Long-term observations of saccharides in remote marine aerosols from the western North Pacific: A comparison between 1990–1993 and 2006–2009 periods

Jing Chen [a,b,d], Kimitaka Kawamura [b,*], Cong-Qiang Liu [a], Pingqing Fu [b,c]

[a] State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, China
[b] Institute of Low Temperature Science, Hokkaido University, Sapporo 060-0819, Japan
[c] State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China
[d] University of Chinese Academy of Sciences, Beijing 100049, China

HIGHLIGHTS

► Analysis of 12 sugar compounds in marine aerosols at Chichi-Jima Island.
► Long-term observations were performed during 1990–1993 and 2006–2009.
► The levels of arabitol and mannitol generally increased from 2006 to 2009.
► Biomass burning is a minor contributor to the remote marine aerosols.

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ABSTRACT

Anhydrosugars (galactosan, mannosan and levoglucosan), sugars (xylose, fructose, glucose, sucrose and trehalose) and sugar alcohols (erythritol, arabitol, mannitol and inositol) were measured in the aerosol samples collected in a remote island (Chichi-Jima, Japan) in the western North Pacific from 1990 to 1993 and from 2006 to 2009. Total concentrations of anhydrosugars, the biomass burning tracers, were $0.01-5.57$ ng m^{-3} (average 0.76 ng m^{-3}) during 1990–1993 versus $0.01-7.19$ ng m^{-3} (0.64 ng m^{-3}) during 2006–2009. Their seasonal variations were characterized by winter/spring maxima and summer/fall minima. Such a seasonal pattern should be caused by the enhanced long-range atmospheric transport of biomass burning products and terrestrial organic matter (such as higher plant detritus and soil dust) from the Asian continent in winter/spring seasons, when the westerly or winter monsoon system prevails over the western North Pacific. Sugars and sugar alcohols showed different seasonal patterns. The monthly mean concentrations of erythritol, arabitol, mannitol, inositol, fructose, glucose and trehalose were found to be higher in spring/summer and lower in fall/winter during both 1990–1993 and 2006–2009 periods, indicating an enhanced biogenic emission of aerosols in warm seasons. Interestingly, saccharides showed a gradual decrease in their concentrations from 1991 to 1993 and an increase from 2006 to 2009. In addition, the monthly averaged concentrations of sugars and sugar alcohols showed maxima in early summer during 1990–1993, which occurred about 1–2 months earlier than those during 2006–2009. Such a clear seasonal shift may be attributable to the changes in the strength of westerly and trade wind systems during two periods.

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1. Introduction

Organic aerosols have been recognized to account for a significant portion of atmospheric particulate matter and to play an important role in regulating regional and global climate (Jacobson et al., 2000; Kanakidou et al., 2005; Pöschl, 2005). Aerosol particles that are enriched with organic compounds can make the aerosol surfaces more hydrophilic or hydrophobic depending on the chemical composition and mixing state, leading to alter the cloud condensation nuclei (CCN) activities of particles. The long-range atmospheric transport of aerosol particles from continental regions to remote oceans is a key process to control the global

* Corresponding author.
E-mail addresses: chenjing406@mails.gucas.ac.cn (J. Chen), kawamura@lowtem.hokudai.ac.jp (K. Kawamura).

distributions of organic aerosols. However, there are very few studies reported on the long-term observations of organic aerosols in the remote marine atmosphere (Kawamura et al., 2003; Mochida et al., 2003).

Saccharides are one of the major classes of water-soluble organic constituents in atmospheric aerosols, which are ubiquitous in urban (Pashynska et al., 2002; Wang et al., 2006; Jia et al., 2010), forest and mountain (Graham et al., 2003; Yttri et al., 2007; Fu et al., 2008; Wang et al., 2009b), marine (Simoneit et al., 2004b; Burshtein et al., 2011; Fu et al., 2011), and the polar regions (Fu et al., 2009). One of the most studied sugar compounds is levoglucosan, which is produced by thermal decomposition of cellulose and hemicellulose. Levoglucosan is emitted during the combustion of biomass such as residential heating and wild/agriculture fires, and has been used as a useful molecular marker of biomass burning (Simoneit et al., 1999, 2004a). Recent studies (Hennigan et al., 2010; Hoffmann et al., 2010) have suggested that levoglucosan is prone to degradation by OH radicals in the atmosphere. Thus, its concentration during long-range transport may be underestimated, especially in summer (Mochida et al., 2010).

The sources of primary sugars in the atmosphere include natural biogenic detritus (such as microorganisms, plants) (Simoneit et al., 2004a; Jaenicke, 2005) and resuspended dusts (Simoneit et al., 2004b). Primary bioaerosols (including plant fragments, airborne pollen, bacteria, etc.) can contribute 20–30% to the total atmospheric particulate matter (PM) ($>$0.2 μm) from Lake Baikal (Russia) and Mainz (Germany) (Jaenicke, 2005). Sugar alcohols, together with glucose, sucrose and trehalose, are more abundant in the coarse than in the fine size of PM (Fuzzi et al., 2007; Pio et al., 2008), attesting to their primary biological sources (Graham et al., 2003; Simoneit et al., 2004a; Bauer et al., 2008). Fungi are important microorganisms in nature, and most of them are known to emit spores whose sizes range between 1.5 and 12 μm in diameter (Elbert et al., 2007). These spores, which account for a major portion of natural bioaerosols, have long residence time in the atmosphere and can travel long distances (Burshtein et al., 2011). Atmospheric levels of mannitol and arabitol are well correlated with the counts of fungal spores in atmospheric PM_{10}, and thus they have been proposed as specific tracers of fungal activities (Bauer et al., 2008).

In East Asia, the outflows of industrial pollutants, biomass burning products, and mineral dusts form a complex mixture of inorganic species, organic compounds, black carbon, minerals, and water (Seinfeld et al., 2004). During the ACE-Asia campaign, a lot of studies have been conducted on the physical and chemical properties of the Asian dusts and polluted aerosols with a function of altitude above the East Asian surface (Huebert et al., 2003). The Chichi-Jima Island, a small island in the western North Pacific, is located on the outflow pathway of the Asian desert dusts and polluted air masses from China (Simoneit et al., 2004b; Wang et al., 2009a). This island is also in the boundary of westerly and trade wind regimes. Previous studies have reported terrestrial lipid compound classes (Kawamura et al., 2003), dicarboxylic acids (Mochida et al., 2003), and levoglucosan (Mochida et al., 2010) in aerosol samples collected at the Chichi-Jima Island, but little is known about the molecular distribution and seasonal variations of saccharides. Such datasets of long-term measurements of organic aerosols in the marine atmosphere are of great importance in evaluating the changes in air quality caused by regional meteorology and the changes in the long-range atmospheric transport of terrestrial organic matter to the ocean.

The objectives of the present study are: (i) to investigate the abundances and annual variations of saccharides in marine aerosols collected at Chichi-Jima using a gas chromatography (GC)/mass spectrometry (MS) technique, and (ii) to compare the seasonal and annual changes between 1990–1993 and 2006–2009 periods. In the data interpretation, we performed positive matrix factorization (PMF) analysis to obtain a better understanding on the source of saccharides in the marine aerosols in the western North Pacific.

2. Experimental

The sampling site (142°13′E, 27°04′N; 254 m, a.s.l.) is located at the Ogasawara Downrange Station of the Japan Aerospace Exploration Agency (JAXA) in the Chichi-Jima Island for observing long-range-transported aerosols in the western North Pacific (Fig. 1) (Kawamura et al., 2003). The Chichi-Jima is an oceanic island, which is located about 2000 km away from the Asian continent and about 1000 km south of Tokyo, Japan. It has never been connected to the land with any continent since its birth. Therefore, a large number of endemic plants are found only in this region. The endemic ratio of vascular plants is 36%. Sclerophyllous scrub woodlands cover large areas of the Chichi-Jima Island (Kawakami, 2010). The island is characterized by a subtropical climate; it is warm (ambient temperature: 7.8–34.1 °C) and humid all year round owing to the warm currents from the North Pacific gyre around the island. Because the population (about 2000) and area (24 km^2) of island are small, the perturbation caused by local biomass-burning emission is insignificant.

Total suspended particles (TSP) were collected on a biweekly basis from April 1990 to November 1993 ($n = 69$) and a weekly basis from January 2006 to January 2010 ($n = 203$) using a high volume air sampler (Shibata HVC-1000 for 1990–1993 and Kimoto AS-810A for 2006–2009) with a flow rate of 1.0 m^3 min^{-1} and quartz fibre filters (QFF, 20 cm × 25 cm, Pallflex). The sampler was placed on the top of the base (5 m, a.g.l.) for the parabola antenna and run without a wind sector control. Before and after sampling, each sample was placed in a pre-combusted (450 °C for 6 h) clean glass jar with a Teflon-lined screw cap. The samples were stored in a dark freezer room at −20 °C prior to analysis.

Filter aliquots (ca. 20 cm^2) were cut off and extracted three times with dichloromethane/methanol (2:1, v/v) under ultrasonication for 10 min. The extracts were concentrated by a rotary evaporator under vacuum, blown down with dry nitrogen gas, and reacted with 50 μl of N,O-bis-(trimethylsilyl)trifluoroacetamide (BSTFA) with 1% trimethylsilyl chloride in the presence of 10 μl of pyridine at 70 °C for 3 h. This procedure derivatizes COOH− and OH− groups to the corresponding trimethylsilyl (TMS) esters and ethers, respectively. After reaction, the derivatives were diluted by adding 140 μl of *n*-hexane that contains 1.43 ng μl^{-1} of internal standard (C_{13} *n*-alkane) prior to GC–MS injection.

GC–MS analyses of the samples were performed on a Hewlett–Packard model 6890 GC coupled to Hewlett–Packard model 5973 mass-selective detector (MSD). The GC was equipped with a split/splitless injector and a DB-5MS fused silica capillary column (30 m × 0.25 mm i.d., 0.25 μm film thickness). The mass spectrometer was operated in the electron impact (EI) mode at 70 eV
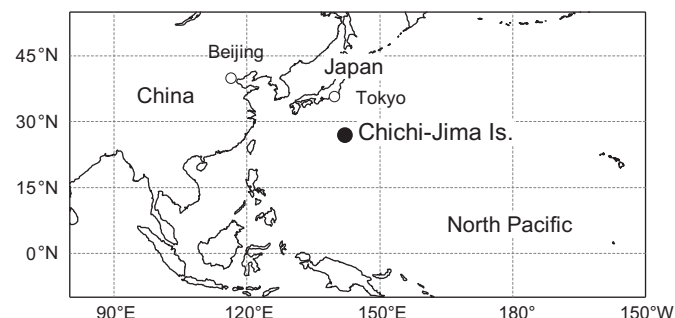

Fig. 1. Map of sampling site (Chichi-Jima Island) in the western North Pacific.

Table 1
Concentrations (ng m^{-3}) of saccharides measured in the marine aerosols collected at Chichi-Jima Island in the western North Pacific during 1990–1993.

Compounds	Winter		Spring		Summer		Autumn	
	(Dec–Feb, n = 12)		(Mar–May, n = 20)		(Jun–Aug, n = 21)		(Sept–Nov, n = 16)	
	Mean	Range	Mean	Range	Mean	Range	Mean	Range
Anhydrosugars								
Galactosan	0.054	0.008–0.13	0.036	0.002–0.14	0.012	0.001–0.043	0.010	0.001–0.06
Mannosan	0.18	0.023–0.41	0.18	0.009–0.65	0.054	0.002–0.22	0.055	0.002–0.23
Levoglucosan	1.35	0.41–2.78	0.79	0.023–4.77	0.37	0.04–1.94	0.18	0.004–1.44
Sugar Alcohols								
Erythritol	0.21	0.07–0.32	0.62	0.14–3.04	1.22	0.11–3.35	0.51	0.02–1.52
Arabitol	0.70	0.07–1.80	3.06	0.20–11.9	3.08	0.57–9.36	1.68	0.28–6.69
Mannitol	0.92	0.12–2.66	2.90	0.16–11.9	4.00	0.21–15.2	2.07	0.32–8.36
Inositol	0.03	0.01–0.06	0.06	0.01–0.16	0.06	0.01–0.20	0.03	0.01–0.08
Sugars								
Xylose	0.19	0.05–0.39	0.19	0.05–0.48	0.14	0.01–0.35	0.06	0.01–0.19
Fructose	1.44	0.32–3.77	2.81	0.18–12.7	2.73	0.06–9.16	0.62	0.04–3.01
Glucose	1.99	0.44–3.99	5.47	0.83–17.6	5.60	0.23–16.5	1.69	0.15–8.69
Sucrose	2.67	0.02–14.2	1.66	0.01–13.2	0.89	0.001–5.68	0.24	0.002–1.54
Trehalose	0.47	0.05–1.11	0.99	0.01–2.91	1.20	0.001–7.43	0.27	0.001–1.36
Total	10.2	3.65–19.6	18.8	3.60–53.6	19.3	2.63–67.5	7.42	1.34–28.6

and scanned from 50 to 650 Da. Data were acquired and processed with the Chemstation software. Individual compounds were identified by comparison of mass spectra with those of authentic standards. Detailed quantification of the organic tracers has been mentioned elsewhere (Fu et al., 2008). Field blank filters were treated as the real samples for quality assurance. No target compounds were found in the blanks. Recoveries for the authentic standards that were spiked into pre-combusted quartz filters were generally better than 90%. Therefore, the data reported here were not corrected for recoveries. Relative standard deviation of the concentrations based on duplicate analysis was generally <15%.

Ten-day air mass back trajectories were calculated using the NOAA Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT4) model (http://www.arl.noaa.gov/ready/hysplit4.html). The starting height of the trajectories present in this study is 500 m a.s.l. Trajectories at 1000 m a.s.l. were also calculated; the horizontal patterns are in general similar to those at 500 m and thus were not shown here.

3. Results and discussion

The concentration ranges of saccharides in four seasons are summarized in Table 1 and Table 2 for the period of 1990–1993 and 2006–2009, respectively. The concentrations of total sugars during 1990–1993 (1.34–67.5 ng m^{-3}, mean 14.8 ng m^{-3}) are lower than those (3.64–247 ng m^{-3}, 38.7 ng m^{-3}) during 2006–2009. The concentrations of biomass burning tracers (galactosan, mannosan and levoglucosan), xylose, erythritol, inositol are similar between two periods. However, the mean concentrations of individual species such as arabitol, mannitol, sucrose and trehalose during 1990–1993 are lower than those during 2006–2009 (Table 1 and Fig. 2).

Fig. 3 presents the temporal variations of the total sugar concentrations in the Chichi-Jima aerosols. Although the concentrations of individual species varied significantly from sample to sample, we found clear seasonal patterns with higher values in warm seasons and lower values in cold seasons. The dominant species that contribute to the higher concentrations of saccharides in warmer seasons are mainly arabitol, mannitol, fructose and glucose (Table 1 and Table 2). These primary saccharides can be abundantly emitted as primary biogenic aerosol particles, that is, fungal spores, pollen, bacteria, and plant fragments (Elbert et al., 2007).

Interestingly, a gradual decrease in the saccharide concentrations was found from 1991 to 1993 whereas a gradual increase was detected from 2006 to 2009. The local ambient temperature showed strong seasonal patterns with no clear inter-annual trends, which suggests that the temperature was not the main contributor

Table 2
Concentrations (ng m^{-3}) of saccharides measured in the marine aerosols collected at Chichi-Jima Island in the western North Pacific during 2006–2009.

| Compounds	Winter		Spring		Summer		Autumn	
	(Dec–Feb, n = 47)		(Mar–May, n = 52)		(Jun–Aug, n = 52)		(Sept–Nov, n = 52)	
	Mean	Range	Mean	Range	Mean	Range	Mean	Range
Anhydrosugars								
Galactosan	0.055	0.001–0.33	0.019	0.002–0.08	0.009	0.001–0.13	0.007	0.001–0.07
Mannosan	0.21	0.006–1.65	0.089	0.003–0.40	0.03	0.002–0.41	0.026	0.003–0.21
Levoglucosan	1.25	0.04–5.20	0.50	0.012–2.53	0.22	0.019–1.43	0.24	0.006–1.14
Sugar Alcohols								
Erythritol	0.31	0.07–0.83	0.33	0.09–0.96	1.65	0.18–4.50	1.15	0.06–5.97
Arabitol	2.11	0.12–21.2	4.67	0.24–21.6	11.4	1.98–45.9	12.9	0.69–83.3
Mannitol	2.52	0.18–24.0	5.23	0.16–31.0	19.4	1.72–88.0	16.2	1.31–84.3
Inositol	0.03	0.01–0.19	0.06	0.01–0.32	0.17	0.03–0.47	0.12	0.02–0.42
Sugars								
Xylose	0.15	0.01–0.76	0.11	0.01–0.31	0.18	0.02–1.02	0.11	0.01–0.55
Fructose	1.20	0.17–5.85	2.07	0.27–12.2	4.14	0.89–14.8	2.63	0.22–7.70
Glucose	2.03	0.27–6.97	4.67	0.79–13.7	8.47	1.94–26.9	6.34	0.75–35.3
Sucrose	3.50	0.02–58.8	10.2	0.01–87.4	6.97	0.02–41.1	3.36	0.002–31.1
Trehalose	1.09	0.13–10.5	2.51	0.29–21.9	5.93	0.34–28.0	5.72	0.04–33.9
Total	14.5	4.12–86.9	30.5	5.18–117	58.5	12.7–196	48.8	3.64–247
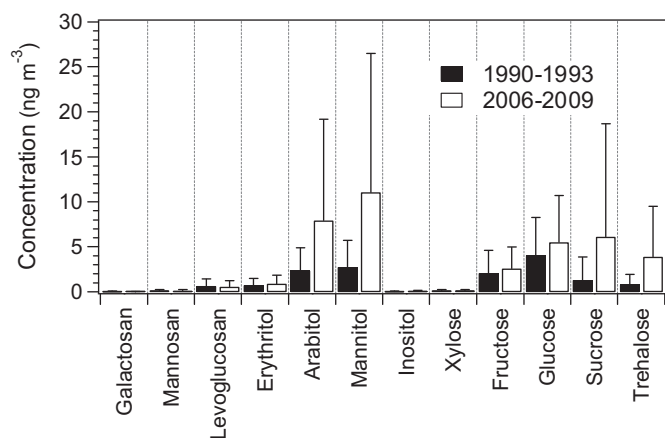
Fig. 2. Averaged concentrations of saccharides in atmospheric aerosols collected at Chichi-Jima Island in the western North Pacific during the 1990–1993 and 2006–2009 periods. Error bars represent the standard deviation.

to such annual variations because there were no significant differences observed during the two sampling periods (Fig. 3). However, the monthly averaged temperatures in 1992 were obviously lower than in 1991. The 1991 volcanic eruption of Pinatubo, which is located in the Philippines, has led to a decrease in average temperatures of 0.5–0.6 °C in the Northern Hemisphere and a global fall of about 0.4 °C in the following years (http://en. wikipedia.org/wiki/Mount_Pinatubo). Lower ambient temperatures may lower the primary biological activities such as the emissions of fungal spores. Thus, the decrease of temperature by such a powerful volcanic eruption may in part contribute to the decreasing concentrations of sugar in the western North Pacific from 1991 to 1993.

3.1. Seasonal variations of anhydrosugars

Levoglucosan and its isomers, galactosan and mannosan, are specific biomass burning tracers in the atmosphere (Simoneit, 2002). These anhydrosugars are derivatives of glucose, galactose, and mannose, respectively (Simoneit et al., 1999; Graham et al., 2003). Their concentration levels and seasonal trends are similar between the 1990–1993 (Fig. 4a–c) and 2006–2009 periods (Fig. 4m–o), indicating that the impacts of biomass burning on the marine aerosols in the recent years were similar to those in early 1990s. Levoglucosan was detected in all the samples with

concentration ranges of 0.004–4.77 ng m^{-3} (0.62 ng m^{-3}) from 1990 to 1993 (Fig. 4c) and 0.006–5.20 ng m^{-3} (0.54 ng m^{-3}) from 2006 to 2009 (Fig. 4o). These values are similar to those reported in marine aerosols over the western North Pacific (0.01–0.52 ng m^{-3}) (Fu et al., 2011) and Chichi-Jima (0.017–15 ng m^{-3}) from 2001 to 2003 (Mochida et al., 2010), representing a background level of levoglucosan in the marine atmosphere.

The seasonally averaged concentrations of levoglucosan are higher in winter (mean 1.35 ng m^{-3}) than in spring (0.79 ng m^{-3}), summer (0.37 ng m^{-3}) and autumn (0.18 ng m^{-3}) during the 1990–1993 period. Similar atmospheric levels and seasonal trends were found during the 2006–2009 period. The seasonal variations of galactosan and mannosan were concurrent to that of levoglucosan. During the 1990–1993 period, the concentration ranges of galactosan and mannosan are 0.001–0.14 ng m^{-3} (mean 0.026 ng m^{-3}) (Fig. 4a) and 0.002–0.66 ng m^{-3} (0.11 ng m^{-3}) (Fig. 4b), respectively. Similarly, their concentration ranges are 0.001–0.34 ng m^{-3} (0.022 ng m^{-3}) (Fig. 4m) and 0.002–1.65 ng m^{-3} (0.085 ng m^{-3}) (Fig. 4n) during 2006–2009. Being similar to levoglucosan, lower concentrations of galactosan and mannosan were generally observed in the samples collected in summer and autumn.

The seasonal variations of anhydrosugars may be controlled by the Asian monsoon system that transports organic aerosols from East Asia to the western North Pacific. In summer, maritime air masses arrived Chichi-Jima under the influence of the Pacific anticyclone. In winter, westerly or monsoon northwesterly winds caused by the Siberian anticyclone dominate East Asia and the western North Pacific, transporting air masses from East Asia to Chichi-Jima (Kawamura et al., 2003; Mochida et al., 2010). Ten-day air mass backward trajectories (Fig. 5) calculated for the sampling period support the above-mentioned transport pathways. As expected, the concentrations of anhydrosugars are high when air masses are transported from northeast China, east Siberia, Korea and Japan during winter/spring seasons. Lower concentrations were observed more frequently when air parcels were delivered from the Pacific Ocean in summer to autumn.

3.2. Seasonal variations of sugars and sugar alcohols

Saccharides have been thought as unique molecular tracers for primary biological aerosol particles (PBAPs) such as pollens and fungal spores (Medeiros et al., 2006). In general, glucose, inositol, sucrose, and trehalose were determined in various representative soils as the common tracers for source evaluation of soil resuspension into the atmosphere (Simoneit et al., 2004a), whereas
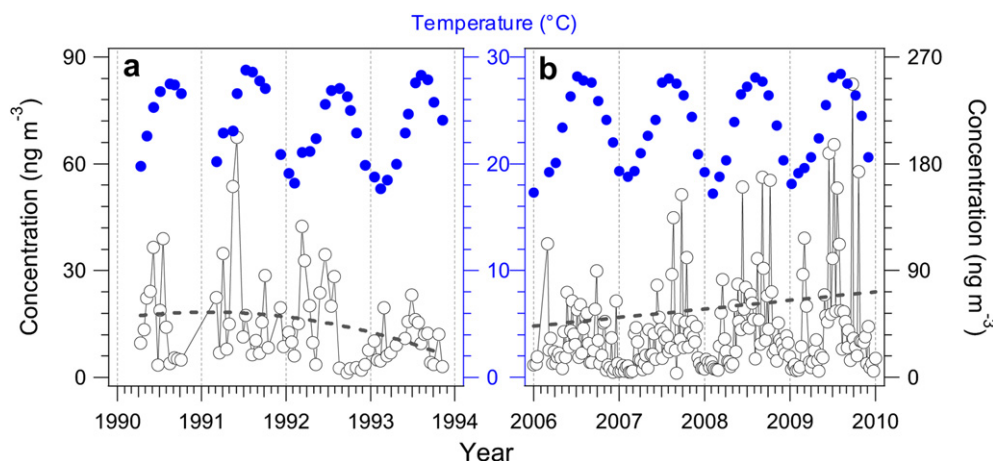

Fig. 3. Temporal variations in the concentrations of total saccharides measured in the marine aerosols collected from Chichi-Jima Island, the western North Pacific during (a) 1990–1993, and (b) 2006–2009. The monthly-averaged ambient temperatures are present in the figure.
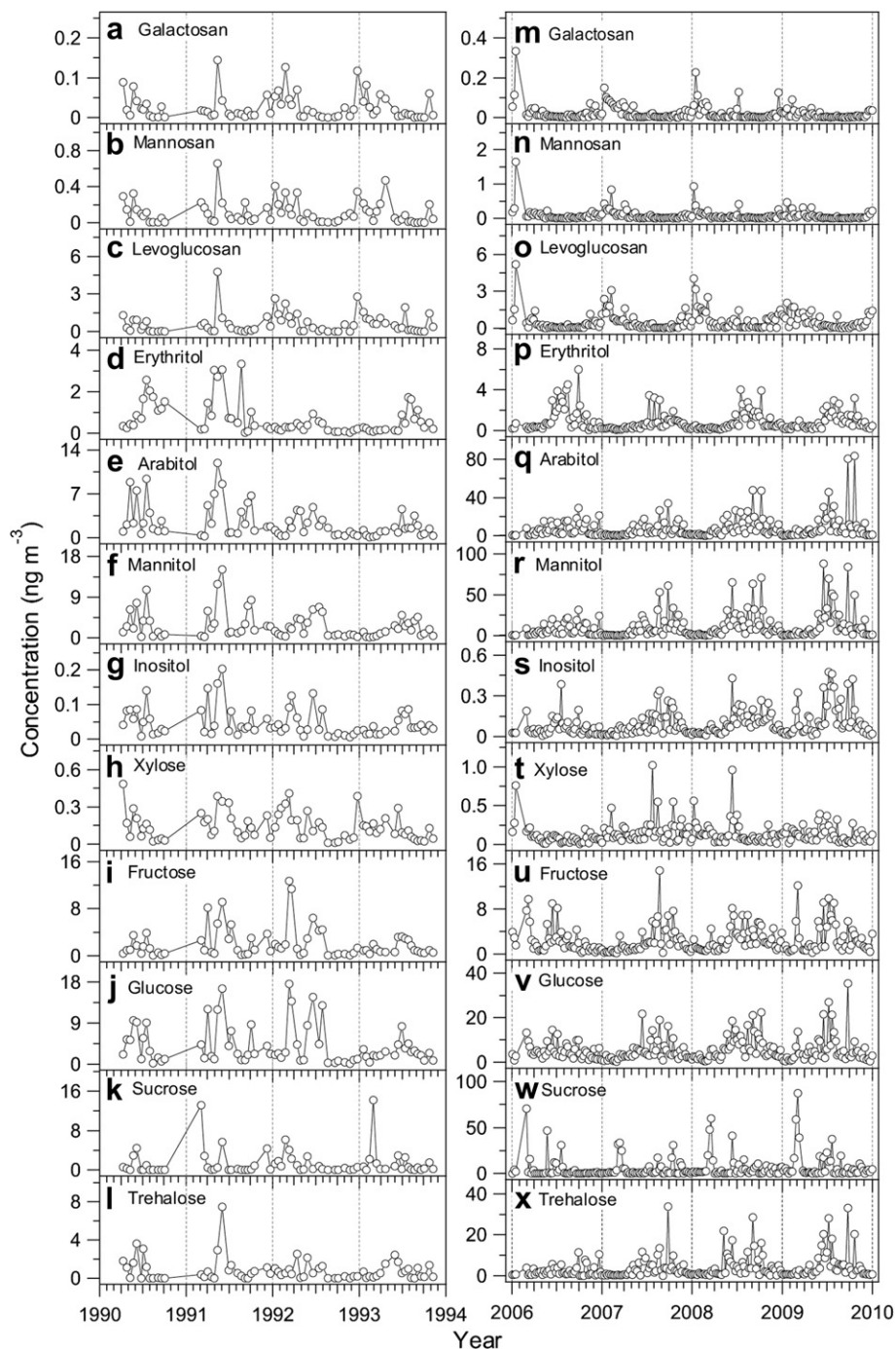
Fig. 4. Temporal variations of anhydrosugars (galactosan, mannosan and levoglucosan), sugar alcohols (erythritol, arabitol, mannitol and inositol), and sugars (xylose, fructose, glucose, sucrose and trehalose) in the marine aerosols from Chichi-Jima Island, the western North Pacific.

arabitol and mannitol are linked to vegetation, mature leaves, fungi and algae (Pashynska et al., 2002). Fig. 2 shows the 4-year averaged concentrations of saccharides in the Chichi-Jima aerosols during the 1990–1993 and 2006–2009 periods. Sugars (fructose, glucose, sucrose and trehalose) and sugar alcohols (erythritol, arabitol and mannitol) were found to be more abundant than anhydrosugars in both periods. During the 1990–1993 period, glucose was found to be the dominant species, followed by mannitol, arabitol, and fructose. During the 2006–2009 period, however, mannitol was the most abundant species, followed by arabitol, sucrose, glucose, trehalose and fructose. A general lack of correlation between the

concentrations of sugars/sugar alcohols and anhydrosugars (Table 3) suggests that sugars and sugar alcohols should be emitted from other sources rather than biomass burning.

The monthly averaged concentrations of sugar alcohols (erythritol, arabitol, mannitol, and inositol) and sugars (fructose, glucose and trehalose) were found to be higher in spring/summer, and lower in winter/autumn during both the periods of 1990–1993 and 2006–2009, whereas sucrose maximized in winter during 1990–1993 but did in spring during 2006–2009, and became the lowest in autumn (Table 1 and Table 2). Arabitol and mannitol are the most abundant sugar alcohols detected in the Chichi-Jima
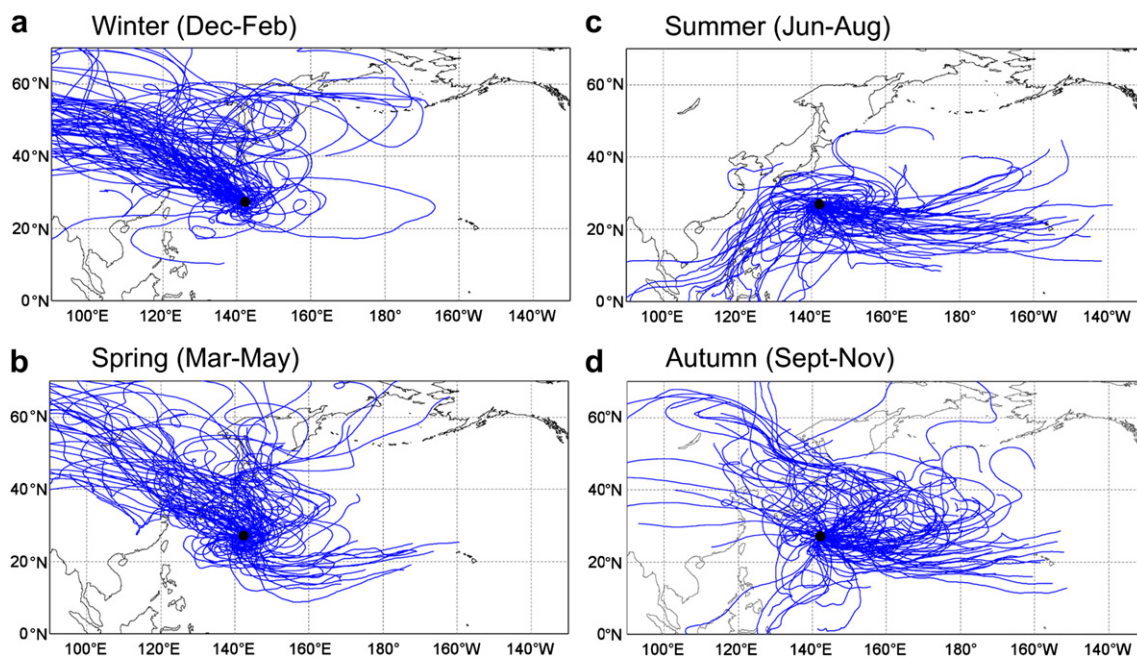
Fig. 5. Ten-day back trajectories of air masses that arrived over the Chichi-Jima Island in (a) winter (December 2008–February 2009), (b) spring (March–May, 2009), (c) summer (June–August, 2009), and (d) autumn (September–November, 2009). The trajectories were calculated every 24 h at 12:00 (Local Time).

samples, ranging from 0.066 to 11.9 ng m^{-3} (mean 2.34 ng m^{-3}) and 0.12–15.2 ng m^{-3} (2.70 ng m^{-3}) during 1990–1993 versus 0.12–83.3 ng m^{-3} (7.90 ng m^{-3}) and 0.16–88.0 ng m^{-3} (11.0 ng m^{-3}) during 2006–2009, respectively. The summertime mean concentrations of arabitol (11.4 ng m^{-3}) and mannitol (19.4 ng m^{-3}) during 2006–2009 are comparable to those (mean 8.43 ng m^{-3} and 21.9 ng m^{-3}) reported in the Mediterranean aerosols (Burshtein et al., 2011), but slightly lower than those (24 ng m^{-3} and 30 ng m^{-3}) at Hyytiälä, Finland in summer (Yttri et al., 2011).

Fructose and glucose are also abundant with concentration ranges of 0.04–12.7 ng m^{-3} (2.04 ng m^{-3}) and 0.15–17.6 ng m^{-3} (4.03 ng m^{-3}) during 1990–1993 versus 0.17–14.8 ng m^{-3} (2.54 ng m^{-3}) and 0.27–35.3 ng m^{-3} (5.46 ng m^{-3}) during 2006–2009, respectively. A positive correlation was found between fructose and glucose ($R > 0.70$) (Table 3). Similar relations between sugars and sugar alcohols have been reported in other studies (Burshtein et al., 2011). Pashynska et al. (2002) found that the concentrations of fructose, glucose and sucrose were highest in June, while both arabitol and mannitol peaked in late summer. In a forest site in suburban Sapporo, Japan, sucrose concentrations peaked in

spring/early summer (May–June, mean 176 ng m^{-3}) due to pollen emission, whereas trehalose levels (128.5–188.3 ng m^{-3}) remained high during summer and autumn (Miyazaki et al., 2012); both sucrose and trehalose in the Sapporo forest aerosols were about 2–7 times more abundant than those in the Chichi-Jima aerosols.

Xylose (wood sugar) is a monosaccharide first isolated from wood (http://en.wikipedia.org/wiki/Xylose). It is the main building block of hemicellulose, one of the main constitutes of biomass. In this study, its concentrations were found to be lowest in autumn (mean 0.06 ng m^{-3} in 1990–1993 and 0.11 ng m^{-3} in 2006–2009) compared to other seasons (Tables 1 and 2 and Fig. 6f). Sullivan et al. (2011) reported higher concentrations of xylose in winter (mean 2.1 ng m^{-3}) than in summer (mean 0.5 ng m^{-3}) in Detroit, Michigan, USA, and interpreted due to biomass burning activities in winter. Interestingly, the positive correlations between xylose and anhydrosugars during 1990–1993 were found to be stronger ($R = 0.61$–0.68) than those ($R = 0.36$–0.43) during the 2006–2009 period (Table 3). This may suggest that xylose was more significantly derived from wood burning smokes in the early 1990s than in more recent years.

Table 3
Pearson correlation coefficients (R) for the dataset of sugars in aerosols collected during 1990–1993 ($n = 69$) and during 2006–2009 ($n = 203$). The R-values >0.5 are displayed in bold character with those of $R > 0.8$ being underlined.

| 2006–2009	1990–1993											
	Galactosan	Mannosan	Levoglucosan	Erythritol	Arabitol	Mannitol	Inositol	Xylose	Fructose	Glucose	Sucrose	Trehalose
Galactosan	1	**0.85**	**0.86**	−0.01	0.14	0.11	0.20	**0.68**	0.17	0.18	0.17	0.32
Mannosan	**0.81**	1	**0.83**	−0.03	0.17	0.15	0.24	**0.63**	0.16	0.20	0.19	0.38
Levoglucosan	**0.82**	**0.87**	1	0.03	0.19	0.19	0.28	**0.61**	0.24	0.22	0.20	0.35
Erythritol	−0.24	−0.22	−0.29	1	**0.63**	**0.53**	0.48	0.01	0.20	0.29	−0.12	0.33
Arabitol	−0.25	−0.24	−0.30	**0.54**	1	**0.88**	**0.70**	0.17	0.33	**0.55**	−0.08	0.43
Mannitol	−0.27	−0.25	−0.31	**0.51**	**0.85**	1	**0.78**	0.25	0.46	**0.66**	−0.01	**0.63**
Inositol	−0.26	−0.24	−0.27	0.47	**0.54**	**0.72**	1	0.42	**0.77**	**0.86**	0.23	**0.57**
Xylose	0.36	0.44	0.43	−0.11	−0.12	0.10	0.22	1	0.51	0.49	0.38	0.49
Fructose	−0.20	−0.17	−0.20	0.40	0.38	**0.57**	**0.71**	0.30	1	**0.90**	0.27	0.41
Glucose	−0.24	−0.21	−0.26	0.44	**0.73**	**0.84**	**0.71**	0.18	**0.74**	1	0.19	**0.55**
Sucrose	−0.05	−0.04	−0.01	−0.03	−0.07	0.05	0.37	0.20	0.43	0.21	1	0.21
Trehalose	−0.22	−0.21	−0.24	0.33	**0.79**	**0.87**	**0.63**	0.12	0.47	**0.77**	0.07	1
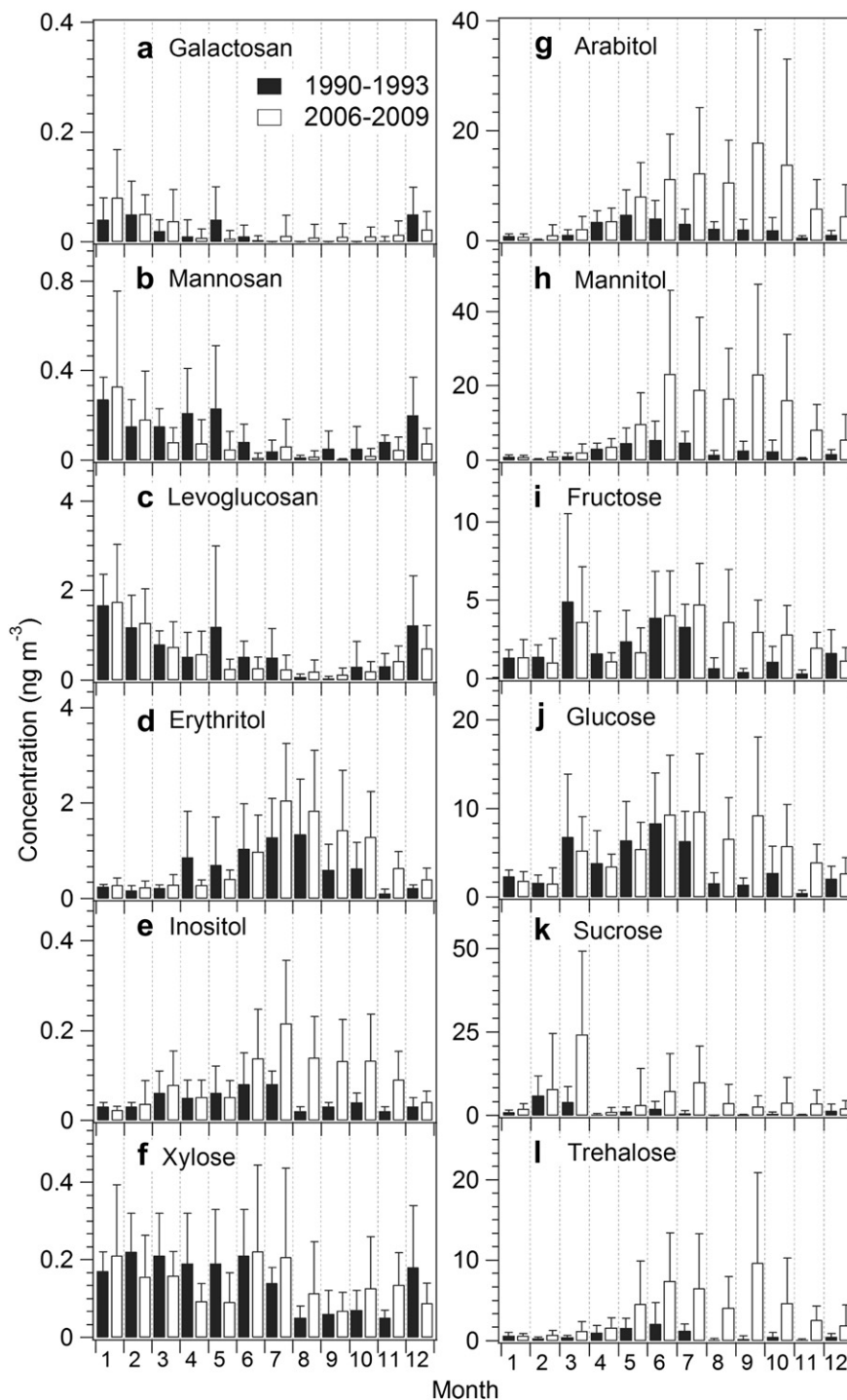
Fig. 6. Monthly averaged concentrations of saccharides in the marine aerosols collected from Chichi-Jima Island. Error bars represent the standard deviation.

The seasonal pattern of sucrose is characterized by two sharp increases in February–March and June–July (Figs. 4k, w, and 6k), which is different from the patterns of other saccharides. Such an enhancement should be influenced by the local emission of airborne pollen grains that contain a large amount of sucrose (Fu et al., 2012). However, long-range atmospheric transport of pollen from the Asian continent to the remote island Chichi-Jima under the influence of westerlies cannot be excluded. Previous studies reported that airborne pollen can be transported from North America to Greenland in spring (Rousseau et al., 2008).

The temporal variations of sugar alcohols (erythritol, arabitol, mannitol, and inositol) are similar each other, which are characterized by concentration peaks during warmer seasons (see Figs. 4d–g, p–s and 6d–e, g–h). Positive correlations ($R > 0.47$) were found among these sugar alcohols (Table 3), indicating that they are mainly derived from fungal spores during warm and humid seasons. Fructose and glucose also showed concentration peaks in warm seasons during both 1990–1993 and 2006–2009 periods (Fig. 4i–j and u–v). As seen in Fig. 6, the monthly aver-aged concentrations of fructose and glucose from January to May in

1990–1993 are higher than those in 2006–2009. However, opposite trends were found in July to November; that is, their concentrations are much higher in 2006–2009 than in 1990–1993.

Such a contrast is more significant for arabitol, mannitol, inositol and trehalose (Fig. 6). For example, the monthly averaged concentrations of arabitol and mannitol peaked in early summer (May–June) during 1990–1993, whereas they remained at high levels from June to September during 2006–2009. Such a difference in the seasonal variation may be attributable to the potential shift in westerly and trade wind regimes although we could not specify the changes in climate systems. In addition to the changes in climatological conditions during the past two decades, another possible reason is that the local vegetation coverage may have been changed at Chichi-Jima Island, a point that warrants further support. Leck and Bigg (2005) have reported that the marine bubble bursting can create aerosols containing microorganisms and lead to primary biological particles in the atmosphere above the central Arctic Ocean. Such a creation of primary bioaerosols may

also contribute to the marine aerosols collected at the remote Chichi-Jima Island in the North Pacific Ocean.

3.3. Positive matrix factorization (PMF) analysis

To better understand the possible sources of the observed saccharides, positive matrix factorization (PMF3.0, USEPA) analysis was used in this study. PMF is a useful approach to verify underlying covariance among chemical parameters (Paatero and Tapper, 1994). The analytical errors estimated for the measured values of saccharides in PMF analysis are 15% uncertainty. Based on Q values (the objective function to be minimized), five interpretable factors appeared to be the optimal solution as to the probable origin of sugars in the atmosphere.

Fig. 7 shows composition profiles for the five factors resolved by PMF. Factor 1 is dominated by sucrose (90.6% during 1990–1993 and 91.3% during 2006–2009), which should be associated with airborne pollen grains (Pacini, 2000; Graham et al., 2003; Fu et al.,
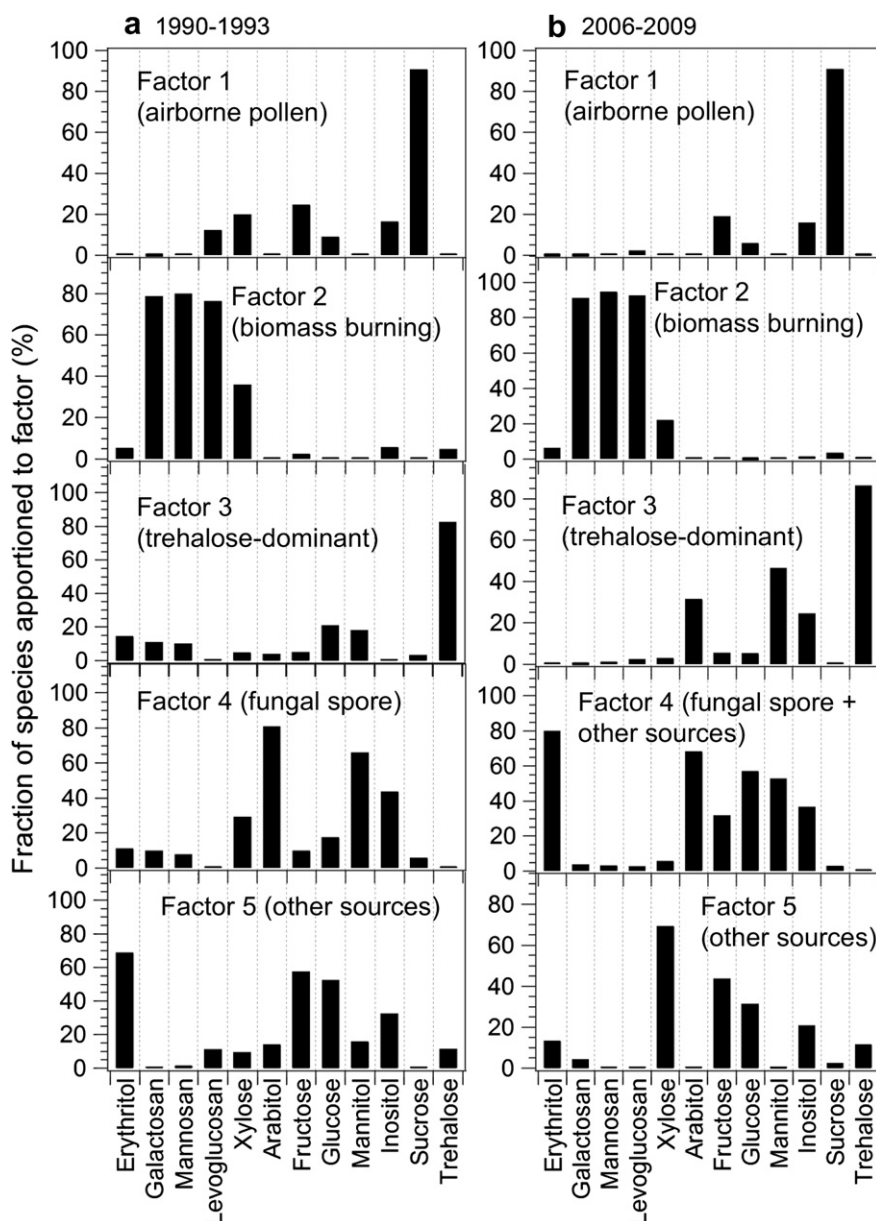

Fig. 7. Composition profiles (% of total of each species) for the five factors resolved by PMF analysis based on the dataset of saccharide compounds.

2012). Factor 2 is characterized by galactosan (78.8%, 91.3%), mannosan (79.9%, 94.7%), levoglucosan (76.5%, 92.5%), and xylose (36.0%, 22.1%) during 1990–1993 and 2006–2009, respectively, which should be associated with biomass burning sources. As mentioned earlier, smoke plumes from biomass burning are often transported over the Chichi-Jima site from the Asian continent during the colder winter period rather than in summer under the control of the Asian monsoon system by long-range atmospheric transport (Kawamura et al., 2003; Mochida et al., 2010). However, factor 2 shows different percent values with the same resource between 1990–1993 and 2006–2009.

As shown in Fig. 4, sharp increases in the concentrations of levoglucosan and its isomers are clearly found in mid-May 1991 (Fig. 4c) and mid-January 2006 (Fig. 4o). To further explain the source regions of the observed biomass burning tracers, 10-day backward air mass trajectory analyses were conducted for the two samples with the highest concentration of levoglucosan during 1990–1993 (sample ID of QFF212) and during 2006–2009 (QFF2871) (Fig. 8). The sample QFF212 was collected on 13–16 May 1991 when the air masses were mainly transported from the Central Pacific to the sampling site (Fig. 8). In the QFF212 sample, the concentrations of other sugars and sugar alcohols were also high, but such an enhancement of other sugars and sugar alcohols cannot be detected in the QFF2871 sample (Fig. 4). A possible explanation is that the QFF212 sample might be involved with a local fire event in Chichi-Jima in early summer; primary saccharides that are generally present in coarse modes can co-transport to the sampling site with smoke aerosols. A recent study also reported that the atmospheric levels of sugar alcohols (arabitol and mannitol) were enhanced by biomass burning events in an urban site in China (Yang et al., 2012). The back trajectory analysis of QFF2871 showed that most of the air masses originated from the arid regions in China, Mongolia and Siberia and then transported to the sampling site through long-range atmospheric transport (Fig. 8), leading to the highest concentration of levoglucosan during 2006–2009 (Fig. 4o).

Factor 3 is dominated by trehalose (82.4% during 1990–1993 and 86.3% during 2006–2009), which is proposed as a tracer for biologically suspended soil dust (Simoneit et al., 2004a; Rogge et al., 2007), and should be associated with fungal metabolism. During 1990–1993, factor 4 is associated with arabitol (80.9%), mannitol (66.1%), and inositol (43.9%), indicating an influence of fungal spores. During 2006–2009, factor 4 is characterized by erythritol (79.9%), arabitol (68.4%), fructose (31.7%), glucose (57.2%), mannitol (52.7%), and inositol (36.8%), suggesting the mixing source emissions from fungal spores and growing plant and so on.

During 1990–1993, factor 5 includes erythritol (68.8%), fructose (57.5%), glucose (52.3%) and inositol (32.7%) (Fig. 7a). During 2006–2009, however, factor 5 includes xylose (69.3%), fructose (43.6%), glucose (31.5%), and inositol (20.9%) (Fig. 7b). Therefore, factor 5 was mainly associated with other sources, for example, the emission of primary saccharides such as fructose and glucose during the growing seasons.

As resolved by the PMF analysis, the average contributions of each factor to the measured saccharides in 1990–1993 and 2006–2009 are shown in Fig. 9. The emission of fungal spore (factor 4) was found to be the dominant contributor to the sugar compounds in both periods, although the average contribution was higher in 2006–2009 (40.4%) than 1990–1993 (33.2%). Similarly, the contribution of airborne pollen was 17.2% in 2006–2009, higher than that (13.9%) in 1990–1993. The contribution of biomass burning to total sugars was only 5.3% in 1990–1993; even a lower value (2.5%) was found in 2006–2009. These results suggest that the emission of primary biological aerosols is the dominant source of water-soluble saccharides, while the contribution of biomass burning was minor in the remote marine atmosphere.

Pearson correlation coefficients (Table 3) are relatively high among the concentrations of sugars/sugar alcohols with a maximum ($R = 0.90$) for the combination of fructose and glucose during 1990–1993. Such a good covariance suggests that terrestrial organic materials are transported over the island from the similar source regions, but the source regions should have been changed according to seasons and meteorological conditions (Kawamura et al., 2003). The years of 1991 and 1992 were also characterized by a strong El Niño event, during which the air over the western equatorial Pacific turned dry and hot, leading to frequent forest fires in Southeast Asia (Kita et al., 2000). These situations may partly interpret the different distribution patterns of the five factors resolved by PMF between the two sampling periods.

Significantly high concentrations of arabitol and mannitol were detected in the aerosol samples collected on 24–25 September 2009 (QFF3267) (Fig. 4q–r), which contributed to the maximum concentration of total sugars during 2006–2009 (Fig. 3b). Relatively high levels of total saccharides in summer–autumn seem to be connected with the diversity of wind directions and source regions. During 2006–2009, high correlation coefficients were found among the concentrations of arabitol, mannitol, glucose and trehalose (Table 3). The aerosol samples collected in autumn are
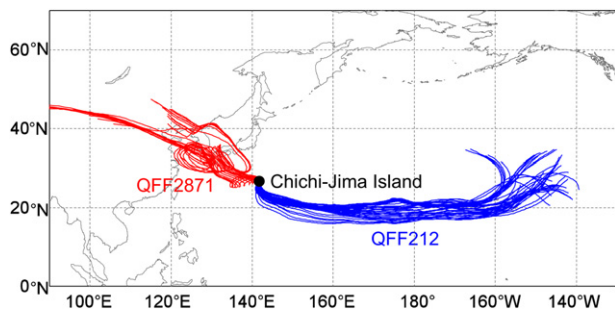

Fig. 8. Ten-day back trajectories of air masses that arrived over Chichi-Jima Island during 13–16 May 1991 (QFF212) and during 16–17 January 2006 (QFF2871). The trajectories were calculated every 3 h. The concentrations of levoglucosan in the QFF212 and QFF2871 samples were the highest during 1990–1993 and 2006–2009, respectively.
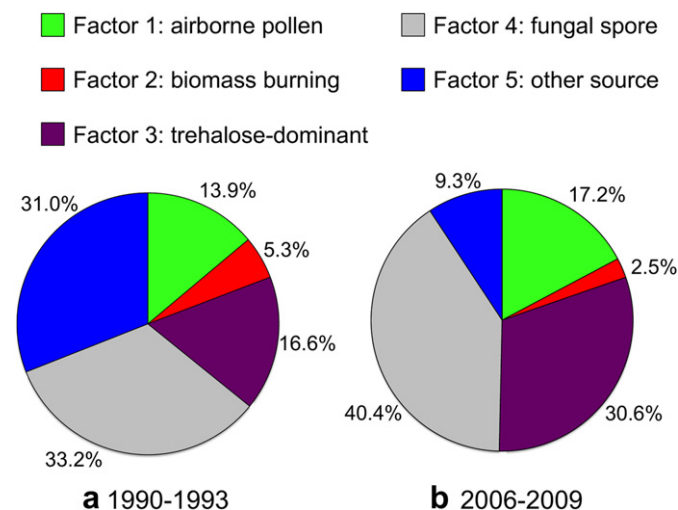

Fig. 9. Pie diagrams showing the estimated average contributions of the five factors resolved by PMF to the measured saccharide compounds in the Chichi-Jima aerosol samples.

heavily influenced by trade winds, whose pathways cover most of the Central North Pacific and a few other source regions such as Southeast Asia and Asian continents (Fig. 5d). Correlation coefficients of glucose and fructose also showed higher values in warmer seasons annually (Table 3). When sucrose is rich in spring aerosols, the atmospheric levels of glucose and fructose are also enhanced, indicating their similar pollen sources (Fu et al., 2012). As a result, it would be difficult to attribute factors 4 and 5 to a specific source (Fig. 7), given that these saccharides were possibly originated from either PBAPs or other sources such as resuspended soil/dust and associated biota (Graham et al., 2003; Simoneit et al., 2004a; Medeiros et al., 2006).

4. Conclusions

Long-term variations of anhydrosugars (galactosan, mannosan and levoglucosan), sugars (xylose, fructose, glucose, sucrose and trehalose), and sugar alcohols (erythritol, arabitol, mannitol and inositol) in marine aerosols were obtained at Chichi-Jima Island in the western North Pacific with distinct seasonality and variability. Although the atmospheric concentrations of anhydrosugars in 2006—2009 were similar to those in 1990—1993, the tracers of fungal spores, arabitol and mannitol, as well as other primary saccharides were much higher in late 2000s than those in early 1990s. The seasonal variations of anhydrosugars were characterized by the concentration peaks in winter/spring due to heavy biomass-burning activities in up-wind Asian continent followed by a long-range atmospheric transport and/or potential local fires in Chichi-Jima. Backward air mass trajectory analysis demonstrated that the air masses in winter/spring were influenced by westerlies and transported from East Asia to the western North Pacific, whereas the air masses in summer/autumn are delivered from the directions of the Central Pacific. The concentration maxima of sugar alcohols (erythritol, arabitol, mannitol, and inositol) were observed in May—June during 1990—1993, while the maxima were recorded in few months later (June—September) during the 2006—2009 periods. PMF analysis demonstrates that primary biological emissions such as pollen grains and fungal spores, trehalose-dominant emission, and biomass burning emission are the major sources of saccharides in the Chichi-Jima aerosols, although the contribution of biomass burning to the measured saccharides was rather minor in the remote marine atmosphere.

Acknowledgements

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