To
Prof. Jason Surratt
Editor
Atmospheric Chemistry and Physics (ACP)

RE: Submission of revised manuscript entitle "**Thirteen years of observations on primary sugars and sugar alcohols over remote Chichijima Island in the western North Pacific**" for publication in Atmospheric Chemistry and Physics

Reference: MS No.: acp-2017-480

Dear Prof. Surratt,

   Thanks for the decision letter on our manuscript. We have revised the manuscript meticulously following the referees' comments and suggestions. The authors greatly appreciate the critical and useful comments by anonymous referee's that significantly improved the quality of our manuscript. Our responses and changes are included in the response letter as blue color. The changes in the revised manuscript are highlighted in yellow. Please find the response letter and revised manuscript.

   We believe that revised manuscript is acceptable for publication in ACP.

Thanking you.

Sincerely yours,

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Authors' responses to the comments Referee # 2

General

This is an interesting time-series paper on aerosol particle measurements of sugars and sugar alcohols at the remote island of Chichijima in the western north Pacific.

The authors should think about the numerical formats of their results. Sometime too many digits are listed which are uncertain in view of the calculated error width.

The English language of the manuscripts should be checked again.

Overall, I think this is an interesting paper for ACP publishable after minor revision

Authors are thankful to the reviewer for the positive recommendation. We carefully checked and improved English writing of through out the manuscripts.

Details

Title: I would suggest 'Thrteen years ...' without hypenation.
Response: We deleted the hypenation from Thrteen years… in the title. (Please see the title in the revised MS).

Abstract: The only numbers given here are the range mass concentrations of 'total sugar compound'. It would be great if more quantitative information could be given in the abstract, e.g. on the observed trends.
Response: According to the reviewer's suggestions, we added some more information in the abstract. (Please see lines 34-36 in the revised MS).

Line 64: For these contribution fractions only Simoneit et al. (2004) is referenced - are there any more recent findings on this, which could be referenced? Can a range be given based on more references?
Response: According to the reviewer's suggestions, we modified the sentences and added new phrase with additional references. (Please see lines 65-73 in the revised MS.)
"Sugar compounds (SCs) contribute 13–26% and 63% of the total compound mass in the 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 WSOC at urban and suburban sites in Norway. Tominaga et al., (2011) analyzed aerosol samples collected from urban and forest suburban sites from Japan and reported that sugars (arabinose, fructose, galactose, glucose, mannose, rhamnose, and xylose) accounted for 2.1% and 4.5% of WSOC in the fine and coarse mode ranges at Yokohama, respectively, and for 3.0% and 7.2% at Mt. Oyama, respectively."

We added a new reference: Tominaga, S., K. Matsumoto, N. Kaneyasu, A. Shigihara, K. Katono, N. Igawa (2011), Measurements of particulate sugars at urban and forested suburban sites, Atmos. Environ. 45, 2335-2339.

Line 183: I feel the section on PMF can considerably be shortened; much of this can be referenced.
Response: According to referee's suggestion, "Section 2.4 Positive matrix factorization (PMF) analysis" was significantly shortened. We added a new sentence: "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)." (Please see lines 206-208 in the revised MS.)

Line 497: The sentence starting 'Thirteen-year...' can be left out.
Response: According to referee's suggestions, we deleted the sentences starting from thirteen-years …..). (Please see line 484 in the revised MS).

Line 669, Summary and conclusion: Please make this consistent with the abstract. What is, finally, the conclusion of the paper?
Response: According to referee's suggestions, we modified the abstract and conclusion section as follows.

We modified sentence: "The primary sugars (glucose and fructose) maximized in summer, possibly due to an increased emission of vegetation products from the local vascular plants in Chichijima." (Please see lines 27-29 in the revised MS).

Added a new sentence: "Sucrose and trehalose were found to present increasing trends from 2001 to 2013, while total sugar components did not show any clear trends during thirteen year periods." (Please see lines 34-36 in the revised MS.)

We significantly modified summary and conclusion sections in the revised MS. (Please see lines 651-687).

1 **Thirteen years of observations on primary sugars and sugar alcohols over remote**

2 **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})

97　　dicarboxylic acids and levoglucosan (biomass burning tracer) have also been reported in

98　　Chichijima aerosols by Mochida et al. (2003a) and Mochida et al. (2010), respectively.

99　　Although seasonal variation of saccharides was reported in Chen et al. (2013), the observation

100　period is rather short.

101　　　　Here, we report thirteen year data set of SCs in remote Chichijima Island. The goal of

102　this study is to characterize seasonal and annual variations of SCs and specify their possible

103　source regions. We will also discuss a potential role of Asian dust to control the distributions

104　of bioaerosols over the western North Pacific. The outcomes of this study will improve our

105　understanding about a possible influence of long-range transport of bioaerosols from the

106　continent to the clean oceanic environment. We will compare the data set of SCs for the

107　periods 1990-1993, 2001-2003 and 2010-2013, which may provide imperative information

108　about decadal changes in the atmospheric conditions over Chichijima. Seasonal source

109　identifications by positive matrix factorization (PMF) analysis will also be discussed for the

110　measured SCs.

111

112　**2 Materials and methods**

113　**2.1 Sampling site and meteorological conditions**

114　The detailed information on the sampling site was reported in Kawamura et al. (2003) and

115　Chen et al. (2013). Briefly, Chichijima Island is located in the western North Pacific (27°04'N;

116　142°13'E), 1000 km south of Tokyo, Japan, and 2000 km east of the Asian continent (Figure 1).

117　Total area of the island is 24 km^2 with a population of 2000 (Verma et al., 2015). The climate

118　of Chichijima is classified as subtropical; it is warm to hot (temperature, 7.8-34.1 °C) and

119　humid (relative humidity, 66-88%) all year round.

120　　　　Figure 2 shows monthly averaged variations in the meteorological parameters of

121　Chichijima during 2001-2013. It receives more precipitation in between April and July,

122　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

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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.

2.3 Backward air mass trajectory analysis

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

2.4 Positive matrix factorization (PMF) analysis

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

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

8

201 on given understanding of sugar sources, 4-7 factors were examined and total five interpretable

202 factors were characterized by the enrichment of each tracer compound, which reproduced more

203 than 94% of SCs. Minimal robust and true Q values of the base run were 3001 and 3413,

204 respectively. Concentrations and percentage of tracers in each factor of bootstrap run were

205 close of those of base run results. The Q values and factor profiles of F_{peak} rotation runs showed

206 no significant changes compared with base run, indicating stable PMF results. The detailed

207 discussions of the determination and application of the PMF are reported in Norris et al. (2008),

208 Paatero et al. (2002) and Zhou et al. (2004).

209   In winter/spring, Chichijima Island receives air masses enriched with anthropogenic

210 aerosols from the Asian continent by strong westerly winds, whereas during summer/autumn it

211 receives clean air masses from the Pacific Ocean under the influences of trade winds. The

212 seasonal changes in the atmospheric circulation over Chichijima may have a significant

213 influence on the seasonal distributions of SCs. Therefore, we performed the seasonal PMF

214 analysis on the thirteen year sugar data set to better understand the seasonal source profile of

215 individual sugar component. For seasonal PMF analysis, 3-5 factors were examined and 4

216 factors were determined for each season. We included the data set of anhydrosugars from

217 Verma et al. (2015) for PMF analysis.

218

219 **3 Results and discussion**

220 **3.1 Ambient concentrations of sugar compounds**

221 Temporal variations of primary sugars and sugar alcohols are shown in Figure 4. Nine sugar

222 compounds (SCs) including five primary sugars and four sugar alcohols were detected in the

223 aerosol samples collected from Chichijima Island. The concentrations of total SCs varied from

224 1.23 to 339 ng m^{-3} (average, 46.7±49.5 ng m^{-3}) during 2001 to 2013 (Table 1). Concentrations

225 of primary sugars and sugar alcohols were in the range of 0.28 to 176 ng m^{-3} (23.3±25.7 ng m^{-}

226 ^{3}) and 0.37 to 231 ng m^{-3} (23.4±30.8 ng m^{-3}), respectively. Average concentration of primary

227   sugars in Chichijima aerosols is several times lower than that of primary sugars (62.0±54.9 ng

228   m^{-3}) reported from Cape Hedo, Okinawa, Japan (Zhu et al., 2015) while that of sugar alcohols

229   is equivalent to or little lower than that from Cape Hedo (29.5±35.5 ng m^{-3}).

230   Interestingly, primary sugars (49.9%) and sugar alcohols (50.1%) were found to

231   contribute almost equal to total SCs during the entire study period. Mannitol (26.7%) and

232   arabitol (21.4%) were the main contributors to total SCs followed by glucose (16.7%), sucrose

233   (13.6%), fructose (10.2%), and trehalose (9.2%). Erythritol (1.6%), inositol (0.3%), and xylose

234   (0.3%) were also present in the aerosols at lower concentration levels. Temporal plots of

235   individual sugars clearly indicate a large variation of SCs (Figure 4). This large variation in the

236   concentrations of SCs might be involved with seasonal changes in the atmospheric circulations

237   over the western North Pacific (Kawamura et al., 2003).

238   **3.1.1 Concentrations of primary sugars in total SCs**

239   Glucose is the dominant sugar species among primary sugars with the concentration range of

240   0.05 to 64.3 ng m^{-3} (average, 7.79±8.80 ng m^{-3}). Similarly, a wide concentration range of

241   fructose (0.03-115 ng m^{-3}; 4.69±8.04 ng m^{-3}) was also observed in Chichijima aerosols.

242   Thirteen year mean concentrations of glucose and fructose were observed to be lower than

243   those (27.2 ng m^{-3} and 16.4 ng m^{-3}, respectively) reported for the aerosol samples (TSP) from

244   Cape Hedo, Okinawa, Japan (Zhu et al., 2015). Glucose and fructose significantly contribute to

245   total primary sugars (33.5% and 20.17%, respectively) in Chichijima aerosols. Primary sugars

246   are abundant in the fragments of vascular plants in vegetated and forest areas (Medeiros et al.,

247   2006). Pacini et al. (2000) reported that primary sugars are synthesized in leaves during

248   photosynthesis and stored in root, stem, flower, pollen and fruit of growing plants. The nectars

249   and fruits of tropical and subtropical plants also contain glucose and fructose abundantly

250   (Backer et al., 1998). Graham et al. (2002) reported significant amounts of glucose and

251   fructose in pollen, fern spores, and insects in aerosol samples collected from the Amazon forest.

252   Chichijima Island is covered with endemic and vascular plants, which may emit glucose and

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

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

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

13

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

357 insignificant via long-range atmospheric transport. Seasonal PMF analysis for autumn showed

358 that more than 85% of trehalose was contributed by microbial factor among four factors

359 (Figure 6d). An indirect contribution of trehalose from soil dust will be discussed later.

360   The seasonal mean concentrations of sucrose are almost equal during spring (8.80±18.0

361 ng m^{-3}), summer (7.31±11.5 ng m^{-3}) and winter (6.60±13.1 ng m^{-3}), except for autumn

362 (2.76±4.35 ng m^{-3}) (Table 1). The similar seasonal distributions suggest multiple sources of

363 sucrose in Chichijima aerosols. Monthly mean concentrations of sucrose show two peaks

364 during February-March and June-July (Figure 5i). The March peak of sucrose was reported in

365 the forest area of Sapporo, Japan to be 3 to 7 times more abundant than other months due to

366 springtime pollen emissions (Miyazaki et al., 2012). Fu et al. (2012) analyzed pollen samples

367 from different plant species (white birch, Chinese willow, Peking willow) for SCs and found

368 the highest concentrations of sucrose followed by fructose and glucose in pollen. The pollen

369 emissions from developing buds of plants may be the reason for the increased concentration of

370 sucrose and fructose in February and March over Chichijima Island in the western North

371 Pacific. Seasonal PMF analysis shows that sucrose contributed 100% in spring and >80% in

372 winter, suggesting a significant pollen contribution for sucrose in those seasons (Figures 6b

373 and 6a).

374   However, the possibilities of pollen transport from East Asia to Chichijima cannot be

375 excluded because pollens can travel long distances with springtime high-speed winds by

376 westerlies (Rousseau et al., 2006). The pollen grains emitted from flowering boreal forest in

377 China, Mongolia, Siberia and Russian Far East, could significantly be delivered to the western

378 North Pacific during spring, which may result in the contribution of sucrose and fructose to

379 Chichijima aerosols. Recent studies have discussed a long-range transport of airborne pollen

380 from North America to Greenland in spring (Rousseau et al., 2008). Lorenzo et al. (2006)

381 reported the long-range transport of airborne allergenic pollen to central Italy. Makra et al.

382 (2010) reported a long-range transport of airborne pollen in three European cities by applying

383 three-dimensional clustering of backward trajectories. Several studies also discussed a long-

384 range transport of pollen to the remote arctic region (Andrews et al., 1980; Bourgeois et al.,

385 2001; Campbell et al., 1999; Hicks et al., 2001; Hjelmroos and Franzen, 1994; Rousseau et al.,

386 2004).

387   These observations may support that westerly winds have delivered pollen grains from

388 the Asian continent including Mongolia, Siberia, and Russian Far East to Chichijima Island in

389 spring. Using a box model and typical settling velocity of pollens (3 cm/sec) with the grain size

390 of 30 μm in diameter (Sosnoskie et al., 2009), we estimated lifetime of pollen grains to be 9.3

391 hours in the atmospheric marine boundary layer (height of 1 km above the ocean surface). The

392 settling velocity of the pollens is ca. 20 times larger than that of typical marine aerosols (Slinn

393 and Slinn, 1980). Because pollen grain sizes range from 10 μm to 100 μm in diameter, the

394 lifetime of pollens may have a large uncertainty. If pollens could be largely transported in the

395 free troposphere (e.g., 5 km high) to the North Pacific from the Asian continent, then lifetime

396 of typical pollen grains would increase upto 2 days. These calculations for the lifetime of

397 pollen grains further support their long-range atmospheric transport from the Asian continent

398 over the western North Pacific. Based on backward air mass trajectories (Figure 3), we can

399 roughly estimate the transport time from East Asia to Chichijima site to be 2-4 days in winter

400 and spring. It is also of interest to note that pollens can rapture under condition of high relative

401 humidity (RH) (Hader et al., 2014; Miguel et al., 2006; Wright et al., 2014), which leads to

402 smaller particles with longer residence time in the atmosphere.

403   In addition, tilling process after wheat crop harvesting in farmland causes an enhanced

404 exposure of wheat root associated with soil particles into the atmosphere. China, India, and

405 USA are three largest countries for wheat production in the world. In China and India there are

406 two seasons (spring and winter) for wheat crops; winter wheat is harvested from mid-May to

407 mid-July. During those periods (early summer), Chichijima Island is highly influenced by trade

408 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).

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).

487   The comparison for three periods indicates that concentrations of anhydrosugars are

488   highest in winter followed by autumn. Their concentrations significantly increased from P-I to

489   P-II/P-III (Figure 8a). The detailed discussions on anhydrosugars were reported in Verma et al.

490   (2015). Here, we refer the data set of anhydrosugars for the decadal comparison with SCs.

491   Interestingly, biomass-burning tracers (BB-tracers; levoglucosan, mannosan and galactosan)

492   showed a significant difference in the decadal trends among three periods (i.e., P-I, P-II, and P-

493   III) during winter and autumn. In winter, BB-tracers showed an increasing trend from P-I to P-

494   III (Figure 8a). Biomass burning is common in winter for house heating (Simoneit et al.,

495   2004a), thus it is obvious that the lower ambient temperature is the more biomass burning

496   activities are. Westerly winds abundantly transport biomass-burning products over Chichijima

497   Island in the western North Pacific with air masses derived from East Asia, Siberia, Mongolia,

498   and Russian Far East during winter (Bendle et al., 2007; Simoneit and Elias, 2010; Verma et al.,

499   2015). In contrast, BB-tracers in autumn show an opposite trend (i.e., higher concentrations in

500   P-I followed by P-II and P-III) compared to those in winter (Figure 8a).

501   The difference in the concentrations of anhydrosugars in winter and autumn during P-I is

502   insignificant while the concentrations are 3 and 5 times higher in winter than autumn for P-II

503   and P-III, respectively. This seasonal shifting in the concentrations of anhydrosugars may be

504   attributable to the changes in the strength of both westerly and trade wind systems from mid

505   autumn to early winter among three periods (Chen et al., 2013). In contrast, the concentrations

506   of primary sugars were 2 to 7 times higher during P-II and P-III than P-I period in summer and

507   autumn (Figure 8b). PMF analysis showed that local emissions from vegetation are important

508   contributor for primary sugars (glucose, fructose and sucrose). Therefore, a drastic increase in

509   the concentrations of primary sugars in summer/autumn for P-II and P-III than P-I may be

510   caused by an increased emission of primary sugars by local vegetation under the influence of

511   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.

641   Interestingly, we found an increasing trend in % contribution of vegetation, pollen, and

642   BB factors to SCs in 2006 to 2013 (see Figure 11). Sugar components, which are contributed

643   from pollen (sucrose, fructose, and inositol), vegetation (glucose and fructose), and biomass

644   burning sources (levoglucosan, galactosan, and mannosan), also show a similar increasing

645   trend for the period of 2006 to 2013 (Figure 7). The increased annual trends of BB and pollen

646   factors might be due to an enhanced long-range transport of airborne pollens and biomass

647   burning products from the Asian continent to the western North Pacific under the influence of

648   strong westerly winds.

649

650   **4 Summary and conclusions**

651   We reported thirteen years of temporal, seasonal and decadal trends of sugar compounds (SCs)

652   measured in the aerosol samples collected at Chichijima Island in the western North Pacific, an

653   outflow region of Asian aerosols. The high abundances of total SCs and primary sugars are

654   found during summer, whereas sugar alcohols are almost equally distributed during summer

655   and autumn. The seasonal distributions of arabitol, mannitol, and trehalose are strongly

656   influenced by long-range atmospheric transport of bioaerosols associated with microbes such

657   as fungi and bacteria and their metabolic activities under the influences of westerly winds and

658   favorable meteorological conditions, including high RH and temperature in summer/autumn.

659   Seasonal variation of sucrose is controlled by both locally emitted and long-range transported

660   pollen from East Asia to the western North Pacific during spring bloom periods. On the other

661   hand, the increased concentrations of sucrose and fructose during summer may be caused by

662   the local activity of vegetation and possibly by long-range transport of plant root-associated

663   soil dust particles from East and Southeast Asia. PMF analysis indicated specific sources for

664   individual SCs during different seasons. The results clearly separated biogenic emissions into

665   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.

Acknowledgements

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References

696   **References**

697   Andrews, J. T., W. N. Mode, and P. T. Davis (1980), Holocene climate based on pollen transfer-
698   functions, Eastern Canadian Arctic, Arct. Alp. Res., 12, 41-64.

699   Baker, H. G., I. Baker, S. A. Hodges (1998), Sugar composition of nectars and fruits consumed
700   by birds and bats in the tropics and subtropics, Biotropica. 30, 559-586.

701   Bauer, H., E. Schueller, G. Weinke, A. Berger, R. Hitzenberger, I. L. Marr, and H. Puxbaum
702   (2008a), Significant contributions of fungal spores to the organic carbon and to the
703   aerosol mass balance of the urban atmospheric aerosol, Atmos. Environ., 42(22), 5542-
704   5549.

705   Bauer, H., M. Claeys, R. Vermeylen, E. Schueller, G. Weinke, A. Berger, and H. Puxbaum
706   (2008b), Arabitol and mannitol as tracers for the quantification of airborne fungal spores,
707   Atmos. Environ., 42(3), 588-593.

708   Bendle, J., K. Kawamura, K. Yamazaki, and T. Niwai (2007), Latitudinal distribution of
709   terrestrial lipid biomarkers and n–alkane compound–specific stable carbon isotope ratios
710   in the atmosphere over the western Pacific and Southern Ocean, Geochim. Cosmochim.
711   Acta, 71(24), 5934–5955.

712   Bieleski, R. L. (1995), Onset of phloem export from senescent petals of Daylily, Plant Physiol.,
713   109(2), 557-565.

714   Bourgeois, J. C., K. Gajewski, and R. M. Koerner (2001), Spatial patterns of pollen deposition
715   in arctic snow, J. Geophys. Res., 106, 5255-5265.

716   Bremer, G. B. (1995), Lower marine fungi (Labyrinthulomycetes) and the decay of mangrove
717   leaf-litter, Hydrobiologia, 304(3), 243-245.

718   Brown, J. K. M., and M. S. Hovmoller (2002), Epidemiology—Aerial dispersal of pathogens on
719   the global and continental scales and its impact on plant disease, Science, 297(5581),
720   537–541.

721   Burshtein, N., N. Lang-Yona, and Y. Rudich (2011), Ergosterol, arabitol and mannitol as tracers
722   for biogenic aerosols in the eastern Mediterranean, Atmos. Chem. Phys., 11(2), 829-839.

723   Bzdusek P.A., E. R. Christensen, C. M. Lee, U. Pakadeesusuk, D. C. Freedman, (2006). PCB
724   congeners and dechlorination in sediments of Lake Hartwell, South Carolina, determined
725   from cores collected in 1987 and 1988. Environ. Sci. Technol., 40, 109–119.

726   Campbell, I. D., K. McDonald, M. Flannigan, and J. Kringayark (1999), Long-distance transport
727   of pollen into the Arctic, Nature, 399, 29– 30.

728   Carvalho, A., C. Pio, and C. Santos (2003), Water-soluble hydroxylated organic compounds in
729   German and Finnish aerosols, Atmos. Environ., 37(13), 1775-1783.

730   Chen, J., K. Kawamura, C. Q. Liu, and P. Q. Fu (2013), Long-term observations of saccharides
731   in remote marine aerosols from the western North Pacific: A comparison between 1990-
732   1993 and 2006-2009 periods, Atmos. Environ., 67, 448-458.

733   Cowie, G. L., and J. I. Hedges (1984), Carbohydrate sources in a coastal marine-environment,
734   Geochim. Cosmochim. Acta, 48(10), 2075-2087.

735   Dahlman, L., J. Persson, T. Nasholm, and K. Palmqvist (2003), Carbon and nitrogen distribution
736   in the green algal lichens Hypogymnia physodes and Platismatia glauca in relation to
737   nutrient supply, Planta, 217(1), 41-48.

738   Duce, R. A., C. K. Unni, B. J. Ray, J.M. Prospero, J.T. Merrill (1980), Long-range atmospheric

739   transport of soil dust from Asia to the tropical North Pacific: temporal variability,
740   Science, 209, 1522–1524.

741 Elbein, A. D. (1974), The metabolism of α, α-trehalose, Adv. Carboh. Chem. and Biochem., 30,
742   227–256.

743 Elbert, W., P. E. Taylor, M. O. Andreae, and U. Pöschl (2007), Contribution of fungi to primary
744   biogenic aerosols in the atmosphere: wet and dry discharged spores, carbohydrates, and
745   inorganic ions, Atmos. Chem. Phys., 7(17), 4569-4588.

746 Elliot S., Blake D.R., Duce R.A., Lai C.A., McCreary I., McNair L.A., Rowland F.S., Russell
747   A.G., Streit G.E., and Turco R.P. Motorization of China implies changes in pacific air
748   chemistry and primary production. Geophys. Res. Letts. 24, 2671 -2674, 1997.

749 Filippo, P. D., D. Pomata, C. Riccardi, F. Buiarelli, and C. Perrino (2013), Fungal contribution
750   to size-segregated aerosol measured through biomarkers, Atmos. Environ., 64,132-140.

751 Fraser, M. P., and K. Lakshmanan (2000), Using levoglucosan as a molecular marker for the
752   long-range transport of biomass combustion aerosols, Environ. Sci. Technol., 34(21),
753   4560-4564.

754 Fu, P. Q., K. Kawamura, M. Kobayashi, and B. R. T. Simoneit (2012), Seasonal variations of
755   sugars in atmospheric particulate matter from Gosan, Jeju Island: Significant
756   contributions of airborne pollen and Asian dust in spring, Atmos. Environ., 55, 234-239.

757 Fuzzi, S., et al. (2007), Overview of the inorganic and organic composition of size-segregated
758   aerosol in Rondonia, Brazil, from the biomass-burning period to the onset of the wet
759   season, J. Geophys. Res.-Atmos., 112(D1), doi:10.1029/2005jd006741.

760 Graham, B., O. L. Mayol-Bracero, P. Guyon, G. C. Roberts, S. Decesari, M. C. Facchini, P.
761   Artaxo, W. Maenhaut, P. Koll, and M. O. Andreae (2002), Water-soluble organic
762   compounds in biomass burning aerosols over Amazonia - 1. Characterization by NMR
763   and GC-MS, J. Geophys. Res.-Atmos., 107(D20), doi:10.1029/2001jd000336.

764 Graham, B., P. Guyon, P. E. Taylor, P. Artaxo, W. Maenhaut, M. M. Glovsky, R. C. Flagan, and
765   M. O. Andreae (2003), Organic compounds present in the natural Amazonian aerosol:
766   Characterization by gas chromatography-mass spectrometry, J. Geophys. Res.-Atmos.,
767   108(D24), doi:10.1029/2003jd003990.

768 Griffin, D. W., et al. (2001), African desert dust in the Caribbean atmosphere: Microbiology and
769   public health, Aerobiologia, 17, 203–213.

770 Griffin, D. W., et al. (2003), Atmospheric microbiology in the northern Caribbean during
771   African dust events, Aerobiologia, 19, 143–157.

772 Griffin, D. W., (2007), Atmospheric movement of microorganisms in clouds of desert dust and
773   implications for human health, Clin. Microbiol. Rev. 20, 459–477.

774 Hackl, E., G. Bachmann, and S. Zechmeister-Boltenstern (2000), Soil microbial biomass and
775   rhizosphere effects in natural forest stands, Phyton.-Ann. Rei. Bot. A, 40(4), 83-90.

776 Hader, J. D., T. P.Wright, and M. D. Petters (2014), Contribution of pollen to atmospheric ice
777   nuclei concentrations, Atmos. Chem. Phys., 14, 5433-5449.

778 Hicks, S., H. Tinsley, A. Huusko, C. Jensen, M. Hättestrand, A. Gerasimides, and E. Kvavadze
779   (2001), Some comments on spatial variation in arboreal pollen deposition: First records
780   from the Pollen Monitoring Programme (PMP), Rev. Palaeobot. Palynol., 117, 183-194.

781 Hirst J. M., O. J. Stedman, and W. H. Hogg (1967), Long-distance spore transport: Methods of
782   measurement, vertical spores profile and the detection of immigrant spores, J. Gen.

783   Microbiol. 48, 329-355.

784 Hjelmroos, M., and L. G. Franzen (1994), Implications or recent long distance pollen transport
785   events for the interpretation of fossil pollen records in Fennoscandia, Rev. Palaeobot.
786   Palynol., 82, 175-189.

787 Hua, N.-P. et al. (2007), Detailed identification of desert-originated bacteria carried by Asian
788   dust storms to Japan, Aerobiologia, 23, 291-298.

789 Hyde, K. D., E. B. G. Jones, E. Leano, S. B. Pointing, A. D. Poonyth, and L. L. P. Vrijmoed
790   (1998), Role of fungi in marine ecosystems, Biodivers. Conserv., 7(9), 1147-1161.

791 Ibrahim, M., A. B. Rabah, B. Liman, and N. T. Ibrahim (2011), Effect of temperature and
792   relative humidity on the growth of Helminthosporium fulvum, Nigerian J. Basic Appli.
793   Sci., 19(1) 127- 129.

794 Ion, A. C., R. Vermeylen, I. Kourtchev, J. Cafmeyer, X. Chi, A. Gelencser, W. Maenhaut, and
795   M. Claeys (2005), Polar organic compounds in rural PM(2.5) aerosols from K-puszta,
796   Hungary, during a 2003 summer field campaign: Sources and diel variations, Atmos.
797   Chem. Phys., 5, 1805-1814.

798 Iwasaka, Y., et al. (1983), The transport and spacial scale of Asian dust-storm clouds: a case
799   study of the dust-storm event of April 1979, Tellus, 35B, 189–196.

800 Jaffe, D., et al. (1999), Transport of Asian air pollution to North America, Geophys. Res. Lett.,
801   26(6), 711–714.

802 Jaffe, D., I. McKendry, T. Anderson, and H. Price (2003), Six "new" episodes of trans-Pacific
803   transport of air pollutants, Atmos. Environ., 37(3), 391–404.

804 Jaffe, D.A., Mahura A., Kelley, J., Atkins J., Novelli P.C. and Merrill J. Impact of Asian
805   Emissions on the Remote North Pacific Atmosphere: Interpretation of CO Data from
806   Shemya, Guam, Midway and Mauna Loa. J. Geophys. Res. 23, 28,627-28,636, 1997.

807 Jeon, E.-M. et al. (2011), Impact of Asian dust events on airborne bacterial community assessed
808   by molecular analyses, Atmos. Environ. 45, 4313–4321.

809 Jia, Y. L., A. L. Clements, and M. P. Fraser (2010), Saccharide composition in atmospheric
810   particulate matter in the southwest US and estimates of source contributions, J. Aerosol
811   Sci., 41(1), 62-73.

812 Jones, E.B.G. (ed.) (1976), Recent Advances in Aquatic Mycology, London: Elek Science.

813 Jones, E.B.G., and J. I. Mitchell (1996), Biodiversity of marine fungi, In Biodiversity:
814   International Biodiversity Seminar (A. Cimerman and N. Gunde-Cimerman, eds), pp. 31-
815   42. Ljubljana: National Inst. Chemistry and Slovenia National Commission for UNESCO.

816 Juntto S and P. Paatero (1994), Analysis of daily precipitation data by positive matrix
817   factorization. Environmetrics, 5, 127–144.

818 Kanakidou, M., et al. (2005), Organic aerosol and global climate modelling: a review, Atmos.
819   Chem. Phys., 5, 1053-1123.

820 Kawamura, K., Y. Ishimura, and K. Yamazaki (2003), Four years' observations of terrestrial
821   lipid class compounds in marine aerosols from the western North Pacific, Global
822   Biogeochem. Cy., 17(1), doi:10.1029/2001gb001810.

823 Kim, V. K., and C.L. Xiao (2005), Influence of culture media and environmental factors on
824   mycelial growth and picnidial production of Sphaeropsis pyriputrescens, Mycologia,
825   97(1): 25-32.

826 Kohlmeyer, J., and B. Volkmann-Kohlmeyer (1991), Illustrated key to the filamentous fungi,
827   Bot. Mar. 34, 1-61.

828 Kohlmeyer, J., and E. Kohlmeyer (1979), Marine Mycology: The Higher Fungi, London:
829   Academic Press.

830 Lacey, M. E., and J. S. West (2006), The Air Spora: a Manual for Catching and Identifying
831   Airborne Biological Particles, pp. 32-33.

832 Lewis, D. H., and D. C. Smith (1967), Sugar alcohols (polyols) in fungi and green plants. I.
833   Distribution physiology and metabolism, New Phytologist, 66(2), 143.

834 Loos, H., R. Kramer, H. Sahm, and G. A. Sprenger (1994), Sorbitol promotes growth of
835   zymomonas mobilis in environments with high concentrations of sugar: evidence for a
836   physiological function of glucose-fructose oxidoreductase in osmoprotection, J.
837   Bacteriol., 176(24), 7688-7693.

838 Lu J., P. Jiang, et al. (2008), Assessing soil quality data by positive matrix factorization.
839   Geoderma, 145, 259-266.

840 Ma, S. X., Z. Z. Wang, X. H. Bi, et al. (2009), Composition and source of saccharides in
841   aerosols in Guangzhou, China, Chinese Sci. Bull., 54, 4500-4506.

842 Makra, L., T. Santa, I. Matyasovszky, A. Damialis, K. Karatzas, K. C. Bergmann, and D. Vokou
843   (2010), Airborne pollen in three European cities: Detection of atmospheric circulation
844   pathways by applying three-dimensional clustering of backward trajectories, J. Geophys.
845   Res.-Atmos., 115, doi:10.1029/2010jd014743.

846 Malik, V. K., and S. Singh (2004), Effect of temperature and relative humidity on teliospore
847   germination in Ustilago hordei, J. Mycol. Plant Pathol., 34, 410-411.

848 Martin, S. T., et al. (2010), Sources and Properties of Amazonian Aerosol Particles, Rev.
849   Geophys., 48, doi:10.1029/2008rg000280.

850 Medeiros, P. M., M. H. Conte, J. C. Weber, and B. R. T. Simoneit (2006), Sugars as source
851   indicators of biogenic organic carbon in aerosols collected above the Howland
852   Experimental Forest, Maine, Atmos. Environ., 40(9), 1694-1705.

853 Medeiros, P. M., and B. R. T. Simoneit, (2008), Source profiles of organic compounds emitted
854   upon combustion of green vegetation from temperate climate forests, Environ. Sci.
855   Technol., 42, 8310-8316.

856 Miguel, A. G., Taylor P. E., House J., Glovsky M. M., and Flagan R. C. (2006), Meteorological
857   influences on respirable fragment release from Chinese elm pollen, Aerosol Sci. Technol.
858   40, 690-696.

859 Mims, S. A., and F. M. Mims (2003), Fungal spores are transported long distances in smoke
860   from biomass fires, Atmos. Environ., 38, 651-655.

861 Miyazaki, Y., J. Jung, P. Fu, Y. Mizoguchi, K. Yamanoi, and K. Kawamura (2012), Evidence of
862   formation of submicrometer water-soluble organic aerosols at a deciduous forest site in
863   northern Japan in summer, J. Geophys. Res., 117, D19213, doi:10.1029/2012JD018250.

864 Mochida, M., A. Kawabata, K. Kawamura, H. Hatsushika, and K. Yamazaki (2003a), Seasonal
865   variation and origins of dicarboxylic acids in the marine atmosphere over the western
866   North Pacific, J. Geophys. Res.-Atmos., 108(D6), doi:10.1029/2002jd002355.

867 Mochida, M., K. Kawamura, N. Umemoto, M. Kobayashi, S. Matsunaga, H. J. Lim, B. J. Turpin,
868   T. S. Bates, and B. R. T. Simoneit (2003b), Spatial distributions of oxygenated organic
869   compounds (dicarboxylic acids, fatty acids, and levoglucosan) in marine aerosols over

870 the western Pacific and off the coast of East Asia: Continental outflow of organic
871 aerosols during the ACE-Asia campaign, J. Geophys. Res.-Atmos., 108(D23),
872 doi:10.1029/2002jd003249.

873 Mochida, M., K. Kawamura, P. Q. Fu, and T. Takemura (2010), Seasonal variation of
874 levoglucosan in aerosols over the western North Pacific and its assessment as a biomass-
875 burning tracer, Atmos. Environ., 44(29), 3511-3518.

876 Morris, C. E., D. G. Georgakopoulos, and D. C. Sands (2004), Ice nucleation active bacteria and
877 their potential role in precipitation, J. Phys. Iv., 121, 87-103.

878 Moss, S.T. (ed.) (1986), The Biology of Marine Fungi, Cambridge: Cambridge University Press.

879 Nakagiri, A., S. Y. Newell, T. Ito, and T. K. Tan (1996), Biodiversity and ecology of the
880 oomycetous fungus, Halophytophthora, In Biodiversity and the Dynamics of Ecosystems.
881 (DIWPA series. Vol.1) (I.M. Turner, C.H. Diong, S.S.L. Lim, and P.K.L. Ng, eds), pp.
882 273-80. Tokyo: International Network for DIVERSITAS in Western Pacific and Asia.

883 Nolte, C. G., J. J. Schauer, G. R. Cass, and B. R. T. Simoneit (2001), Highly polar organic
884 compounds present in wood smoke and in the ambient atmosphere, Environ. Sci.
885 Technol., 35(10), 1912-1919.

886 Paatero, P., and U. Tapper (1994), Positive Matrix Factorization - a Nonnegative Factor Model
887 with Optimal Utilization of Error-Estimates of Data Values, Environmetrics, 5(2), 111-
888 126.

889 Paatero, P. et al. (2002), Understanding and controlling rotations in factor analytic models,
890 Chemom. Intell. Lab. Syst., 60, 253-264.

891 Pacini, E. (2000), From anther and pollen ripening to pollen presentation, Plant Syst. Evol. 222,
892 19-43.

893 Pashynska, V., R. Vermeylen, G. Vas, W. Maenhaut, and M. Claeys (2002), Development of a
894 gas chromatographic/ion trap mass spectrometric method for the determination of
895 levoglucosan and saccharidic compounds in atmospheric aerosols. Application to urban
896 aerosols, J. Mass Spectrom., 37(12), 1249-1257.

897 Pavuluri, C. M., K. Kawamura, E. Tachibana, and T. Swaminathan (2010), Elevated nitrogen
898 isotope ratios of tropical Indian aerosols from Chennai: Implication for the origins of
899 aerosol nitrogen in South and Southeast Asia, Atmos. Environ., 44(29), 3597-3604.

900 Polissar A.V., P. K. Hopke, P. Paatero, Y. J. Kaufmann, D. K. Hall, B.A. Bodhaine, E.G. Dutton,
901 J. M. Harris, (1999). The aerosol at Barrow, Alaska: long-term trends and source
902 locations. Atmos. Environ., 33, 2441–2458.

903 Prospero, J.M. and D.L. Savoie (1989), Effect of continental sources of nitrate concentrations
904 over the Pacific Ocean, Nature, 339, 687- 689.

905 Prospero, J. M. et al. (2005), Interhemispheric transport of viable fungi and bacteria from Africa
906 to the Caribbean with soil dust, Aerobiologia, 21, 1–19.

907 Rogge, W. F., P. M. Medeiros, and B. R. T. Simoneit (2006), Organic marker compounds for
908 surface soil and fugitive dust from open lot dairies and cattle feedlots, Atmos. Environ.,
909 40(1), 27-49.

910 Rogge, W. F., P. M. Medeiros, and B. R. T. Simoneit (2007), Organic marker compounds in
911 surface soils of crop fields from the San Joaquin Valley fugitive dust characterization
912 study, Atmos. Environ., 41, 8183-8204.

913 Rousseau, D.-D., P. Schevin, J. Ferrier, D. Jolly, T. Andreasen, S. E. Ascanius, S. E. Hendriksen,

914 and U. Poulsen (2008), Long-distance pollen transport from North America to Greenland
915 in spring, J. Geophys. Res.-Biogeo., 113(G2), doi:10.1029/2007jg000456.

916 Rousseau, D.-D., D. Duzer, J.-L. Etienne, G. Cambon, D. Jolly, J. Ferrier, and P. Schevin (2004),
917 Pollen record of rapidly changing air trajectories to the North Pole, J. Geophys. Res., 109,
918 D06116, doi:10.1029/2003JD003985.

919 Rousseau, D.-D., P. Schevin, D. Duzer, G. Cambon, J. Ferrier, D. Jolly, and U. Poulsen (2006),
920 New evidence of long distance pollen transport to southern Greenland in late Spring, Rev.
921 Palaeobot. Palynol., 141, 277–286.

922 Schmidl, C., H. Bauer, A. Dattler, R. Hitzenberger, G. Weissenboeck, I. L. Marr, and H.
923 Puxbaum (2008), Chemical characterisation of particle emissions from burning leaves,
924 Atmos. Environ., 42(40), 9070-9079.

925 Seinfeld, J. H., et al., (2004), ACE-ASIA - Regional climatic and atmospheric chemical effects
926 of Asian dust and pollution, Bull. Amer. Meteorol. Soc., 85(3), 367-380.

927 Sharma, R. and R. C. Rajak (2003), Keratinophilic fungi: Nature's keratin degrading machines,
928 Resonance, 8, 28-30.

929 Simoneit, B. R. T. (2002), Biomass burning - A review of organic tracers for smoke from
930 incomplete combustion, Appl. Geochem., 17(3), 129-162.

931 Simoneit, B. R. T. (2004), Biomarkers (molecular fossils) as geochemical indicators of life, Adv.
932 Space. Res., 33(8), 1255-1261.

933 Simoneit, B. R. T., and V. O. Elias (2000), Organic tracers from biomass burning in
934 atmospheric particulate matter over the ocean, Mar. Chem., 69(3-4), 301-312.

935 Simoneit, B. R. T., V. O. Elias, M. Kobayashi, K. Kawamura, A. I. Rushdi, P. M. Medeiros, W.
936 F. Rogge, and B. M. Didyk (2004a), Sugars-dominant water-soluble organic compounds
937 in soils and characterization as tracers in atmospheric particulate matter, Environ. Sci.
938 Technol., 38(22), 5939-5949.

939 Simoneit, B. R. T., M. Kobayashi, M. Mochida, K. Kawamura, M. Lee, H. J. Lim, B. J. Turpin,
940 and Y. Komazaki (2004b), Composition and major sources of organic compounds of
941 aerosol particulate matter sampled during the ACE-Asia campaign, J. Geophys. Res.-
942 Atmos., 109(D19), doi:10.1029/2004jd004598.

943 Slinn, S. A., and W. G. N. Slinn (1980), Predictions for particle deposition on natural waters,
944 Atmos. Environ., 14, 1013-1016.

945 Soonthornnonda P. and E. R. Christensen (2008). Source apportionment of pollutants and flows
946 of combined sewer wastewater. Water Res., 42, 1989–1998.

947 Sosnoskie, L. M., T. M. Webster, D. Dales, G. C. Rains, T. L. Grey, and A. S. Culpepper (2009),
948 Pollen grain size, density, and settling velocity for Palmer amaranth (Amaranthus
949 palmeri), Weed Science, 57, 404-409. http://dx.doi.org/10.1614/WS-08-157.1.

950 Speranza, A., G. L. Calzoni, and E. Pacini (1997), Occurrence of mono- or disaccharides and
951 polysaccharide reserves in mature pollen grains, Sex. Plant Reprod., 10(2), 110–115.

952 Sullivian, A. P., N. Frank, D. M. Kenski, Jr., and J. L. Collett (2011), Application of high-
953 performance anion-exchange chromatography-pulsed amperometric detection for
954 measuring carbohydrates in routine daily filter samples collected by a national network: 2.
955 Examination of sugar alcohols/polyols, sugars, and anhydrosugars in the upper Midwest,
956 J. Geophys. Res. 116. http://dx.doi.org/10.1029/2010JD014169.

957 Talbot R.W. et al. (1997), Chemical characteristics of continental outflow from Asia to the

958 troposphere over the western Pacific Ocean during February-March 1994: Results from
959 PEM -West B. J. Geophys. Res., 102, 28,255-28,274, 1997.

960 Tominaga, S., K. Matsumoto, N. Kaneyasu, A. Shigihara, K. Katono, N. Igawa (2011),
961 Measurements of particulate sugars at urban and forested suburban sites, Atmos. Environ.
962 45, 2335-2339.

963 Uno, I., K. Eguchi, K. Yumimoto, T. Takemura, A. Shimizu, M. Uematsu, Z. Y. Liu, Z. F.
964 Wang, Y. Hara, and N. Sugimoto (2009), Asian dust transported one full circuit around
965 the globe, Nat. Geosci., 2(8), 557-560.

966 Verma, S. K., K. Kawamura, J. Chen, P. Q. Fu and C. Zhu (2015), Thirteen years observation of
967 biomass-burning organic tracers over Chichijima Island in the western North Pacific: An
968 outflow region of Asian aerosols, J. Geophys. Res. Atmos., 120,
969 doi:10.1002/2014JD022224. .

970 Wan, E. C. H., and J. Z. Yu (2007), Analysis of sugars and sugar polyols in atmospheric
971 aerosols by chloride attachment in liquid chromatography/negative ion electrospray mass
972 spectrometry, Environ. Sci. Technol., 41(7), 2459-2466.

973 Wang, G. H., C. L. Chen, J. J. Li, B. H. Zhou, M. J. Xie, S. Y. Hu, K. Kawamura, and Y. Chen
974 (2011), Molecular composition and size distribution of sugars, sugar-alcohols and
975 carboxylic acids in airborne particles during a severe urban haze event caused by wheat
976 straw burning, Atmos. Environ., 45(15), 2473-2479,

977 Wang, G. H., K. Kawamura, and M. Lee (2009), Comparison of organic compositions in dust
978 storm and normal aerosol samples collected at Gosan, Jeju Island, during spring 2005,
979 Atmos. Environ., 43(2), 219-227.

980 Wiemken, A. (1990), Trehalose in Yeast, Stress Protectant Rather Than Reserve Carbohydrate,
981 Anton. Leeuw. Int. J. G, 58(3), 209-217.

982 Wright, T. P., J. D. Hader, G. R. McMeeking, and M. D. Peters (2014), High relative humidity
983 as a trigger for widespread release of ice Nuclei, Aerosol Sci. Technol., 48, 11, i-v, DOI:
984 10.1080/02786826.2014.968244.

985 Xie Y. and C. M. Berkowitz (2006), The use of positive matrix factorization with conditional
986 probability functions in air quality studies: an application to hydrocarbon emissions in
987 Houston, Texas. Atmos. Environ., 40, 3070– 3091.

988 Xu, W. Y., et al. (2011), Characteristics of pollutants and their correlation to meteorological
989 conditions at a suburban site in the North China Plain, Atmos. Chem. Phys., 11(9), 4353-
990 4369.

991 Yamaguchi, N., T. Ichijo, A. Sakotani, T. Baba, and M. Nasu (2012), Global dispersion of
992 bacterial cells on Asian dust, Sci. Rep., 2, DOI: 10.1038/srep00525.

993 Yang, Y. H., C. Y. Chan, J. Tao, M. Lin, G. Engling, Z. S. Zhang, T. Zhang, and L. Su (2012),
994 Observation of elevated fungal tracers due to biomass burning in the Sichuan Basin at
995 Chengdu City, China, Sci. Total Environ., 431, 68–77.

996 Yttri, K. E., C. Dye, and G. Kiss (2007), Ambient aerosol concentrations of sugars and sugar-
997 alcohols at four different sites in Norway, Atmos. Chem. Phys., 7(16), 4267-4279.

998 Zhang, T., Engling, G., Chan, C.-Y., Zhang, Y.-N., Zhang, Z.-S., Lin, M., Sang, X.-F., Li, Y.D.,
999 Li, Y.-S., 2010. Contribution of fungal spores to particulate matter in a tropical rainforest.
1000 Environ. Res. Lett. 5. doi:10.1088/1748-9326/1085/1082/ 024010.

1001 Zhou, L., E. Kim et al., (2004), Advanced factor analysis on Pittsburgh particle size distribution
1002 data, Aerosol Sci. Technol., 38, 118-132.

Zhu, C., K. Kawamura, and B. Kunwar (2015), Organic tracers of primary biological aerosol particles at subtropical Okinawa Island in the western North Pacific Rim, J. Geophys. Res. Atmos., 120, 5504–5523.

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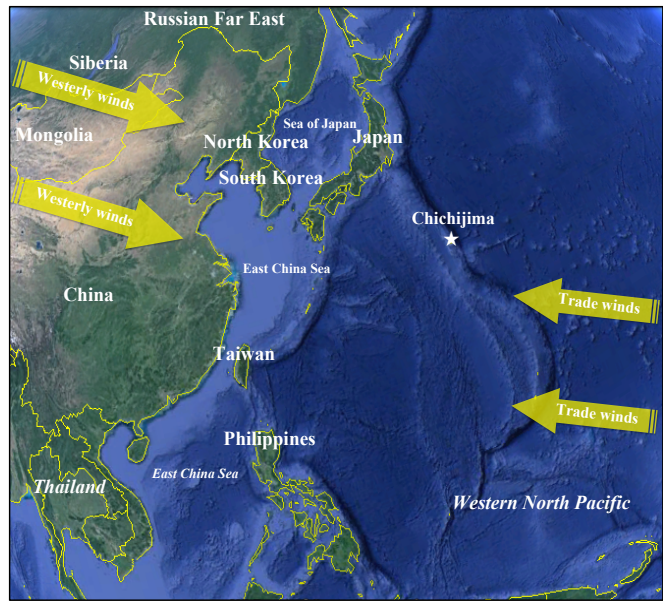
Figure 11. Annual trends in % contributions of five source factors: (a) vegetation, (b) fungal and microbial, (c) mixed, (d) biomass burning (BB), and (e) pollen factors to SCs in Chichijima aerosols. The data of 2005 are not plotted due to limited data points.

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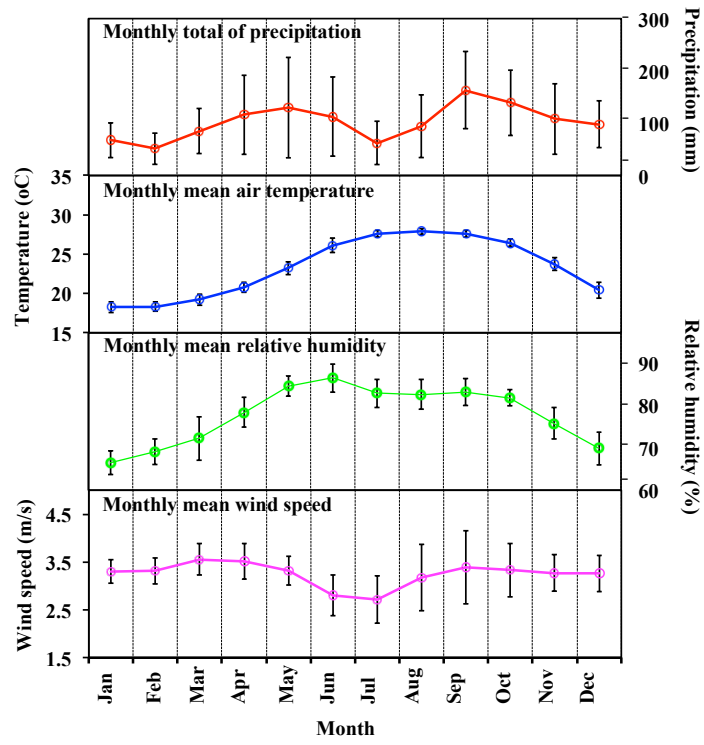1075   **Figure 1.**

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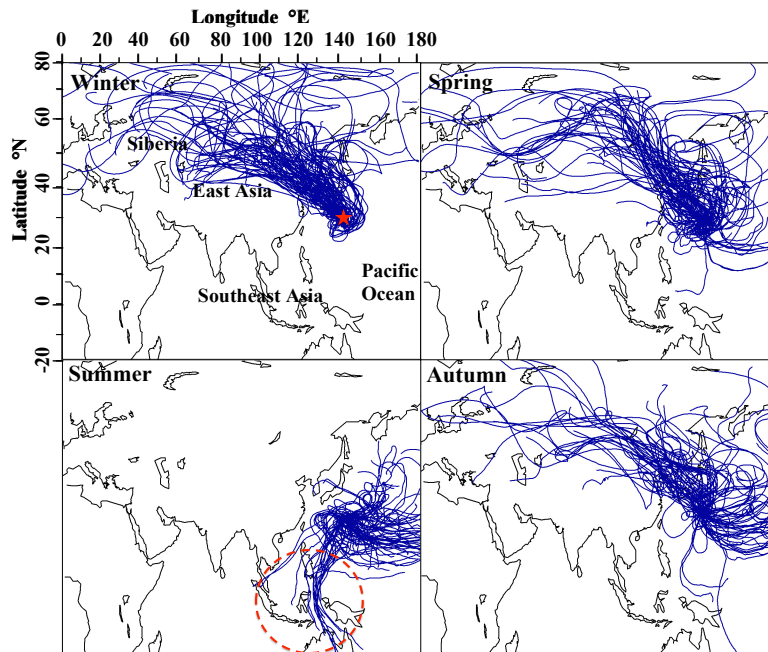1108   **Figure 2.**

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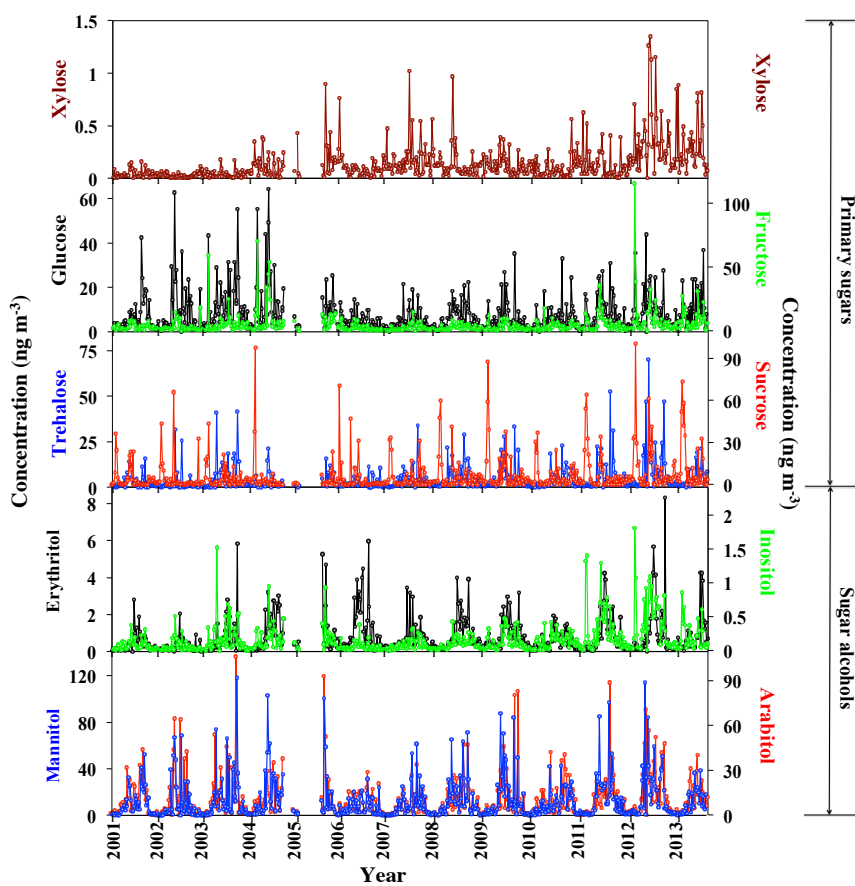1124

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1126 **Figure 3.**

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1146 **Figure 4.**

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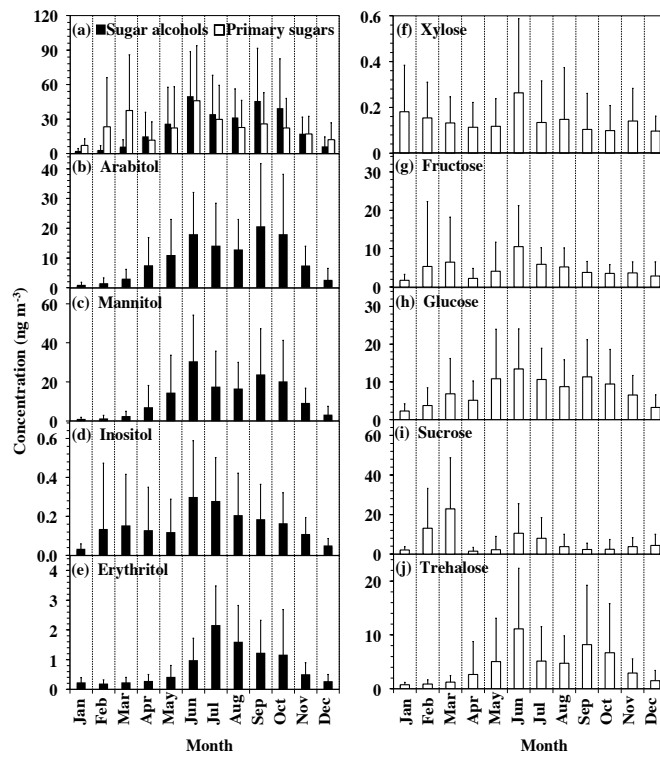
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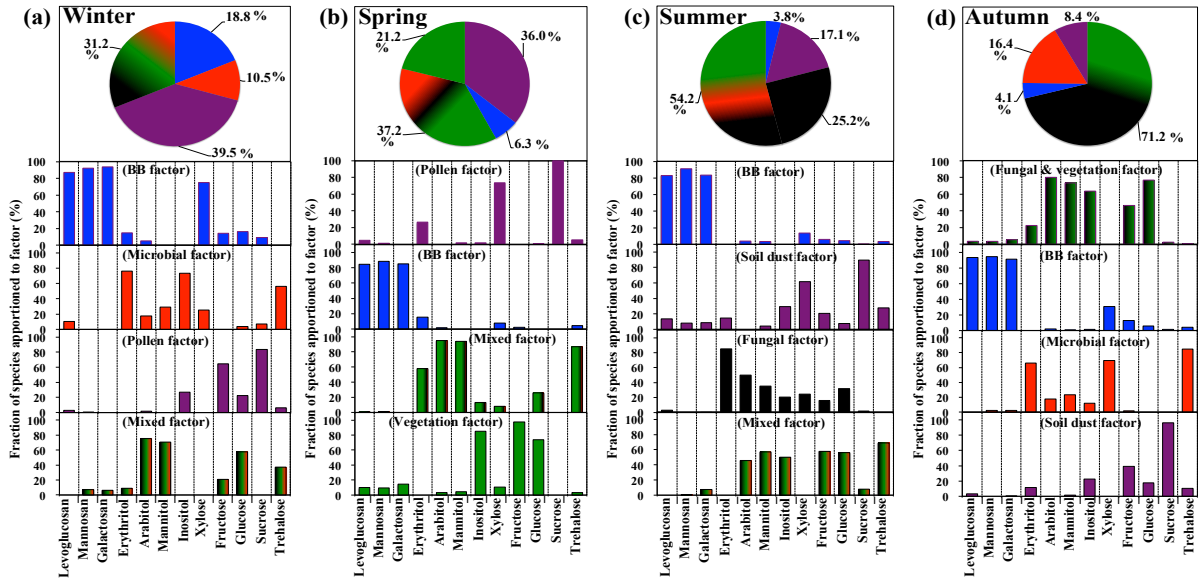
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1204   **Figure 7.**
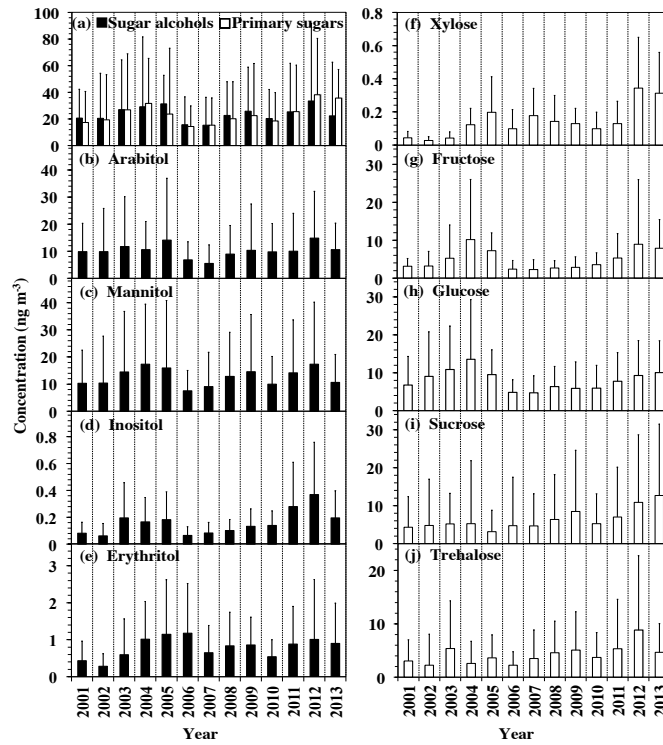
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1226 **Figure 8.**
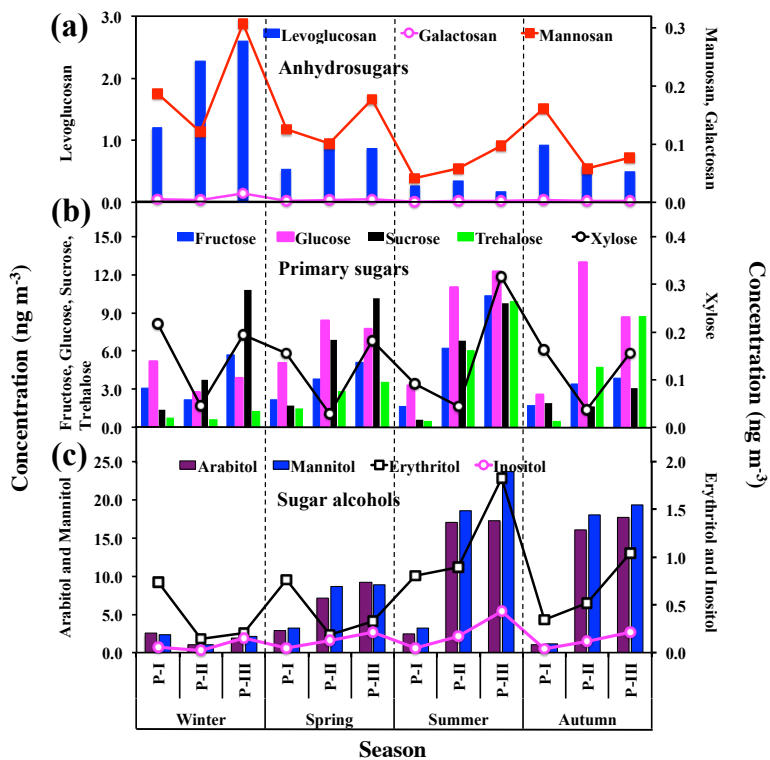
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1250 **Figure 9.**
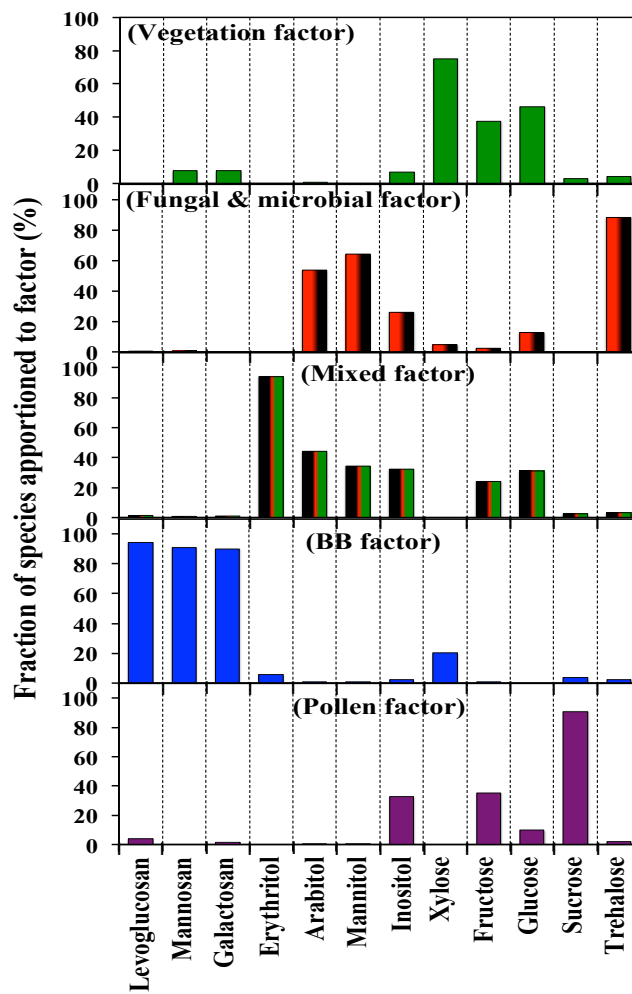
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1271   **Figure 10.**

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

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1284   **Figure 11.**

1286 **Table 1.** Seasonal concentrations (ng m^{-3}) of sugar compounds (SCs) in the aerosol samples collected at ChichiJima Island in the western North
1287 Pacific during 2001-2013.

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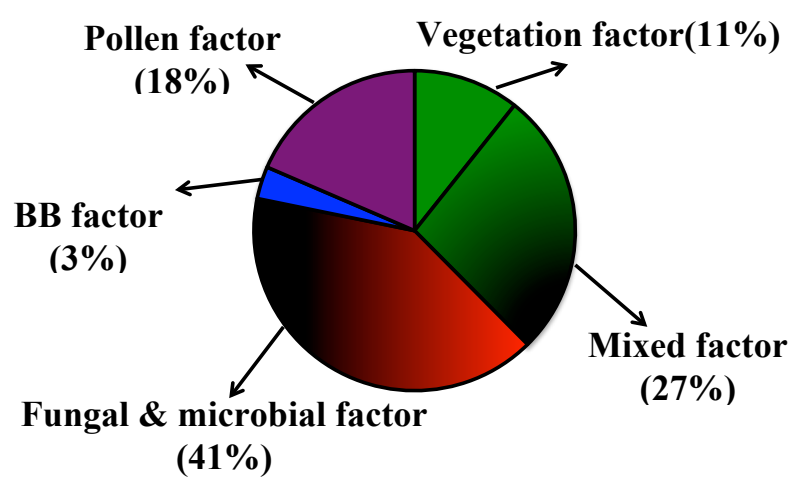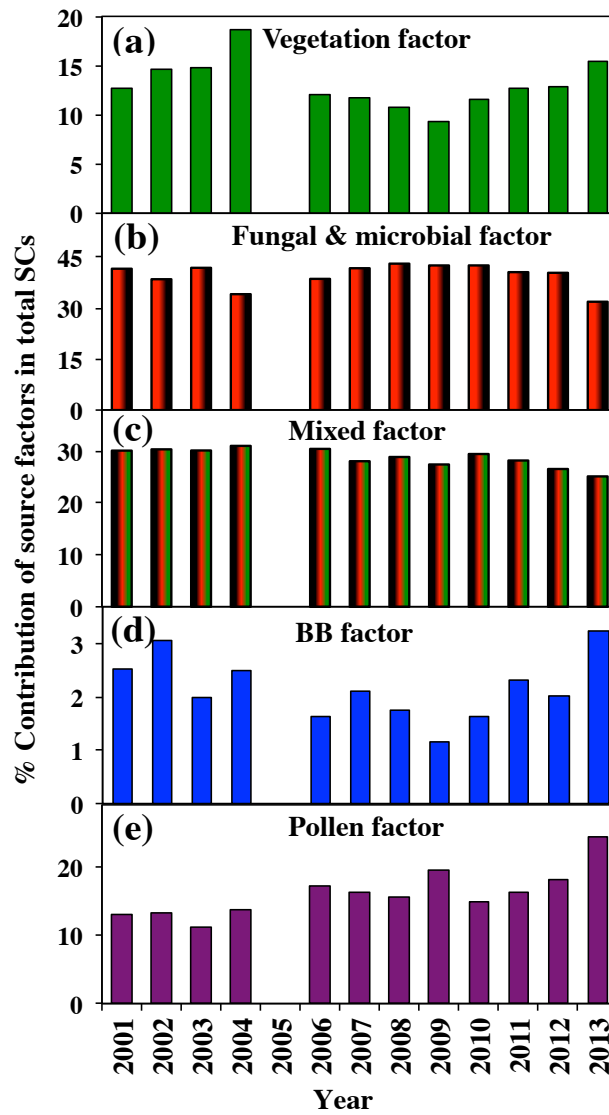| Sugars	Winter[a] (n=139)			Spring[b] (n=155)			Summer[c] (n=146)			Autumn[d] (n=150)			2001-2013 (n=590)		
	Range	Mean±SD	Med[e]	Range	Mean±SD	Med[e]	Range	Mean±SD	Med[e]	Range	Mean±SD	Med[e]	Range	Mean±SD	Med[e]
Primary sugars															
Xylose	0.05-0.89	0.14±0.15	0.12	0.001-0.56	0.12±0.11	0.08	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

1289 [a] Winter (December-February), [b]Spring (March-May), [c]Summer (June-August) and [d]Autumn (September-November); Med.=Median
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1298 **Table 2.** Pearson correlation coefficients (r) for the dataset of sugars in Chichijima aerosols during 2001-2013 (n = 590).

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	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

1300 [a] data from Verma et al. (2015)

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1315 **Table 3.** Comparisons of seasonal concentrations (ng m^{-3}) of primary sugars and relative contributions (%) of sugar compounds (SCs) in total
1316 SCs in Chichijima aerosols among 1990-1993[a], 2001-2003 and 2010-2013.
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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			

1319 [a] data from Chen et al. (2013), [b] data from Verma et al. (2015).
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