Thirteen years of observations on primary sugars and sugar alcohols over remote Chichijima Island in the western North Pacific

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Abstract. In order to understand the atmospheric transport of bioaerosols, we conducted long-term observations of primary sugars and sugar alcohols over remote Chichijima Island in the western North Pacific from 2001 to 2013. Our results showed that concentrations of total sugar compounds for 13 years ranged from 1.2 to 310 ng m^{-3} (average, 46±49 ng m^{-3}). We found that atmospheric circulations significantly affect the seasonal variations of bioaerosol distributions over the western North Pacific. The primary sugars (glucose and fructose) maximized in summer, possibly due to an increased emission of the vegetation products from local vascular plants in Chichijima. We also found higher concentrations of sugar components (arabitol, mannitol and trehalose) in more recent years during summer/autumn, suggesting an enhanced emission of fungal and microbial species over the island. Sucrose peaked in late winter to early spring, indicating a springtime pollen contribution by long-range atmospheric transport, while elevated concentrations of sucrose in early summer could be explained by long-range transport of soil dust from Southeast Asia to Chichijima. Sucrose and trehalose were found to present increasing trends from 2001 to 2013, while total sugar components did not show any clear trends during the thirteen years periods. Positive matrix factorization analyses suggested the locally emitted sugar compounds as well as long-range transported air borne pollen grains, microbes and fungal spores are the major contributors to total sugar compounds in the Chichijima aerosols. Backward air mass trajectories support the atmospheric transport of continental aerosols from the Asian continent during winter/spring over Chichijima.

Keywords: Sugar compounds, fungal and microbial tracer, pollen tracer, bioaerosols, the western North Pacific.

1 Introduction

East Asia has experienced rapid economic developments and population growth since last several decades (Elliot et al., 1997; Jaffe et al., 1999, 2003), whose activities emit organic and bioaerosols into the atmosphere (Xu et al., 2011). The atmospheric particles are transported to downwind region in the Pacific, associated with Asian desert dust from the Taklamakan and Gobi Deserts, and Loess plateau (Duce et al., 1980; Iwasaka et al., 1983; Jaffe et al., 1997; Prospero and Savoie, 1989; Talbot et al., 1997). The transported dust contains bacterial cells, fungal spores, and microbial cells, which fall out over the Pacific and remote islands in the Pacific Ocean (Lacey and West, 2006; Mims and Mims, 2003). The microbes associated with bioaerosols significantly affect the natural environment of marine and land ecosystem in downwind regions (Graffin et al., 2003, 2007; Prospero et al., 2005). Long-range atmospheric transport plays a key role for the global distribution of microbes from source regions to receptor site (Graffin et al., 2001). Fungi and bacteria are often attached to dust particles, which can propagate diseases to human and plants (Brown and Hovmoller, 2002). Therefore, the transported organic- and bio-aerosols have been the focus of extensive studies for the past years (Yamaguchi et al., 2012).

Organic aerosols are composed of a complex mixture of different types of molecules, in which water-soluble organic compounds (WSOCs) are enriched (Graham et al., 2002). WSOCs play an important role in climate change and global radiative forcing by scattering or absorbing light directly or indirectly (Fuzzi et al., 2007). They can act as cloud condensation nuclei (CCN) (Kanakidou et al., 2005; Martin et al., 2010). Sugar compounds (SCs) contribute 13–26% and 63% of total WSOCs identified in continental and marine aerosol samples, respectively (Simoneit et al., 2004a; 2004b). Yttri et al. (2007) reported that sugars (fructose, glucose, sucrose, trehalose) accounted for 0.6-3.1% of the WSOC at urban and suburban sites in Norway. Tominaga et al. (2011) analyzed aerosol samples collected from urban and forest suburban site from Japan and reported that the sugars (arabinose, fructose, galactose, glucose,

mannose, rhamnose, and xylose) accounted for 2.1% and 4.5% of the WSOC in the fine and coarse mode ranges at Yokohama, respectively, and for 3.0% and 7.2% at Mt. Oyama, respectively. SCs are directly emitted from biological sources such as fungi, algae, pollen, spores and bacteria (Carvalho et al., 2003; Wang et al., 2009) and transported long distances in the atmosphere (Wang et al., 2011). They are also derived from suspended soil particles and associated biota (Rogge et al., 2006; Simoneit et al., 2004b; Wang et al., 2009), and biomass burning (Schmidl et al., 2008; Simoneit et al., 2002).

Primary sugars are emitted from biological sources (Medeiros et al., 2006). Glucose and fructose are emitted from terrestrial plant fruits, pollen, and detritus of vascular plants (Cowie and Hedges, 1984; Speranza et al., 1997). Sucrose is dominant sugar component in airborne pollen grains and plays a significant role in plant blossoming activity (Bieleski, 1995; Fu et al., 2012; Pacini, 2000). Trehalose is emitted from fungal metabolic activities and resuspension of soil particles and unpaved road dust (Rogge et al., 2007; Simoneit et al., 2004). Sugar alcohols are also emitted from biological sources like fungi and microbes via metabolic activities (Bauer et al., 2008). Sugar alcohols, i.e., arabitol and mannitol, are tracers for fungal spores (Jia and Fraser; 2011; Yang et al., 2012). Di Filippo et al. (2013) reported that arabitol and mannitol are key sugar components in fungal spores.

Chichijima Island is located in the western North Pacific: an outflow region of Asian dust (Mochida et al., 2003a). It is one of the best remote islands to study a long-range transport of Asian aerosols, because local pollutants in Chichijima are insignificant due to low population density and no major sources from industrial or anthropogenic activities (Chen et al., 2013). Kawamura et al. (2003) reported that concentrations of lower molecular weight fatty acids (C_{12}-C_{19}) derived from marine organisms became higher in summer, while those of higher molecular weight fatty acids (C_{21}-C_{34}), n-alkanes (C_{25}-C_{35}), n-alcohols (C_{20}-C_{34}) and dicarboxylic acids (C_{20}-C_{28}) derived from terrestrial higher plants and soil organic matter maximized in winter to spring. Seasonal variations of low molecular weight (C_2-C_{10})

dicarboxylic acids and levoglucosan (biomass burning tracer) have also been reported in Chichijima aerosols by Mochida et al. (2003a) and Mochida et al. (2010), respectively. Although seasonal variation of saccharides was reported in Chen et al. (2013), the observation period is rather short.

Here, we report thirteen year data set of SCs in remote Chichijima Island. The goal of this study is to characterize seasonal and annual variations of SCs and specify their possible source regions. We will also discuss a potential role of Asian dust to control the distributions of bioaerosols over the western North Pacific. The outcomes of this study will improve our understanding about a possible influence of long-range transport of bioaerosols from the continent to the clean oceanic environment. We will compare the data set of SCs for the periods 1990-1993, 2001-2003 and 2010-2013, which may provide imperative information about decadal changes in the atmospheric conditions over Chichijima. Seasonal source identifications by positive matrix factorization (PMF) analysis will also be discussed for the measured SCs.

2 Materials and methods

2.1 Sampling site and meteorological conditions

The detailed information on the sampling site was reported in Kawamura et al. (2003) and Chen et al. (2013). Briefly, Chichijima Island is located in the western North Pacific (27°04'N; 142°13'E), 1000 km south of Tokyo, Japan, and 2000 km east of the Asian continent (Figure 1). Total area of the island is 24 km^2 with a population of 2000 (Verma et al., 2015). The climate of Chichijima is classified as subtropical; it is warm to hot (temperature, 7.8-34.1 °C) and humid (relative humidity, 66-88%) all year round.

Figure 2 shows monthly averaged variations in the meteorological parameters of Chichijima during 2001-2013. It receives more precipitation in between April and July, September and October during the thirteen year period. The sampling site is less influenced by

123 the East Asian monsoon to receive heavy rainfall compared to Northeast Asia. The climate
124 over Chichijima is strongly influenced by the seasonal changes in wind system. In
125 winter/spring, the westerly winds are dominant with the air masses being enriched with Asian
126 dust, industrial pollutants, biomass burning products, organic compounds and black carbon as
127 well as bioaerosols emitted from East Asia and Eurasia (Figure 3) (Seinfeld et al., 2004;
128 Simoneit et al., 2004b; Wang et al., 2009). Trade winds are dominant in summer/autumn,
129 which transport clean and pristine marine air masses from the central Pacific to Chichijima
130 (Kawamura et al., 2003, Mochida et al., 2010).

131 **2.2 Aerosol sampling and chemical analysis**

132 The details on aerosol sampling and chemical analysis are reported elsewhere (Chen et al.,
133 2013; Mochida et al., 2010). Briefly, total suspended particle (TSP) samples were collected at
134 the Ogasawara Downrange Station of the Japan Aerospace Exploration Agency (JAXA) in
135 Chichijima Island (254 m, above sea level, asl). The samples were collected on weekly basis
136 (January 2001 to November 2013) using a high-volume air sampler (Kimoto AS-810A) at a
137 flow rate of 1.0 m^3 min^{-1} and pre-combusted (450°C for 6 h) quartz fiber filters (20 x 25 cm,
138 Pallflex). Filter sample was placed in a pre-combusted glass jar with a Teflon-lined screw cap
139 and stored in a dark freezer room at -20 °C prior to analysis in order to inhibit fungal growth.
140 Due to the maintenance of the JAXA facility at sampling site, TSP samples were not collected
141 for November–December 2004 and March–August 2005.

142 Total 590 aerosol samples were analyzed to determine primary sugars (xylose, fructose,
143 glucose, sucrose and trehalose) and sugar alcohols (erythritol, arabitol, mannitol and inositol)
144 during 2001 to 2013. An aliquot (21 cm^2) of the filters were extracted three times with
145 dichloromethane/methanol (2:1, v/v) mixture using ultrasonic agitation for 10 minutes. A
146 Pasture pipette packed with quartz wool was used to remove particles and filter debris in the
147 extracts. Filtrates were then concentrated using a rotary evaporator under vacuum and blown
148 down with a stream of pure nitrogen gas. The total extracts were derivatized using 60 µl of

149 N,O-bis-(trimethylsilyl)trifluoroacetamide (BSTFA) with 1% trimethylsilyl chloride in the
150 presence of 10 μl of pyridine in a sealed vial at 70 °C for 3 hours to convert hydroxyl groups to
151 corresponding trimethylsilyl (TMS) ethers. The derivatized fractions were diluted with n-
152 hexane containing internal standard of C_{13} n-alkane (1.43 ng μl^{-1}), prior to injection to gas
153 chromatography-mass spectrometer (GC-MS).

154 Identification of SCs have been confirmed by the comparison of GC retention times and
155 mass spectra with those of authentic standards as well as literature and library data. SCs were
156 characterized by their common base peak at m/z 217 and 204 with specific fragment ions for
157 individual sugars, i.e., m/z = 307 (arabitol), 205 and 319 (mannitol), 205 (erythritol), 305 and
158 318 (inositol), 361 (sucrose and trehalose), 191 (glucose), and 437 (fructose). The selected ion
159 peak area and relative response factors determined by injection of authentic standards have
160 been used for the quantification of sugar compounds. Field blank filters were analyzed as a real
161 sample, but no target compounds were detected in the field blanks. The recoveries of the target
162 compounds were better than 90%. Therefore, the data reported here were not corrected for
163 recoveries. Analytical errors of SCs were generally <15% based on duplicate analysis. The
164 detection limits of primary sugars and sugar alcohols were 105-557 pg μl^{-1}, which corresponds
165 to ambient concentrations of 0.0015-0.0081 ng m^{-3} under a typical sampling volume of 9000
166 m^3 (Zhu et al., 2015).

167 The derivatized fractions were introduced into GC-MS using an Agilent model 7890 GC
168 coupled to an Agilent model 5975 mass selective detector (MSD) operated in an electron
169 impact mode at 70 eV and scanned from 40 to 650 Dalton. The GC separation was carried out
170 on a DB-5MS fused silica capillary column (30 m long, 0.25 mm i.d., 0.25 μm film thickness),
171 with a temperature program of 50 °C for 2 min at a rate of 15 °C min^{-1} from 50 to 120 °C, then
172 from 120 to 305 °C at a rate of 5 °C min^{-1} with a final isotherm hold at 305 °C for 15 min. The
173 sample was injected on a splitless mode at an injector temperature of 280 °C. GC-MS data
174 were acquired and processed with the Agilent GC/MSD ChemStation software.

175 **2.3 Backward air mass trajectory analysis**

176 In order to identify the source regions of sugar compounds in Chichijima aerosols, ten-day
177 backward trajectories were calculated at 00:00 UTC of each sampling period for thirteen years
178 using the NOAA Hybrid Single-Particle Lagrangian Integrated Trajectory
179 (http://ready.arl.noaa.gov/HYSPLIT.php) (Figure 3). The starting height of the trajectories
180 presented in this study is 500 m asl. We plotted thirteen year trajectories for each sampling day
181 but there are no significant year-to-year changes in the atmospheric circulations. Therefore, we
182 presented seasonal trajectories for recent year (December, 2011 to November, 2012) in Figure
183 3 to understand the seasonal aerosol mass transport from the source regions to Chichijima
184 Island. Backward trajectories significantly supported a long-range transport of air mass under
185 the influence of existing meteorological parameters (Figure 3). The trajectories clearly show
186 the influences of continental air masses during mid-autumn to mid-spring and of marine air
187 masses during mid-spring to mid-autumn.

188 **2.4 Positive matrix factorization (PMF) analysis**

189 Positive matrix factorization (PMF 3.0, Environmental Protection Agency, USA) has
190 been used as a powerful statistical tool that may resolve potential sources contributing to
191 atmospheric levels of particle (as presented by %) when appropriate source profiles are not
192 available (Paatero and Tapper, 1994). At the beginning PMF has been used in precipitation
193 study (Juntto and Paatero, 1994) as well as air pollution and source apportionment studies
194 (Polissar et al., 1999). Recently, it is widely used for the air quality and source apportionment
195 (Xie and Berkowitz, 2006). In addition, PMF has been applied to the wastewater
196 (Soonthornnonda and Christensen, 2008), lake sediments (Bzdusek et al., 2006) and soils (Lu
197 et al., 2008). One of the main features of PMF results is their quantitative nature; it is possible
198 to obtain the composition of the sources determined by the model.

199 PMF analysis was performed for quantitative estimation of sources for the collected
200 samples using tracer compounds for primary sugars, sugar alcohols, and anhydrosugars. Based

on given understanding of sugar sources, 4-7 factors were examined and total five interpretable factors were characterized by the enrichment of each tracer compound, which reproduced more than 94% of SCs. Minimal robust and true Q values of the base run were 3001 and 3413, respectively. Concentrations and percentage of tracers in each factor of bootstrap run were close of those of base run results. The Q values and factor profiles of F_{peak} rotation runs showed no significant changes compared with base run, indicating stable PMF results. The detailed discussions of the determination and application of the PMF are reported in Norris et al. (2008), Paatero et al. (2002) and Zhou et al. (2004).

In winter/spring, Chichijima Island receives air masses enriched with anthropogenic aerosols from the Asian continent by strong westerly winds, whereas during summer/autumn it receives clean air masses from the Pacific Ocean under the influences of trade winds. The seasonal changes in the atmospheric circulation over Chichijima may have a significant influence on the seasonal distributions of SCs. Therefore, we performed the seasonal PMF analysis on the thirteen year sugar data set to better understand the seasonal source profile of individual sugar component. For seasonal PMF analysis, 3-5 factors were examined and 4 factors were determined for each season. We included the data set of anhydrosugars from Verma et al. (2015) for PMF analysis.

3 Results and discussion

3.1 Ambient concentrations of sugar compounds

Temporal variations of primary sugars and sugar alcohols are shown in Figure 4. Nine sugar compounds (SCs) including five primary sugars and four sugar alcohols were detected in the aerosol samples collected from Chichijima Island. The concentrations of total SCs varied from 1.23 to 339 ng m^{-3} (average, 46.7±49.5 ng m^{-3}) during 2001 to 2013 (Table 1). Concentrations of primary sugars and sugar alcohols were in the range of 0.28 to 176 ng m^{-3} (23.3±25.7 ng m^{-3}) and 0.37 to 231 ng m^{-3} (23.4±30.8 ng m^{-3}), respectively. Average concentration of primary

sugars in Chichijima aerosols is several times lower than that of primary sugars (62.0±54.9 ng m^{-3}) reported from Cape Hedo, Okinawa, Japan (Zhu et al., 2015) while that of sugar alcohols is equivalent to or little lower than that from Cape Hedo (29.5±35.5 ng m^{-3}).

Interestingly, primary sugars (49.9%) and sugar alcohols (50.1%) were found to contribute almost equal to total SCs during the entire study period. Mannitol (26.7%) and arabitol (21.4%) were the main contributors to total SCs followed by glucose (16.7%), sucrose (13.6%), fructose (10.2%), and trehalose (9.2%). Erythritol (1.6%), inositol (0.3%), and xylose (0.3%) were also present in the aerosols at lower concentration levels. Temporal plots of individual sugars clearly indicate a large variation of SCs (Figure 4). This large variation in the concentrations of SCs might be involved with seasonal changes in the atmospheric circulations over the western North Pacific (Kawamura et al., 2003).

3.1.1 Concentrations of primary sugars in total SCs

Glucose is the dominant sugar species among primary sugars with the concentration range of 0.05 to 64.3 ng m^{-3} (average, 7.79±8.80 ng m^{-3}). Similarly, a wide concentration range of fructose (0.03-115 ng m^{-3}; 4.69±8.04 ng m^{-3}) was also observed in Chichijima aerosols. Thirteen year mean concentrations of glucose and fructose were observed to be lower than those (27.2 ng m^{-3} and 16.4 ng m^{-3}, respectively) reported for the aerosol samples (TSP) from Cape Hedo, Okinawa, Japan (Zhu et al., 2015). Glucose and fructose significantly contribute to total primary sugars (33.5% and 20.17%, respectively) in Chichijima aerosols. Primary sugars are abundant in the fragments of vascular plants in vegetated and forest areas (Medeiros et al., 2006). Pacini et al. (2000) reported that primary sugars are synthesized in leaves during photosynthesis and stored in root, stem, flower, pollen and fruit of growing plants. The nectars and fruits of tropical and subtropical plants also contain glucose and fructose abundantly (Backer et al., 1998). Graham et al. (2002) reported significant amounts of glucose and fructose in pollen, fern spores, and insects in aerosol samples collected from the Amazon forest. Chichijima Island is covered with endemic and vascular plants, which may emit glucose and

253 fructose. Moreover, different sources such as soil dust (Rogge et al., 2007; Simoneit et al.,
254 2004), lichens (Dahlman et al., 2003) and biomass burning (Medeiros et al., 2006; Nolte et al.,
255 2001) have also been reported as dominate sources for glucose and fructose.

256 Among all the SCs detected in the Chichijima aerosols, sucrose is the second most
257 abundant sugar species (0.002-100 ng m^{-3} ; 6.43±12.9 ng m^{-3}), accounting for 27.3% of total
258 primary sugars. The average sucrose concentration observed in Chichijima is twice lower than
259 that (13.2 ng m^{-3}) from Cape Hedo, Okinawa, Japan (Zhu et al., 2015). Sucrose is synthesized
260 in plant leaves and circulated by phloem to different plant sections, which is accumulated in
261 root cells as well as developing flower buds (Bieleski, 1995; Jia et al., 2010). Sucrose is a
262 dominant component in airborne pollen grains derived from flowering plants (Bieleski, 1995;
263 Pacini, 2000). Simoneit et al. (2004a and 2004b) reported the presence of sucrose in surface
264 soil and paved road dust. Sucrose was also observed in dry plant materials during harvesting
265 period (Ma et al., 2009).

266 Thirteen year mean concentration of trehalose ranged from 0.01 to 70.2 ng m^{-3}
267 (4.30±7.28 ng m^{-3}), whose average concentration accounts for 18.4% of total primary sugars
268 detected in Chichijima aerosols for 13 years. Microbes (bacterial cell), fungal spores, yeast,
269 algae, invertebrates, suspended soil dust, as well as plant species, contribute significantly to
270 trehalose in the atmosphere (Elbein, 1974; Graham et al., 2003; Medeiros et al., 2006; Rogge et
271 al., 2007; Simoneit et al., 2004; Wiemken, 1990). Xylose is a less abundant primary sugar,
272 accounting for 0.60% of total primary sugars observed in Chichijima aerosols. The
273 concentration range of xylose was 0.001-1.35 ng m^{-3} (0.14±0.18 ng m^{-3}) during sampling
274 period of thirteen years. Biomass burning activities emit xylose to the atmosphere. Cowie and
275 Hedges (1984) reported that xylose is produced by angiosperm and gymnosperm plants,
276 phytoplankton, and groups of microorganisms. Simoneit et al. (2004a) have reported xylose in
277 soil dust from various locations in the United States and Japan. Wan and Yu (2007) also
278 observed xylose in soils and associated microbiota.

3.1.2 Concentrations of sugar alcohols in total SCs

Thirteen year mean concentrations of arabitol and mannitol were found to be 9.99±13.6 ng m^{-3} and 12.5±17.5 ng m^{-3}, which contribute to 42.7% and 53.3% of total sugar alcohols, respectively. The concentration ranges of arabitol (0.04–106 ng m^{-3}) and mannitol (0.10–118 ng m^{-3}) are comparable to those from the Mediterranean region, Israel (arabitol, 1.85–58.3 ng m^{-3} and mannitol, 5.57–138 ng m^{-3}) (Burshtein et al., 2011). Yttri et al. (2007) also reported that arabitol and mannitol were main contributors of sugar alcohols in aerosol samples collected from the different background sites in Norway. Sugar alcohols (arabitol, mannitol) can be used as tracers for various fungal and algal species (Bauer et al., 2008a,b; Pashanska et al., 2002; Zhang et al., 2010). Loos et al. (1994) discussed arabitol and mannitol as potential sources of bacteria and other microbes. High levels of detritus from the spring bloom and autumn decomposition have been reported as significant sources for arabitol and mannitol in the vegetated region (Burshtein et al., 2011; Pashynska et al., 2002). Good positive correlations of arabitol ($r = 0.63$) and mannitol ($r = 0.72$) with glucose indicate a vegetation contribution to both sugar alcohols in Chichijima aerosols. Erythritol and inositol are less abundant sugar species, accounting for 3.29% and 0.66% of total sugar alcohols. Their concentration ranges are 0.01-8.32 ng m^{-3} and 0.01-1.81 ng m^{-3}, respectively. Significant positive correlations of both sugar species with arabitol and mannitol indicate similar sources for these SCs in Chichijima aerosols (Table 2).

3.2 Seasonal variations of total sugar compounds

Seasonal concentration range, mean and median values of individual sugars during the study periods of thirteen years are presented in Table 1. The concentrations of individual sugars were extensively fluctuated from season to season in aerosol samples collected at Chichijima (Figures 4 and 5a). The seasonally averaged concentrations of total SCs are higher in summer (71.5±70.9 ng m^{-3}) and autumn (57.0±64.2 ng m^{-3}) than spring (39.8±67.6 ng m^{-3}) and winter (18.2±34.0 ng m^{-3}) over Chichijima Island. Zhu et al. (2015) measured sugar components in

aerosol samples collected from Cape Hedo, Okinawa, Japan and reported 2 to 3 times higher concentrations in summer (136 ng m^{-3}) and spring (133 ng m^{-3}) than autumn (86 ng m^{-3}) and winter (40 ng m^{-3}), whose seasonal trends are similar to Chichijima. Wan and Wu (2007) reported different seasonal variations with the highest concentration in autumn (375 ng m^{-3}), followed by winter (292 ng m^{-3}) and spring (84 ng m^{-3}) for the continental urban aerosols collected from Hong Kong. These concentrations in Hong Kong are 16 and 6 times higher than those of the remote Chichijima samples for winter and autumn, respectively. Interestingly, the different seasonal trends between the continental urban sites and two islands in the western North Pacific may be associated with different sources and transport pathways between the urban and marine environments.

3.2.1 Seasonal variations of primary sugars

Glucose maximized in summer (11.0±9.02 ng m^{-3}) followed by autumn (9.25±8.63 ng m^{-3}), spring (7.68±10.3 ng m^{-3}) and winter (3.11±3.53 ng m^{-3}) (Table 1 and Figure 5h). Glucose is the most abundant primary sugar in Chichijima aerosols. In winter/spring, Chichijima is influenced by strong westerly winds that deliver the air masses from the Asian continent including Mongolia, Russian Far East and North China, where vegetation is active. Consequently, declined concentrations of glucose in winter mean a depressed transport of glucose associated with continental bioaerosols from Asia despite long-range transport of Asian dusts due to strong westerly winds. The local vegetation (vascular plants) in Chichijima Island might be responsible to the enhanced concentration of glucose during growing season (spring and summer) and decaying periods of plant leaves (autumn). Seasonal PMF analysis also supports dominant sources of vegetation for glucose among four factors, which contributed >75% for mixed factor in summer (Figure 6c), >80% for fungal and vegetation factor in autumn (Figure 6d), and >75% for vegetation factor in spring (Figure 6b).

Fructose shows the highest concentrations in summer (7.25±7.63 ng m^{-3}) followed by spring (4.51±9.21 ng m^{-3}), autumn (3.70±2.68 ng m^{-3}) and winter (3.36±10.2 ng m^{-3}). As

shown in Table 2, a significant correlation (r=0.57) was obtained between glucose and fructose. Burshtein et al. (2011) reported similar correlations for both sugar species, suggesting a contribution of glucose and fructose from the local vegetation in summer (Baker et al., 1998; Pacini, 2000). Monthly mean concentrations of fructose show two prominent peaks in February-March and June-July, the latter peak may be due to the local vegetation in Chichijima (Figure 5g). The fructose peak in February-March may be influenced by air borne pollen grains in the spring bloom of flowering plants. High concentration of fructose was observed in spring followed by summer, indicating an input of this sugar compound from pollen grains (Fu et al., 2012). The positive correlation of fructose with sucrose (pollen tracer) supports the similar sources. Seasonal PMF analysis further supports the identical source for fructose and sucrose; that is, among four factors, fructose contributes >70% and >60% for pollen factor in spring (Figure 6b) and winter (Figure 6a), respectively.

Seasonal mean concentrations of trehalose showed a maximum in summer (7.06±8.49 ng m^{-3}) followed by autumn (6.09±8.81 ng m^{-3}), spring (2.93±6.08 ng m^{-3}) and winter (1.03±1.26 ng m^{-3}) (Table 1). Monthly mean concentrations of SCs for 13 years show that concentrations of trehalose are higher during June to October (Figure 5j). Ma et al. (2009) reported higher concentrations of trehalose at the urban site of Guangzhou, China during summer and autumn. Similarly, Wan and Wu (2007) reported a similar autumn maximum in Hong Kong. On the other hand, different seasonal trends of trehalose were reported for the aerosol samples (TSP) collected in USA (Medeiros et al., 2006), China (Wang et al., 2011), Australia (Hackl et al., 2000), and Gosan, Jeju Island in the western North Pacific Rim (Fu et al., 2012). In the above studies, highest concentrations of trehalose were reported in early spring due to the re-suspension of soil particles during agricultural practice. Hackl et al. (2000) also obtained abundant trehalose in spring, and proposed that trehalose can be used as a tracer of soil dust emission to the atmosphere. However, we did not detect a spring peak of trehalose in Chichijima aerosols, suggesting that soil dust contribution of trehalose over Chichijima is

insignificant via long-range atmospheric transport. Seasonal PMF analysis for autumn showed that more than 85% of trehalose was contributed by microbial factor among four factors (Figure 6d). An indirect contribution of trehalose from soil dust will be discussed later.

The seasonal mean concentrations of sucrose are almost equal during spring (8.80±18.0 ng m^{-3}), summer (7.31±11.5 ng m^{-3}) and winter (6.60±13.1 ng m^{-3}), except for autumn (2.76±4.35 ng m^{-3}) (Table 1). The similar seasonal distributions suggest multiple sources of sucrose in Chichijima aerosols. Monthly mean concentrations of sucrose show two peaks during February-March and June-July (Figure 5i). The March peak of sucrose was reported in the forest area of Sapporo, Japan to be 3 to 7 times more abundant than other months due to springtime pollen emissions (Miyazaki et al., 2012). Fu et al. (2012) analyzed pollen samples from different plant species (white birch, Chinese willow, Peking willow) for SCs and found the highest concentrations of sucrose followed by fructose and glucose in pollen. The pollen emissions from developing buds of plants may be the reason for the increased concentration of sucrose and fructose in February and March over Chichijima Island in the western North Pacific. Seasonal PMF analysis shows that sucrose contributed 100% in spring and >80% in winter, suggesting a significant pollen contribution for sucrose in those seasons (Figures 6b and 6a).

However, the possibilities of pollen transport from East Asia to Chichijima cannot be excluded because pollens can travel long distances with springtime high-speed winds by westerlies (Rousseau et al., 2006). The pollen grains emitted from flowering boreal forest in China, Mongolia, Siberia and Russian Far East, could significantly be delivered to the western North Pacific during spring, which may result in the contribution of sucrose and fructose to Chichijima aerosols. Recent studies have discussed a long-range transport of airborne pollen from North America to Greenland in spring (Rousseau et al., 2008). Lorenzo et al. (2006) reported the long-range transport of airborne allergenic pollen to central Italy. Makra et al. (2010) reported a long-range transport of airborne pollen in three European cities by applying

three-dimensional clustering of backward trajectories. Several studies also discussed a long-range transport of pollen to the remote arctic region (Andrews et al., 1980; Bourgeois et al., 2001; Campbell et al., 1999; Hicks et al., 2001; Hjelmroos and Franzen, 1994; Rousseau et al., 2004).

These observations may support that westerly winds have delivered pollen grains from the Asian continent including Mongolia, Siberia, and Russian Far East to Chichijima Island in spring. Using a box model and typical settling velocity of pollens (3 cm/sec) with the grain size of 30 μm in diameter (Sosnoskie et al., 2009), we estimated lifetime of pollen grains to be 9.3 hours in the atmospheric marine boundary layer (height of 1 km above the ocean surface). The settling velocity of the pollens is ca. 20 times larger than that of typical marine aerosols (Slinn and Slinn, 1980). Because pollen grain sizes range from 10 μm to 100 μm in diameter, the lifetime of pollens may have a large uncertainty. If pollens could be largely transported in the free troposphere (e.g., 5 km high) to the North Pacific from the Asian continent, then lifetime of typical pollen grains would increase upto 2 days. These calculations for the lifetime of pollen grains further support their long-range atmospheric transport from the Asian continent over the western North Pacific. Based on backward air mass trajectories (Figure 3), we can roughly estimate the transport time from East Asia to Chichijima site to be 2-4 days in winter and spring. It is also of interest to note that pollens can rapture under condition of high relative humidity (RH) (Hader et al., 2014; Miguel et al., 2006; Wright et al., 2014), which leads to smaller particles with longer residence time in the atmosphere.

In addition, tilling process after wheat crop harvesting in farmland causes an enhanced exposure of wheat root associated with soil particles into the atmosphere. China, India, and USA are three largest countries for wheat production in the world. In China and India there are two seasons (spring and winter) for wheat crops; winter wheat is harvested from mid-May to mid-July. During those periods (early summer), Chichijima Island is highly influenced by trade winds (Figure 3). However, air mass trajectories clearly show the occasional atmospheric

409 transport from Southeast Asia to Chichijima in summer (Pavuluri et al., 2010). PMF results of
410 sucrose for summer (Figure 6c) and autumn (Figure 6d) account for >85% and >90%,
411 respectively, for soil dust factor among four source factors, suggesting an additional source of
412 soil dust for sucrose in Chichijima aerosols (Simoneit et al., 2004). The elevated sucrose
413 concentrations in June and July (summer; non-flowering season) suggest the long-range
414 transport of sucrose associated with soil particles under the influence of occasional air mass
415 transport from Southeast Asia in summer (Figure 3).

416 Xylose was found as the least abundant sugar compound in the aerosol samples. The
417 maximum concentration of xylose (1.35 ng m^{-3}) was found in summer whereas minimum
418 (0.001 ng m^{-3}) in spring (Table 1). Summer mean concentration (0.18±0.26 ng m^{-3}) was highest
419 (Table 1). The PMF analyses showed that xylose contributed >75% for BB factor in winter
420 (Figure 6a) and >70% in autumn (Figure 6d) for microbial factor. These results suggest
421 different sources and seasons for xylose; i.e., biomass burning in winter (Sullivian et al., 2011)
422 and groups of microorganisms in summer (Cowie and Hedges, 1984).

423 **3.2.2 Seasonal variations of sugar alcohols**

424 The seasonal mean concentrations of arabitol and mannitol are higher in summer/autumn than
425 spring/winter (Table 1). The concentrations of arabitol are equally distributed between summer
426 (15.1±12.9 ng m^{-3}) and autumn (15.8±18.3 ng m^{-3}) with lower levels in spring (7.13±9.50 ng
427 m^{-3}) and winter (1.73±2.60 ng m^{-3}). Mannitol maximized in summer (21.7±19.7 ng m^{-3})
428 followed by autumn (18.2±19.9 ng m^{-3}), spring (7.95±13.8 ng m^{-3}) and winter (1.89±2.81 ng
429 m^{-3}). Arabitol and mannitol strongly co-varied throughout the study period. As depicted in
430 thirteen year monthly mean concentrations of total SCs (Figure 5b,c), we found elevated
431 concentrations of sugar alcohols from May to October. Similar seasonal trends were reported
432 for the aerosol samples collected from Gosan, Jeju Island in the western North Pacific Rim (Fu
433 et al. 2012) and urban aerosol samples from Ghent, Belgium (Pashynska et al., 2002). In above
434 studies, higher relative abundances of arabitol and mannitol in total sugar alcohols were

435 reported during late summer to autumn. The higher concentration of arabitol in autumn was
436 also reported for aerosol samples from the Mediterranean region in Israel (Burshtein et al.,
437 2011). Erythritol and inositol showed the similar seasonal trend, but their concentrations are
438 lower than the former two sugar species.

439 Sugar alcohols are emitted to the atmosphere from a variety of bacteria, few green algal
440 lichens and fungi (Dahlman et al., 2003; Filippo et al., 2013). Arabitol and mannitol are
441 abundant in fungal spores (Lewis and Smith, 1967; Yttri et al., 2007). Arabitol ($r = 0.73$) and
442 mannitol ($r = 0.80$) showed a strong co-variance with trehalose, suggesting identical sources of
443 sugar species in Chichijima. The PMF analysis showed that fungal factor and mixed factor
444 (fungal, vegetation, and microbial) accounted for 25% and 54.2% of total SCs observed in
445 summer, respectively (Figure 6c). In autumn, fungal and vegetation factor contributed 71% of
446 total SCs detected in Chichijima aerosols (Figure 6c). In winter (Figure 6a) and spring (Figure
447 6b) fungal and vegetation factor and mixed factor account for 31.2% and 37.2% of total SCs,
448 respectively. This is reasonable because fungal and microbial activities are lower during
449 winter/spring as compared to summer/autumn. The meteorological factors such as RH and
450 temperature significantly affect fungal and bacterial activity (Kim and Xiao, 2005; Malik and
451 Singh, 2004). Higher RH and temperature are crucial in increasing fungal and bacterial growth
452 (Sharma and Razak, 2003). Their maximum growth was observed under the condition of 92-
453 100% RH (Ibrahim et al., 2011). Higher concentrations of arabitol and mannitol in summer and
454 autumn may be caused by the increased fungal and bacterial activities in Chichijima Island.

455 Several studies have described the occurrence of fungi in marine environment (Jones,
456 1976; Kohlmeyer and Kohlmeyer, 1991; Moss, 1986). The fungal species eject spores from
457 hard materials like coral and sand grains. Some fungi also eject spores from woods associated
458 with sand in summer/autumn when higher ambient temperature and RH are available (Jones
459 and Mitchell, 1996). Marine fungal growths are observed on several mediums of substrates
460 such as wood, sediments, muds, soil, sand, algae, corals, decaying leaves of mangroves and

461 living animals in marine environment (Bremer, 1995; Kohlmeyer and Kohlmeyer, 1979;
462 Nagakiri et al., 1996). Although the above-mentioned studies have claimed the occurrence and
463 growth of marine fungi on several mediums of substrates, the knowledge of the role of marine
464 fungi in sediments and decaying dead animals are still insufficient due to a lack of appropriate
465 data set. It is still unclear if these fungi are active in sediments (Hyde et al., 1998). Therefore,
466 due to the inadequate data set, we doubt the marine contribution of sugar alcohols (arabitol,
467 mannitol) to Chichijima aerosols.

468 Thirteen year monthly mean concentrations of SCs clearly show slightly decreased
469 concentrations of arabitol, mannitol, and erythritol in July and August; a similar trend was
470 observed for trehalose (Figure 5b,c,e,j). These sugar compounds are derived from the microbial
471 activities in source regions. The thirteen year precipitation record over Chichijima Island
472 shows that precipitations were lowered in July and August (Figure 2). The lower precipitation
473 amount decreases the RH (Figure 2) and thus depresses the fungal and microbial activities. The
474 lower precipitation also suppresses the moisture contents in the surface soil of Chichijima,
475 which should result in a significant decline of local fungal and other microbial activities on the
476 ground of Chichijima Island. Decreased precipitation might be a possible reason for the lower
477 concentrations of arabitol, mannitol, and trehalose in July and August.

478 **3.3 Annual variations and decadal comparisons of SCs**

479 The annual variations in the concentrations of primary sugars and sugar alcohols are shown in
480 Figure 7a. The annual mean concentrations of total SCs varied randomly during 2001 to 2013.
481 As shown in Figure 7 (i, j, f and d), concentrations of sucrose, trehalose, xylose, and inositol
482 increase from 2001 to 2013 in Chichijima aerosols. Similarly, arabitol (Figure 7b), glucose
483 (Figure 7h), and fructose (Figure 7g) show clear increasing trends from 2006 to 2013 whereas
484 mannitol (Figure 7c) and erythritol (Figure 7e) show a random trend. Here, we compare the
485 data set of SCs for the periods of 1990-1993 (Period, P-I) with the current observations from
486 2001 to 2003 (P-II) and 2010 to 2013 (P-III) (Table 3) (Figures 8a, b, c).

The comparison for three periods indicates that concentrations of anhydrosugars are highest in winter followed by autumn. Their concentrations significantly increased from P-I to P-II/P-III (Figure 8a). The detailed discussions on anhydrosugars were reported in Verma et al. (2015). Here, we refer the data set of anhydrosugars for the decadal comparison with SCs. Interestingly, biomass-burning tracers (BB-tracers; levoglucosan, mannosan and galactosan) showed a significant difference in the decadal trends among three periods (i.e., P-I, P-II, and P-III) during winter and autumn. In winter, BB-tracers showed an increasing trend from P-I to P-III (Figure 8a). Biomass burning is common in winter for house heating (Simoneit et al., 2004a), thus it is obvious that the lower ambient temperature is the more biomass burning activities are. Westerly winds abundantly transport biomass-burning products over Chichijima Island in the western North Pacific with air masses derived from East Asia, Siberia, Mongolia, and Russian Far East during winter (Bendle et al., 2007; Simoneit and Elias, 2010; Verma et al., 2015). In contrast, BB-tracers in autumn show an opposite trend (i.e., higher concentrations in P-I followed by P-II and P-III) compared to those in winter (Figure 8a).

The difference in the concentrations of anhydrosugars in winter and autumn during P-I is insignificant while the concentrations are 3 and 5 times higher in winter than autumn for P-II and P-III, respectively. This seasonal shifting in the concentrations of anhydrosugars may be attributable to the changes in the strength of both westerly and trade wind systems from mid autumn to early winter among three periods (Chen et al., 2013). In contrast, the concentrations of primary sugars were 2 to 7 times higher during P-II and P-III than P-I period in summer and autumn (Figure 8b). PMF analysis showed that local emissions from vegetation are important contributor for primary sugars (glucose, fructose and sucrose). Therefore, a drastic increase in the concentrations of primary sugars in summer/autumn for P-II and P-III than P-I may be caused by an increased emission of primary sugars by local vegetation under the influence of meteorological conditions in the western North Pacific. However, a possible soil dust

512 contribution of primary sugar (sucrose) associated with an occasional air mass transport from
513 Southeast Asia cannot be excluded.

514 Similar to primary sugars, a drastic increase in the concentrations of sugar alcohols was
515 observed for P-II and P-III compared to P-I period (Figure 8c). The concentrations of sugar
516 alcohols in P-II and P-III are 6 to 19 times higher than those of P-I in summer/autumn (Table
517 3; Figure 8c). Arabitol and mannitol are key sugar alcohols and are reported as fungal and
518 microbial tracers, which contribute significantly to total SCs (Bauer et al., 2008b; Lewis and
519 Smith, 1967; Zhang et al., 2010). Microbes such as fungi and bacteria are significantly
520 increasing in the Asian and European countries (Yamaguchi et al., 2012). They are largely
521 transported towards downwind regions in the Pacific Ocean from the Asian continent in
522 winter/spring under the influence of strong westerlies (Griffin et al., 2001, 2003, 2007; Hua et
523 al., 2007; Uno et al., 2009) and settled down by wet and dry deposition in the western North
524 Pacific according to air mass trajectories (Figure 3).

525 Hirst et al. (1967) studied the lifetime of fungal spores and long-range transport of spores.
526 They reported that the fungal spores are in between 2 and 200 μm in diameter (mostly 5 to 30
527 μm), whose settling velocity is between 0.05 and 3.0 cm/sec with a mode of less than 1 cm/sec,
528 and that different fungal spores vary significantly in shape and ornamentation. This study
529 suggested the possibilities of long-range transport of fungal spores. Jeon et al. (2011) and
530 Yamaguchi et al. (2012) have analyzed aerosol samples collected during the Asian dust event
531 over the Sea of Japan, and identified similar groups of microbes (bacterial cells) transported
532 from the source regions of Asian dust. Consequently, bacteria and fungi associated with
533 bioaerosols grow extensively during summer/autumn, when the climate conditions are
534 favorable (i.e., higher RH and temperature) for their metabolic activities (Morris et al., 2004).
535 Accordingly, an increased transport of bioaerosols for the last decade may have caused a
536 drastic increase in the concentrations of sugar alcohols during P-II and P-III compared to P-I
537 period over the western North Pacific.

3.4 Source apportionment of SCs

To investigate the source apportionment of sugar components, the data sets of Chichijima aerosols were subjected to positive matrix factorization (PMF) analysis. Based on the PMF analysis, total five factors were determined to be significant to classify the sources of sugar compounds (SCs). Five factors successfully explored the source profile for the individual sugar component. Factor profiles resolved by PMF analysis are shown in Figures 6 and 9-11, where percentages of each component summed for factors 1 to 5 are calculated to be 100%.

Vegetation factor (Figure 9) was dominated by xylose (75%), glucose (48%), and fructose (36%). Xylose is significantly produced by gymnosperm and angiosperm (Cowie and Hegdes, 1984; Sjostrom, 1981). Fructose and glucose are highly water-soluble sugar species, and present in the bark and leaves of plants (Fu et al., 2012). Glucose is the second most abundant sugar that contributed to this factor. Cowie and Hegdes (1984) reported higher concentration of glucose in vascular plants and phytoplankton in the marine environment. The SCs emitted by the vegetation during growing season significantly contribute to vegetation factor. In Chichijima aerosols, glucose and fructose are significant contributors in spring (Figure 6b), summer (Figure 6c) and autumn (Figure 6d), Therefore, the respective factors in Figure 6 are termed as a vegetation source for both sugar species. This is reasonable because plants started growing in spring and summer seasons. In autumn, leaf senescence and decay result in the emission of glucose and fructose to the atmosphere.

Fungal and microbial factor (Figure 9) was characterized by trehalose (88%), mannitol (64%), and arabitol (54%). These sugars that contribute to fungal and microbial factor are associated with fungal spores, bacteria and yeast (Bauer et al., 2008a; Medeiros et al., 2006; Wiemken, 1990). The three sugars are good tracers of fungal spores and microbes (Loos et al., 1994; Rogge et al., 2007). Arabitol and mannitol are produced by a large variety of fungal species (Ion et al., 2005; Medeiros et al., 2006), and considered as a suitable tracer for fungal and bacterial metabolic activities (Bauer et al., 2008b; Elbein et al., 1974; Rogge et al., 2007).

564 Arabitol is strongly correlated with mannitol (r=0.88), suggesting similar sources for both
565 species (Table 2) (Elbert et al., 2007). Fungi, bacteria, and other microbes in soils are the main
566 sources for trehalose (Graham et al., 2003; Rogge et al., 2007; Simoneit et al., 2004). An
567 excellent correlation of trehalose with arabitol and mannitol suggested similar sources in the
568 marine environment (Table 2) (Lewis and Smith, 1967).

569 Sugar alcohols have been proposed as tracers for microbes and fungal spores (Bauer et
570 al., 2008b; Ion et al., 2005; Medeiros et al., 2006; Rogge et al., 2007). The fungal and
571 microbial activities are considered higher during summer and autumn due to higher
572 temperature and RH. The above discussions for the sources of arabitol, mannitol and trehalose
573 are well supported by the seasonal PMF analysis. Arabitol and mannitol are well contributed in
574 summer (Figure 6c, fungal factor and mixed factor) and autumn (Figure 6d, fungal and
575 vegetation factor). Correspondingly, trehalose also contributed in summer (Figure 6c, mixed
576 factor) and autumn (Figure 6d, microbial factor). Therefore, significant contributions of
577 arabitol, mannitol and trehalose are observed during the respective seasons in Chichijima
578 aerosols.

579 Mixed factor (Figure 9) is associated with erythritol (94%), arabitol (44%), mannitol
580 (34%), inositol (32%), glucose (24%), and fructose (31%). Due to the highly miscellany
581 characteristics of fungi, other microbes, and plant debris, it is quite difficult to specify the
582 particular source for individual sugar species (Percival, 1970). Arabitol and mannitol are also
583 attributed to the vegetation (photosynthesized by mature leaves) (Burshtein et al., 2011;
584 Pashynska et al., 2002). The contributions of arabitol and mannitol in winter (Figure 6a, mixed
585 factor) and spring (Figure 6b, mixed factor) indicate other sources than fungal spores in the
586 Chichijima aerosols. Because the fungal and microbial growth is less important in winter and
587 spring compared to summer/autumn. Therefore, this factor (Figure 9) should be associated with
588 mixed sources from microbial and vegetational activities. Contemplating of mixed sources is

589 very likely because sugar species that are highly apportioned to vegetation factor contribute, to
590 some extent, to the same sources that are responsible to fungal and microbial factor (Figure 9).
591 Biomass burning (BB) factor (Figure 9) is loaded significantly with levoglucosan (94%),
592 mannosan (91%), and galactosan (90%), and moderately with xylose (20%). These species are
593 associated with biomass burning (Fraser and Lakshmanan, 2000; Graham et al., 2002;
594 Simoneit, 2002). Nolte et al. (2001) and Medeiros et al. (2006) also reported that biomass
595 burning-influenced aerosols are enriched with levoglucosan, mannosan, and galactosan.
596 Kawamura et al. (2003) and Mochida et al. (2010) reported that biomass-burning products are
597 abundantly transported to Chichijima under the influence of westerly winds during
598 winter/spring (Figure 3). These results are well supported by the fact that BB factor is
599 associated with SCs, which are derived from biomass burning in East Asia. The seasonal PMF
600 analysis (Figure 6) also supports the above explanation; BB products are contributed highest in
601 winter/spring (18.8% and 6.3%) under the influence of westerly winds, followed by
602 summer/autumn (3.8% and 4.1%) for total sugars in the aerosol samples collected from
603 Chichijima Island.
604 Pollen factor (Figure 9) is characterized by high loading of sucrose (91%), fructose
605 (35%), and inositol (33%); these sugar species are associated with airborne pollen sources.
606 Previous studies have also reported that sucrose is an excellent tracer for airborne pollen grains
607 of flowering plants (Pacini, 2000; Graham et al., 2003; Wang et al., 2008; Medeiros et al.,
608 2006). Fructose is well correlated with inositol (r=0.57), indicating a similar origin for both
609 sugar species (Table 2). Two prominent peaks of sucrose, fructose, and inositol, which
610 appeared in late winter to early spring or summer, also indicate a similar source of those sugars.
611 Contributions of sucrose (pollen factor) in winter (39.5%; Figure 6a) and spring (36%; Figure
612 6b) are supported the sources of airborne pollen for sucrose in Chichijima Island. The sucrose
613 contribution (soil dust factor) in non-flowering seasons, i.e., summer (17.1%; Figure 6c) and
614 autumn (8.4%; Figure 6d), indicates different sources for sucrose in Chichijima aerosols.

615 According to the seasonal PMF analysis (Figure 6), we termed additional sources of sucrose in
616 Chichijima Island as soil dust factor as well as pollen factor.

617 As seen in Figure 6a, b, c and d, PMF analysis for seasonal source identification
618 indicated variable contributions of individual SCs in different seasons according to their
619 seasonal source origin. In winter (Figure 6a), airborne pollen (39.5%) contributed highest
620 followed by vegetation, microbial, fungal (mixed) (31.2%), biomass burning (18.8%), and
621 microbial (10.5%) sources. However, in spring, vegetation, microbial, and fungal (mixed)
622 sources (37.2%) contributed almost equal to the airborne pollen (36.0%) followed by
623 vegetation (21.2%) and biomass burning (6.3%) sources. The vegetation, microbial, fungal
624 (mixed) (54.2%), fungal (25.2%), soil particles (17.1%), and biomass burning (3.8%) sources
625 are characterized to maximize in summer. Similar to summer, vegetation and fungal (mixed)
626 sources (71.2%) are also leading to the contribution of total SCs followed by microbial
627 (16.4%), soil particles (8.4%), and biomass burning (4.1%) in total SCs observed in autumn for
628 Chichijima aerosols.

629 Overall, average contributions of each factor to measured SCs as resolved by the PMF
630 analyses are shown in Figure 10. Fungal and microbial factor accounts for 41% of total SCs
631 measured. The emission from microbes including fungal spores was found as a dominant
632 contributor to total SCs. Mixed factor (27%) indicates a common involvement of fungal,
633 microbial, and vegetation sources. Figure 11 shows annual trends in % contributions of five
634 source factors to SCs in Chichijima aerosols. Fungal and microbial factor, and mixed factor
635 contributed higher than other sources for SCs during 2001 to 2013. However, no clear trends in
636 annual % contributions were observed for both source factors during thirteen-year study period.
637 The sugar species assigned as pollen tracers were found to contribute 18% in pollen factor.
638 Vegetation accounts for 11% of total SCs, indicating less emission from vegetation as
639 compared to fungi and microbes. As indicated by BB factor, biomass-burning source
640 contributes only 3% of total SCs.

Interestingly, we found an increasing trend in % contribution of vegetation, pollen, and BB factors to SCs in 2006 to 2013 (see Figure 11). Sugar components, which are contributed from pollen (sucrose, fructose, and inositol), vegetation (glucose and fructose), and biomass burning sources (levoglucosan, galactosan, and mannosan), also show a similar increasing trend for the period of 2006 to 2013 (Figure 7). The increased annual trends of BB and pollen factors might be due to an enhanced long-range transport of airborne pollens and biomass burning products from the Asian continent to the western North Pacific under the influence of strong westerly winds.

4 Summary and conclusions

We reported thirteen years of temporal, seasonal and decadal trends of sugar compounds (SCs) measured in the aerosol samples collected at Chichijima Island in the western North Pacific, an outflow region of Asian aerosols. The high abundances of total SCs and primary sugars are found during summer, whereas sugar alcohols are almost equally distributed during summer and autumn. The seasonal distributions of arabitol, mannitol, and trehalose are strongly influenced by long-range atmospheric transport of bioaerosols associated with microbes such as fungi and bacteria and their metabolic activities under the influences of westerly winds and favorable meteorological conditions, including high RH and temperature in summer/autumn. Seasonal variation of sucrose is controlled by both locally emitted and long-range transported pollen from East Asia to the western North Pacific during spring bloom periods. On the other hand, the increased concentrations of sucrose and fructose during summer may be caused by the local activity of vegetation and possibly by long-range transport of plant root-associated soil dust particles from East and Southeast Asia. PMF analysis indicated specific sources for individual SCs during different seasons. The results clearly separated biogenic emissions into two parts as vegetation and microbes, including fungal species. The emissions from vegetation,

pollen, and microbial activities contributed about 97% of total measured SCs, with the remaining fraction being derived from biomass burning activities.

The concentration and seasonal variations of SCs at Chichijima Island are well regulated by atmospheric circulations, i.e., the westerly winds passing from the Asian continent during winter/spring and trade winds originated from the central Pacific Ocean during summer/autumn dominate over Chichijima Island in the western North Pacific. The meteorological parameters also significantly affect the concentrations and seasonal variations of SCs over Chichijima. Based on a decadal observation at Chichijima, we conclude that drastic increases in the concentrations of sugar alcohols and primary sugars during 2001-2003 and 2010-2013 can be caused by an enhanced atmospheric transport of bioaerosols from East Asia to the western North Pacific.

Sugar components (SCs) are important compositions of organic aerosols worldwide and it is recognized as a significant factor affecting air quality and possibly climate. The outcomes of the thirteen-year study of SCs at Chichijima Island have an implication for global radiative forcing by scattering or absorbing light and also the activity of cloud condensation nuclei (CCN) in the western North Pacific that have a high sensitivity to global climate change due to an outflow region of the Asian dust and bioaerosols. The NASA Global Climate Change (http://climate.nasa.gov/vital-signs/global-temperature/) has reported a continuous increase in the global land/ocean temperature. The increasing annual trends in % contribution of vegetation factor to SCs suggested a significantly increased activity of local vegetation in Chichijima Island from 2006 to 2013, which could be involved with a recent global warming especially in the western North Pacific region.

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Figure Captions

Figure 1. Geographical location of Chichijima Island (27°04'N; 142°13'E; 254 m, asl) in the western North Pacific.

Figure 2. The monthly variation of the meteorological parameters over Chichijima Island during 2001-2013 (The error bars denote the standard deviations).

Figure 3. The seasonal ten-day air mass backward trajectories over Chichijima for 2012 (winter: Dec-Feb, spring: Mar-May, summer: Jun-Aug, autumn: Sep-Nov). The trajectory calculations were performed everyday starting at Chichijima Island.

Figure 4. Temporal plots for the concentrations (ng m^{-3}) of sugar compounds in Chichijima aerosol samples collected for 2001-2013 in the western North Pacific.

Figure 5. Monthly mean concentrations (ng m^{-3}) of sugar compounds in aerosol samples from Chichijima Island in the western North Pacific during 2001-2013.

Figure 6. Seasonal source contributions to sugar compounds from various sources based on PMF analyses. (BB – biomass-burning; Mixed – vegetation, fungal and microbial sources).

Figure 7. Annual mean concentrations (ng m^{-3}) of sugar compounds in aerosol samples collected from Chichijima Island in the western North Pacific during 2001-2013.

Figure 8. The seasonal concentrations of anhydrosugars (biomass burning tracers), primary sugars and sugar alcohols measured in Chichijima aerosols during three periods, i.e., P-I (1990-1993), P-II (2001-2003) and P-III (2010-2013).

Figure 9. PMF analyses of sugar compounds in Chichijima aerosols based on the 2001-2013 data set. (BB – biomass-burning; Mixed – vegetation, fungal and microbial sources).

Figure 10. Source contributions to sugar compounds from various sources based on PMF analyses. (BB – biomass-burning; Mixed – vegetation, fungal and microbial sources).

Figure 11. Annual trends in % contributions of five source factors: (a) vegetation, (b) fungal and microbial, (c) mixed, (d) biomass burning (BB), and (e) pollen factors to SCs in Chichijima aerosols. The data of 2005 are not plotted due to limited data points.

Figure 1.

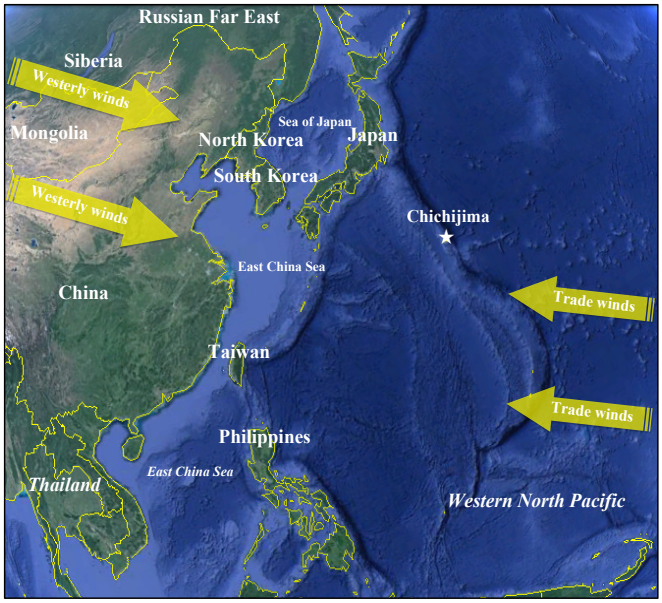
Russian Far East
Siberia
Westerly winds
Mongolia
Sea of Japan
North Korea
Japan
South Korea
Westerly winds
Chichijima
East China Sea
China
Trade winds
Taiwan
Trade winds
Philippines
East China Sea
Thailand
Western North Pacific

Figure 2.

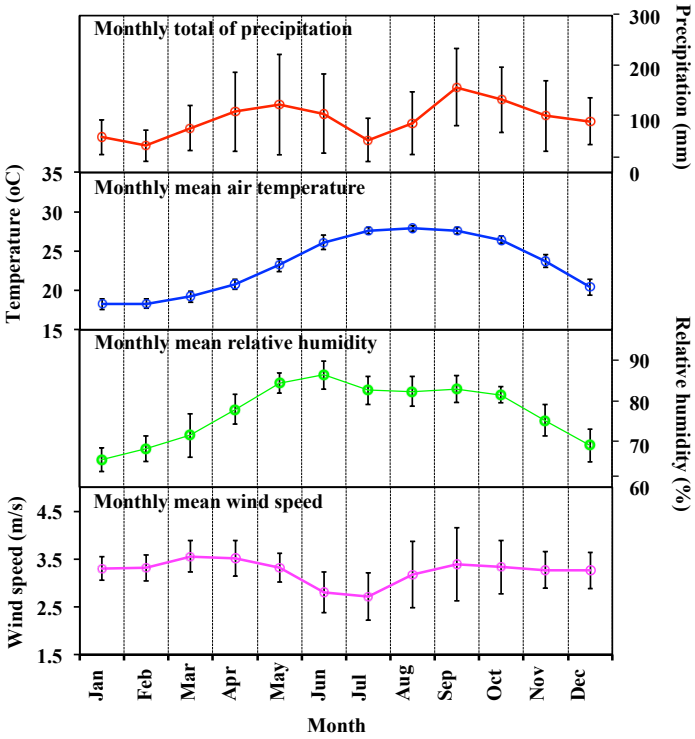
Monthly total of precipitation
Monthly mean air temperature
Monthly mean relative humidity
Monthly mean wind speed
Precipitation (mm)
Temperature (oC)
Relative humidity (%)
Wind speed (m/s)
Month
Jan
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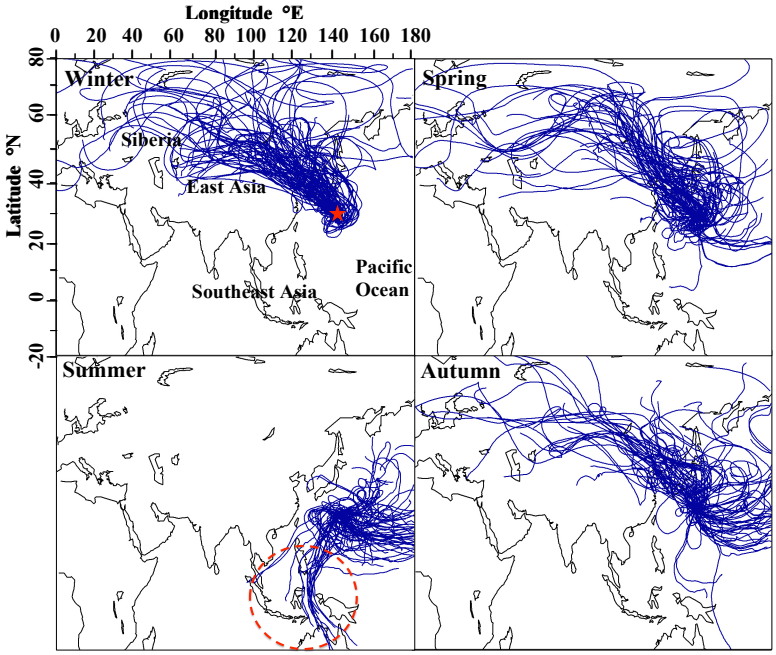
Figure 3.


Longitude °E
0 20 40 60 80 100 120 140 160 180
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Winter
Siberia
East Asia
Southeast Asia
Pacific Ocean
Spring
Summer
Autumn

Figure 4.

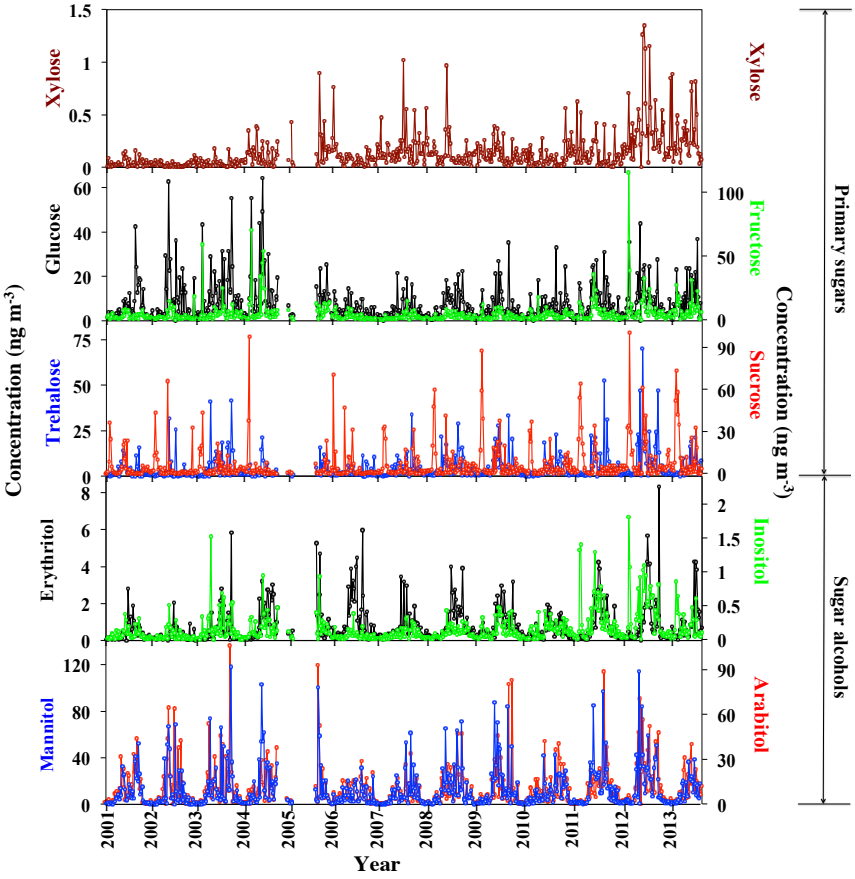
Xylose
Glucose
Trehalose
Erythritol
Mannitol
Fructose
Sucrose
Inositol
Arabitol
Concentration (ng m-3)
Concentration (ng m-3)
Primary sugars
Sugar alcohols
Year
2001
2002
2003
2004
2005
2006
2007
2008
2009
2010
2011
2012
2013

Figure 5.

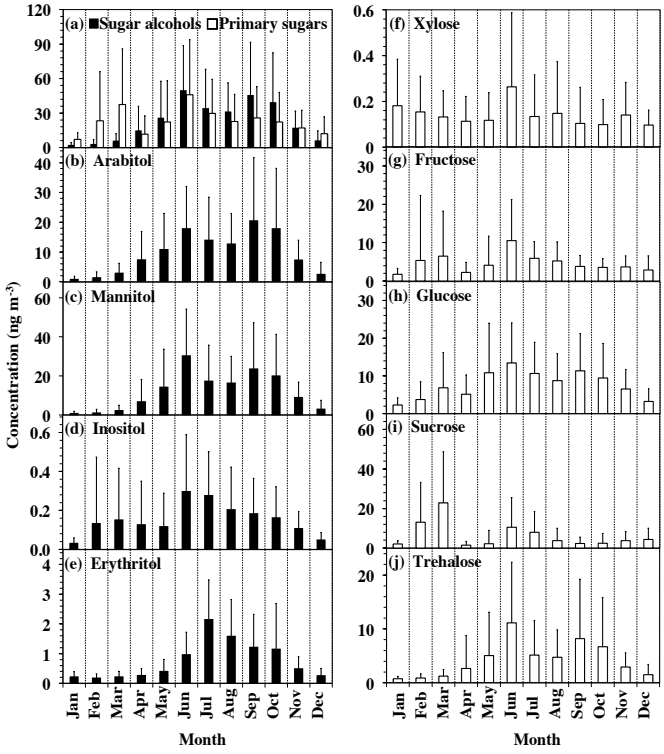
(a) Sugar alcohols Primary sugars
(b) Arabitol
(c) Mannitol
(d) Inositol
(e) Erythritol
(f) Xylose
(g) Fructose
(h) Glucose
(i) Sucrose
(j) Trehalose
Concentration (ng m-3)
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec
Month

Figure 6.

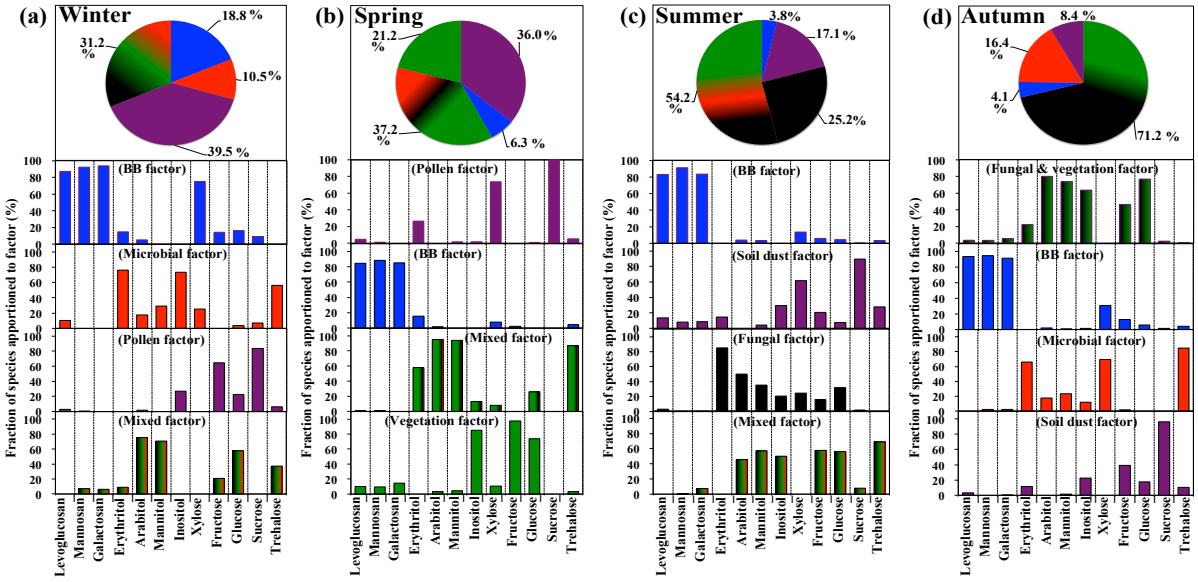
(a) Winter
18.8 %
31.2 %
10.5 %
39.5 %
(b) Spring
21.2 %
36.0 %
37.2 %
6.3 %
(c) Summer
3.8%
17.1 %
54.2 %
25.2%
(d) Autumn
8.4 %
16.4 %
4.1 %
71.2 %
Fraction of species apportioned to factor (%)
(BB factor)
(Microbial factor)
(Pollen factor)
(Mixed factor)
(Pollen factor)
(BB factor)
(Mixed factor)
(Vegetation factor)
(BB factor)
(Soil dust factor)
(Fungal factor)
(Mixed factor)
(Fungal & vegetation factor)
(BB factor)
(Microbial factor)
(Soil dust factor)
Levoglucosan
Mannosan
Galactosan
Erythritol
Arabitol
Mannitol
Inositol
Xylose
Fructose
Glucose
Sucrose
Trehalose

Figure 7.

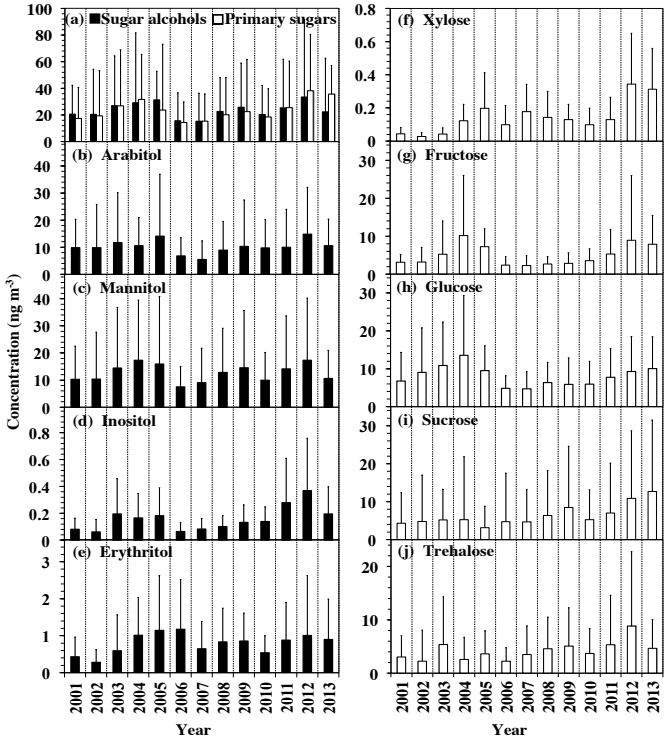
(a) Sugar alcohols Primary sugars
(b) Arabitol
(c) Mannitol
(d) Inositol
(e) Erythritol
(f) Xylose
(g) Fructose
(h) Glucose
(i) Sucrose
(j) Trehalose
Concentration (ng m-3)
Year
Year

Figure 8.

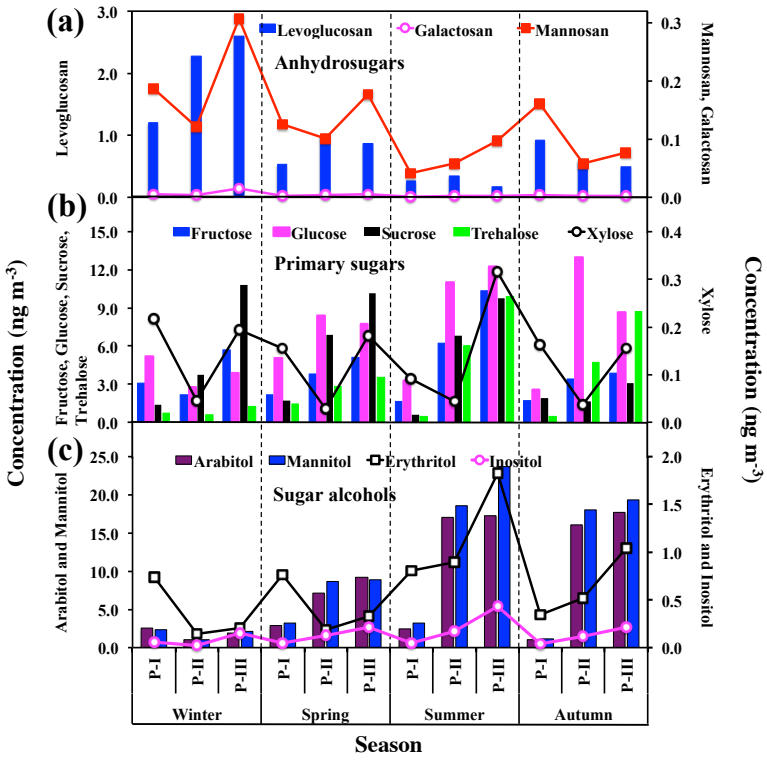
(a)
Levoglucosan
Galactosan
Mannosan
Anhydrosugars
Levoglucosan
Mannosan, Galactosan
(b)
Fructose
Glucose
Sucrose
Trehalose
Xylose
Primary sugars
Fructose, Glucose, Sucrose, Trehalose
Xylose
(c)
Arabitol
Mannitol
Erythritol
Inositol
Sugar alcohols
Arabitol and Mannitol
Erythritol and Inositol
Concentration (ng m-3)
Concentration (ng m-3)
P-I
P-II
P-III
Winter
Spring
Summer
Autumn
Season

Figure 9.

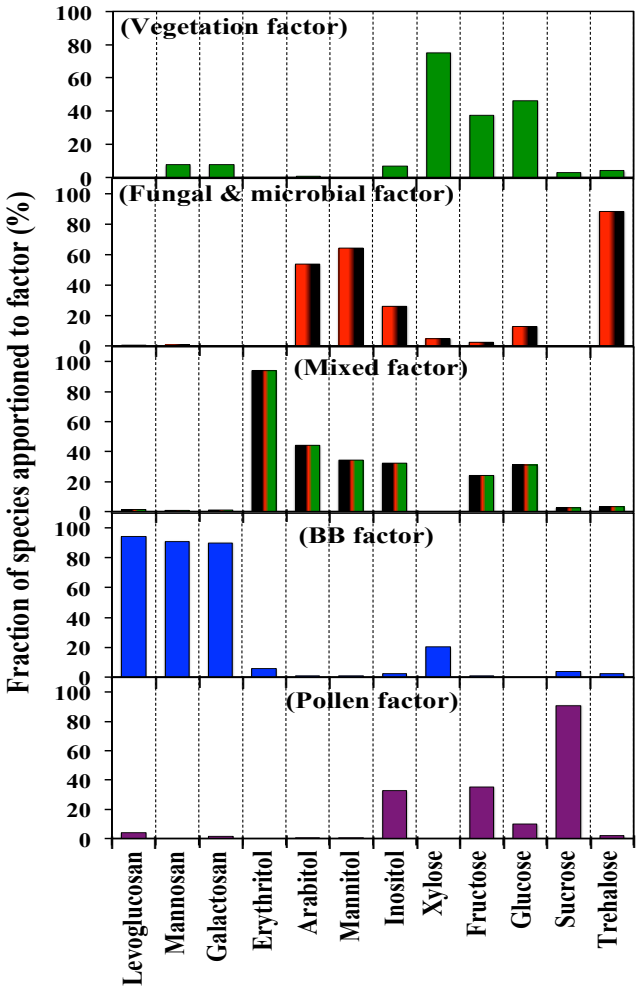
(Vegetation factor)
(Fungal & microbial factor)
(Mixed factor)
(BB factor)
(Pollen factor)
Fraction of species apportioned to factor (%)
0
20
40
60
80
100
Levoglucosan
Mannosan
Galactosan
Erythritol
Arabitol
Mannitol
Inositol
Xylose
Fructose
Glucose
Sucrose
Trehalose

Figure 10.

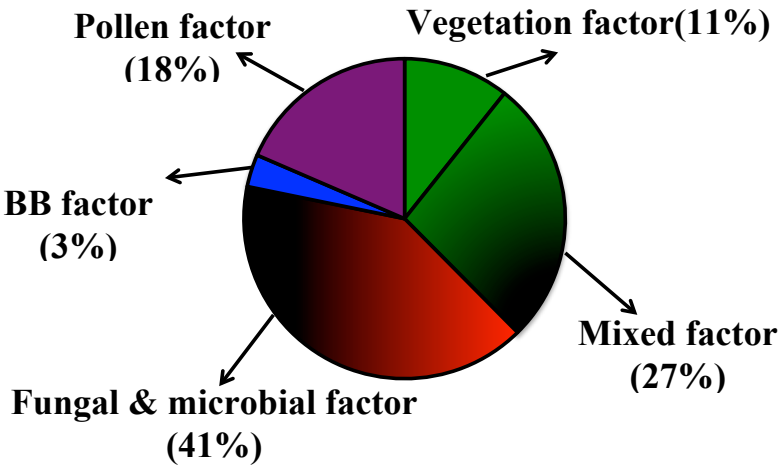
Pollen factor
(18%)
Vegetation factor(11%)
BB factor
(3%)
Mixed factor
(27%)
Fungal & microbial factor
(41%)

Figure 11.

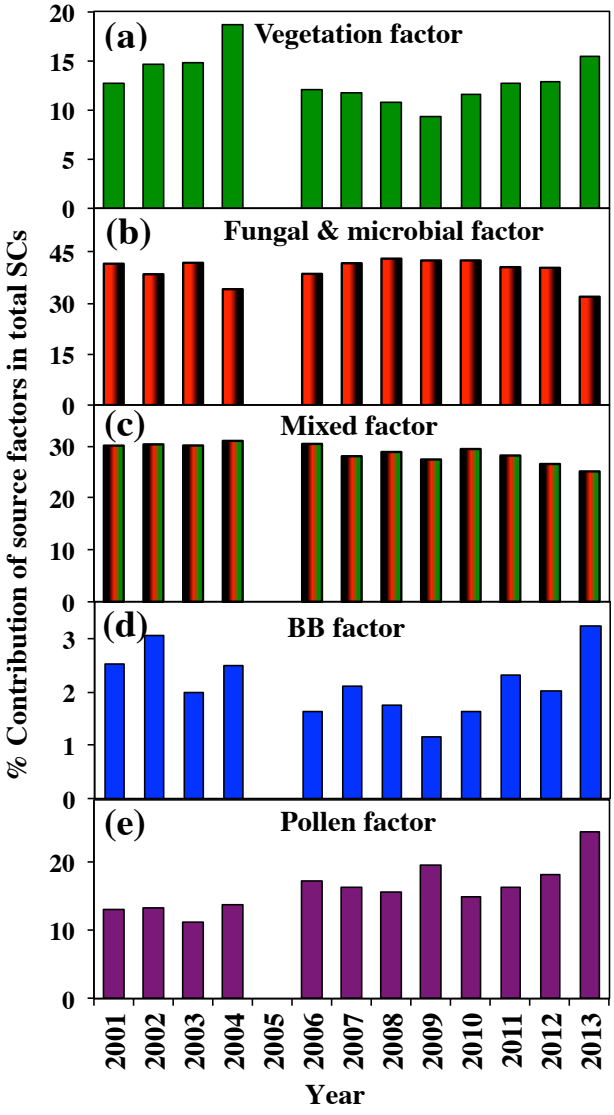
(a) Vegetation factor
(b) Fungal & microbial factor
(c) Mixed factor
(d) BB factor
(e) Pollen factor
% Contribution of source factors in total SCs
Year
2001
2002
2003
2004
2005
2006
2007
2008
2009
2010
2011
2012
2013

Table 1. Seasonal concentrations (ng m^{-3}) of sugar compounds (SCs) in the aerosol samples collected at ChichiJima Island in the western North Pacific during 2001-2013.

Sugars	Winter[a] (n=139)			Spring[b] (n=155)			Summer[c] (n=146)			Autumn[d] (n=150)			2001-2013 (n=590)		
	Range	Mean±SD	Med[e]	Range	Mean±SD	Med[e]	Range	Mean±SD	Med[e]	Range	Mean±SD	Med[e]	Range	Mean±SD	Med[e]
Primary sugars															
Xylose	0.05-0.89	0.14±0.15	0.12	0.001-0.56	0.12±0.11	0.08	0.002-1.35	0.18±0.26	0.08	0.004-0.89	0.11±0.14	0.07	0.001-1.35	0.14±0.18	0.08
Fructose	0.17-115	3.36±10.2	1.38	0.03-70.4	4.51±9.21	1.90	0.24-54.0	7.25±7.63	5.23	0.22-13.0	3.70±2.68	2.82	0.03-115	4.69±8.04	2.53
Glucose	0.27-23.3	3.11±3.53	2.13	0.05-62.6	7.68±10.3	4.36	0.23-64.3	11.0±9.02	7.99	0.72-55.5	9.25±8.63	5.77	0.05-64.3	7.79±8.80	4.73
Sucrose	0.02-73.4	6.60±13.1	2.08	0.005-100	8.80±18.0	1.30	0.003-66.0	7.31±11.5	2.58	0.002-31.1	2.76±4.35	1.14	0.002-100	6.43±12.9	1.71
Trehalose	0.03-10.5	1.03±1.26	0.72	0.006-47.2	2.93±6.08	1.23	0.03-70.2	7.06±8.49	4.46	0.04-52.5	6.09±8.81	2.46	0.01-70.2	4.30±7.28	1.61
Σ Primary sugars	**0.49-223**	**14.2±28.2**		**0.09±281**	**24.2±43.8**		**0.51-256**	**32.8±36.9**		**0.99-153**	**22.0±24.6**		**0.28-176**	**23.3±25.7**	
Sugar alcohols															
Erythritol	0.03-1.17	0.23±0.18	0.18	0.008-2.25	0.31±0.28	0.23	0.07-5.70	1.55±1.21	1.18	0.05-8.32	0.99±1.16	0.56	0.01-8.32	0.77±1.01	0.37
Arabitol	0.12-21.2	1.73±2.60	0.96	0.04-70.8	7.13±9.50	4.44	0.24-64.5	15.1±12.9	12.0	0.57-106	15.8±18.3	8.83	0.04-106	9.99±13.6	4.97
Mannitol	0.10-23.9	1.89±2.81	1.11	0.16-114	7.95±13.8	4.07	0.25-104	21.7±19.7	16.9	0.59-118	18.2±19.9	9.15	0.10-118	12.5±17.5	5.54
Inositol	0.01-1.81	0.07±0.20	0.03	0.008-1.51	0.13±0.22	0.06	0.01-1.29	0.26±0.25	0.17	0.01-0.93	0.16±0.15	0.10	0.01-1.81	0.16±0.22	0.08
Σ Sugar alcohols	**0.26-48.2**	**3.93±5.79**		**0.22-188**	**15.5±23.8**		**0.56-175**	**38.6±34.1**		**1.22-234**	**35.1±39.5**		**0.37-231**	**23.4±30.8**	
Σ SCs	**0.75-272**	**18.2±34.0**	**10.6**	**0.31-469**	**39.8±67.6**	**18.2**	**1.07-431**	**71.5±70.9**	**50.7**	**2.21-387**	**57.0±64.2**	**31.1**	1.23-339	46.7±49.5	30.9

[a] Winter (December-February), [b]Spring (March-May), [c]Summer (June-August) and [d]Autumn (September-November); Med.=Median

Table 2. Pearson correlation coefficients (r) for the dataset of sugars in Chichijima aerosols during 2001-2013 (n = 590).

	Levoglucosan[a]	Mannosan[a]	Galactosan[a]	Erythritol	Arabitol	Mannitol	Inositol	Xylose	Fructose	Glucose	Sucrose	Trehalose
Levoglucosan	1.00											
Mannosan	0.79	1.00										
Galactosan	0.55	0.58	1.00									
Erythritol	-0.18	-0.12	-0.16	1.00								
Arabitol	-0.16	-0.09	-0.16	0.48	1.00							
Mannitol	-0.18	-0.12	-0.17	0.49	0.88	1.00						
Inositol	-0.06	-0.02	0.10	0.35	0.49	0.58	1.00					
Xylose	0.20	0.32	0.34	0.15	0.18	0.23	0.42	1.00				
Fructose	0.02	0.08	0.26	0.17	0.16	0.28	0.57	0.31	1.00			
Glucose	-0.11	0.00	-0.06	0.32	0.63	0.72	0.53	0.19	0.57	1.00		
Sucrose	0.05	0.07	0.18	-0.02	-0.06	0.01	0.40	0.26	0.30	0.14	1.00	
Trehalose	-0.10	-0.05	-0.10	0.33	0.73	0.80	0.55	0.33	0.22	0.54	0.13	1.00

[a] data from Verma et al. (2015)

Table 3. Comparisons of seasonal concentrations (ng m^{-3}) of primary sugars and relative contributions (%) of sugar compounds (SCs) in total SCs in Chichijima aerosols among 1990-1993[a], 2001-2003 and 2010-2013.

Season	Anhydrosugars			Sugar alcohols			Primary sugars			Total Sugars		
	1990-93 [a]	2001-03[b]	2010-13 [b]	1990-93 [a]	2001-03	2010-13	1990-93 [a]	2001-03	2010-13	1990-93 [a]	2001-03	2010-13
Winter	1.44	2.44	3.05	5.64	2.27	4.37	10.6	9.34	21.8	17.7	14.0	29.2
%	11.4	23.7	16.6	31.6	16.5	19.8	57.0	59.9	63.6			
Spring	0.68	1.12	1.10	6.91	16.2	18.6	10.5	21.3	26.7	18.1	38.6	46.4
%	5.08	5.17	4.04	39.2	39.7	42.0	55.7	55.1	53.9			
Summer	0.31	0.43	0.29	6.50	36.7	43.1	6.07	30.0	42.5	12.9	67.1	86.0
%	2.92	0.91	0.43	55.6	51.3	51.1	41.4	47.8	48.5			
Autumn	1.12	0.65	0.60	2.57	34.7	38.3	6.79	22.6	24.5	10.5	57.9	63.4
%	12.9	3.45	1.73	29.2	50.4	57.2	57.9	46.2	41.1			

[a] data from Chen et al. (2013), [b] data from Verma et al. (2015).