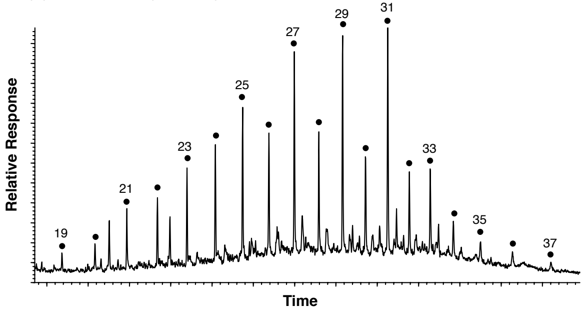
(a) Total extracts (TIC)



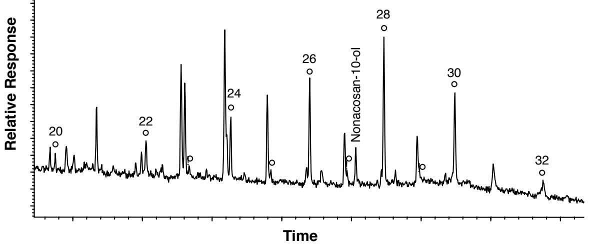
(b) n-Alkanoic acids (m/z 117)



(c) n-alkanes (m/z 85)



(d) n-alkanols (m/z 75)



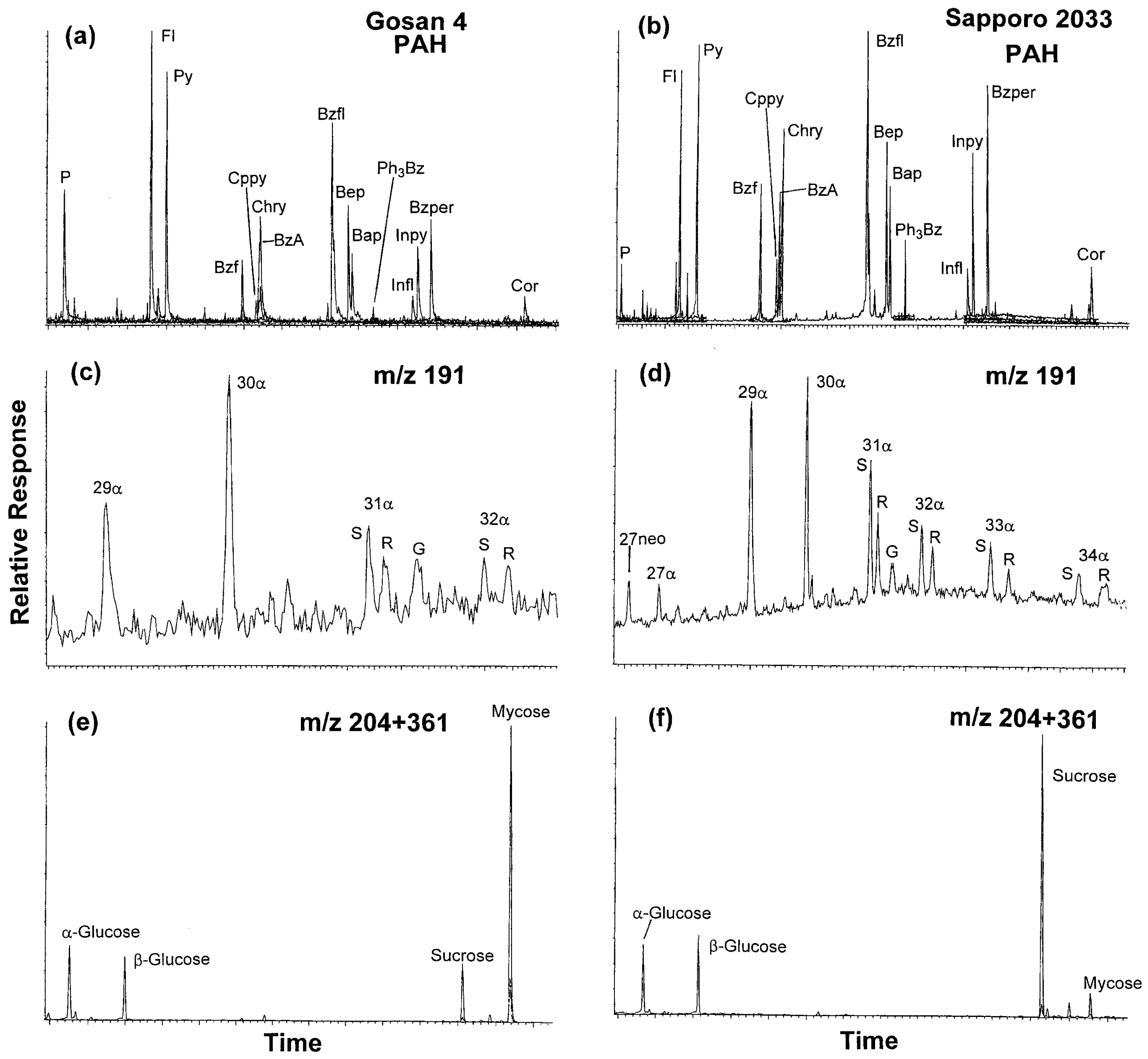

(a)
Gosan 4
PAH
P
Fl
Py
Bzf
Cppy
Chry
BzA
Bzfl
Bep
Bap
Ph3Bz
Infl
Inpy
Bzper
Cor
(b)
Sapporo 2033
PAH
P
Fl
Py
Bzf
Cppy
Chry
BzA
Bzfl
Bep
Bap
Ph3Bz
Infl
Inpy
Bzper
Cor
Relative Response
(c)
m/z 191
29α
30α
31α
S
R
G
32α
S
R
(d)
m/z 191
27neo
27α
29α
30α
31α
S
R
G
32α
S
R
33α
S
R
34α
S
R
(e)
m/z 204+361
α-Glucose
β-Glucose
Sucrose
Mycose
Time
(f)
m/z 204+361
α-Glucose
β-Glucose
Sucrose
Mycose
Time

Fig. 4

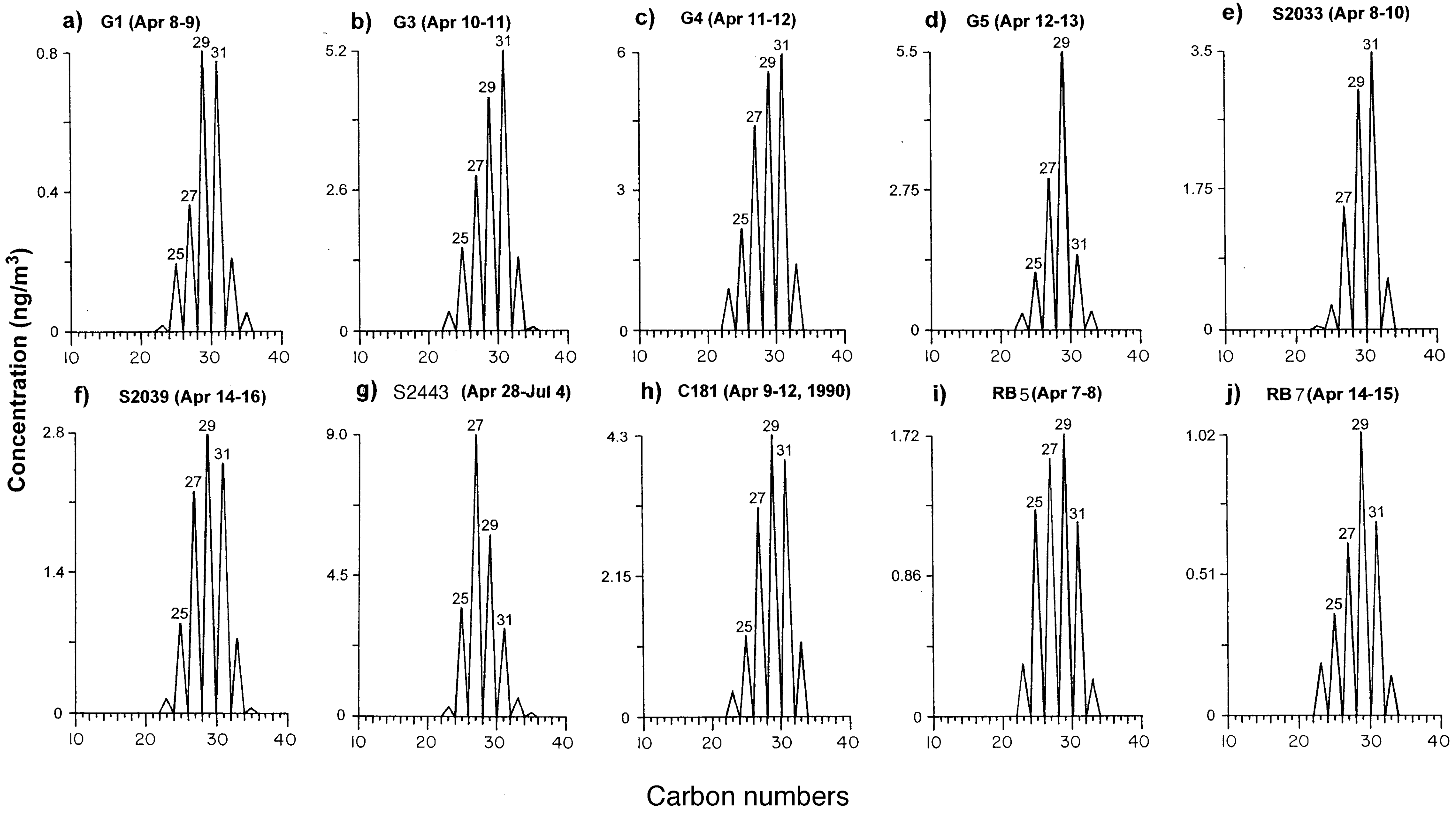
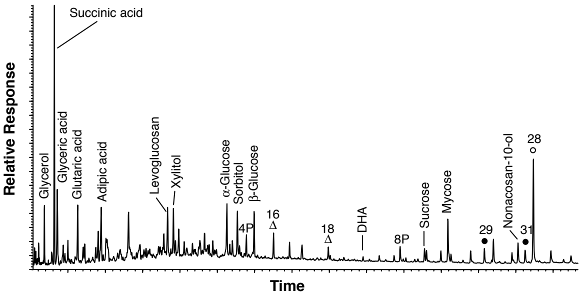

a) G1 (Apr 8-9)
b) G3 (Apr 10-11)
c) G4 (Apr 11-12)
d) G5 (Apr 12-13)
e) S2033 (Apr 8-10)
f) S2039 (Apr 14-16)
g) S2443 (Apr 28-Jul 4)
h) C181 (Apr 9-12, 1990)
i) RB 5 (Apr 7-8)
j) RB 7 (Apr 14-15)
25
27
29
31
0.8
0.4
5.2
2.6
6
3
5.5
2.75
3.5
1.75
2.8
1.4
9.0
4.5
4.3
2.15
1.72
0.86
1.02
0.51
0
10
20
30
40
Concentration (ng/m^3)
Carbon numbers

Fig. 5

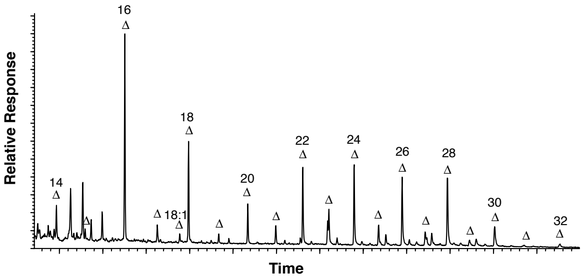
Fig. 6

Chichi-jima (181)
April 9-12, 1990

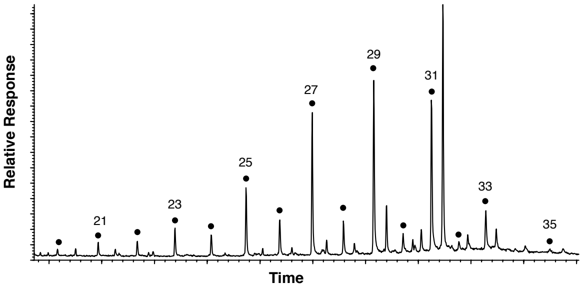
(a) Total extracts (TIC)



(b) n-Alkanoic acids (m/z 117)



(c) n-alkanes (m/z 85)



(d) n-alkanols (m/z 75)

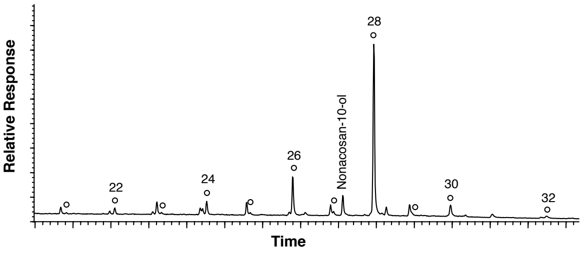
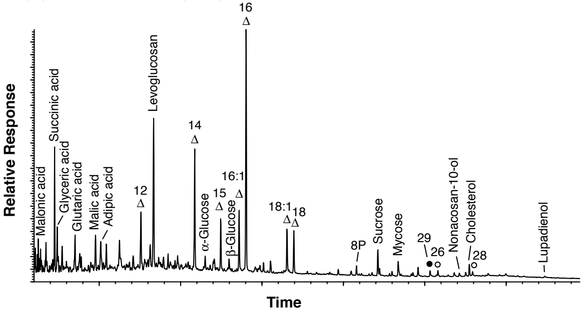


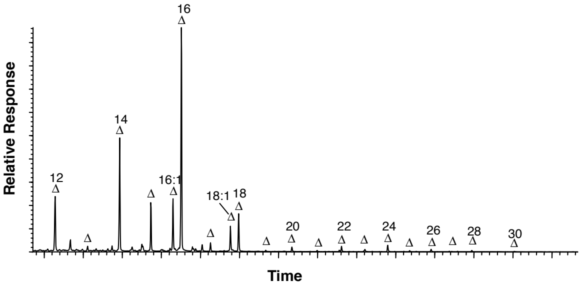
Fig. 7

Ron Brown 2376
April 14-15, 2001

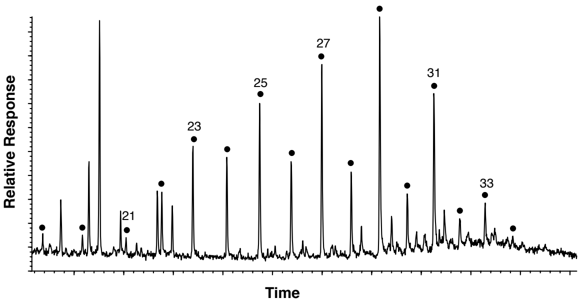
(a) Total extracts (TIC)



(b) n-Alkanoic acids (m/z 117)



(c) n-Alkanes (m/z 85)



(d) n-Alkanols (m/z 75)

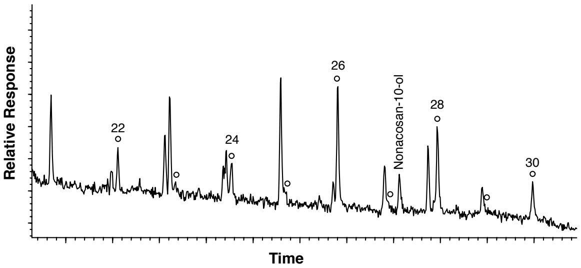


Fig.8

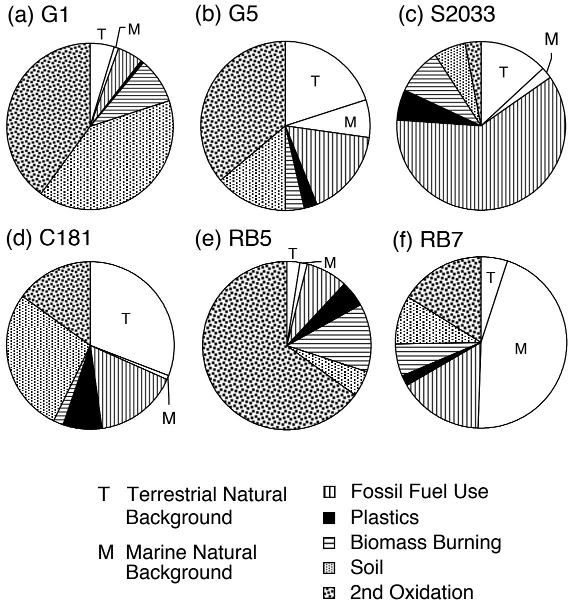
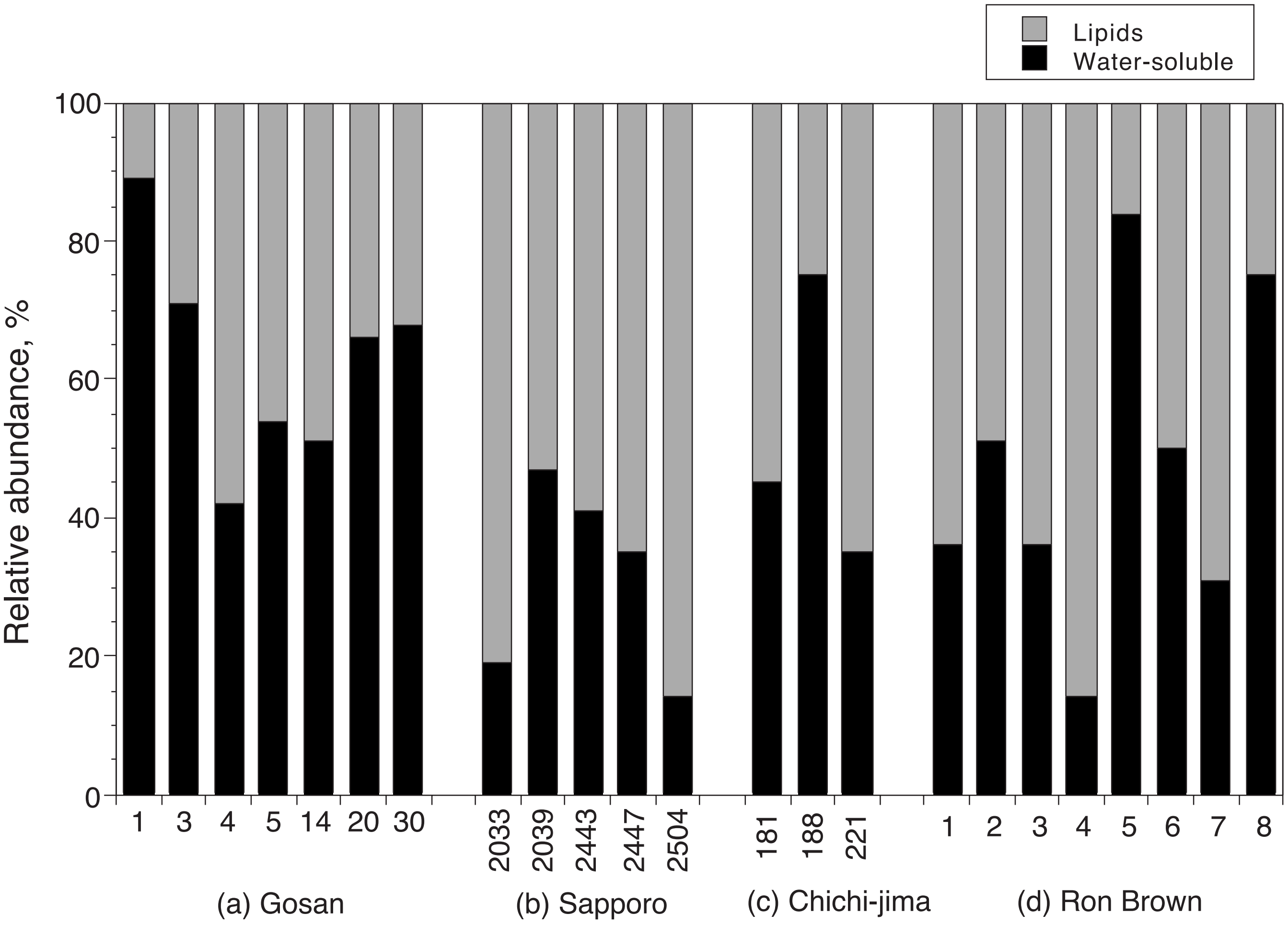
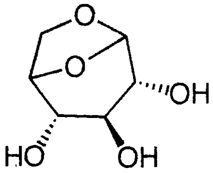

(a) G1
T
M
(b) G5
T
M
(c) S2033
T
M
(d) C181
T
M
(e) RB5
T
M
(f) RB7
T
M
T Terrestrial Natural Background
M Marine Natural Background
Fossil Fuel Use
Plastics
Biomass Burning
Soil
2nd Oxidation

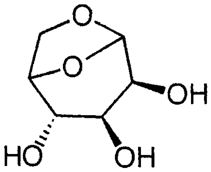
Fig. 9


Lipids
Water-soluble
Relative abundance, %
100
80
60
40
20
0
1
3
4
5
14
20
30
2033
2039
2443
2447
2504
181
188
221
1
2
3
4
5
6
7
8
(a) Gosan
(b) Sapporo
(c) Chichi-jima
(d) Ron Brown

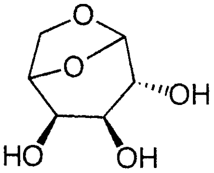
Appendix I - Chemical Structures Cited



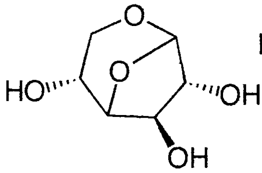
I. Levoglucosan



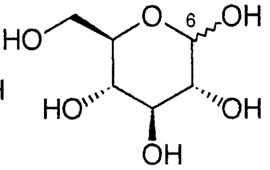
II. Mannosan



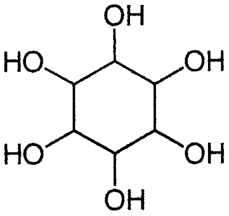
III. Galactosan



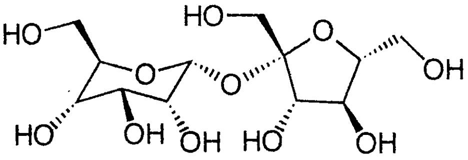
IV. 1,6-Anhydro-β-glucofuranose



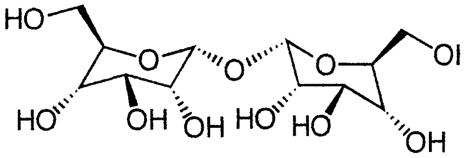
V. Glucose (α=6αOH, β=6β



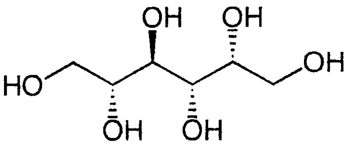
VI. Inositols



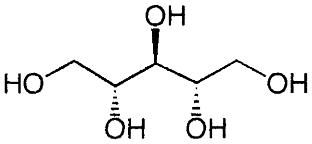
VII. Sucrose



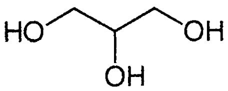
VIII. Mycose (trehalose)



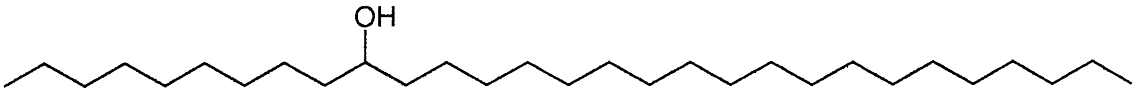
IX. Sorbitol (glucitol)



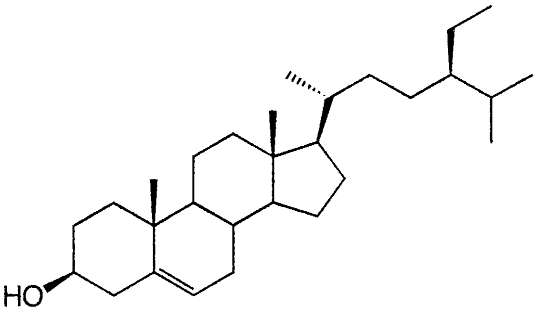
X. Xylitol



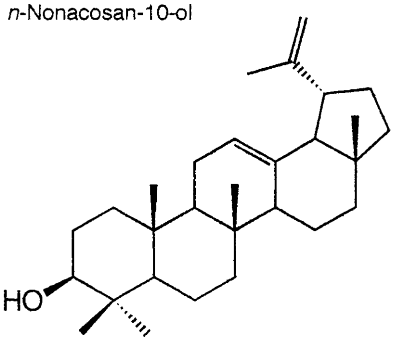
XI. Glycerol



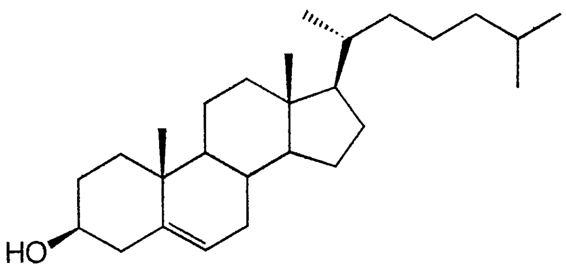
XII. *n*-Nonacosan-10-ol



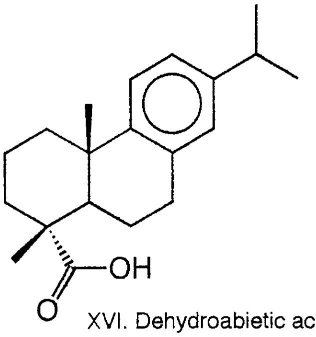
XIII. β-Sitosterol



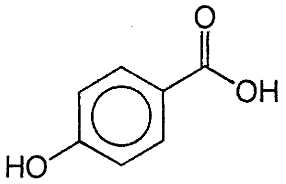
XIV. Lupadienol



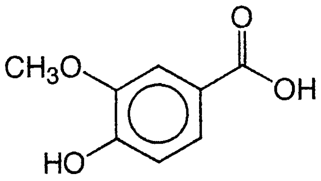
XV. Cholesterol



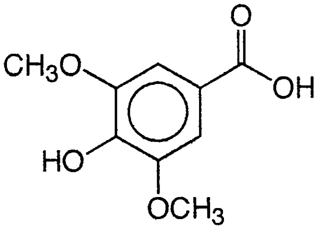
XVI. Dehydroabietic acid



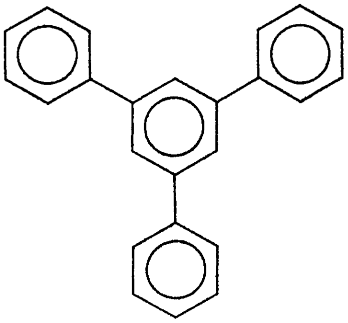
XVII. 4-Hydroxybenzoic acid



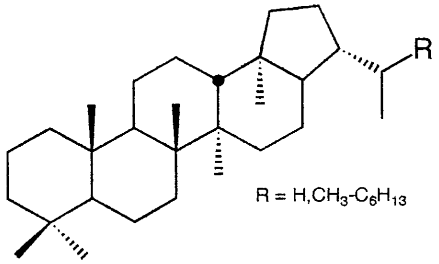
XVIII. Vanillic acid



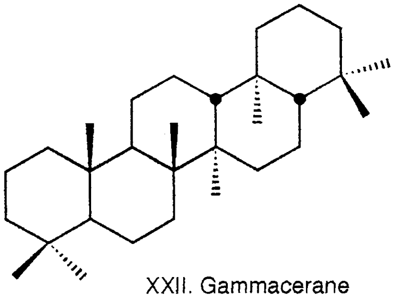
XIX. Syringic acid



XX. 1,3,5-Triphenylbenzene



XXI. 17α(H)-Hopanes



XXII. Gammacerane