Response to the Editor’s comments

Editor’s Evaluations

The authors have reasonably addressed the comments of the two anonymous referees and they have improved the language and grammar of the main text, but additional improvement is needed as indicated below. More important, as explained in detail below, there are some serious problems with the Main text of this manuscript and there also alterations and corrections needed in both the Main text and Supporting information. Consequently, substantial revisions are needed before the manuscript can be published in ACP.

Response: Thanks for the editorial review to improve the quality of this paper. Following the Editor’s recommendation, we have carefully made corrections and revised the main text as well as supporting information. Our responses to all comments made by the Editor are given below. Please refer to the revised MS, in which changes are highlighted in yellow.

Main text:

1. Comment: Water-soluble organic carbon (WSOC) is mentioned for the first time in line 551. No information is given on how WSOC was obtained, although this information is definitely needed; it should be described within Methods. Furthermore, it should be discussed within the text what fraction of the organic carbon (OC) was WSOC.

Response: Following the Editor’s suggestion, we have added the WSOC analysis procedure in the method section. We also added the WSOC fraction in OC in the revised MS. Please see lines 186-195 and 571-580.

2. Comment: I have serious problems with the authors' application of PMF. Their number of samples (21 as mentioned in line 128) is insufficient for a PMF analysis. Henry et al. state on page 1512 of their seminal 1984 paper (full reference: Henry, R.C., Lewis, C.W., Hopke, P.K., Williamson, H.J, 1984. Review of receptor model fundamentals. Atmos. Environ. 18, 1507-1515) that, in order for a multivariate receptor model to be acceptable, the number of samples N should be larger than 30 + (V + 3)/2, with V being the number of variables. As V is 20 (as can be seen in Figure 7), this means that the number of samples in the authors’ sample set should be larger than 42. It is only 21 and thus only half the needed number. Furthermore, many researchers suggest to have at least 50 samples (and some even 100) for PMF; see also G. P. Norris et al., "EPA Positive Matrix Factorization (PMF) 5.0

Fundamentals and User Guide", April 2014, in which there is written on page 10: "PMF is often used on speciated $PM_{2.5}$ data sets with 10 to 20 species over 100 samples". Consequently, I very strongly recommend to remove section 3.5 and Figure 7 from the manuscript unless the authors can come up with convincing arguments to retain them.

Response: We comply with the Editor's view. The numbers of samples are less (n = 21) as each TSP sample was collected for 10–14 days to get enough signals for the radiocarbon measurements. We have removed PMF analysis result (section 3.5, Figure 7) in the revised MS.

3. **Comment**: Lines 94-95: This statement is in clear contradiction with lines 88-90 where it says that monoterpenes account for 11% of the monoterpene emissions. It looks like the authors misinterpreted what is written in Griffin et al. (1999); it says in Table 3 of the latter reference that α-pinene accounts for 35% of the global monoterpene emissions.

Response: We are sorry for this mistake. This sentence has been rephrased in the revised MS. Please see lines 94-95.

4. **Comment**: Abbreviations and acronyms should be defined (written full-out) when first used. Consequently, on line 186, "TC and EC" should be replaced by "total carbon (TC) and elemental carbon (EC) and on line 187 "total carbon (TC)" should be replaced by "TC". WSOC, first mentioned in line 551, is not defined within the text, but it should be in the section on the measurement of WSOC; see my comment 1 above.

Response: According to the Editor’s suggestion, we have made these corrections. Please see lines 187, 193, and 196-197 in the revised MS.

Alterations and corrections:

Comment: Line 39: Replace "the Gosan" by "Gosan".

Response: Replaced. Please see line 39 in the revised MS.

Comment: Line 71: Replace "primarily" by "primary".

Response: Replaced. Please see line 70 in the revised MS.

Comment: Line 99: Replace "Korean" by "the Korean".

Response: Replaced. Please see line 99 in the revised MS.

Comment: Line 163: Replace "Sigmaaldrich" by "Sigma-Aldrich".

Response: Replaced. Please see line 163 in the revised MS.

Comment: Line 187: Replace "in TSP samples were analyzed" by "in the TSP samples were determined".

Response: Replaced. Please see line 197 in the revised MS.

Comment: Line 193: Replace "isolated it" by "isolated".

Response: Replaced. Please see line 203 in the revised MS.

Comment: Line 197: Replace "using Swiss" by "using the Swiss".

Response: Replaced. Please see line 207 in the revised MS.

Comment: Line 198: Replace "MICDAS" by "MICADAS".

Response: Replaced. Please see line 208 in the revised MS.

Comment: Line 226: Replace "in spring" by "in the spring".

Response: Replaced. Please see line 236 in the revised MS.

Comment: Line 233: Replace "spring" by "the spring".

Response: Replaced. Please see line 243 in the revised MS.

Comment: Line 237: Replace "continental" by "the continental".

Response: Replaced. Please see lines 246-247 in the revised MS.

Comment: Lines 253 and 273: Replace "and Russian" by "and the Russian".

Response: Replaced. Please see lines 263 and 283 in the revised MS.

Comment: Lines 261, 507, 595, 596, 605, 714, and 1127 (within Table 1): Replace "monoterpene SOA" by "monoterpene-SOA".

Response: Replaced. Please see lines 271, 518, 583, 584, 593, 702, and Table 1 in the revised MS.

Comment: Line 278: Replace "similar" by "the similar".

Response: Replaced. Please see line 288 in the revised MS.

Comment: Line 285: Replace "anhydrosugars" by "the anhydrosugars".

Response: Replaced. Please see line 295 in the revised MS.

Comment: Line 287: Replace "fungal" by "the fungal".

Response: Replaced. Please see line 297 in the revised MS.

Comment: Line 288: Replace "spring" by "the spring".

Response: Replaced. Please see line 298 in the revised MS.

Comment: Line 300: Replace "similar" by "a similar".

Response: Replaced. Please see lines 310-311 in the revised MS.

Comment: Line 306: Replace "from other" by "from that of the other".

Response: Replaced. Please see line 316 in the revised MS.

Comment: Line 311: Replace "in other" by "in the other".

Response: Replaced. Please see line 322 in the revised MS.

Comment: Line 321: "Sheesley et al., 2003" is missing in the Reference list.

Response: Added in the revised MS. Please see in the Reference list.

Comment: Line 332: Replace "of atmospheric" by "of the atmospheric".

Response: Replaced. Please see line 343 in the revised MS.

Comment: Line 338: Replace "sum of" by "sum of the".

Response: Replaced. Please see line 349 in the revised MS.

Comment: Lines 343, 480, 488, and 1127 (within Table 1): Replace "isoprene SOA" by "isoprene-SOA".

Response: Replaced. Please see lines 354, 491, 499, and Table 1 in the revised MS.

Comment: Line 347: Replace "in Gosan samples (Table 1). Total" by "in the Gosan samples (Table 1). The total".

Response: Replaced. Please see line 358 in the revised MS.

Comment: Line 351: Replace "somewhat" by "a somewhat".

Response: Replaced. Please see line 362 in the revised MS.

Comment: Line 367: Replace "Molecular" by "molecular".

Response: Replaced. Please see line 378 in the revised MS.

Comment: Line 380: Replace "that air" by "that the air".

Response: Replaced. Please see line 391 in the revised MS.

Comment: Line 383: Replace "KOS: 979" by "KOS979".

Response: Replaced. Please see line 394 in the revised MS.

Comment: Line 385: Replace "masses are" by "masses were".

Response: Replaced. Please see line 396 in the revised MS.

Comment: Line 419: Replace "of latter" by "of the latter".

Response: Replaced. Please see line 430 in the revised MS.

Comment: Line 427: Replace "and fossil-/nonfossil" by "and the fossil-/nonfossil".

Response: Replaced. Please see line 438 in the revised MS.

Comment: Line 431: Replace "found the" by "found that the".

Response: Replaced. Please see line 442 in the revised MS.

Comment: Line 432: Replace "and somewhat" by "and are somewhat".

Response: Replaced. Please see line 443 in the revised MS.

Comment: Line 443: Replace "fact that" by "fact that the" and replace "from the" by "ratio from the".

Response: Replaced. Please see line 454 in the revised MS.

Comment: Line 444: Replace "than hardwood" by "than from hardwood".

Response: Replaced. Please see line 455 in the revised MS.

Comment: Line 448: Replace "with hardwood" by "with those from hardwood".

Response: Replaced. Please see line 459 in the revised MS.

Comment: Line 450: Replace "than pine" by "ratios than those of pine" and replace "is on the lower side than" by "ratio is on the lower side than for".

Response: Replaced. Please see lines 461 and 462 in the revised MS.

Comment: Line 453: Replace "and, this" by "and this".

Response: Replaced. Please see line 464 in the revised MS.

Comment: Line 478: Replace "showed significant" by "showed a significant".

Response: Replaced. Please see line 489 in the revised MS.

Comment: Line 491: Replace "contrary," by "contrary, the".

Response: Replaced. Please see line 502 in the revised MS.

Comment: Line 493: Replace "with air" by "with the air".

Response: Replaced. Please see line 504 in the revised MS.

Comment: Lines 528 and 545: Replace "monoterpene" by "monoterpenes".

Response: Following the Editor's suggestion, this section has been removed.

Comment: Line 530: Replace "loaded by" by "loaded by the".

Response: This section has been removed.

Comment: Line 548: Replace "PMF" by "The PMF".

Response: This section has been removed.

Comment: Line 580: Replace "Contributions" by "The contributions".

Response: Replaced. Please see line 559 in the revised MS.

Comment: Line 581: Replace "lower than" by "lower than those of".

Response: Replaced. Please see line 560 in the revised MS.

Comment: Line 586: Replace "highest" by "the highest".

Response: Replaced. Please see line 565 in the revised MS.

Comment: Line 588: Replace "to carbonaceous" by "to the carbonaceous".

Response: Replaced. Please see line 567 in the revised MS.

Comment: Line 589: Replace "was occurred" by "occurred".

Response: Replaced. Please see line 568 in the revised MS.

Comment: Line 601: "Kleindienst et al. (2007)" is missing in the Reference list.

Response: Added. Please see in the Reference list.

Comment: Line 606: Replace "isoprene" by "that of isoprene-SOA".

Response: Replaced. Please see line 594 in the revised MS.

Comment: Line 623: Replace "showed poor" by "showed a poor".

Response: Replaced. Please see line 611 in the revised MS.

Comment: Line 624: Replace "but significant" by "but a significant".

Response: Replaced. Please see line 612 in the revised MS.

Comment: Line 628: Lev was already defined in line 420; it should not be defined again; thus replace "Levoglucosan (Lev)" by "Lev".

Response: Replaced. Please see line 616 in the revised MS.

Comment: Lines 639 and 652: Replace "in Gosan" by "in the Gosan".

Response: Replaced. Please see lines 627 and 640 and in the revised MS.

Comment: Line 658: Replace "Jun'2013" by "June 2013".

Response: Replaced. Please see line 646 in the revised MS.

Comment: Line 677: Replace "likely being" by "likely".

Response: Replaced. Please see line 665 in the revised MS.

Comment: Lines 691-692: Replace "P-value" by "p-value".

Response: Replaced. Please see lines 679-680 in the revised MS.

Comment: Lines 695-696: It should be specified whether this is 2012a or 2012b.

Response: Corrected. Please see line 684 in the revised MS.

Comment: Line 724: Replace "; however, our observations are" by ", our observations".

Response: Replaced. Please see line 712 in the revised MS.

Comment: Line 866: Replace "L., and" by "L. and".

Response: Replaced. Please see line 860 in the revised MS.

Comment: Line 960: Replace "Tec.," by "Technol.,".

Response: Replaced. Please see line 964 in the revised MS.

Comment: Lines 995-1006: "Simoneit et al., 1999" should come before "Simoneit et al., 2004a".

Response: Replaced. Please see lines 1003-1014 in the revised MS.

Comment: Line 131, within Table 2: Replace "MGA/MLTs" by "2-MGA/2-MLTs".

Response: Replaced. Please see Table 2 in the revised MS.

Supporting information, alterations and corrections:

Comment: Lines 38-39: The unit for the arrival height should be specified.

Response: Added. Please see line 38 in the supporting information.

Comment: Line 75: Replace "isoprene SOA" by "isoprene-SOA" and replace "monoterpene SOA" by "monoterpene-SOA".

Response: Replaced. Please see line 75 in the supporting information.

Comment: Lines 85-87: In the ordinate of Figure S5b and in line 87, replace "P+P/MBTCA" by "P/MBTCA".

Response: Corrected. Please see line 87 and Figure S5b in the supporting information.

Regional heterogeneities in the emission of airborne primary sugar compounds and biogenic secondary organic aerosols in the East Asian outflow: Evidence for coal combustion as a source of levoglucosan

M. Mozammel Haque[1,2,3], Yanlin Zhang[1,2*] Srinivas Bikkina[4], Meehye Lee[5], and Kimitaka Kawamura[3,4*]

[1]*Yale-NUIST Center on Atmospheric Environment, International Joint Laboratory on Climate and Environment Change (ILCEC), Nanjing University of Information Science & Technology, Nanjing, 210044, China*
[2]*School of Applied Meteorology, Nanjing University of Information Science & Technology, Nanjing 210044, China*
[3]*Institute of Low Temperature Science, Hokkaido University, Sapporo 060-0819, Japan*
[4]*Chubu Institute for Advanced Studies, Chubu University, Kasugai 487-8501, Japan*
[5]*Department of Earth and Environmental Sciences, Korea University, Anam-dong, Sungbuk-gu, Seoul 136-701, South Korea*

Corresponding author

E-mail: dryanlinzhang@outlook.com (Yan-Lin Zhang)

E-mail: kkawamura@isc.chubu.ac.jp (Kimitaka Kawamura)

ABSTRACT

Biomass burning (BB) significantly influences the chemical composition of organic aerosols (OA) in the East Asian outflow. Source apportionment of BB-derived OA is an influential factor for understanding their regional emissions, which is crucial for reducing uncertainties in their projected climate and health-effects. We analyzed here three different classes of atmospheric sugar compounds (anhydrosugars, primary sugars, and sugar alcohols) and two types of biogenic secondary organic aerosol (BSOA) tracers (isoprene- and monoterpene derived SOA products) in a year-long collected total suspended particulate matter (TSP) from an island-based receptor site in South Korea, Gosan. We investigate seasonal variations in the source-emissions of BB-derived OA using mass concentrations of anhydrosugars and radiocarbon (^{14}C-) isotopic composition of organic carbon (OC) and elemental carbon (EC) in ambient aerosols. Levoglucosan (*Lev*) is the most abundant anhydrosugar, followed by galactosan (*Gal*) and mannosan (*Man*). Strong correlations of *Lev* with *Gal* and *Man*, along with their ratios (*Lev*/*Gal:* 6.65 ± 2.26; *Lev*/*Man*: 15.1 ± 6.76) indicate the contribution from hardwood burning emissions. The seasonal trends revealed that the BB impact is more pronounced in winter and fall, as evidenced by the high concentrations of anhydrosugars. Likewise, significant correlations among three primary sugars (i.e., glucose, fructose, and sucrose) emphasized the contribution of airborne pollen. The primary sugars showed higher concentrations in spring/summer than winter/fall. The fungal spore tracer compounds (i.e., arabitol, mannitol, and erythritol) correlated well with trehalose (i.e., a proxy for soil organic carbon), suggesting the origin from airborne fungal spores and soil microbes in the East Asian outflow. These sugar alcohols peaked in summer, followed by spring/fall and winter. Monoterpene-derived SOA tracers were most abundant compared to isoprene-SOA tracers. Both BSOA tracers were dominant in summer, followed by fall, spring, and winter. The source apportionment based on multiple linear regressions and diagnostic mass ratios together revealed that BB emission mostly contributed from hardwood and crop-residue burning. We also found significant positive linear relationships of ^{14}C-based nonfossil- and fossil-derived organic carbon fractions with *Lev-C* along with the comparable regression slopes, suggesting the importance of BB and coal combustion sources in the East Asian outflow.

Keywords: Biomass burning tracers, primary biological aerosol particles, biogenic SOA tracers, radiocarbon-based source apportionment, organic aerosols, East Asian outflow

1. Introduction

Organic aerosols (OA), which account for a major fraction of up to 50% of airborne total suspended particulate matter, have considerable effects on regional and global climate by absorbing or scattering sunlight (Kanakidou et al., 2005). However, the climate effects of OA are involved with large uncertainties due to our limited understanding of the contributing sources. OA can be derived from both primary emissions and secondarily formed species. Sugars are an important group of water-soluble, primary organic compounds whose concentrations are significant in atmospheric aerosols over the continent (Jia and Fraser, 2011; Fu et al., 2008; Yttri et al., 2007; Graham et al., 2003). Anhydrosugars such as levoglucosan, galactosan, and mannosan are the key tracers of biomass burning (BB) emissions (Simoneit, 2002). Sugar alcohols, along with glucose, trehalose and sucrose are mostly originated from primary biological particles such as fungal spores, pollen, bacteria, and viruses, and vegetative debris (Graham et al., 2003; Simoneit et al., 2004a; Bauer et al., 2008; Deguillaume et al., 2008). Primary sugars and sugar alcohols are predominantly present in the coarse mode aerosols, accounting for 0.5-10% of atmospheric aerosol carbon matter (Yttri et al., 2007; Pio et al., 2008).

Secondary organic aerosol (SOA) is a large fraction of OA, while there were only limited studies about the key factors controlling SOA formation. The SOA formation significantly increased with the enhancement of the ambient aerosol mass (Liu et al., 2018). SOA is formed by both homogenous and heterogeneous reactions of volatile organic compounds (VOCs) in the atmosphere (Surratt et al., 2010; Robinson et al., 2007; Claeys et al., 2004). On a global estimation, biogenic VOCs (BVOCs) such as isoprene, monoterpenes (e.g., *α*/*β*-pinene), and sesquiterpenes (e.g., *β*-caryophyllene) are one order of magnitude higher than those of anthropogenic VOCs (e.g., toluene) (Guenther et al., 2006). The global emissions of annual BVOCs were estimated to be 1150 TgC yr^{-1}, accounting for 44% isoprene and 11% monoterpenes (Guenther et al., 1995). Isoprene is highly reactive and promptly reacts with oxidants such as O_3, OH, and NO_x in the atmosphere to form SOA (Kroll et al., 2005, 2006; Ng et al., 2008; Surratt et al., 2010; Bikkina et al., 2021), estimated to be 19.2 TgC yr^{-1}, consisting of ~70% of the total SOA budget (Heald et al., 2008). Monoterpenes are important sources of biogenic secondary organic aerosol (BSOA), considering *α*-pinene as major species, accounting for ~35% of the global monoterpenes emissions (Griffin et al., 1999).

Anthropogenic activities such as coal and biofuel combustion over East Asia, including China, are responsible for the vast emission of OA (Huebert et al., 2003; Zhang et al., 2016). Understanding the ambient levels OA in the East Asian outflow is crucial for assessing their regional climatic effects. As part of this effort, the Korean Climate Observatory at Gosan (KCOG), a super site located in South Korea, is an ideal location for investigating the atmospheric outflow characteristics from East Asia (Fu et al., 2010a; Kundu et al., 2010; Ramanathan et al., 2007; Kawamura et al., 2004; Arimoto et al., 1996). For instance, primary OA associated with soil/desert dust in East Asia, along with forest fires in Siberia/northeastern China, are transported over Gosan in spring (Wang et al., 2009a). BSOA during long-range transport from the continent and Open Ocean, as well as local vegetation, can significantly contribute to Gosan aerosols. Although these investigations were carried out almost a decade ago, no such observations are available in contemporary times from Gosan. Here, we attempt to understand the current states of East Asian OA using both the molecular marker approach and radiocarbon data of carbonaceous components.

The KCOG, located on the western side of Jeju Island adjacent to the Yellow Sea and the East China Sea, is facing the Asian continent but is isolated from public areas of the island (Kawamura et al., 2004). Simoneit et al. (2004b) have documented during the ACE-Asia campaign that OA from the BB and fossil fuel combustion sources are transported along with desert dust to the KCOG during continental outflow. An intensive campaign was organized at the KCOG during spring 2005 to observe the physical properties of East Asian aerosols while two dust events were detected (Nakajima et al., 2007). Here, we focus on the characterization of airborne anhydrosugars, primary sugars, sugar alcohols, and BSOA tracers from the KCOG. Gosan is influenced by the continental outflow from East Asia during winter, spring and fall, whereas the site is influenced by the maritime air masses from the Pacific Ocean and other marginal seas. This makes the KCOG ideal for characterizing the regional heterogeneities in the emissions of organic compounds in the East Asian outflow based on the TSP samples collected during April 2013-April 2014.

2. Methods

2.1. Aerosol sampling and prevailing meteorology

Total suspended particles (TSP) were collected on pre-combusted (450°C for 6 h) quartz fiber filters (20 cm × 25 cm, Pallflex) at the KCOG (33.17 °N, 126.10 °E, see Figure 1), South Korea. To get enough signal for the radiocarbon measurements, each TSP sample was collected for 10–14 days from April 2013 to April 2014. Twenty-one samples were collected using a high-volume air sampler (Kimoto AS-810, ~65 m^3 h^{-1}) installed on the rooftop of a trailer house (~3 m above the ground). After the collection, aerosol filters were transferred to a pre-combusted (450°C for 6 h) glass jar (150 mL) equipped with a Teflon-lined screw cap and transported to the laboratory in Sapporo. These TSP samples were stored in a dark freezer room at -20°C until the analysis. Three field blank filters were also collected during the campaign.

The ambient temperatures at the Gosan site were on average 6.9°C in winter, 14.1°C in spring, 27.0°C in summer, and 17.1°C in fall. Likewise, the average relative humidity was found to be highest in summer (71.3%), followed by spring (64.9%), fall (63.5%), and winter (54.7%). Gosan is influenced by the pollution sources in East Asia during winter as well as other transition periods (spring and fall) due to the prevailing westerlies. In contrast, winds in summer blew mostly from the western North Pacific (WNP) by the easterly winds. The spring season is, in particular, important for the transport of mineral dust mixed with polluted OA to Gosan (Kundu et al., 2010).

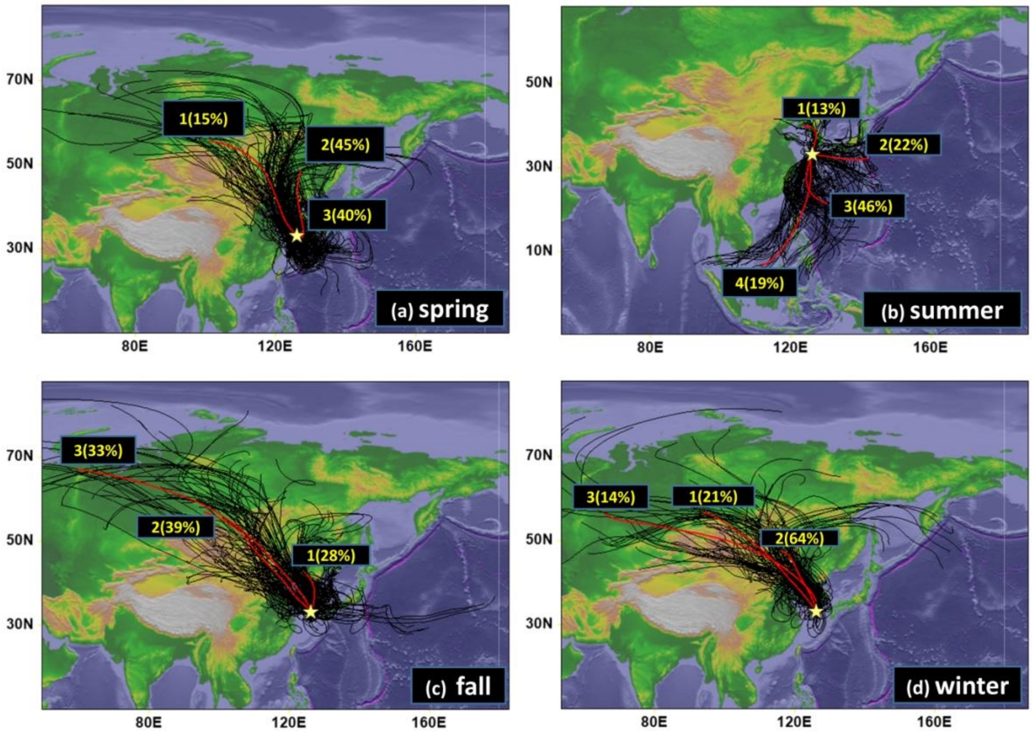

Figure 1. Cluster analysis of backward air mass trajectories over Gosan (indicated by a star symbol) for the TSP collected during (a) spring, (b) summer, (c) fall, and (d) winter seasons.

2.2. Extraction and analysis of organic compounds

Approximately 3.14 cm^2 filter cuts were extracted with dichloromethane/methanol (2:1; v/v). The extracts were concentrated using a rotary evaporator under vacuum and then blown down to near dryness with pure nitrogen gas. The dried residues were subsequently reacted with N, O-bis(trimethylsilyl)trifluoroacetamide containing 1% trimethylchlorosilane (BSTFA+1% TMCS, SUPELCO©, Sigma-Aldrich ®) and pyridine at 70 °C for 3 h to derive OH and COOH groups of polar organic compounds to trimethylsilyl ethers and esters, respectively. After the derivatization followed by the addition of a known amount of internal standard solution (Tridecane; 1.43 ng L^{-1} in n-hexane), the derivatized extracts were injected onto a gas chromatograph (Hewlett-Packard model 6890 GC) coupled to a mass spectrometer (Hewlett-Packard model 5973, MSD) (GC-MS). More details on the quantification of polar organic compounds using GC-MS are described in Haque et al. (2019).

The target compounds (anhydrosugars, primary sugars, sugar alcohols, and BSOA tracers) were separated on a DB-5MS fused silica capillary column (30 m x 0.25 mm i.d., 0.5 μm film thickness) using helium as a carrier gas at a flow rate of 1.0 ml min^{-1}. The GC oven temperature was programmed from 50°C for 2 min and then increased from 50 to 120°C at 30°C min^{-1} and to 300°C at 6°C min^{-1} with a final isotherm hold at 300°C for 16 min. The sample was injected in a splitless mode with the injector temperature at 280°C. The MS was operated at 70 eV and scanned from 50 to 650 Da on an electron impact (EI) mode. Mass spectral data were acquired and processed using the Chemstation software. The organic compounds were identified individually by comparison with retention times and mass spectra of authentic standards and NIST library and literature data of mass fragmentation patterns (Medeiros and Simoneit, 2007). For assessing the recoveries, ~100-200 ng of the standard solution was spiked on the blank filter and analyzed as a real sample. Overall, the average recoveries were found to be 80-104% for target compounds. The field and laboratory blank filters (n = 3) were also analyzed by the same procedures as a real sample. Target compounds were not found in the field blanks. The analytical errors based on concentrations by replicate sample analyses (n = 3) were less than 15%.

2.3. Carbon fractions analysis

Organic carbon (OC) and elemental carbon (EC) were analyzed using a thermal-optical transmittance method with a Sunset Laboratory carbon analyzer following the NIOSH protocol (Birch and Cary, 1996), and detailed procedures were given elsewhere (Zhang et al., 2016). Furthermore, a portion of 2.54 cm^2 of each sample filter was extracted with 15 mL ultrapure water (resistivity > 18.2 MΩcm, Sartorius arium 611 UV) with ultrasonication for 30 min. The water extracts were then filtered through a membrane disc filter for water-soluble organic carbon (WSOC) analysis by a total organic carbon (TOC) analyzer (Shimadzu, TOC-Vcsh) (Boreddy et al., 2018). The concentrations of WSOC were corrected by field blanks. The analytical errors in the triplicate analyses were less than 5% for WSOC.

2.4. Radiocarbon isotopic composition of total carbon (TC) and EC

The concentrations of TC in the TSP samples were determined using an elemental analyzer. For the radiocarbon isotopic composition ($\Delta^{14}C$), the aerosol filter punches were exposed for ~12 h to HCl fumes in a vacuum desiccator. Subsequently, these filters were analyzed for $\Delta^{14}C$ on a modified elemental analyzer coupled via a gas interface to Accelerator Mass Spectrometer Mini Carbon Dating System (MICADAS) at the University of Bern, Switzerland (Salazar et al., 2015). The evolved CO_2 of TC from the elemental analyzer was passed through a moisture trap (Sicapent, Merck) and isolated from other residual gasses using a temperature-controlled zeolite trap. The purified CO_2 was introduced through a gas interface system to MICADAS, where $^{14}C/^{12}C$ ratios are measured according to the analytical procedures detailed in Zhang et al. (2016). Likewise, the evolved CO_2 of elemental carbon from the Sunset Lab OC/EC analyzer using the Swiss 4S protocol (Zhang et al., 2012), was directed to the MICADAS and measured for the $^{14}C/^{12}C$ ratio relative to standard calibration gas. These results were expressed as fractions of modern carbon (f_M) by normalizing with a $\delta^{13}C$ value of the reference standard in the year 1950 (–25‰) according to Stuiver and Polach (1997) for the fractionation effects. The f_M(OC) can be estimated by using the f_M(TC) and f_M(EC) in an isotope mass balance equation (Zhang et al., 2015). Additionally, we estimated the relative contributions of OC and EC from the nonfossil and fossil sources ($f_{nonfossil}$ and f_{fossil}, respectively) using the following equations.

$$f_{\text{nonfossil-OC}} = f_M(\text{OC-sample})/f_M(\text{OC-ref});\ f_M(\text{OC-ref}) \approx 1.07\pm0.04 \quad (1)$$

$$f_{\text{nonfossil-EC}} = f_M(\text{EC-sample})/f_M(\text{EC-ref});\ f_M(\text{EC-ref}) \approx 1.10\pm0.05 \quad (2)$$

$$f_{\text{fossil-OC}} = 1 - f_{\text{nonfossil-OC}} \quad (3)$$

$$f_{\text{fossil-EC}} = 1 - f_{\text{nonfossil-EC}} \quad (4)$$

The reference values of OC and EC were obtained from Mohn et al. (2008). Using the fractions of $f_{\text{fossil-OC}}$ and $f_{\text{nonfossil-OC}}$, we can, therefore, estimate the mass concentration of ambient organic carbon (OC-ambient) from fossil and nonfossil sources (OC_{fossil} and $OC_{nonfossil}$, respectively).

$$OC_{nonfossil} = f_{\text{nonfossil-OC}} \times [OC]_{ambient} \quad (5)$$

$$OC_{fossil} = f_{\text{fossil-OC}} \times [OC]_{ambient} \quad (6)$$

More details on the radiocarbon isotopic composition data over Gosan were reported elsewhere (Zhang et al., 2016).

3. Results and discussion

3.1. Trajectory and cluster analysis

Backward air mass trajectories are useful for assessing the impact of local versus regional source emissions over Gosan. Seven-day isentropic backward air mass trajectories were computed using the hybrid single-particle Lagrangian integrated trajectory model (HYSPLIT, version 4: Stein et al., 2015) over the KCOG for the sampling period using the meteorological datasets of the Global Data Assimilation System (GDAS) network. The trajectory endpoint files from the HYSPLIT model were further used for the cluster analysis using the Trajstat package (Wang et al., 2009b) for all four seasons (Figure 1). Although cluster analysis revealed the predominance of continental transport in the spring, fall, and winter seasons, the air masses over the KCOG in summer mostly originated from the WNP. Since spring is a transition of winds switching from westerlies to easterlies, Gosan is likely influenced by the long-range transport of dust, pollution, and sea-salt aerosols.

The vertical mixing of pollutants within the boundary layer height also plays an important role in controlling the strength of continental outflow alongside regional meteorology. For instance, the mixing height of air parcels from the HYSPLIT model is mostly confined to 1000 m in winter but somewhat increased towards the spring and fall seasons (Figure S1). This vertical enhancement in the boundary layer height facilitates the transport of mineral dust particles from the arid and semiarid regions in East Asia along with urban pollutants to Gosan in spring and fall compared to winter. However, the strength of the continental outflow somewhat depends on several factors, including source emissions, meteorology, and mixing height of air parcels.

Gosan is influenced by three types of air masses in spring (Figure 1a), from the Mongolian Desert (cluster 1: 15%), North China (cluster 2: 45%), and from the Yellow Sea (cluster 3: 40%). In contrast, the easterlies from the WNP in summer mostly influenced the composition of TSP over the KCOG. This inference is based on the cluster analysis for summer samples (Figure 1b), which showed four regimes, including transport from the Sea of Japan (cluster 1: 13%), WNP (cluster 2: 22%), South China Sea (cluster 3: 46%), and East China Sea (cluster 4: 19%). In contrast, cluster analysis revealed three major transport regimes from East Asia in fall and winter (Figure 1c-d). However, there are subtle differences that exist between winter and fall in terms of influence from nearby versus distant pollution sources. For instance, long-range transport of air masses from west Mongolia (cluster 1: 21%) and the Russian Far East (cluster 3: 14%) exerted a weak influence on the TSP sampled over Gosan in winter. Besides, we observed a somewhat larger impact of air masses from the North China Plain over Gosan (cluster 2: 64%) in winter. In contrast, Gosan is less influenced by air masses originating from the North China Plain, contributing ~28% (cluster 1) than those from Mongolia (cluster 2: 39%) and the Russian Far East (cluster 3: 33%) in fall. Therefore, the impact of East Asian outflow is stronger in winter than in spring and fall.

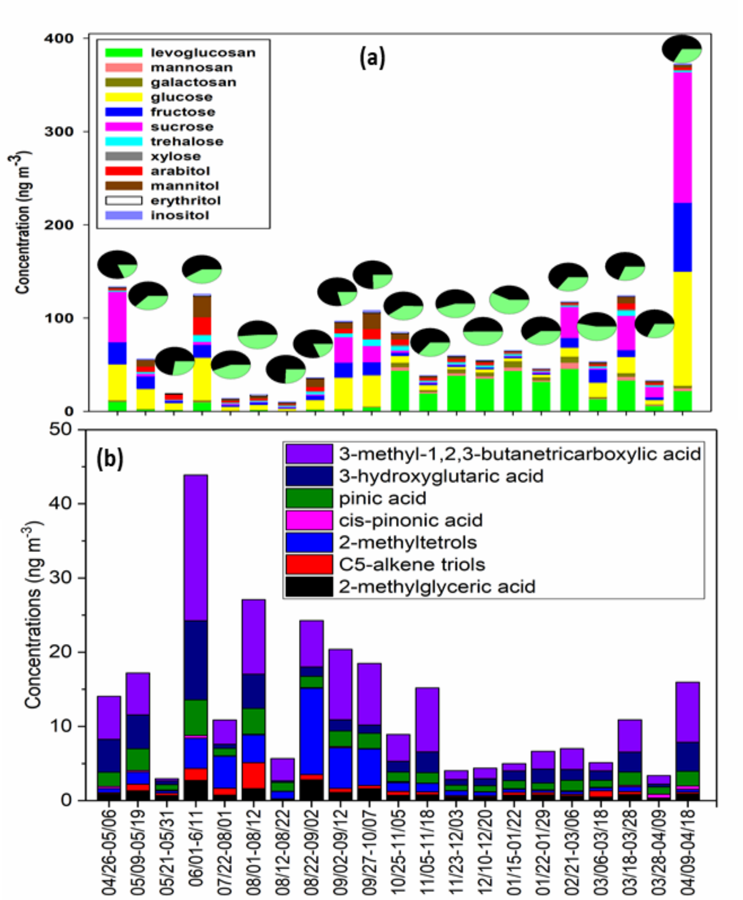

Figure 2. (a) Cumulative concentration levels of anhydrosugars, primary sugars, and sugar alcohols (i.e., represented by bars), and depicting the contributions of nonfossil (green color) and fossil (black color) organic carbon (i.e., pie charts), (b) Cumulative concentration levels of isoprene- and monoterpene-SOA tracers in each TSP sample collected over Gosan.

3.2. Temporal and seasonal variability of sugars

The temporal/seasonal trends of sugar compounds over the KCOG provide useful information on the emission strengths of various sources in the East Asian outflow. All three anhydrosugars showed similar temporal and seasonal trends with higher concentrations in winter and fall than spring and summer (Figures 2a and S2). As levoglucosan and other two anhydrosugars (mannosan and galactosan) are the pyrolysis products of cellulose/hemicellulose, their higher concentrations along with an increase in nonfossil fraction of OC (Figure 2a; pie charts) in TSP from winter and fall revealed the impact of BB emissions. The MODIS satellite-based fire counts (Figure S1) together with cluster analysis in winter and fall (Figure 1) have revealed an influence of active BB emissions in the North China Plain, Mongolia, and the Russian Far East. The temporal trends of glucose, fructose, and sucrose exhibited less variability throughout the sampling period; however, we observed a slight increase in their concentration towards spring/summer (Figure 2a). Glucose and fructose have origins from leaf fragments and pollen species (Fu et al., 2012a). Sucrose is a potential tracer for airborne pollen (Fu et al., 2012a) and late spring/early summer is often regarded as a season of “pollen-allergies”. Therefore, the similar temporal trends of glucose and fructose with sucrose indicate their common source, pollens (Figure 2a). Since glucose, fructose, and sucrose showed moderately significant correlations ($R^2 = 0.44$-0.48, $p<0.01$) with levoglucosan in winter, it is somewhat possible that BB source emission could also influence the concentrations of these saccharides in this season (Haque et al., 2019; Fu et al., 2008).

BB also contributes to xylose and, hence, the temporal variability of xylose is mimicking that of the anhydrosugars. Trehalose is a primary sugar and a useful tracer for organic carbon associated with soil dust particles (Fu et al., 2012a). The temporal variability of trehalose closely resembled that of the fungal spore tracers (arabitol, mannitol, and erythritol), showing high concentrations in the spring, summer, and fall seasons (Figures 2a and S2) (Zhu et al., 2015a; Fu et al., 2012a). The KCOG is under the influence of a large-scale advection of mineral dust from East Asia to the WNP during these three seasons (Tyagi et al., 2017; Huebert et al., 2003). The mineral dust transport from East Asia to the WNP can be traced by the high concentrations of non-sea-salt Ca^{2+} in the TSP samples from Gosan (Arimoto et al., 1996). Similar temporal trends of trehalose and nss-Ca^{2+}, particularly in spring samples (Figure S3), suggest that the abundance of OA specific to fungal spores over Gosan is likely associated with the Kosa (Asian dust) events.

The major sources of arabitol and mannitol are airborne fungal spores (Bauer et al., 2008), accompanying detritus from mature leaves (Pashynska et al., 2002). Heald and Spracklen (2009) reported that mannitol and arabitol are considerably associated with terrestrial biosphere activity. Inositol is largely derived from the developing leaves in summer (Pashynska et al., 2002) and BB in winter (Fu et al., 2010b). Zhu et al. (2015b) found a similar seasonal behavior of inositol with those of other sugar alcohols with the predominance in summer, associated with microbial activities in local forests from Okinawa. Inositol showed a moderately significant correlation with levoglucosan ($R^2 = 0.33$, $p< 0.01$)

in winter; however, there were no positive linear relationships between levoglucosan and other sugar alcohols, implying a partial emission of inositol from the BB during winter in Gosan aerosols. Therefore, the temporal variability of inositol differs from that of the other sugar alcohols (Figure S2). The sources of sugar compounds are further discussed in section 3.4.

The seasonally averaged concentrations of all the anhydrosugars and xylose are higher in winter/fall than spring/summer (Figure 3a-d), possibly due to a greater influence of long-range transport from East Asia. In contrast, glucose, fructose, and sucrose peaked in spring but decreased in the other seasons (Figure 3e-g), mainly because of the contribution of airborne pollen. Trehalose showed higher concentrations in fall and summer, followed by spring and winter (Figure 3h). Arabitol, mannitol, and erythritol showed higher concentrations in summer/fall than in winter and spring (Figure 3i-k). This seasonal trend is consistent with those of soil-derived fungal spores. This feature is consistent with earlier observations from a remote oceanic island in the WNP (Okinawa) during the impact of East Asian outflow (Zhu et al., 2015a). The seasonally averaged mass concentrations of inositol are highest in spring, followed by summer, fall, and winter (Figure 3l). Overall, the molecular compositions of anhydrosugars showed the predominance of levoglucosan followed by galactosan and mannosan (Figure S4). Galactosan is more abundant in crop-residue burning emissions than mannosan (Engling et al., 2009; Sheesley et al., 2003). It is very much likely that the impact of crop-residue burning emissions in East Asia over Gosan is more prominent in winter/spring. Such high abundances of galactosan over mannosan were found in the North China Plain (Fu et al., 2008) and in the Indo-Gangetic Plain outflow sampled over the Bay of Bengal (Bikkina et al., 2019). Although the temporal variability of primary sugars in the TSP samples from Gosan showed a characteristic peak of glucose and sucrose (Figure 2a), the seasonally averaged distributions are different (Figure S4). The molecular distributions of sugar alcohols are characterized by high loadings of arabitol and mannitol, followed by erythritol and inositol (Figure S4).

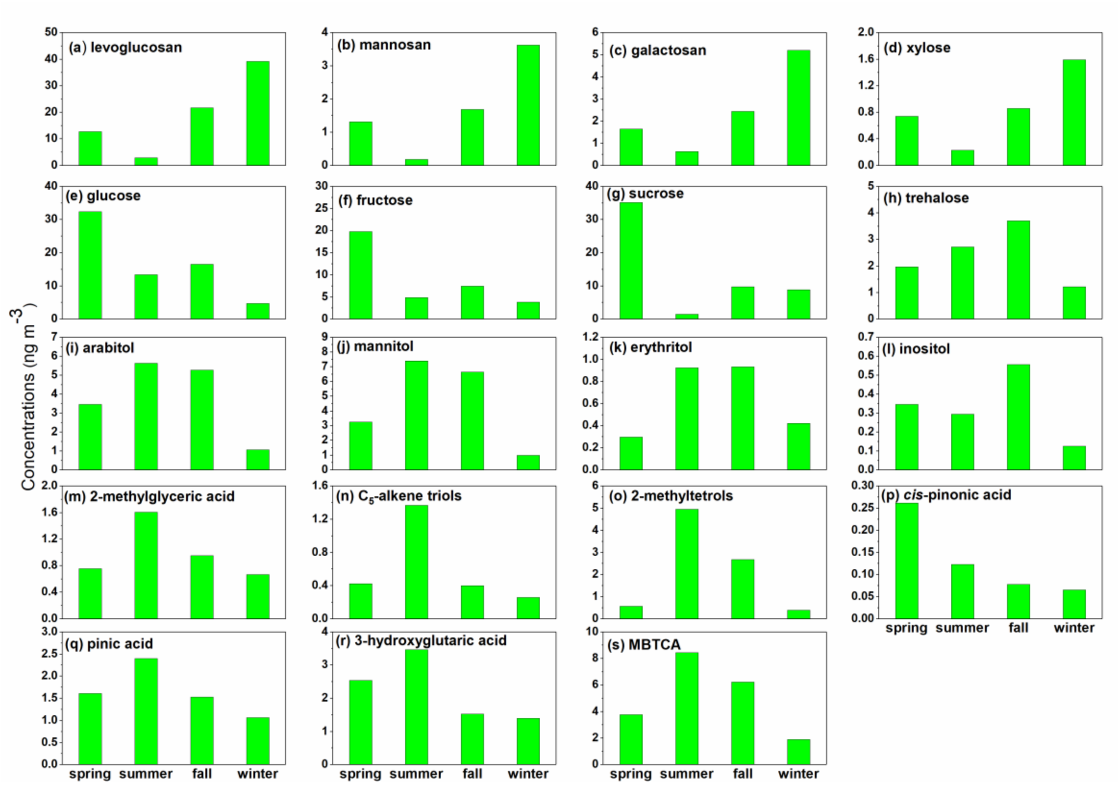

Figure 3. Seasonal variability of the atmospheric levels of sugar compounds and BSOA tracers in TSP samples from Gosan during April 2013-April 2014.

3.3. Temporal and seasonal variability of BSOA tracers

We identified six isoprene-SOA tracers such as 2-methylglyceric acid (2-MGA), three C_5-alkene triols, and two 2-methyltetrols (2-methylthreitol and 2-methylerythritol) (2-MTs) in Gosan aerosol samples (Table 1). The sum of the isoprene-SOA tracers ranged from 0.35 to 15.1 ng m^{-3} (avg. 3.69 ng m^{-3}) with the predominance of 2-MTs (avg. 2.09 ng m^{-3}). 2-MGA is the second most abundant isoprene-SOA tracer (avg. 0.99 ng m^{-3}), a high-generation product probably formed by further photooxidation of methacrolein and methacrylic acid. A similar molecular composition was observed over the North Pacific and California Coast (Fu et al., 2011). All the isoprene-SOA tracers exhibited similar temporal variations with higher concentrations in the summer/spring months compared to autumn and winter (Figure 2b). Conversely, four monoterpene-SOA tracers, i.e., *cis*-pinonic acid, pinic acid, 3-hydroxyglutaric acid (3-HGA), and 3-methyl-1,2,3-butanetricarboxylic acid (MBTCA), were detected in the Gosan samples (Table 1). The total concentrations of monoterpene-SOA tracers were found to be 1.65 to 35.5 ng m^{-3} (avg. 9.24 ng m^{-3}) with a high concentration of MBTCA (avg. 5.11 ng m^{-3}). All the monoterpene-SOA tracers showed similar temporal trends with high values in summer/spring periods than autumn/winter (Figure 2b).

Nevertheless, *cis*-pinonic acid was ascribed a somewhat different temporal variability with other monoterpene-SOA tracers. It is likely that *cis*-pinonic acid might be further photo-oxidized to form MBTCA (Szmigielski et al., 2007).

The seasonal average of isoprene-SOA tracers showed high concentrations in summer, followed by spring/fall and winter (Figure 3m-o). One key feature of the data presented here is that two fall samples (KOS984; 2-12 September and KOS986; 27 September to 07 October) exhibited high concentrations for 2-MTs over Gosan (Figure 2b). We presumed that local vegetation might contribute significantly to the formation of 2-MTs as it is a first-generation product. Moreover, 2-MTs can be derived from the open ocean under low NO_x conditions (Hu et al., 2013). 3-HGA and pinic acid showed somewhat higher concentrations in summer/spring than fall/winter due to the growing vegetation (Figure 3q-r). *Cis*-pinonic acid was more abundant in spring compared to summer (Figure 3p) because of its photo-degradation, as discussed above. In contrast, MBTCA was dominant in summer/fall than spring/winter (Figure 3s). Here, the formation of MBTCA could be enhanced in fall during atmospheric transport from East Asia. The molecular distributions of isoprene-SOA tracers were characterized by a high loading of 2-MTs, followed by 2-MGA and C_5-alkene triols in all seasons (Figure S4). The molecular composition of monoterpene-SOA tracers was dominated by MBTCA, followed by 3-HGA, pinic acid, and *cis*-pinonic acid in all seasons (Figure S4). Overall, BSOA tracers were found to be most abundant in summer, followed by fall, spring, and winter (Table 1). Interestingly, it is likely that secondary OA undergoes much faster cycling than the primary sugar compounds, considering the feasibility of photooxidation. This would mean a slight underestimation of BSOA over the KCOG in the East Asian outflow and, hence, their atmospheric abundances over Gosan reflect a lower limit.

Kang et al. (2018b) reported that monoterpene-SOA tracers were more abundant than isoprene-SOA tracers in spring-summer over the East China Sea, which is consistent with this study. Although the mass budget calculations showed that isoprene and monoterpenes are largely emitted by terrestrial plants; however, the open ocean can also contribute to isoprene and monoterpenes significantly (Conte et al., 2020; Shaw et al., 2010; Broadgate et al., 1997). Air mass back trajectory and cluster analysis (Figure 1) implied that the air masses mostly originated from the ocean during summer. This means the open ocean significantly contributed to isoprene-SOA production. However, terrestrial sources from the continent also substantially enhanced the formation of BSOA. For example, one sample (KOS979; 1-11

June 2013) during summer showed the highest loading of BSOA tracers (Figure 2b) when air masses were transported from the continent (Zhang et al., 2016).

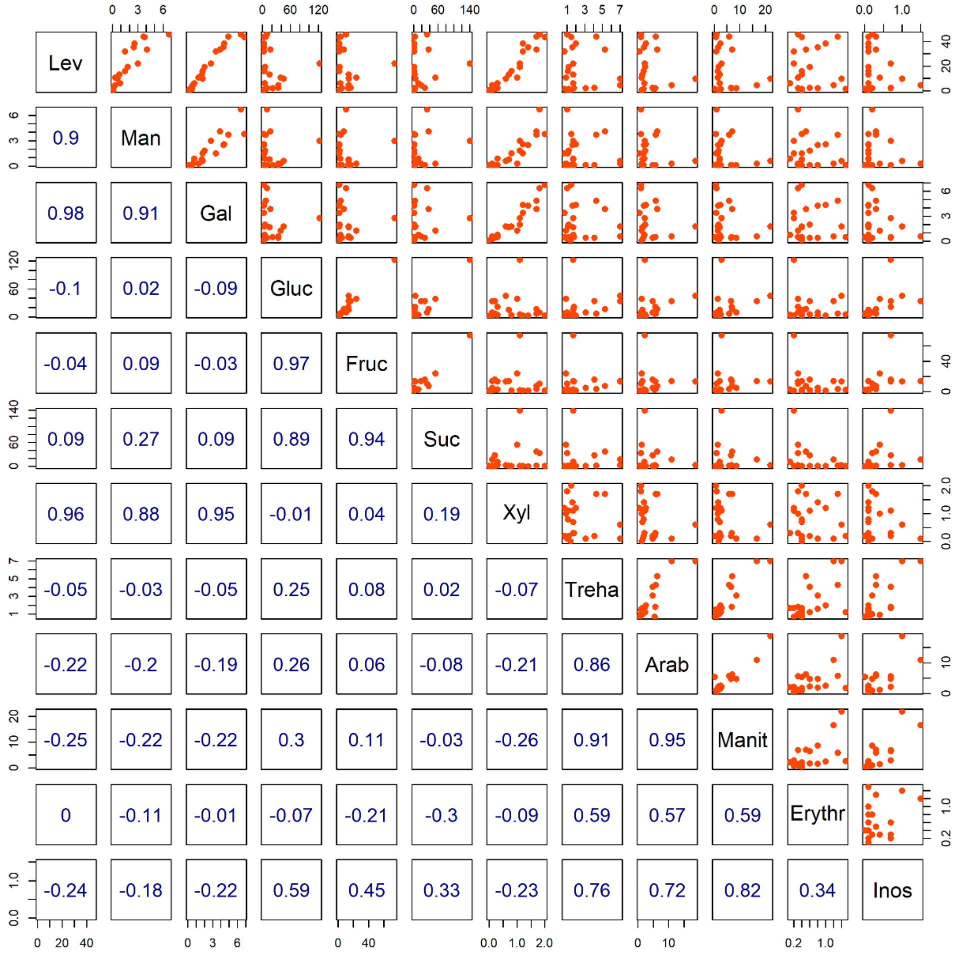

Figure 4. Multiple linear regression (MLR) analysis of airborne sugar compounds in TSP collected over Gosan.

3.4. Source apportionment- regression analysis and diagnostic ratios

Anhydrosugars strongly correlated with xylose (Figure 4), suggesting their common source as BB emission in East Asia. Fu et al. (2012a) analyzed pollen from different tree species (*e.g.*, White birch, Chinese willow, Peking willow, Sugi, Hinoki), which are enriched in sucrose (182-37,300 μg g^{-1}), glucose (378-3601 μg g^{-1}) and fructose (162-1813 μg g^{-1}). In our samples, sucrose strongly correlated with glucose and fructose (Figure 4), suggesting their origin from plant-derived airborne pollen. Likewise, a strong correlation was found between arabitol and mannitol, indicating a mutual origin from a similar type of fungal spores (Zhu et al., 2015a; Fu et al., 2012a; Bauer et al., 2008; Yttri et al., 2007). Bauer et al. (2008) ascribed weak correlations between arabitol and mannitol to the diverse nature of ambient fungal spores. Furthermore, both sugar alcohols correlated well with trehalose, a tracer for soil organic carbon (Fu et al., 2012a). This observation suggests their common origin from soil organic matter associated with fungal spores. Erythritol also originates from fungal spores; however, its abundance is affected by the atmospheric aging process. Kessler et al. (2010) reported that erythritol is degraded during long-range transport in 12.7 days. Consequently, arabitol and mannitol were moderately correlated with erythritol in the Gosan samples due to the degradation of the latter sugar alcohol in the East Asian outflow.

The linear relationship of levoglucosan (*Lev*) with mannosan (*Man*), galactosan (*Gal*), and nss-K^+ provide useful information on the type of BB emissions (hardwood, softwood, or crop-residue). Ratios of *Lev*/*Man* and *Lev*/(*Man* + *Gal*) can be useful to distinguish BB and coal combustion contributions. The average ratios of *Lev*/*Man* (15.1 ± 6.76) and *Lev*/(*Man* + *Gal*) (4.27 ± 1.23) in Gosan aerosols are much closer to those from wood burning and coal combustion sources emissions, respectively (Yan et al., 2018). It reveals that *Lev* could originate from both biomass and coal burning source emissions, which is consistent with the linear relationship between *Lev*-C and the fossil-/nonfossil carbon fraction (section 3.6). Furthermore, different types of biomass are characterized by distinct *Lev/Man* ratios. For instance, *Lev/Man* ratios from the softwood burning emissions (3-10) differ from those of hardwood (15-25) and crop-residues (>40) (Singh et al., 2017; Schmidl et al., 2008a, b; Fu et al., 2008; Engling et al., 2006, 2009; Fine et al., 2001, 2004). We found that the *Lev/Man* ratios (Table 2) over the KCOG overlap between seasons and are somewhat close to that of hardwood burning emissions in northern China, Mongolia, and the Russian Far East, as corroborated by the backward air mass trajectories and MODIS fire counts (Figures 1 and S1). Besides, *Lev*/K^+ and *Man/Gal* ratios in summer differ from those of other seasons (Table 2). Cheng et al. (2013) have apportioned qualitatively the source contributions of anhydrosugars over a receptor site based on the comparison of *Lev*/K^+ and *Lev/Man* ratios in aerosols to those from various sources profiles compiled from the literature. This approach of using the mass ratios of *Lev/Man* and *Lev*/K^+ has been proven useful for deciphering the difference in BB-derived OA (Bikkina et al., 2019).

Here, we adopted this methodology to ascertain the likely contributing sources of anhydrosugars, which are BB tracers from different seasons (Figure 5). This source apportionment relies on the fact that the *Lev*/K^+ ratio from the softwood burning (10-1000) is higher than from hardwood (1-100) (Fine et al., 2004). In contrast, *Lev*/*Man* ratios for softwood are lower than those of hardwood burning (10-100) (Fine et al., 2004). Likewise, the *Lev/Man* and *Lev*/K$^+$ ratios from grasses and crop-residues are 10-100 and 0.01-1.0, respectively (Bikkina et al., 2019). On a similar note, the *Lev*/K$^+$ ratios from the burning of pine needles (0.1-1.0) somewhat overlap with those from hardwood burning emissions but are characterized by distinct *Lev/Man* ratios (Bikkina et al., 2019). The burning of dead leaves (duff) showed higher *Lev*/K$^+$ ratios than those of pine needles and grasses, but their *Lev/Man* ratio is on the lower side than for the former biomass type and the softwood burning emissions. However, *Lev* is more susceptible to degradation by photooxidation with OH radicals during atmospheric transport (half-life: <2.2 days) (Hennigan et al., 2010) and this would cause lower abundances of this anhydrosugar. Hence, the hardwood *Lev*/K$^+$ ratios could slightly shift downwards. Therefore, caution is required while interpreting the ambient data from a receptor site (Bikkina et al., 2019). Overlapping the seasonal data on this scatter plot of *Lev*/K$^+$ versus *Lev/Man* (Figure 5) clearly revealed a mixed contribution of burning of hardwood and crop-residue in the East Asian outflow. It should be noted that the photooxidation process during atmospheric transport is also applicable for low concentrations and poor correlations of other primary saccharides.

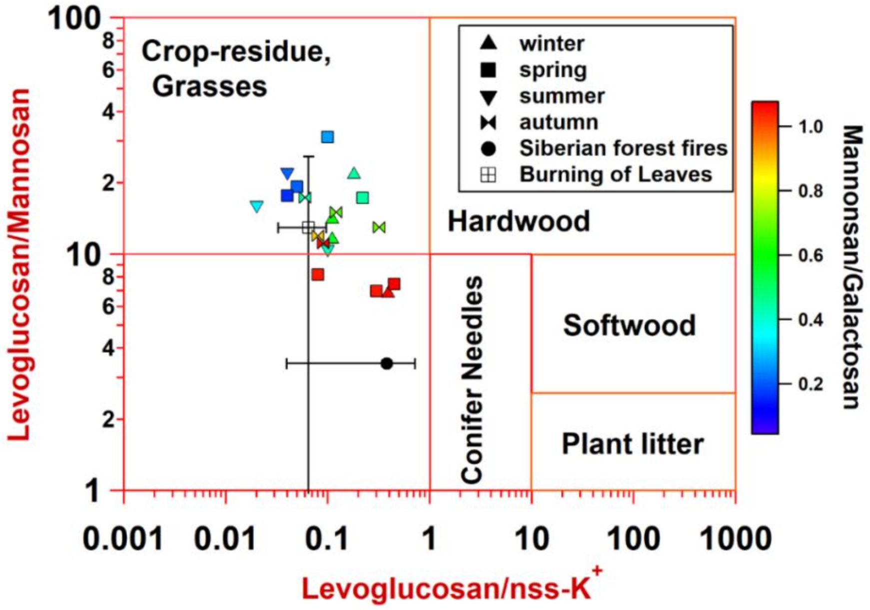

Figure 5. Scatter plot of levoglucosan/K$^+$ versus levoglucosan/mannosan ratios in TSP collected over Gosan during April 2013-April 2014. The data for Siberian forest fires and burning leaves were adopted from Sullivan et al. (2008).

The correlation coefficients and diagnostic ratios of BSOA tracers specify their source origin or formation pathway. Nevertheless, atmospheric stability or reactivity of BSOA tracers through the photooxidation during long-range transport may bias the correlation coefficients in Gosan aerosols. 2-MGA showed a significant correlation with 2-MTs ($r = 0.79$, $p<0.01$) and C_5-alkene triols ($r = 0.50$, $p<0.05$) (Figure 6a-b), suggesting their similar formation pathway or common sources of isoprene-SOA tracers. However, a poor correlation coefficient between 2-MTs and C_5-alkene triols ($r = 0.33$, $p = 0.13$) (Figure 6c) indicates their different formation process over the Gosan atmosphere. Wang et al. (2005) documented that polyols are formed from isoprene through diepoxy derivatives, which further convert into 2-MTs by acid-catalyzed hydrolysis. On the other hand, C_5-alkene triols are produced from the precursor of hydroxyperoxy radicals that are initially derived from isoprene through rearrangement reactions (Surratt et al., 2006). It can be noted that the formation mechanisms of 2-MGA and 2-MTs are different while depending on the NO_x concentrations (Surratt et al., 2010). Thus, the ratio of 2-MGA/2-MTs attributes to the influence of NO_x on isoprene-SOA formation. We found a low ratio of 2-MGA/2-MTs (0.67) (Figure S5a, Table 2) in summer, implying enhancement of 2-MTs formation over the open ocean due to the low-NO_x environment in the ocean atmosphere. On the contrary, the 2-MGA/2-MTs ratios for other seasons were >1.0 (Figure S5a, Table 2), indicating an elevated formation of 2-MGA through continental high NO_x condition, which is consistent with the air masses back trajectory.

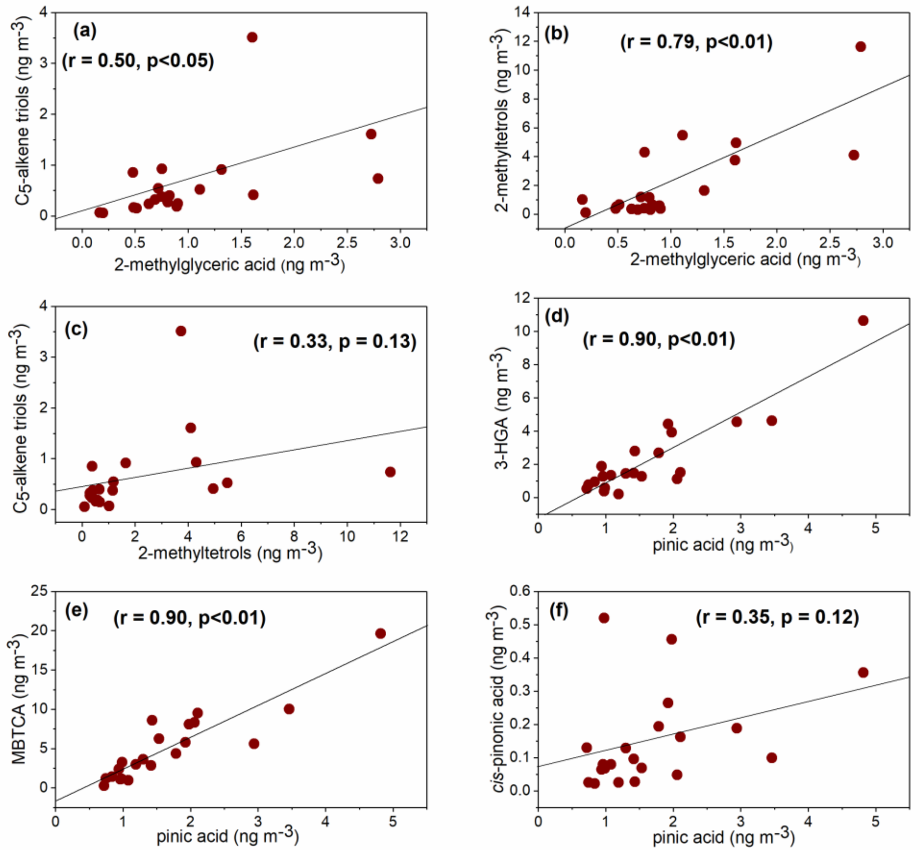

Figure 6. Pearson linear correlation coefficient analysis of BSOA tracers in Gosan TSP aerosols during April 2013-April 2014.

Cis-pinonic acid showed a weak correlation with pinic acid (r = 0.35, p = 0.12) (Figure 6f), suggesting that different atmospheric reactivity of cis-pinonic/pinic acids during transport would cause such poor correlation. In contrast, pinic acid exhibited a strong positive linear correlation with 3-HGA (r = 0.90, p<0.01) and MBTCA (r 0.90, p<0.01) (Figure 6d-e), implying their similar sources. It should be noted that the formation processes of pinic acid, 3-HGA, and MBTCA are different because pinic acid is a first-generation product, and 3-HGA and MBTCA are high-generation products (Claeys et al., 2007; Müller et al., 2012; Szmigielski et al., 2007). The ratio of *cis*-pinonic acid + pinic acid to MBTCA (P/M) is used to evaluate the aging of monoterpene-SOA.

A low P/M ratio suggests the transformation of *cis*-pinonic and pinic acids to MBTCA and thus relatively aged monoterpene-SOA, whereas a high ratio reflects relatively fresh monoterpene-SOA (Gómez-González et al., 2012; Ding et al., 2014). Gómez-González et al. (2012) reported aged monoterpene-SOA (P/M = 0.84) from a Belgian forest site while fresh chamber-produced α-pinene-SOA tracers showed P/M ratios of 1.51 to 3.21 (Offenberg et al., 2007). The average ratio of P/M in this study showed 0.62 with the low value in summer (Figure S5b, Table 2), which is lower than those of Guangzhou (fresh monoterpene-SOA; 28.9) while the air masses originated from southern China (Ding et al., 2014). This observation indicates that monoterpenes SOA have undergone substantial aging during transport to Gosan, particularly in summer when extensive photochemical oxidation occurred due to the high temperature and intense solar radiation.

3.5. Relative abundances in WSOC and OC

Levoglucosan is the most abundant anhydrosugar, contributing 0.05-1.54% of WSOC and 0.03-1.02% of OC. Likewise, sucrose, glucose, and fructose were more abundant primary sugars, contributing 0.01-2.83%, 0.03-1.22%, and 0.02-0.74% of WSOC, respectively. Contributions of the three primary sugars varied from 0.05% to 3.41% of OC. Arabitol and mannitol are the most abundant sugar alcohols, whose contribution to WSOC and OC ranged from 0.02% to 0.93% and 0.01% to 0.85%, respectively. Figure 7 depicts the contribution of sugar compounds to WSOC and OC in TSP collected over Gosan during the study period. We also compared the atmospheric abundances of sugar compounds from Gosan with the literature data (Table 3). This comparison revealed the less influence of BB tracer compounds (i.e., anhydrosugar levels) over Gosan, a factor of 5-10 times lower than those reported for the BB-influenced source regions in China and East Asia (Fu et al., 2012b; Kang et al., 2018a; Wang and Kawamura, 2005; Wang et al., 2012). However, the levels of anhydrosugar over Gosan are higher than those observed over the remote Canadian High Arctic site (Fu et al., 2009a). In contrast, Gosan is characterized by high concentrations of primary sugars compared to other remote sampling sites in Table 3. This is because of the overwhelming contribution of primary sugars associated with soil dust particles over Gosan during the East Asian outflow. Such high loadings of primary sugars were observed from other remote island receptor sites in the WNP (Okinawa) during the spring season (Zhu et al., 2015a). Likewise, the concentrations of sugar alcohols from Gosan are similar to those from other receptor sites influenced by the East Asian outflow (Verma et al., 2018; Zhu et al., 2015a).

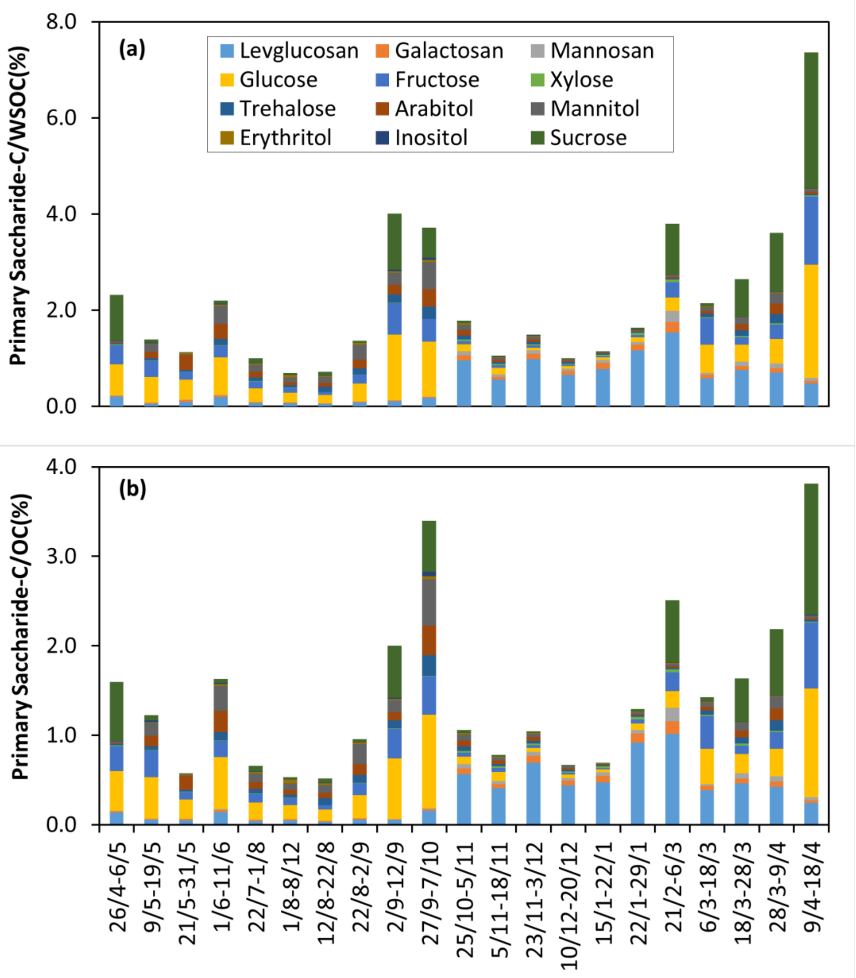

Figure 7. Contribution of primary saccharide-C in (a) WSOC(%) and (b) OC(%) in TSP collected over Gosan during April 2013-April 2014.

The Contributions of isoprene-SOA tracers to ambient OC (0.01-0.40%, avg. 0.09%) and WSOC (0.02-0.57%, 0.13%) were lower than those of monoterpene-derived SOA (0.04-0.63%, 0.23% for OC and 0.06-0.82%, 0.33% for WSOC). The contributions of isoprene oxidation products to OC and WSOC were found to be highest in summer (0.23% and 0.32%, respectively), followed by fall (0.09% and 0.13%), spring (0.04% and 0.06%), and winter (0.02% and 0.03%). Likewise, the contributions of monoterpene-SOA products to aerosol OC and WSOC exhibited the highest value in summer (0.40% and 0.55%, respectively), followed by fall (0.24% and 0.35%), spring (0.18% and 0.27%), and winter (0.10% and 0.14%). We found that the contribution of BSOA products to the carbonaceous components was twice in summer (Figure 8, Table 2). This means BSOA formation occurred in summer to a greater extent due to the intensive BVOCs emission with key factors of meteorological parameters (higher temperature and radiation), i.e., higher concentrations of ozone, and other oxidizing agents (NO_x, OH, etc.). It should be pointed out that the fraction of WSOC in OC (WSOC/OC) is often prone to photochemical aging and, hence, contributes to SOA. More specifically, the WSOC/OC > 0.4 over a receptor site indicates aged aerosols with a significant SOA contribution (Haque et al., 2019; Boreddy et al., 2018). In our study, the average WSOC/OC ratio was observed 0.68 with the highest in summer (0.72), although subtle difference exhibited in winter/autumn (> 0.68), implying the presence of aged OA over the KCOG. Such higher abundances of SOA over Gosan during transport from East Asia is mainly due to photochemical aging of anthropogenic (fossil fuel/biomass combustion) emissions. Huang et al. (2014) reported significant SOA formation from the fossil fuel/biomass combustion precursor VOCs in winter over China.

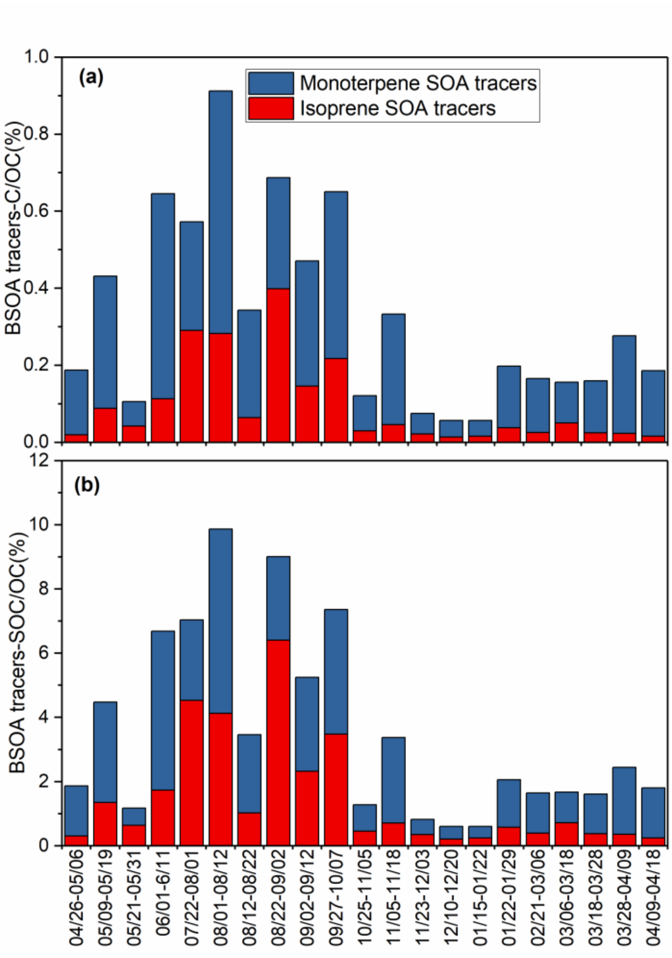

Figure 8. Contribution of (a) isoprene- and monoterpene-SOA tracers-C in ambient OC(%) and (b) isoprene- and monoterpene-SOA tracers-SOC in ambient OC(%) in Gosan aerosol samples during April 2013-April 2014.

We estimated secondary organic carbon (SOC) derived from isoprene and monoterpenes, using the measured values of BSOA tracers and following the SOA tracer-based method first proposed by Kleindienst et al. (2007). A summary of the estimated SOC is provided in Table 2. The contribution of isoprene to SOC was calculated 2.26 to 97.4 ngC m^{-3} (avg. 23.7 ngC m^{-3}), accounting for 1.45% of OC and 35.5% of total BSOC with the predominance in summer (3.56% and 46.6% for OC and SOC, respectively). The estimation of monoterpene-SOA to SOC (avg. 40.1 ngC m^{-3}) was observed around two times higher than that of isoprene-SOA (Table 2). Interestingly, the contribution of monoterpene-derived SOC to ambient OC (3.65%) was dominant in summer, but monoterpene SOC to total SOC (72.1%) was most abundant in spring (Figure 8b). The seasonal distributions of biogenic SOC (Table 2) implied that a substantial amount of SOC was formed from monoterpenes in spring. The estimated biogenic SOC at the KCOG is almost one order of magnitude lower than those from other continental sites in China urban areas (e.g., Pearl River Delta: 446 ngC m^{-3}) (Ding et al., 2012). However, the estimated biogenic SOC load from the KCOG is much higher than that reported from a remote site in the Canadian High Arctic (Alert: 9.4 ngC m^{-3}) (Fu et al., 2009b) and comparable with over the East China Sea (Kang et al., 2018b).

It should be noted that concentrations of SOA tracers cannot always provide the actual contribution of the biogenic source to ambient organic aerosol mass. For example, loadings of monoterpene-SOA tracers were lower in sample KOS999 (28 March - 9 April 2014) (3.02 ng m^{-3}) compared to sample KOS1000 (9 - 18 April 2014) (14.4 ng m^{-3}), whereas the estimated contribution of SOC to ambient OC showed an opposite trend (KOS999: 2.08%; KOS1000: 1.56%). This result demonstrates that the estimation of SOC is an important factor in evaluating the contribution of BSOA to organic aerosol mass. We calculated biogenic OC using radiocarbon (^{14}C) data following the method proposed by Szidat et al. (2006). Biogenic OC showed a poor correlation with biogenic SOC ($r = 0.36$, $p = 0.09$) but a significant linear relationship with primary sugars (i.e., glucose, fructose and sucrose) ($r = 0.54$, $p<0.5$), suggesting that primary bioaerosols from plant-derived airborne pollen dictate on biogenic OC over Gosan.

3.6. Significance of fossil fuel as a source for levoglucosan

The ambient *Lev* levels showed a significant linear relationship with the OC_{fossil}, suggesting the fossil source contribution of this molecular marker (Figure 9a). However, such a significant correlation was not evident between OC_{fossil} and other major sugar compounds. Until recently, *Lev* has been thought to originate primarily from the hemicellulose/cellulose pyrolysis of vegetation and, hence, can be employed as a powerful tracer for biomass smoke particles (Fraser and Lakshmanan, 2000; Simoneit et al., 1999). Nevertheless, residential coals (*e.g.*, lignite and bituminous coal) have been shown to contain high concentrations of ‘*Lev*’ but also emit traces of *Man and Gal* (Kourtchev et al., 2011; Fabbri et al., 2008). Recently, Yan et al. (2018) found a significant linear relationship between the ^{14}C-based fossil fraction of WSOC and *Lev-C* in the aerosols generated from coal combustion and the ambient aerosol samples. Therefore, the prevailing linear relationship between OC_{fossil} and *Lev*-C in the Gosan samples (Figure 9a) is likely due to a common source contribution from coal combustion in East Asia.

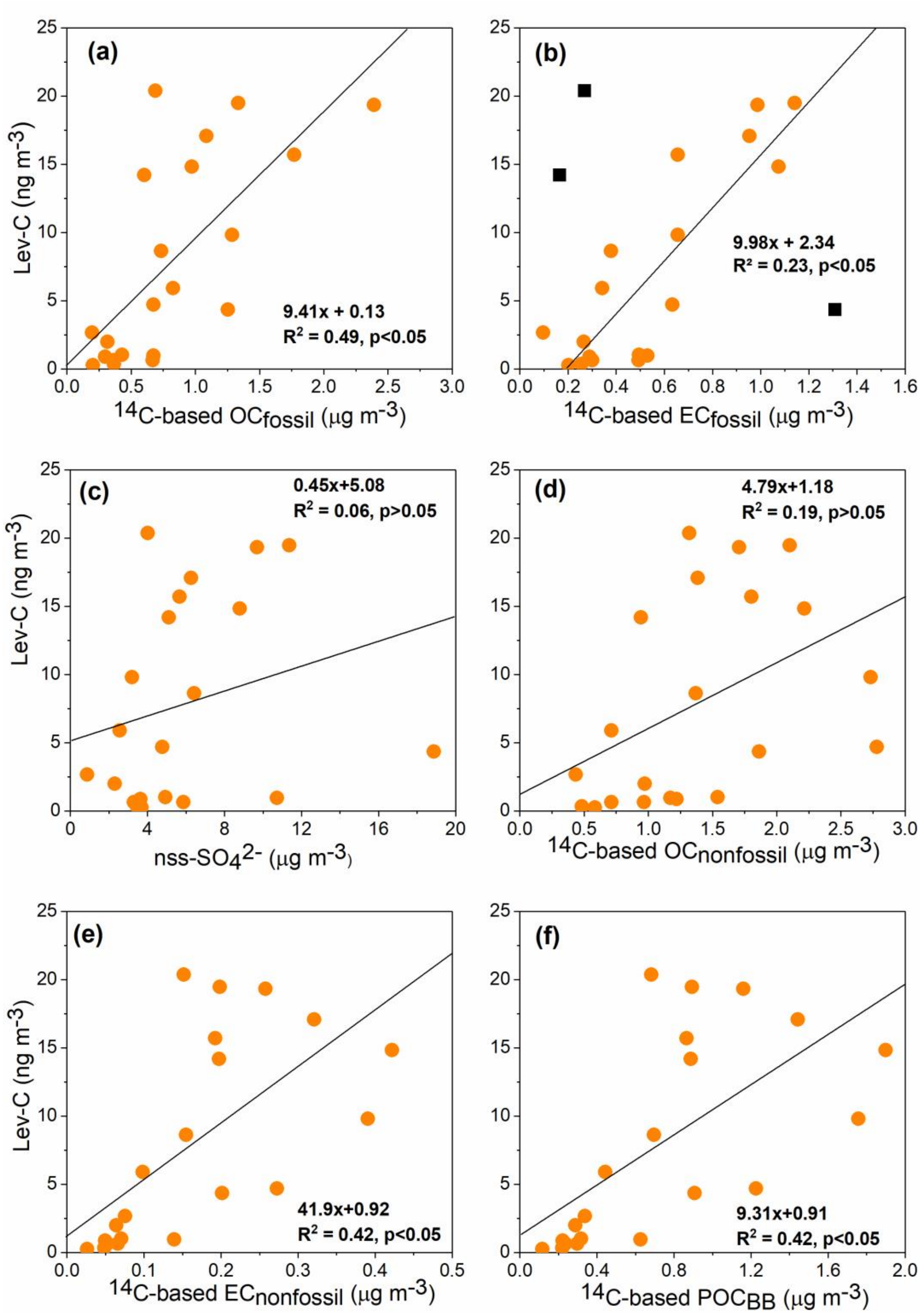

Figure 9. Linear regression analysis between levoglucosan in terms of their carbon content (*Lev*-C) and ^{14}C-based mass concentrations of (a) organic carbon and (b) elemental carbon of fossil origin (OC_{fossil} and EC_{fossil}, respectively), (c) nss-SO_4^{2-}, (d) nonfossil derived organic carbon ($OC_{nonfossil}$), (e) nonfossil derived elemental carbon ($EC_{nonfossil}$), and (f) biomass burning derived primary OC (POC_{BB}) in TSP collected over Gosan during April 2013-April 2014. In panel (b), the squares represent three outliers (i.e., samples with rather high and low *Lev*/EC ratios; please see text for more details).

The slope of the linear regression between *Lev-C* and OC_{fossil} (0.0094; Figure 9a) is higher than those documented for the coal combustion source in China (~0.004±0.007) (Yan et al., 2018). Moreover, *Lev-C* moderately correlated with the EC_{fossil} with the regression slope (~0.01) in the Gosan samples (Figure 9b), being comparable to that observed for the coal combustion in China (0.044±0.076) (Yan et al., 2018). It should be noted that excluding the three outliers as shown in Figure 9b (black square), *Lev-C* showed a stronger correlation with EC_{fossil} (R^2 = 0.74, $p < 0.05$). Of these, two outliers in winter (KOS995: 22-29 January 2014; KOS996: 21 February - 3 March 2014) have higher *Lev*-C levels over that of EC_{fossil}, when air mass trajectories showed the impact of BB emissions in the North China Plain. In contrast, the third outlier in summer (KOS979: 1-11 June 2013) has a lower *Lev*-C/EC_{fossil}, while air parcels transported from nearby cities in China, Korea, and Japan, thus, have more contribution from vehicular emissions. Overall, both the regression slopes are, thus, the representative nature of *Lev*-C/OC_{fossil} and *Lev*-C/EC_{fossil} in the East Asian outflow. *Lev*-C and nss-SO_4^{2-} exhibited a poor correlation (Figure 9c), although both were transported from East Asia.

Lev-C exhibited a rather weak correlation with $OC_{nonfossil}$ (R^2 = 0.19) than that with $EC_{nonfossil}$ (R^2 = 0.42) over Gosan during the study period (Figure 9d-e). This could be likely because $OC_{nonfossil}$ has contributions from the BB and the secondary formation process or the primary biogenic sources. The contribution of primary OC generated from BB (POC_{BB}) to $OC_{nonfossil}$ was taken from Zhang et al. (2016). In their study, the ^{14}C-based $EC_{nonfossil}$ levels were scaled by a factor to constrain the POC_{BB} (Zhang et al., 2016). Here the conversion factor is '4.5' (range: 3-10), which is a median value representing the primary OC/EC ratio from BB emissions ($(POC/EC)_{BB}$).

$$POC_{BB} = EC_{nonfossil} \times (POC/EC)_{BB} \qquad (7)$$

Lev-C showed a somewhat improved linear correlation with POC_{BB} than with the $OC_{nonfossil}$ (Figure 9f). It is apparent from Figure 9 that the regression slopes are comparable, indicating their contribution to *Lev* from both coal combustion and BB emissions over Gosan. The prevailing weak linear relationship (moderate correlation) of *Lev* with nonfossil and fossil carbon fractions is likely the result of photo-degradation of *Lev* during atmospheric transport. This result would mean that the higher atmospheric abundance of *Lev* and its pronounced linear relationships with the nonfossil and fossil carbon fractions implies a much stronger impact of both source emissions in East Asia during the continental outflow in winter and spring.

Overall, we present a new finding on the contribution of coal combustion sources in East Asia in controlling the atmospheric levels of *Lev* apart from the traditional biomass/biofuel burning emissions. This is based on the prevailing linear relationship between the radiocarbon based nonfossil-EC and *Lev* in the year-round TSP samples collected from the KCOG site in Jeju Island. The Gosan supersite is the best location to understand how the chemical composition of source-emissions from East Asia affects the outflow regions in winter and spring. Recent studies have highlighted the potential contribution of *Lev* from residential coal combustion in China (Yan et al., 2018), with estimated annual emission of ~2.2 Gg of *Lev* from domestic coal combustion (Wu et al., 2021). Given this background information, the prevailing significant linear relationship between *Lev* and nonfossil-EC (p-value < 0.05) over the KCOG clearly emphasizes the need for reconsideration of the previous assessments on the impact of BB in East Asian outflow to the WNP. Additionally, this dataset is further compared with the molecular distributions and relative abundances of organic tracers in the TSP samples collected over Gosan during 2001, a decade ago (Fu et al., 2012a). This comparison allows us to better understand the regional changes in the emission sources (*e.g.*, fugitive dust, BB, and fossil-fuel combustion) on a decadal basis.

4. Conclusions

We investigated seasonal variations of primary organic components such as anhydrosugars, primary sugars, and sugar alcohols and biogenic secondary organic aerosol (BSOA) tracers (isoprene- and monoterpene-derived SOA products) in ambient aerosols from Gosan, Jeju Island. Among the detected sugar compounds, levoglucosan was dominant in winter/fall, whereas glucose and sucrose were more abundant in spring/summer. The seasonal trends documented that BB impact is more significant in winter/fall and the primary bioaerosol particles are important in spring/summer. Diagnostic ratios of levoglucosan, galactosan, and mannosan reflect that emissions from BB are mostly dominated by hardwood. The significant linear relationship of sucrose with glucose and fructose suggests their origin from airborne pollen. On a similar note, trehalose showed a significant positive correlation with arabitol, mannitol, and erythritol, implying their contribution from airborne fungal spores and soil microbes over the KCOG.

Distributions of biogenic SOA tracers were characterized by a predominance of monoterpene- than isoprene-derived oxidation products in Gosan aerosols. The BSOA tracers were formed in summer to a greater extent, followed by fall/spring and then winter. The low ratio of *cis*-pinonic acid + pinic acid to MBTCA (P/M) demonstrated that monoterpene-SOA was relatively aged over Gosan aerosols. The estimated secondary organic carbon (SOC) with the predominance in summer shows that substantial BSOA formation occurred in summer due to favorable meteorological conditions. The backward air mass trajectories and source apportionment studies entirely demonstrated that emission from East Asia significantly dominates the ambient OA mass over KCOG. Interestingly, levoglucosan-*C* exhibited a significant positive correlation with nonfossil and fossil organic carbon fractions, along with the comparable regression slopes. This result reveals that BB and coal (lignite) combustion both are prominent sources for levoglucosan in the East Asian outflow.

Although there is some evidence that levoglucosan could originate from the combustion of brown coals (*e.g.,* lignite) in China, our observations from the KCOG (receptor site) also hinted at the fossil source contribution of this molecular marker in the regional influx of the East Asian outflow. Therefore, attribution of ambient levoglucosan levels over the WNP to the impact of BB emission may cause large uncertainty.

Data availability

The data used in this paper are available upon request from the corresponding author.

Author contributions

KK and YLZ designed the research. ML collected the aerosol samples. MMH and SB performed the analysis of aerosol samples. MMH wrote the paper under the guidance of YLZ and KK. All authors were actively involved in the discussion of the paper.

Competing interests

The authors declare that they have no conflict of interest.

Acknowledgments

We acknowledge the financial supports of the Japan Society for the Promotion of Science (JSPS) through Grant-in-Aid No. 24221001 and the National Natural Science Foundation of China (Grant No 41977305).

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Table 1. Concentrations of identified sugar compounds and BSOA tracers (ng m^{-3}) in the atmospheric aerosol samples from Gosan.

Species	Annual	Summer	Fall	Winter	Spring
	Avg.[a] ± S.D.[b] Min.[c], Max.[d]	Avg. ± S.D. Min., Max.	Avg. ± S.D. Min., Max.	Avg. ± S.D. Min., Max.	Avg. ± S.D. Min., Max.
Anhydrosugars					
Levoglucosan (Lev)	17.6 ± 16.8	2.92 ± 3.89	21.7 ± 19.0	39.2 ± 6.60	12.7 ± 11.6
	0.60, 45.9	0.60, 9.81	2.30, 43.9	32.0, 45.9	1.45, 33.4
Mannosan (Man)	1.57 ± 1.82	0.18 ± 0.24	1.69 ± 1.49	3.63 ± 2.28	1.31 ± 1.57
	0.05, 6.74	0.05, 0.61	0.13, 3.66	1.47, 6.74	0.08, 4.08
Galactosan (Gal)	2.28 ± 2.10	0.64 ± 0.68	2.45 ± 2.13	5.21 ± 1.64	1.65 ± 1.26
	0.14, 6.78	0.14, 1.82	0.35, 4.92	3.40, 6.78	0.50, 3.88
Primary Sugars					
Glucose	18.8 ± 27.1	13.4 ± 18.2	16.5 ± 15.7	4.74 ± 3.14	32.4 ± 41.1
	2.45, 122	2.45, 45.6	2.68, 33.6	2.88, 9.44	4.87, 122
Fructose	10.3 ± 15.9	4.90 ± 5.15	7.48 ± 6.99	3.82 ± 4.45	19.8 ± 25.0
	0.97, 74.0	0.97, 13.7	1.71, 16.2	1.56, 10.5	2.69, 74.0
Sucrose	16.1 ± 32.2	1.46 ± 1.05	9.74 ± 12.1	8.87 ± 16.3	35.1 ± 50.5
	0.26, 140	0.68, 3.28	0.76, 27.2	0.42, 33.3	0.26, 140
Trehalose	2.42 ± 1.97	2.72 ± 2.53	3.71 ± 2.29	1.21 ± 0.42	1.98 ± 1.55
	0.65, 7.03	0.97, 6.98	1.18, 7.03	0.72, 1.63	0.65, 5.33
Xylose	0.81 ± 0.65	0.23 ± 0.23	0.86 ± 0.68	1.59 ± 0.37	0.74 ± 0.55
	0.04, 2.03	0.04, 0.63	0.14, 1.70	1.21, 2.03	0.16, 1.68
Sugar alcohols					
Arabitol	3.96 ± 4.24	5.64 ± 7.46	5.27 ± 3.51	1.06 ± 0.60	3.47 ± 2.19
	0.47, 18.7	1.20, 18.7	2.27, 10.9	0.47, 1.91	1.18, 6.30
Mannitol	4.61 ± 5.54	7.39 ± 8.65	6.64 ± 6.03	0.99 ± 0.45	3.24 ± 2.67
	0.25, 22.0	1.71, 22.0	1.69, 16.7	0.55, 1.60	0.25, 7.03
Erythritol	0.62 ± 0.43	0.92 ± 0.53	0.93 ± 0.33	0.42 ± 0.27	0.30 ± 0.12
	0.12, 1.52	0.42, 1.52	0.55, 1.27	0.16, 0.80	0.12, 0.48
Inositol	0.34 ± 0.39	0.29 ± 0.41	0.56 ± 0.60	0.13 ± 0.07	0.35 ± 0.28
	0.04, 1.53	0.08, 1.03	0.10, 1.53	0.08, 0.23	0.04, 0.73
Isoprene-SOA tracers					
2-MGA	0.99 ± 0.70	1.61 ± 1.17	0.95 ± 0.43	0.67 ± 0.14	0.76 ± 0.35
	0.17, 2.79	0.17, 2.79	0.51, 1.61	0.49, 0.81	0.20, 1.32
Σ2-MLTs	1.04 ± 1.40	2.48 ± 1.98	1.34 ± 1.16	0.20 ± 0.05	0.29 ± 0.25
	0.05, 5.81	0.51, 5.81	0.33, 2.74	0.15, 0.26	0.05, 0.82
ΣC5-alkene triols	0.20 ± 0.25	0.46 ± 0.44	0.13 ± 0.05	0.09 ± 0.03	0.14 ± 0.11
	0.02, 1.17	0.02, 1.17	0.05, 0.18	0.05, 0.13	0.02, 0.30
Monoterpene-SOA tracers					
cis-pinonic acid	0.15 ± 0.14	0.12 ± 0.13	0.08 ± 0.06	0.07 ± 0.03	0.26 ± 0.17
	0.02, 0.52	0.03, 0.36	0.02, 0.16	0.02, 0.10	0.08, 0.52
pinic acid	1.67 ± 1.01	2.40 ± 1.67	1.53 ± 0.57	1.07 ± 0.25	1.61 ± 0.78
	0.72, 4.81	0.99, 4.81	0.75, 2.11	0.84, 1.42	0.72, 2.94
3-HGA	2.30 ± 2.38	3.46 ± 4.39	1.52 ± 0.77	1.40 ± 0.39	2.54 ± 1.83
	0.19, 10.6	0.19, 10.6	0.76, 2.79	0.94, 1.88	0.38, 4.57
MBTCA	5.11 ± 4.54	8.44 ± 6.88	6.24 ± 3.64	1.90 ± 0.85	3.77 ± 2.96
	0.29, 19.6	3.00, 19.6	1.17, 9.49	0.97, 2.83	0.29, 8.08

[a] Average, [b] Standard deviation, [c] Minimum, [d] Maximum. 2-MGA: 2-methylglyceric acid, 2-MLTs: 2-methyltetrols, 3-HGA: 3-hydroxyglutaric acid, MBTCA: 3-methyl-1,2,3-butanetricarboxylic acid.

Table 2. Statistical summary of diagnostic ratios and carbonaceous components contribution in Gosan aerosols.

Species	Annual	Summer	Fall	Winter	Spring
	Avg.[a] ± S.D.[b] Min.[c], Max.[d]	Avg. ± S.D. Min., Max.	Avg. ± S.D. Min., Max.	Avg. ± S.D. Min., Max.	Avg. ± S.D. Min., Max.
Diagnostic ratios					
Lev/Man	15.1 ± 6.76 6.81, 31.3	17.1 ± 8.21 8.58, 28.4	13.7 ± 2.51 11.1, 17.3	13.5 ± 6.24 6.81, 21.7	15.4 ± 8.76 6.98, 31.3
Man/Gal	0.55 ± 0.32 0.10, 1.07	0.28 ± 0.13 0.10, 0.42	0.64 ± 0.20 0.37, 0.90	0.66 ± 0.27 0.43, 1.05	0.61 ± 0.43 0.16, 1.07
Lev/(Man + Gal)	4.27 ± 1.23 2.18, 6.56	3.12 ± 0.78 2.18, 4.03	5.08 ± 0.32 4.71, 5.51	4.85 ± 1.35 3.49, 6.56	4.17 ± 1.33 2.50, 6.45
Lev/K^+ × 10^{-2}	5.73 ± 5.65 0.65, 23.2	1.00 ± 0.52 0.65, 1.91	3.94 ± 2.51 1.26, 6.30	12.3 ± 7.87 6.42, 23.2	6.65 ± 4.48 1.77, 15.3
Lev/OC × 10^{-2}	0.73 ± 0.66 0.08, 2.29	0.14 ± 0.10 0.08, 0.31	0.84 ± 0.61 0.12, 1.56	1.60 ± 0.67 0.99, 2.29	0.56 ± 0.40 0.11, 1.05
Lev/WSOC × 10^{-2}	1.09 ± 0.97 0.10, 3.46	0.20 ± 0.13 0.10, 0.42	1.25 ± 0.94 0.23, 2.21	2.33 ± 0.90 1.48, 3.46	0.92 ± 0.65 0.13, 1.69
2-MGA/2-MLTs	2.18 ± 1.59 0.33, 5.40	0.67 ± 0.43 0.33, 1.33	1.04 ± 0.49 0.41, 1.55	3.60 ± 1.43 1.90, 5.40	3.27 ± 1.17 1.60, 4.75
[e]P/MBTCA	0.62 ± 0.61 0.17, 5.90	3.22 ± 0.62 2.47, 3.89	3.62 ± 1.67 1.52, 5.90	1.68 ± 0.64 0.83, 2.38	1.73 ± 1.07 0.34, 3.32
Carbonaceous components					
Isoprene derived SOC (µgC m^{-3})	23.7 ± 23.9 2.26, 97.4	51.0 ± 32.4 8.08, 97.4	25.9 ± 17.9 8.54, 45.7	8.52 ± 1.07 7.46, 9.87	11.3 ± 6.80 2.26, 24.9
Isoprene SOC to OC (%)	1.45 ± 1.74 0.21, 6.40	3.56 ± 2.18 1.03, 6.40	1.46 ± 1.38 0.35, 3.48	0.35 ± 0.17 0.21, 0.57	0.57 ± 0.38 0.24, 1.35
Isoprene SOC to total SOC (%)	35.5 ± 15.5 13.5, 71.1	46.6 ± 20.3 26.0, 71.1	38.1 ± 10.4 21.1, 47.3	31.6 ± 7.05 24.0, 39.7	27.9 ± 15.5 13.5, 54.0
Monoterpene SOC (µgC m^{-3})	40.1 ± 33.3 7.18, 154	62.7 ± 56.5 19.2, 154	40.7 ± 20.0 11.8, 57.7	19.3 ± 5.61 14.0, 25.2	35.5 ± 23.5 7.18, 62.7
Monoterpene SOC to OC (%)	2.0 ± 1.47 0.37, 5.74	3.65 ± 1.58 2.44, 5.74	2.15 ± 1.45 0.48, 3.88	0.87 ± 0.58 0.37, 1.48	1.58 ± 0.84 0.54, 3.13
Monoterpene SOC to total SOC (%)	64.5 ± 15.5 28.9, 86.5	53.4 ± 20.3 28.9, 74.0	61.9 ± 10.4 52.7, 78.9	68.4 ± 7.05 60.3, 76.1	72.1 ± 15.5 46.0, 86.5

[e] P: *cis*-pinonic acid + pinic acid

Table 3. Comparisons of the mean concentration (ng m^{-3}) of anhydrosugars, sugar, and sugar alcohols in Gosan aerosols with those from different sites around the world.

Sampling sites	Sampling type	Sampling time	Anhydro-sugars	Primary sugars	Sugar alcohols	References
Gosan, South Korea	**TSP**	Summer	**3.74**	**22.8**	**14.3**	**This study**
		Fall	**25.9**	**38.3**	**13.4**	
		Winter	**48.0**	**20.2**	**2.60**	
		Spring	**15.7**	**90.0**	**7.36**	
Chennai, India	PM$_{10}$	Summer	127	15.5	7.44	Fu et al., 2010b
		Winter	134	11.4	4.81	
Mt. Tai, China	TSP	Summer (June)	224	61.1	125	Fu et al., 2012b
Alert, Canada	TSP	Winter	0.32	1.14	0.25	Fu et al., 2009a
		Spring	0.02	0.18	0.36	
Okinawa, western North Pacific	TSP	Summer	0.93	73.5	62.9	Zhu et al., 2015a, b
		Autumn	2.58	56.0	30.7	
		Winter	6.04	34.4	6.03	
		Spring	3.44	101	31.2	
Chichijima, western North Pacific	TSP	Summer	0.32	32.8	38.6	Verma et al., 2015, 2018
		Autumn	0.85	22.0	35.1	
		Winter	2.40	14.2	3.93	
		Spring	0.94	24.2	15.5	
Mt. Hua, China (Non-dust storm)	PM$_{10}$	April	57.8	92.5	22.4	Wang et al., 2012
Mt. Hua, China (Dust storm)	PM$_{10}$	April	44.5	162	25.7	Wang et al., 2012
Nanjing, China	PM$_{2.5}$	Summer	151 (Lev)	59.3	11.8	Wang and Kawamura, 2005
		Winter	268 (Lev)	42.3	13.4	
Beijing, China	PM$_{2.5}$	Summer	14.5	6.63	3.31	Kang et al., 2018a
		Autumn	129	17.2	13.7	
		Winter	254	41.5	17.8	
		Spring	81.4	33.9	12.3	
Belgrade, Serbia	TSP	Autumn	425 (Lev)	116	98.4	Zangrando et al., 2016
Maine, USA	PM$_1$	May-October	13.9	28	8.31	Medeiros et al., 2006
Crete, Greece	PM$_{10}$	Year-round	14.4	32.3	6.53	Theodosi et al., 2018

Supporting information for

Regional heterogeneities in the emission of airborne primary sugar compounds and biogenic secondary organic aerosols in the East Asian outflow: Evidence for coal combustion as a source of levoglucosan

Md. Mozammel Haque[1,2,3], Yanlin Zhang[1,2*] Srinivas Bikkina[4], Meehye Lee[5], and Kimitaka Kawamura[3,4*]

[1]Yale-NUIST Center on Atmospheric Environment, International Joint Laboratory on Climate and Environment Change (ILCEC), Nanjing University of Information Science & Technology, Nanjing, 210044, China
[2]School of Applied Meteorology, Nanjing University of Information Science & Technology, Nanjing 210044, China
[3]Institute of Low Temperature Science, Hokkaido University, Sapporo 060-0819, Japan
[4]Chubu Institute for Advanced Studies, Chubu University, Kasugai 487-8501, Japan
[5]Department of Earth and Environmental Sciences, Korea University, Anam-dong, Sungbuk-gu, Seoul 136-701, South Korea

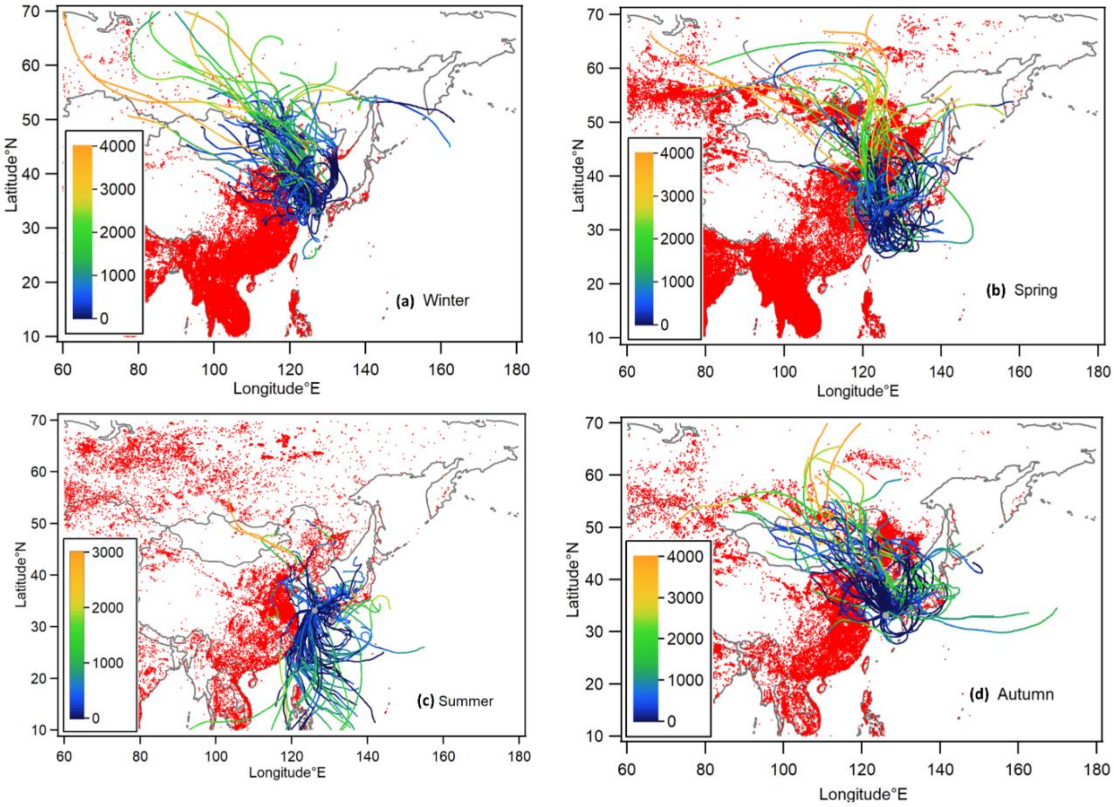

Figure S1. HYSPLIT-based backward air mass trajectories as a function of arrival height (m) (see color bar in each panel) and MODIS-firecounts for the TSP collected over Gosan according the four seasons, (a) winter, (b) spring, (c) summer, and (d) autumn.

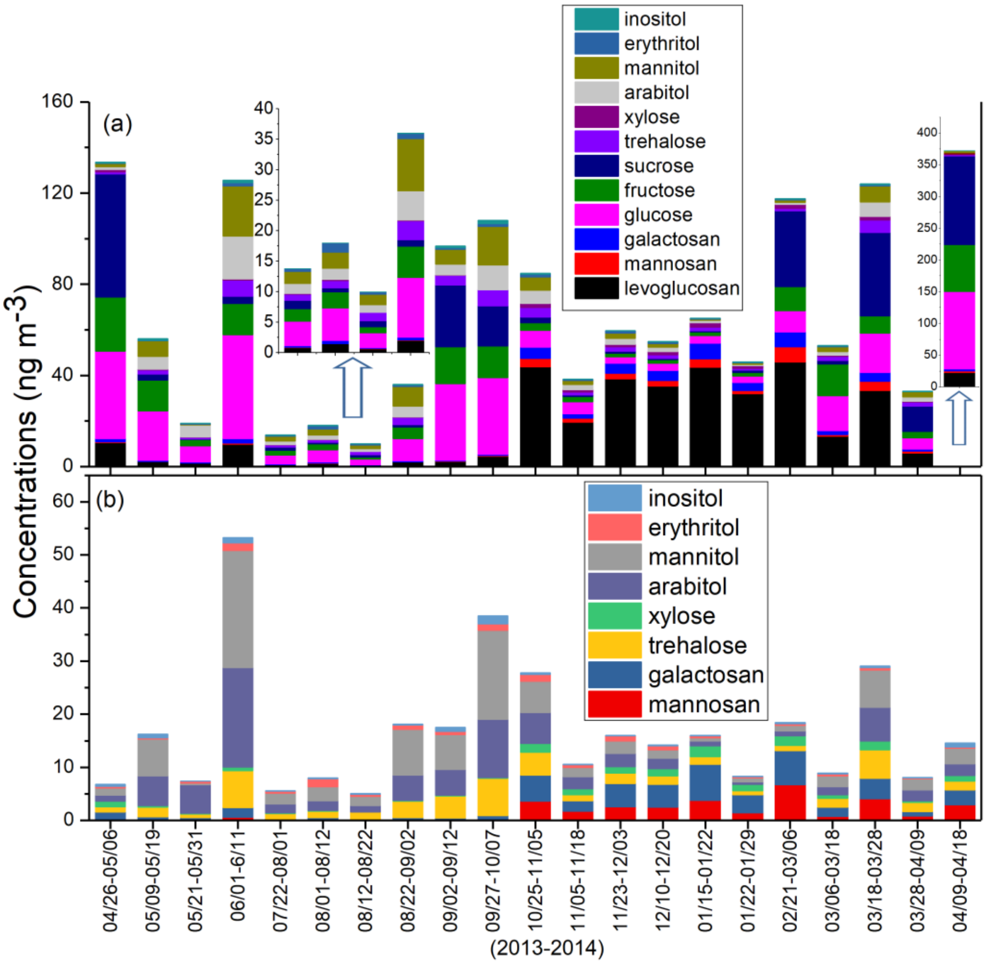

Figure S2. Time series of the concentrations of anhydrosugars, primary sugars, and sugar alcohols during the period of study in Gosan.

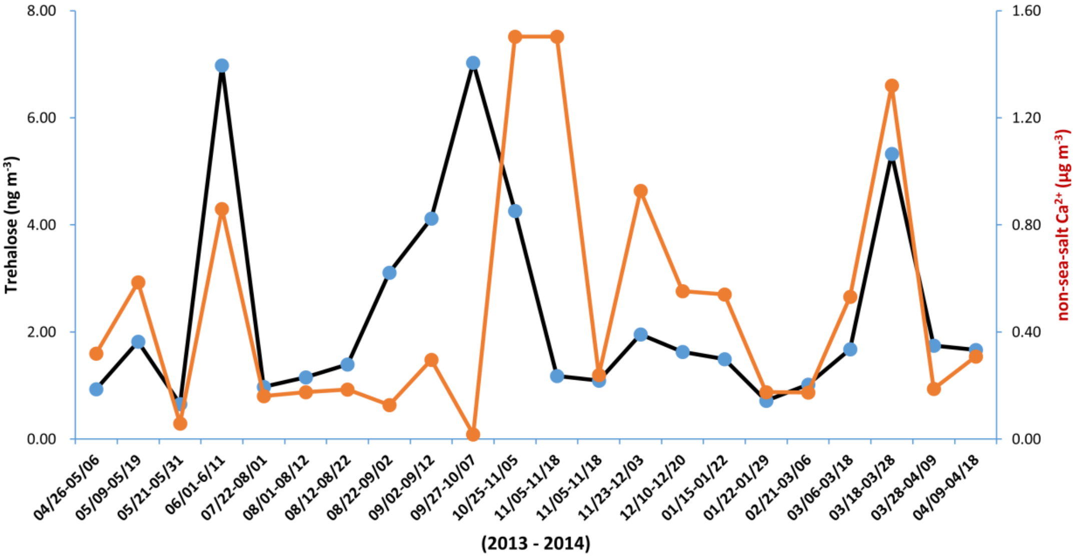

Figure S3. Temporal variability of mass concentrations of trehalose and non-sea-salt Ca^{2+} in TSP collected over Gosan during April 203-April 2014.

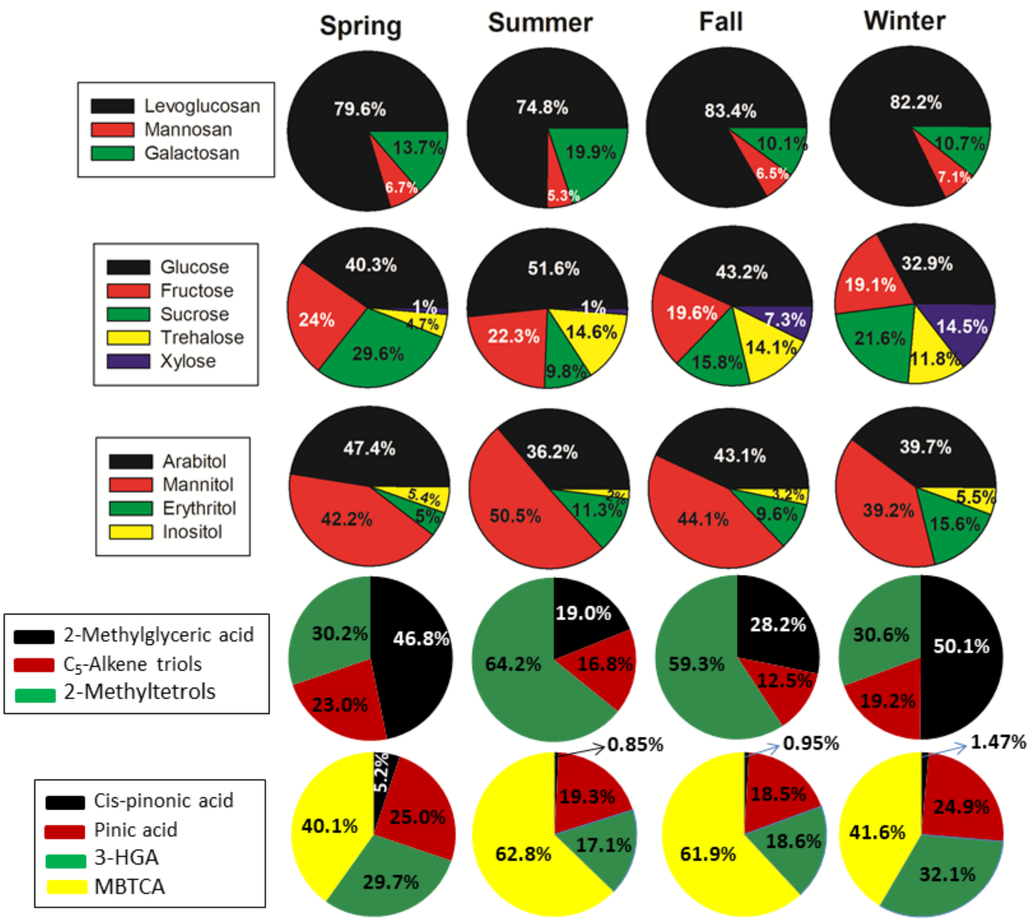

Figure S4. Seasonal variability of molecular distributions of anhydrosugars, primary sugars, sugar alcohols, isoprene-SOA tracers and monoterpene-SOA tracers in their total mass concentration of respective group-types in TSP samples collected over Gosan during April 2013-April 2014.

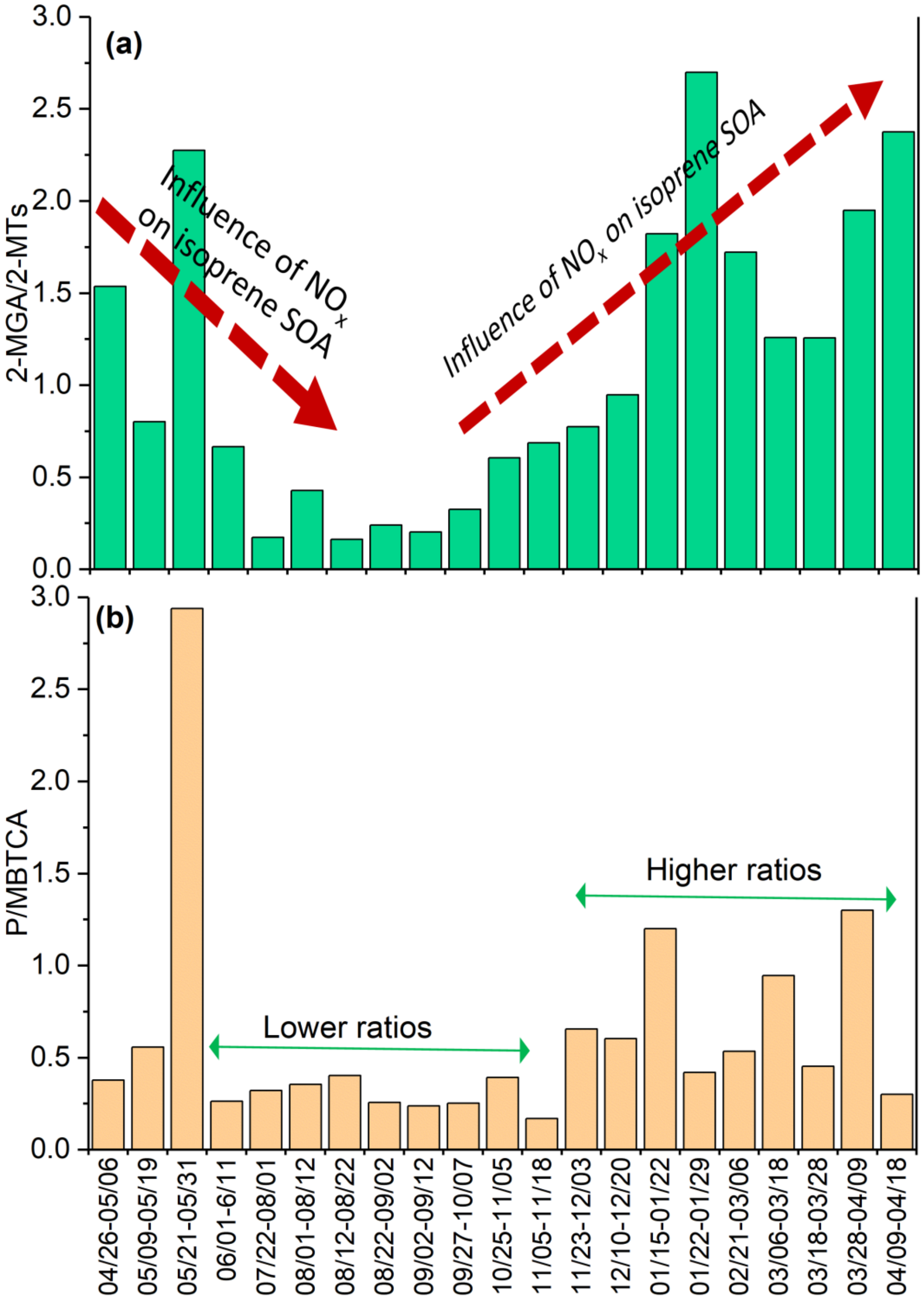

Figure S5. Diagnostic ratios of 2-methylglyceric acid/2-methyltetrols (2-MGA/2-MTs) and *cis*-pinonic acid+pinic acid/3-methyl-1,2,3-butanetricarboxylic acid (P/MBTCA) in Gosan TSP samples collected during April 2013-April 2014.