Arabitol, mannitol and glucose as tracers of primary biogenic organic aerosol: influence of environmental factors on ambient air concentrations and spatial distribution over France

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Abstract. The primary sugar compounds (SC, defined as glucose, arabitol and mannitol) are widely recognized as suitable molecular markers to characterize and apportion primary biogenic organic aerosol emission sources. This work improves our understanding of the spatial behavior and distribution of these chemical species and evidences their major effective environmental drivers. We conducted a large study focusing on the daily (24 h) PM_{10} SC concentrations for 16 increasing space scale sites (local to nation-wide), over at least one complete year. These sites are distributed in several French geographic areas of different environmental conditions. Our analyses, mainly based on the examination of the short-term evolutions of SC concentrations, clearly show distance-dependent correlations. SC concentration evolutions are highly synchronous at an urban city-scale and remain well correlated throughout the same geographic regions, even if the sites are situated in different cities. However, sampling sites located in two distinct geographic areas are poorly correlated. Such pattern indicates that the processes responsible for the evolution of the atmospheric SC concentrations present a spatial homogeneity over typical areas of at least tens of kilometers. Local phenomena, such as resuspension of topsoil and associated microbiota, do no account for the major emissions processes of SC in urban areas not directly influenced by agricultural activities. The concentrations of SC and cellulose display remarkably synchronous temporal evolution cycles at an urban site in Grenoble, indicating a common source ascribed to vegetation. Additionally, higher concentrations of SC at another site located in a crop field region occur during each harvest periods, pointing out resuspension processes of plant materials (crop detritus, leaf debris) and associated microbiota for agricultural and nearby urbanized areas. Finally, ambient air temperature, relative humidity and vegetation density constitute the main effective drivers of SC atmospheric concentrations.

1. Introduction

Primary biogenic organic aerosols (PBOA), which notably comprise bacterial and fungal cells or spores; viruses; or microbial fragments such as endotoxins and mycotoxins; and pollens and plant debris, are ubiquitous particles released from the biosphere to the atmosphere (Amato et al., 2017; Fang et al., 2018; Martin et al., 2010; Perrino and Marcovecchio, 2016; Wéry et al., 2017). PBOA can contribute significantly to the total coarse aerosol mass (Amato et al., 2017; Bozzetti et al., 2016; Coz et al., 2010; Fröhlich-Nowoisky et al., 2016; Jaenicke, 2005; Manninen et al., 2014; Morris et al., 2011; Samaké et al., 2019; Vlachou et al., 2018; Yue et al., 2017). Besides their expected negative human health effects (Fröhlich-Nowoisky et al., 2009, 2016; Humbal et al., 2018; Lecours et al., 2017; Zamfir et al., 2019), they substantially influence the carbon and water cycles at the global scale, notably acting as cloud and ice nuclei (Ariya et al., 2009; Elbert et al., 2007; Fröhlich-Nowoisky et al., 2016; Hill et al., 2017; Humbal et al., 2018; Morris et al., 2014; Rajput et al., 2018). While recent studies have revealed highly relevant information on the abundance and size partitioning of PBOA (Fröhlich- Nowoisky et al., 2017; Huffman and Santarpia, 2017), their emission sources and contribution to total airborne particles are still poorly documented, partly due to the analytical limitations to distinguish PBOA from other types of carbonaceous particulate matter (Bozzetti et al., 2016; China et al., 2018; Di Filippo et al., 2013; Perrino and Marcovecchio, 2016; Yan et al., 2019). Notably, the global emissions of fungal spore emitted into the atmosphere are still poorly constrained and range from 8 Tg.y^{-1} to 186 Tg.y^{-1} (Després et al., 2012; Elbert et al., 2007; Jacobson and Streets, 2009; Sesartic and Dallafior, 2011; Tanarhte et al., 2019).

Recently, source-specific tracer methodologies have been introduced to estimate their contribution to aerosol loadings (Di Filippo et al., 2013; Gosselin et al., 2016; Li et al., 2018; Medeiros et al., 2006b; Verma et al., 2018; Wang et al., 2018). Indeed, atmospheric organic aerosols (OA) contain specific chemical species that can be used as reliable biomarkers in tracing the sources and abundance of PBOA (Bauer et al., 2008; Gosselin et al., 2016; Holden et al., 2011; Jia et al., 2010; Li et al., 2018; Medeiros et al., 2006b; Wang et al., 2018). For instance, among sugar alcohols, arabitol and mannitol (two common storage soluble carbohydrates in fungi)have been recognized as tracers for airborne fungi, and their concentrations are widely used to estimate PBOA contributions to OA mass (Amato et al., 2017; Bauer et al., 2008; Buiarelli et al., 2013; Golly et al., 2018; Medeiros et al., 2006b; Samaké

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et al., 2019; Srivastava et al., 2018; Verma et al., 2018; Weber et al., 2018, 2019). Similarly, glucose has also been used as a tracer for plant materials (such as pollen, leaves, and their fragments) or soil emissions within various studies around the world (Chen et al., 2013; Medeiros et al., 2006b; Pietrogrande et al., 2014; Rathnayake et al., 2017; Rogge et al., 2007; Wan et al., 2019; Xiao et al., 2018; Zhu et al., 2015).

In this context, atmospheric concentrations of specific sugar alcohols and/or primary monosaccharides (including glucose) have been previously quantified at sites in several continental, agricultural, coastal or polar regions (Barbaro et al., 2015; Chen et al., 2013; Glasius et al., 2018; Li et al., 2018; Pietrogrande et al., 2014; Verma et al., 2018; Wan et al., 2019; Yan et al., 2019; Yttri et al., 2007). However, large datasets investigating their (multi)annual cycles, seasonal and simultaneous short-term variations at multiple spatial scale resolutions (i.e. from local to continental) are still lacking (Liang et al., 2013; Nirmalkar et al., 2018; Pietrogrande et al., 2014; Yan et al., 2019). Such records are essential to better understand the spatial behavior of primary sugar compound (SC) concentrations (i.e., glucose, arabitol and mannitol) and PBOA emission processes, and to isolate their potential key drivers (e.g., vegetation type and density, topography, weather conditions, etc.), which are still unclear (Bozzetti et al., 2016). This information would be essential for further implementation into chemical transport models (Heald and Spracklen, 2009; Myriokefalitakis et al., 2017; Tanarhte et al., 2019).

It is commonly acknowledged that SC (particularly arabitol and mannitol) originate from primary biogenic derived sources such as bacterial, fungal spores, and plant materials (Di Filippo et al., 2013; Golly et al., 2018; Gosselin et al., 2016; Holden et al., 2011; Kang et al., 2018; Medeiros et al., 2006b; Wan et al., 2019; Yan et al., 2019; Yttri et al., 2007; Zhu et al., 2018a). Some studies have characterized the composition of SC in topsoil samples (for fractions larger than PM_{10}) from both, natural (i.e., uncultivated) and agricultural regions (Medeiros et al., 2006a; Rogge et al., 2007; Simoneit et al., 2004; Wan and Yu, 2007). The authors suggested that the particulate arabitol, mannitol and glucose are introduced into the atmosphere mainly through resuspended soils or dust particles and associated biota derived from natural soil erosion, unpaved road dust or agricultural practices. Conversely, Jia and Fraser (2011) reported higher concentrations of SC relative to PBOA in size-segregated aerosol samples collected at a suburban site (Higley, USA) compared to the local size-fractionated soils (equivalent to atmospheric $PM_{2.5}$ and PM_{10}). This suggested that direct emissions from biota (microbiota, vascular plant materials) could also be a significant atmospheric input process for SC at this suburban site.

A large database on SC concentrations was obtained over France in the last decade. It already allowed the investigation of the size distribution and seasonal variabilities of SC concentrations in aerosols at 28 French sites, notably showing that SC are ubiquitous primary aerosols, accounting for a significant proportion of PM_{10} organic matter (OM) mass (Samaké et al., 2019). Results confirmed that their ambient concentrations display a well-marked seasonality, with maximum concentrations from late spring to early autumn, followed by an abrupt decrease in late autumn, and a minimum concentration during wintertime in France. This study also showed that the mean PBOA chemical profile is largely dominated by organic compounds, with only a minor contribution of dust particle fraction. The latter result indicated that ambient polyols could most likely be associated with direct biological particle emissions (e.g. active spore discharge, microbiota released from phylloplane or phyllosphere, etc.) rather than with the microorganism-containing soil resuspension. These observations call for more investigations of the predominant SC (and PBOA) emission sources.

Cellulose, a linear polymer composed of D-glucopyranose units linked by β-1,4 bonds, is the most frequent polysaccharide occurring in terrestrial environments (Ramoni and Seiboth, 2016). Plant materials contain cellulose

86 which has been reported as a suitable proxy to evaluate the vegetative debris contribution to OM mass (Bozzetti
87 et al., 2016; Daellenbach et al., 2017; Glasius et al., 2018; Hiranuma et al., 2019; Puxbaum and Tenze-Kunit, 2003;
88 Sánchez-Ochoa et al., 2007; Yttri et al., 2011b). The ambient PM_{10} cellulose has been shown to be abundant in the
89 European semi-rural or background environments (accounting for 2 to 10 % of OM mass) (Glasius et al., 2018;
90 Sánchez-Ochoa et al., 2007) and Nordic rural environments in Norway (contributing to 12 to 18 % of total carbon
91 mass) (Yttri et al., 2011b). Thus, simultaneous concentration measurements of cellulose and SC can provide
92 essential information into their emission source dynamics.

93 As the continuation of our previous work (Samaké et al., 2019), the present paper aims to delineate the processes
94 that drive the atmospheric concentrations of SC and then PBOA. This is achieved through (i) the analysis of
95 simultaneous annual short-term time series of particulate SC concentrations over pairs of sites across multiple
96 space ranges, including local, regional and nationwide sites, and (ii) the investigation of links between
97 concentrations and series key parameters such as meteorological and phenological ones. Simultaneous annual
98 short-term concentration measurements of SC and cellulose was performed to better understand of their sources
99 correlations.

100 **2. Material and methods**

101 **2.1 Sampling sites**

102 Daily PM_{10} concentrations reported in the present work were obtained from different research and monitoring
103 programs conducted over the last six years in France. Within the framework of the present study, we carefully
104 selected sites sharing at least one complete year of concurrent monitoring with another one, to be representative
105 of the annual variation cycles. The final dataset includes data from 16 sites, which are distributed in different
106 regions of France (Fig. 1) and cover several main types of environmental conditions in terms of site topography,
107 local vegetation, and climate. The characteristics and data available at each sampling site are listed in Table S1 of
108 the supplementary material (SM), together with the information on the annual average concentrations of aerosol
109 chemical composition (Table S2). Detailed information on the sampling conditions can be found in Samaké et al.
110 (2019), such as the campaign periods, number of collected PM samples, sampling flow rates, sample storage and
111 handling, etc. Note that, the previous database (Samaké et al., 2019) has been updated here with arabitol and
112 mannitol in PM_{10} collected at the suburban site of Nogent-sur-Oise for a series covering the years 2013 to 2017.

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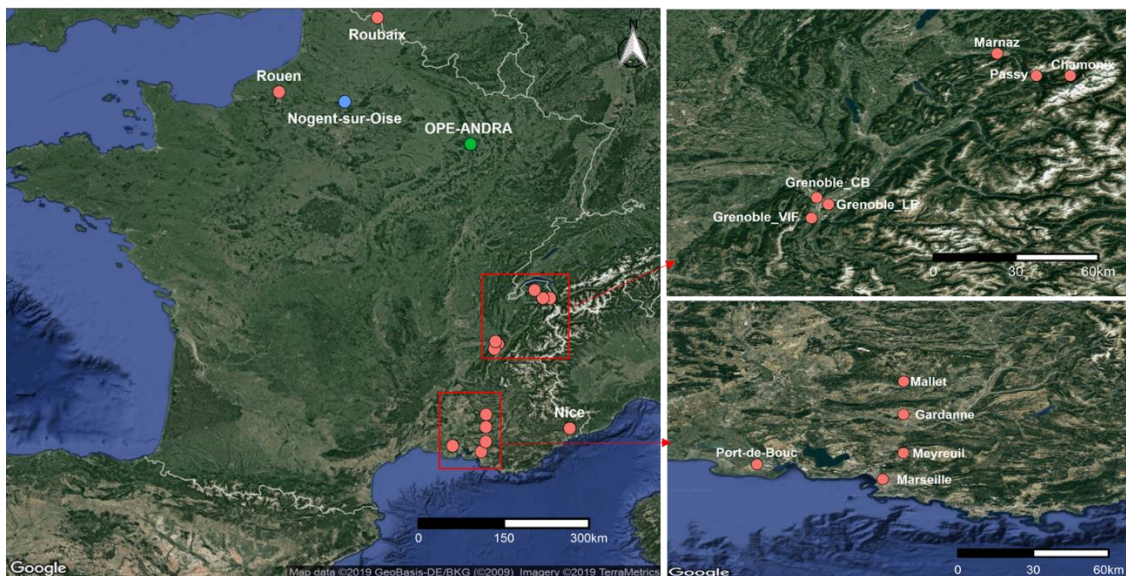
Figure 1: Geographical location of the selected sampling sites. The red and blue dots indicate respectively urban and suburban sites while the green one corresponds to a rural site, surrounded by field crop areas.

2.2 Chemical analyses

Daily (24 h) PM_{10} samples were collected onto prebaked quartz fiber filter (Tissuquartz PALL QAT-UP 2500 150 mm diameter) every third or sixth day, but not concurrently at all sites. They were then analyzed for various chemical species using subsampled fractions of the collection filters and a large array of analytical methods. Details of all the chemical analysis procedures are reported elsewhere (Golly et al., 2018; Samaké et al., 2019; Waked et al., 2014; Weber et al., 2018). Briefly, primary sugar compounds were extracted from filter aliquots (punches typically about 10 cm²) into ultrapure water. The extracts are then filtered using a 0.22 µm Acrodisc filter. Depending on the site, analyses were conducted either by the IGE (Institut des Géosciences de l'Environnement) or by the LSCE (Laboratoire des Sciences du Climat et de l'Environnement) (Samaké et al., 2019). At the IGE, extraction was performed during 20 min in a vortex shaker and analyses were achieved using high-performance liquid chromatography with pulsed amperometric detection (HPLC-PAD). A first set of equipment was used until March 2016, consisting of a Dionex DX500 equipped with three columns Metrosep (Carb 1-Guard + A Supp 15-150 + Carb 1-150), the analytical program was isocratic with 70 mM sodium hydroxide (NaOH) as eluent for 11 min, followed by a gradient cleaning step with a 120 mM NaOH as eluent for 9 min. This procedure allows the analysis of arabitol, mannitol and glucose (Waked et al., 2014). A second set of equipment was used after March 2016, with a Thermo-Fisher ICS 5000+ HPLC equipped with 4 mm diameter Metrosep Carb 2×150 mm column and 50 mm pre-column. The analytical run was isocratic with 15 % of an eluent of sodium hydroxide (200 mM) and sodium acetate (4 mM) and 85 % water, at 1 mL min^{-1}. At the LSCE, extraction was performed for 45 min by sonication and analyses were achieved using ion chromatography instrument (IC, DX600, Dionex) with Pulsed Amperometric Detection (ICS3000, Thermo- Fisher). In addition, a CarboPAC MA1 column has been used (4×250 mm, Dionex) along with an isocratic analytical run with 480 mM sodium hydroxide eluent. This analytical technique allows to quantify arabitol, mannitol and glucose (Srivastava et al., 2018).

For cellulose quantification, we used an optimized protocol based on that described by (Kunit and Puxbaum, 1996; Puxbaum and Tenze-Kunit, 2003), in which the cellulose contained in the lignocellulosic material is enzymatically hydrolyzed into glucose units before analysis. Since the alkaline peroxide pretreatment step used to remove lignin in the original protocol results in a loss of sample material, it has been avoided in this study. Therefore, only the "free cellulose" is reported in our samples. Note that Sánchez-Ochoa et al., (2007) consider that this free cellulose could represent only about 70 % of the total cellulose in air samples and that the total cellulose could represent only about 50 % of the "plant debris" content of atmospheric PM. Very few other results are available on this topic (Bozzetti et al., 2016; Glasius et al., 2018; Vlachou et al., 2018; Yttri et al., 2011b). The protocol has been improved to increase sensitivity and accuracy, by reducing the contribution of glucose in the blanks and by using an HPLC-PAD as the analytical method for the determination of glucose concentrations. Trichoderma reesei cellulase (>700 u g-1, Sigma Aldrich) and Aspergilus Niger glucosidase (>750 u g^{-1}, Sigma Aldrich) have been used as saccharification enzymes. The protocol is detailed in Section 2 of the SM.

Field blank filters (about 10 % of samples) were handled as real samples for quality assurance. The present data have been corrected from field blanks. The reproducibility of the analysis of primary sugar compounds (polyols, glucose) and cellulose, estimated from the analysis of sample extracts from 10 punches of the same filters were in

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154 the range of 10-15 %. The quantification limits primary sugar compounds and cellulose ranged from 0.63 to 0.89

155 ng m^{-3}. About 2 800 samples are considered in this work for the polyols and glucose series, while 290 samples

156 (from the sites of Grenoble_LF and OPE-ANDRA) are considered for the cellulose series. Hereafter, the term

157 "Polyols" is used to refer uniquely to the sum of arabitol and mannitol concentrations.

158 **2.3 Meteorological data and LAI measurements**

159 Ambient weather data were not available at all monitoring sites (see Table S1). In this study, data including daily

160 relative humidity (%), night-time temperature (°C), average and maximum temperatures (°C), wind speed (m s^{-1}),

161 solar radiation (W m^{-2}), and rainfall level (mm) for the sites of Marnaz and OPE-ANDRA (Fig. 1), representing

162 different climatic regions and environmental conditions, were obtained from the French meteorological data

163 sharing service system (Météo-France) and ANDRA (French national radioprotective agency, in charge of the

164 OPE-ANDRA site), respectively.

165 The leaf area index (LAI), which is defined as the projected area of leaves over a unit of land, is an important

166 measure of the local vegetation density variation (Heald and Spracklen, 2009; Yan et al., 2016a, 2016b). For this

167 study, we used the MODIS Collection 6 LAI product because it is considered to have the highest quality among

168 all the MODIS LAI products (Yan et al., 2016a, 2016b). The MCD15A3H product uses both Terra and Aqua

169 reflectance observations as inputs to estimate daily LAI at 500 m spatial resolution, and a 4-day composite is

170 calculated to reduce the noise from abiotic factors. Using a 2×2 km grid box around the monitoring site, the local

171 vegetation density variation was retrieved from LP DAAC (https://lpdaac.usgs.gov/, last accessed: 15 March 2019)

172 for the sites of Marnaz, OPE-ANDRA, and Grenoble_LF.

173 **2.4 Data analyses**

174 All the statistical analyses were carried out using the open-source R software (R studio interface, version 3.4.1).

175 Several statistical analyses were performed on the concentrations to identify the spatial patterns of emission

176 sources and the potential parameters of influence as explained below.

177 The normalized cross-correlation (NCC) test was chosen to examine the potential similarities among the

178 monitoring sites for particulate SC concentrations, in terms of short-term temporal trends (e.g. synchronized

179 periods of increase or decrease, simultaneous fluctuations during specific episodes). The main advantage of NCC

180 over the traditional correlation tests is that it is less sensitive to linear changes in the amplitudes of the two-time

181 series compared. Therefore, to reduce the possibility of spurious "anti-correlation" due to highly variable

182 concentration ranges, data were amplitude-normalized prior to correlation analysis. A thorough discussion on the

183 normalized cross-correlation method can be found elsewhere (Bardal and Sætran, 2016; Dai and Zhou, 2017;

184 Eisner et al., 2009; Kaso, 2018; Lainer et al., 2016; Le Pichon et al., 2019). To achieve pair-wise correlation

185 analysis between the sampling sites collected during the same periods, the original raw daily measurements were

186 processed as follows: starting on identical days for each pairs of sites, arrangement on the original daily data into

187 consecutive 3-day intervals (or 6-day intervals in the case of OPE-ANDRA) and calculation of the average

188 concentration values for the middle-day were performed. The resultant data were used for correlation analysis

189 between site pairs (Table S3).

190 Multiple linear regression (MLR) was used to assess the strength of the relationships between atmospheric

191 concentrations of particulate SC and local environmental factors including the daily mean relative humidity, night-

192 time temperature, average and maximum temperature, wind speed, solar radiation, rain levels and LAI. Because

193 the LAI is a 4-day composite, daily values of the other variables were re-scaled into consecutive 4-day averaged

194 values. The linear regression (linear model or lm) package in R was employed for multiple regression analyses.
195 The concentration data were log-transformed to obtain regression residual distributions as close as possible to the
196 normal Gaussian one (Fig. S1). Stepwise forward selection was used to select the predictors that explain well the
197 temporal variation of SC concentrations at the site of Marnaz.

198 It should be noted that due to the limited availability of external parameters, the environmental factors driving SC
199 atmospheric levels have been extensively investigated for only two monitoring sites with contrasted
200 characteristics: the urban background site of Marnaz located in an Alpine valley, and the rural OPE-ANDRA site
201 surrounded by field crop areas spreading over several tens of km.

202

3. Results and discussion

3.1 Example of spatial coherence of the concentrations at different scales

205 Our previous work (Samaké et al., 2019) showed that particulate polyols and glucose are ubiquitous primary
206 compounds with non-random spatial and seasonal variation patterns over France. Here, an inter-site comparison
207 of their short-term concentration evolutions has been carried out at different space scales (from local to national)
208 for the pairs that can be investigated in our data base. Figure 2 presents some of these comparisons for 3 spatial
209 scales (15, 120, and 205 km).

210 The daily average concentrations of polyols (defined as sum of arabitol and mannitol) and glucose display highly
211 synchronous evolutional trends (i.e., homogeneity in the concentrations, the timing of concentration peaks,
212 simultaneity of the daily specific episodes of increase/decrease of concentrations) over 3 neighboring monitoring
213 sites located 15 km apart in the Grenoble area (Figs. 2A and B). Interestingly, remarkable synchronous patterns
214 both for short term (near-daily) and longer term (seasonal) still occur for sites located 120 km apart, as exemplified
215 for 2 sites in Alpine environments (Grenoble and Marnaz) (Figs. 2C and D). However, as shown in Figs. 2E and
216 F, the evolutions of concentrations become quite dissimilar and asynchronous in terms of seasonal and daily
217 fluctuations for more distant sites (Grenoble and Nice, 205 km apart), that are located in different climatic regions
218 (Alpine for Grenoble, Mediterranean for Nice). This is contrasting with results from the rural background site of
219 OPE-ANDRA and the suburban site of Nogent-sur-Oise, both located in a large field crop region of extensive
220 agriculture, and about 230 km apart from each other (Fig. 2G). Indeed, they present very similar variations of daily
221 concentrations for multi-year series, despite their distance apart, with concentration peaks generally more
222 pronounced at the rural site of OPE-ANDRA.

223 The following sections are dedicated to the investigation of the processes that can lead to these similarities and
224 differences according to these spatial scales.
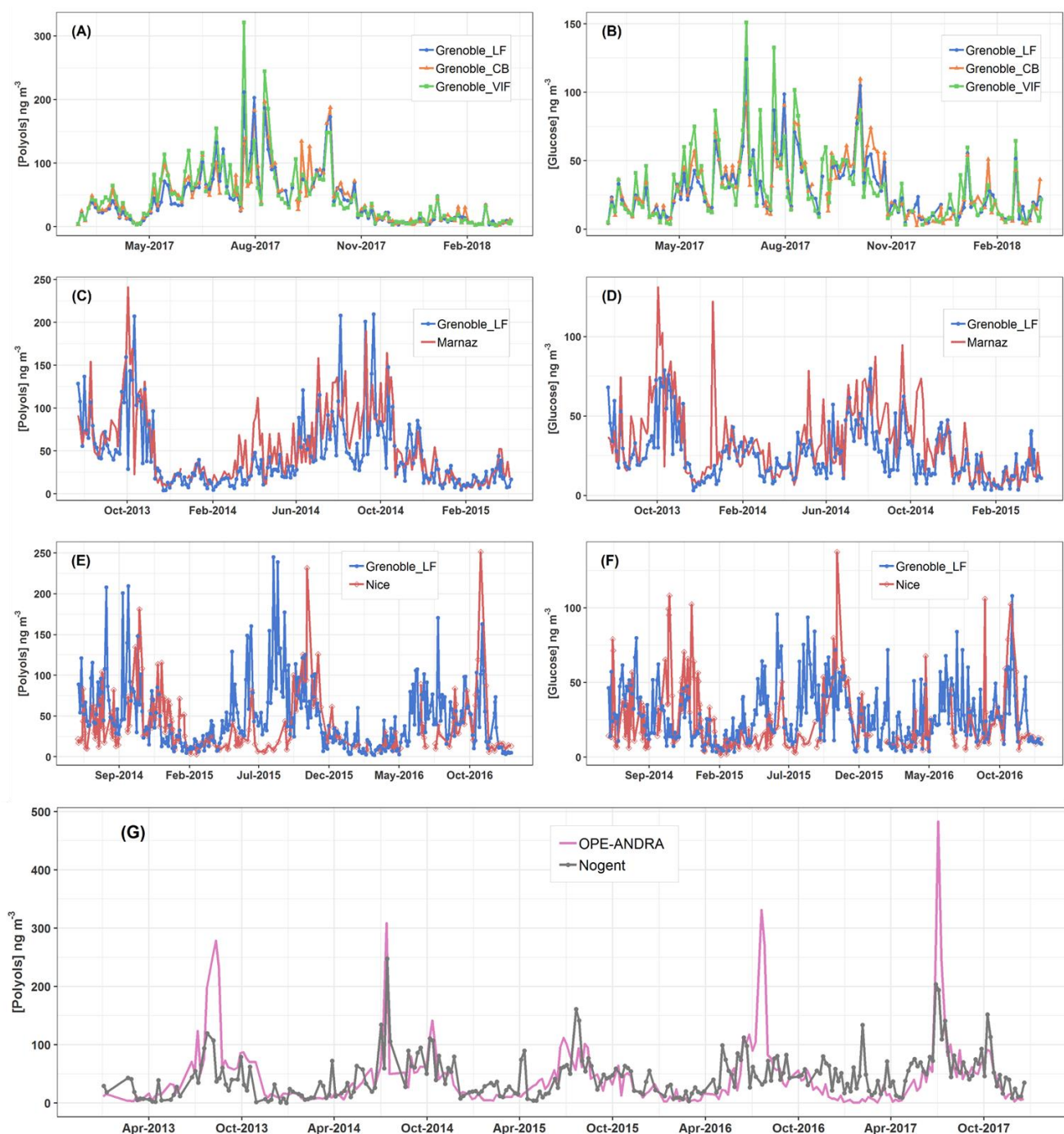
Figure 2: Concentrations (in ng m⁻³) of (left) ambient particulate polyols (defined as the sum of arabitol and mannitol) and glucose (right) over different monitoring sites in France. Since PM₁₀ were collected every 3-days at Nogent-sur-Oise and 6-days at OPE-ANDRA, the original data sets are averaged over consecutive 6-day intervals (bottom graph).

3.2 Inter-site correlations and spatial scale variability

Figures 3A and 3B provide an overview of the cross-correlation coefficients for the daily evolution of concentrations (for polyols and glucose (SC)) between pairs of sites located at multiple increasing space scales across France (Table S3). Time series of concentrations for both SC show a clear distance-dependent correlation. The strength of the correlations is highly significant for distances up to 150-190 km (R > 0.72, p < 0.01) and gradually decreases with increasing inter-site distances. One exception is the pair OPE-ANDRA and Nogent-sur-

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236 Oise (high correlation for a distance above 230 km), both sites being located in highly-impacted agricultural areas.

237 This overall pattern suggests that the processes responsible for the atmospheric concentrations of SC present a

238 spatial homogeneity over typical areas of at least several tens of km.
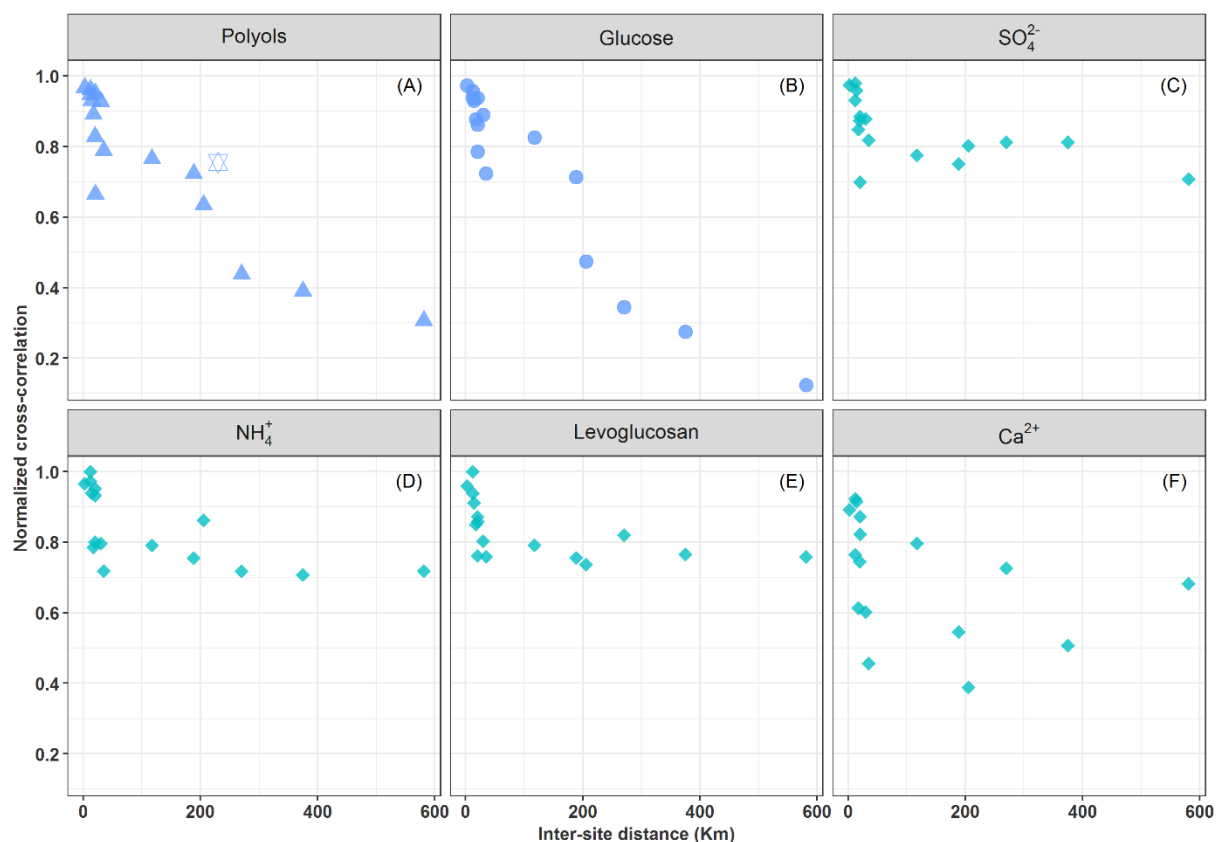
239

240 **Figure 3: Normalized cross-correlation values for the daily evolution of particulate polyols (A), glucose (B), sulfate (C),**
241 **ammonium (D), levoglucosan (E) and calcium (F) concentrations over pairs of sites located at multiple increasing space**
242 **scales across France. The hexagram corresponds to the correlation between the sites of OPE-ANDRA and Nogent-sur-**
243 **Oise, both sites being surrounded by crop field areas.**

244 Unlike SC, ambient air concentrations of sulfate (Fig. 3C) and ammonium (Fig. 3D), associated with long-range

245 aerosol transport (Abdalmogith and Harrison, 2005; Amato et al., 2016; Coulibaly et al., 2015; Pindado and Perez,

246 2011; Waked et al., 2014) and levoglucosan ((Fig. 3E), associated with biomass burning in cold season (Weber et

247 al., 2019; Xiao et al., 2018), display stronger positive correlations ($R > 0.72$-0.98, $p < 0.01$) at all pairs of sites

248 considered in the present work. Moreover, ambient concentrations of calcium (Fig. 3F), associated with local

249 fugitive dust sources or/and long-range aerosol transport (Ram et al., 2010; Wan et al., 2019) display random

250 correlation patterns. These results are in agreement with Zhu et al. (2018) who also reported non-significant

251 correlations between SC and sulfate in $PM_{2.5}$ aerosols measured at Shanghai, China. The distinct spatial behaviors

252 between sulfate (or Ca^{2+}) and SC in the present work further suggest a dominant regional influence for atmospheric

253 SC, as opposed to processes associated with either local sources for calcium or long-range transport for sulfate.

254 Mannitol and arabitol are well-known materials of fungal spores, serving as osmo-regulatory solutes (Medeiros et

255 al., 2006b; Simoneit et al., 2004; Verma et al., 2018; Xiao et al., 2018; Zhang et al., 2015). Based on parallel

256 measurements of spore counts and PM_{10} polyol concentrations at three sites within the area of Vienna (Austria),

257 Bauer et al. (2008a) found an average arabitol and mannitol content per fungal spores of respectively 1.2 pg spore[-1]

258 (range 0.8-1.8 pg spore[-1]) and 1.7 pg spore[-1] (range 1.2-2.4 pg spore[-1]). Mannitol and arabitol have also been

259 often identified in the green algae and lower plants (Buiarelli et al., 2013; Di Filippo et al., 2013; Gosselin et al.,

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260 2016; Vélëz et al., 2007; Xu et al., 2018; Zhang et al., 2010). Gosselin et al., 2016 observed a relatively low (R^2 =
261 0.31) to high (R^2 = 0.84) coefficient of determination between mannitol and arabitol for total suspended particles
262 (TSP) collected at a pine-forested area during dry and rainy periods, respectively. High correlation in rainy periods
263 possibly suggested that both chemical species in the TSP fraction in this pine-forested area could have been derived
264 mainly from the same sources, i.e., actively wet-discharged ascospores and basidiospores, while the relatively poor
265 correlation in dry periods could have been likely due to more complex sources, i.e., dry discharged spores, plants,
266 algae, etc. Being important chemical species for the metabolism of the microorganisms (Shcherbakova, 2007), it
267 may well be that the concentration ratio of mannitol-to-arabitol could deliver some information on the spatial or
268 temporal evolution of their emission processes (Gosselin et al., 2016). The annual average mannitol-to-arabitol
269 ratio at all sites is about 1.15 ± 0.59, with ratios for the warm period (Jun-Sept) being 1 to 2 times higher than
270 those in the cold period (Dec-May) (Table S1). These ratios are within the range of those previously reported for
271 PM_{10} aerosols collected at various urban and rural background sites in Europe (Bauer et al., 2008; Yttri et al.,
272 2011b). Similarly, Burshtein et al. (2011) also reported comparable ratios for PM_{10} aerosols collected during
273 autumn and winter from a Mediterranean region in Israel.

274 Similarly, the annual average glucose-to-polyols ratio at all sites is about 0.79 ± 0.77. No literature data are
275 currently available for comparison. Further work is needed to relate these variations with microorganism
276 communities and plant growing stages.

277 However, as evidenced in Fig. 4, both mannitol-to-arabitol and glucose-to-polyols ratios show a clear distance-
278 dependent correlation, with higher correlations (R = 0.64 to 0.98, p < 0.01) observed for pairs of sites within 150-
279 190 km distance. This spatial consistency highlights once again that the dominant emission processes should be
280 effective regionally, rather than being specific local input processes, and that atmospheric dynamics of the
281 concentration levels (i.e., driven by the interplay of emission and removal processes) are determined by quite
282 similar environmental factors (e.g. meteorological conditions, vegetation, land use, etc.) at such a regional scale.
283 This implies that local events and phenomena, such as the mechanical resuspension of topsoil and associated biota
284 (like bacteria, fungi, plant materials, etc.) might not be their major atmospheric input processes, particularly in
285 urban background areas typically characterized by less bare soil, and with a variable nature of the unpaved topsoil
286 at the regional scale (Karimi et al., 2018). Furthermore, Karimi et al. (2018) also recently reported heterogeneous
287 topsoil microbial structure within patches of 43 to 260 km across different regions of France. It follows that the
288 hypotheses of emissions related to mechanical resuspension of topsoil particles and associated biota, or microbiota
289 emitted actively from surface soil into the air generally assumed in most pioneering reports (Medeiros et al., 2006b;
290 Rogge et al., 2007; Simoneit et al., 2004; Wan and Yu, 2007) are most probably not valid.

291 Alternatively, the vegetation leaves have also been suggested as sources of atmospheric SC (Bozzetti et al., 2016;
292 Golly et al., 2018; Jia et al., 2010; Myriokefalitakis et al., 2017; Pashynska et al., 2002; Sullivan et al., 2011;
293 Verma et al., 2018; Wan et al., 2019). In fact, vascular plant leaf surfaces is an important habitat for endophytic
294 and epiphytic microbial communities (Kembel and Mueller, 2014; Lindow and Brandl, 2003; Lymperopoulou et
295 al., 2016; Mhuireach et al., 2016; Whipps et al., 2008). Our results are more in agreement with a dominant
296 atmosphere entrance process closely linked to vegetation, which is more homogeneous than topsoil at the climatic
297 regional scale. Consistent with this, Sullivan et al. (2011) also observed evident distinct regional patterns for daily
298 $PM_{2.5}$ polyols and glucose concentrations at ten urban and rural sites located in the upper Midwest (USA). The
299 authors attributed such a spatial pattern to the differences in vegetation types and microbial diversity over distinct

geographical regions. Accordingly, the vegetation structure and composition have been previously shown to play essential roles on airborne microbial variabilities in nearby areas (Bowers et al., 2011; Laforest-Lapointe et al., 2017; Lymperopoulou et al., 2016; Mhuireach et al., 2016).
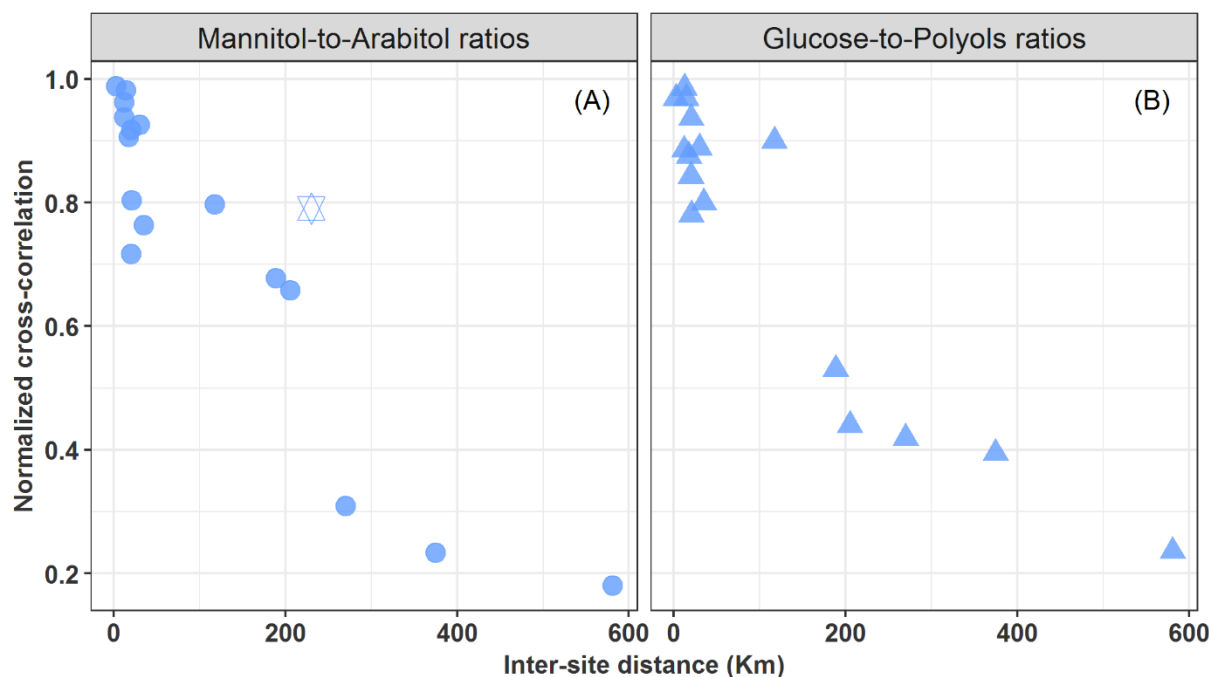
Figure 4: Normalized cross-correlation values for da (A) olution of particulate mannitol-to-arabitol (A (B) se-to-polyols (B) and ratios over pairs of sites located at multiple increasing space scales across France. The hexagram corresponds to the correlation between the sites of OPE-ANDRA and Nogent-sur-Oise, both sites being surrounded by crop field areas.

3.3 Influence of the vegetation on polyols and glucose concentrations

The relationships between SC PM_{10} concentrations and vegetation (plant materials) can be examined at the site of Grenoble Les Frênes (Grenoble_LF) by comparing the annual evolutions of SC and the free atmospheric cellulose concentrations, together with LAI ones.

The daily ambient concentration levels of SC and cellulose range respectively from 5.0 to 301.9 ng m^{-3} (with an average of 41.2 ± 39.9 ng m^{-3}) and 0.7 to 207.2 ng m^{-3} (with an average of 52.9 ± 44.2 ng m^{-3}), which correspond to respectively to 0.1 to 6.6 % and 0.01 to 5.3 % of total organic matter (OM) mass in PM_{10}. These values are comparable to those previously reported for various sites in Europe (Daellenbach et al., 2017; Sánchez-Ochoa et al., 2007; Vlachou et al., 2018; Yttri et al., 2011b). Thus, a major part of PBOA could possibly be ascribed cellulose and SC derived sources.

As evidenced in Fig. 5A, ambient free cellulose concentrations vary seasonally, with maximum seasonal average values observed in summer (81.4 ± 47.6 ng m^{-3}) and autumn (64.2 ± 49.2 ng m^{-3}), followed by spring (52.6 ± 37.8 ng m^{-3}), and lower levels in winter (23.0 ± 19.9 ng m^{-3}). This is the same global pattern for polyols, that are also more abundant in summer (82.4 ± 47.4 ng m^{-3}) and autumn (48.7 ± 41.6 ng m^{-3}), followed by spring (24.9 ± 16.3 ng m^{-3}), and winter (10.2 ± 9.6 ng m^{-3}) in the Grenoble area. On a daily scale, the episodic increases or decreases of polyols in PM_{10} are very often well synchronized with that of cellulose (Fig. 5A). Moreover, the maximum atmospheric concentrations of polyols also mainly occur when the vegetation density (LAI) is at its highest in late summer (Fig. 5B). Similar global behaviors are also observed for atmospheric particulate glucose

10

and LAI (Figs. 5A and B). To further assess the relationships between SC PM_{10} concentrations and vegetation at a rural area, a two-year measurement of cellulose concentrations at the highly-impacted agricultural rural site of OPE-ANDRA has been conducted. The average concentration of cellulose at OPE-ANDRA (197.9 ± 217.8 ng m^{-3}) is 3.5 times higher than that measured in the urban area of Grenoble. In terms of temporal dynamics, the evolution cycles (i.e., peaks and decreases) of both polyols and glucose are also very often well synchronized with that of cellulose at OPE-ANDRA (Fig. 5C).

Altogether, these findings highlight that SC in PM_{10} and cellulose in both urban background and rural agricultural areas most probably share a common source related to the vegetation. This is an additional evidence in support of the hypothesis suggested in previous studies (Bozzetti et al., 2016; Burshtein et al., 2011; Daellenbach et al., 2017; Pashynska et al., 2002; Verma et al., 2018; Vlachou et al., 2018; Wan and Yu, 2007; Yttri et al., 2007). It is also in line with studies indicating that the PBOA source profile identified using offline aerosol mass spectrometry (offline-AMS) correlates very well with coarse cellulose concentrations (Bozzetti et al., 2016; Vlachou et al., 2018). Noticeable contribution of cellulose to PBOA mass (26 %) at the rural background site of Payerne (Switzerland), during summer 2012 and winter 2013, was reported by Bozzetti et al. (2016).

As also evidenced in Fig. 5, the cellulose concentration peaks are not systematically correlated to those of polyols. The development stage of the plants (developing or mature leaves, flowering plants) in addition to the metabolic activities of endophytic and epiphytic biota (growth, sporulation), all closely related to meteorological conditions (Bodenhausen et al., 2014; Bringel and Couée, 2015; Lindow and Brandl, 2003; Pirttilä and Frank, 2011; Reddy et al., 2017), could explain such observations. The influence of local meteorological conditions for an urban Alp valley site is discussed in Section 3.4. Consistent with our observations, previous studies conducted at various urban background sites in Europe have suggested that particulate polyols are associated to mature plant leaves and microorganisms (bacterial and fungal spores) while glucose, which is a monomer of cellulose, would most likely be linked to the developing leaves (Bozzetti et al., 2016; Burshtein et al., 2011; Pashynska et al., 2002; Yttri et al., 2007; Zhu et al., 2015).
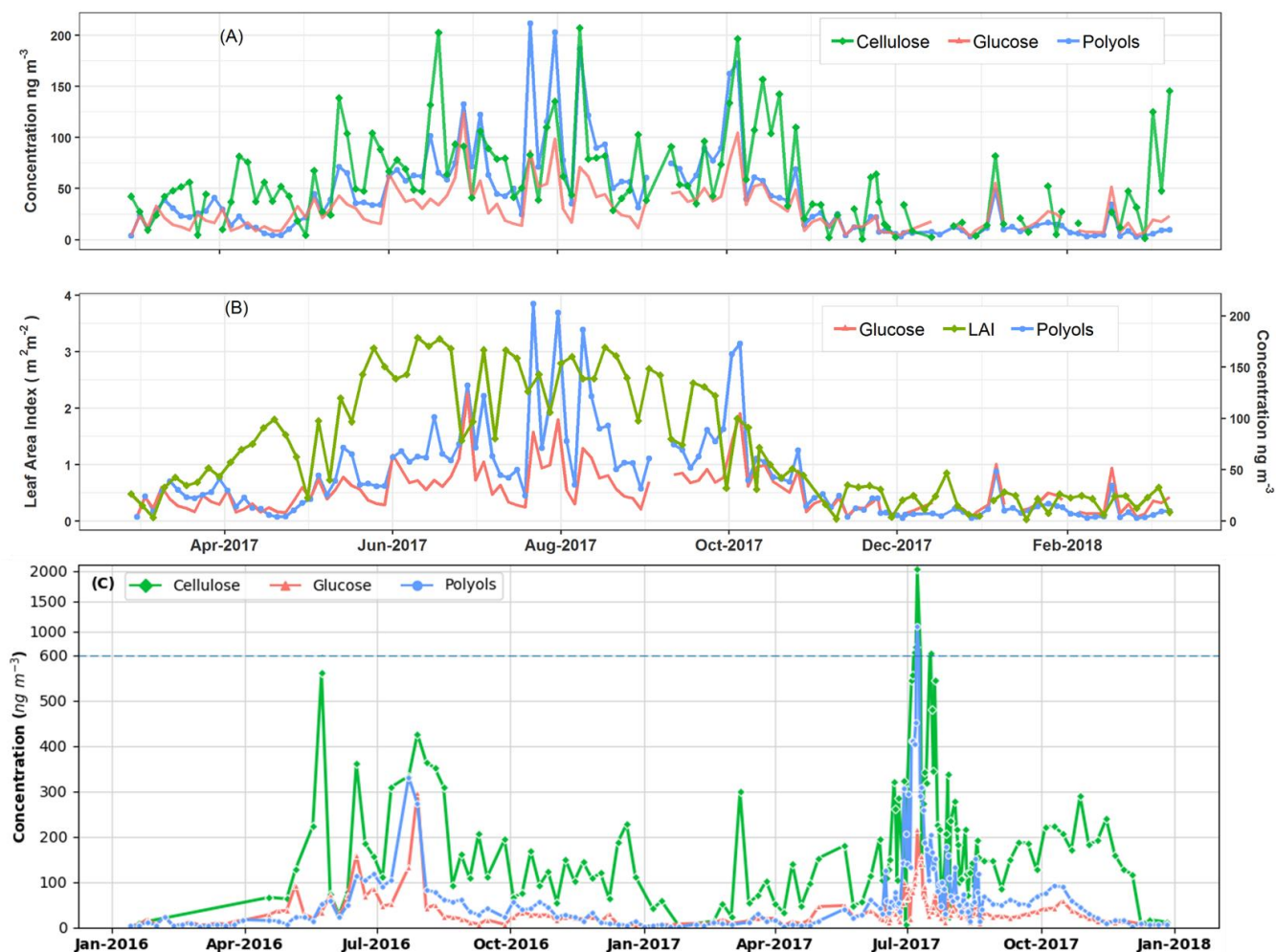
Figure 5: Temporal covariation cycles of the daily particulate polyols and glucose concentrations along with vegetation indicators at the urban background site of Grenoble (A and B) and the rural agricultural background site of OPE-ANDRA (C), respectively. Note that PM$_{10}$ aerosols are intensively collected at OPE-ANDRA every day (24-h) from 12 June 2017 to 22 August 2017, and that the concentration scale is changing above 600 ng m^{-3} in Figure C, due to extreme concentration peak in July 2017.

3.4 Influence of meteorological parameters on ambient concentrations of polyols and glucose

We used here a multiple linear regression analysis (MLR) approach to gain further insight about the environmental factors influencing the annual and short time variation cycles of atmospheric SC concentrations. This tentative MLR analysis is focused on the urban background site of Marnaz only since meteorological and other data are readily available for this site and are not influenced too much by some large city effects. Several variables were tested, that are already mentioned in the literature as drivers of SC concentrations. It includes the ambient relative humidity, rainfall level, wind speed, solar radiation, night-time temperature, average (or maximum) temperature, and LAI. Night-time temperature was selected since the time series in Marnaz and Grenoble indicate that the major drop of concentrations in late fall (Fig. 2C) is related to the first night of the season with night-time temperature below 5°C. The use of the night-temperature is also consistent with the bi-modal distribution of polyols during night and day time found in previous studies (Claeys et al., 2004; Graham et al., 2003; Yan et al., 2019; Yttri et al., 2011a).

370 Overall, the environmental factors including the mean night-time temperature, relative humidity, wind speed and

371 the leaf area index explain up to 82 % (adjusted $R^2 = 0.82$, see Table 1) of the annual temporal variation cycles of

372 SC concentrations. The mean night-time temperature and LAI contribute respectively to 54 % and 37 % of the

373 observed annual variabilities of SC concentrations. The atmospheric humidity is also a driver for these chemical

374 species (3 % of the explained variation). These results are consistent with previous studies showing that

375 concentrations of mannitol (in both PM_{10} and $PM_{2.5}$ size fractions) linearly correlate best with the LAI, atmospheric

376 water vapor and temperature (Heald and Spracklen, 2009; Hummel et al., 2015; Myriokefalitakis et al., 2017). All

377 of these drivers have been previously shown to induce the initial release and influence the long-term airborne

378 microbial (i.e. bacteria, fungi) concentrations (China et al., 2016; Elbert et al., 2007; Grinn-Gofroń et al., 2019;

379 Jones and Harrison, 2004; Rathnayake et al., 2017; Zhang et al., 2015).

380 Besides, the wind speed (range of 0.2 to 5.6 m s^{-1}) seems an additional effective driver affecting the contribution

381 of the local vegetation to SC concentrations in the atmosphere. Albeit enough air movement is required to passively

382 release microorganisms along with plant debris into the atmosphere, strong air motions induce higher dispersion.

383 These observations are in good agreement with those previously reported (Jones and Harrison, 2004; Liang et al.,

384 2013; Zhang et al., 2010, 2015; Zhu et al., 2018b). For instance Liang et al. (2013) have found a negative

385 correlation between wind speed and polyols concentrations, and the highest atmospheric fungal spores

386 concentrations were observed for a wind speed range of 0.6 to 1.0 m s^{-1}.

387 **Table 1: Multiple linear regression for ambient polyols and glucose concentrations and their effective environmental**
388 **factors at the Marnaz site. Contributions of predictor are normalized to sum 1. "Relaimpo package under R" was**
389 **used to compute bootstrap confidence intervals for importance of effective predictors (n=1000) (Grömping, 2006).**

	Dependent variable	*Variability explained by effective predictors*
	log(Polyols + Glucose)	
Night-time temperature (°C)	0.112*** (0.090, 0.133)	0.538 (0.453, 0.604)
Relative Humidity (%)	0.017*** (0.005, 0.030)	0.030 (0.018, 0.067)
Leaf Area Index	0.386** (0.034, 0.737)	0.372 (0.286, 0.444)
Wind speed (m s^{-1})	0.226 (-0.203, 0.655)	0.021 (0.015, 0.058)
Leaf Area Index × Wind Speed[a]	-0.596*** (-1.001, -0.191)	0.039 (0.014, 0.085)
Constant	2.023*** (0.787, 3.260)	
Observations	87	
R^2	0.837	
Adjusted R^2	0.824	
Residual Std. Error	0.297 (df = 81)	
F Statistic	66.677*** (df = 5; 81)	
Note	**p < 0.01; ***p < 0.001	[a] stands for interaction between predictors

390 .

391 One of the limitations of this study is that 4-day averaged observations do not allow to evaluate the driver

392 contributions that might explain some short term events for which the influence of meteorological parameters such

393 as rainfall or solar radiation could also be significant (Grinn-Gofroń et al., 2019; Heald and Spracklen, 2009; Jones

394 and Harrison, 2004). However, such simple parameterizations could be a first step in considering SC

395 concentrations in CTM models, and further work is required in this direction in order to generate a robust

396 parametrization of the emissions.

397 **3.5 Specific case of a highly-impacted agricultural area**

398 This section focuses on evidencing the environmental drivers of PM_{10} SC concentrations specific to agricultural

399 areas. To achieve this objective, the site of OPE-ANDRA has been selected because it is extensively impacted by

agricultural activities, without being too prone to influences by other sources. OPE-ANDRA is a specific rural background observatory located at about 230 km east of Paris at an altitude of 392 m. It is characterized by a low population density (< 22 inhabitants km^{-2} within an area of 900 km^2), with no surrounding major transport road or industrial activities. The air monitoring site itself lies in a "reference sector" of 240 km^2, in the middle of a field crop area (tens of kilometers in all directions). The daily agricultural practices within this reference sector are recorded and made available by ANDRA. The parcels within the agricultural area are submitted to a 3-year crop-rotation system. The major crops are wheat, barley, rape, pea and sunflower. Additionally, OPE-ANDRA is also characterized by a homogeneous type of soil, with a predominance of superficial clay-limestone.

Figure 6 shows the daily evolution of polyols concentrations in the PM$_{10}$ fraction at OPE-ANDRA from 2012 to 2018, together with the agricultural activities recorded daily and averaged over 12 days.

Although the concentration of polyols fluctuates from a year to another, they display clear annual variation cycles, with higher values in the warm periods (Jun. to Nov.) and lower concentration values in the cold periods (Oct. to May). Interestingly, the annual concentrations of polyols in 2015 (4.2-111.7 ng m^{-3}; annual average: 37.0 ± 29.1 ng m^{-3}) are significantly lower than those observed for the other years (0.6-1084.6 ng m^{-3}; annual average: 62.9 ± 96.8 ng m^{-3}). Similar inter-annual evolution trends, but with variable intensities, are also observed for glucose concentrations (Fig. 6). Year 2015 has been found to be particularly hot and dry at OPE-ANDRA (Fig. 7) whereas the local averaged wind conditions are quite stable over the years within the period of study, suggesting that the wind conditions are not the main driver of the observed inter-annual variability. These results highlight that ambient air temperature and humidity are key meteorological drivers of the annual variation cycles of polyols and glucose concentrations. Hot and dry ambient air conditions may decrease the metabolic activity of the microorganisms (e.g. microbial growth and sporulation) (Fang et al., 2018; Liang et al., 2013; Meisner et al., 2018). Finally, maximum ambient concentration levels for both SC and cellulose are observed in excellent temporal agreement with the harvest periods (late summer) at the OPE-ANDRA site (Fig. 6). Harvesting activities have been previously reported as the major sources for particulate polyols and glucose to the atmosphere in agricultural and nearby urbanized areas (Golly et al., 2018; Rogge et al., 2007; Simoneit et al., 2004). Hence, the resuspension of plant materials (crop detritus, leaves debris) and associated microbiota (e.g., bacteria, fungi) originating from cultivated lands are most-likely major input processes of PM$_{10}$ polyols and glucose at field crop sites.
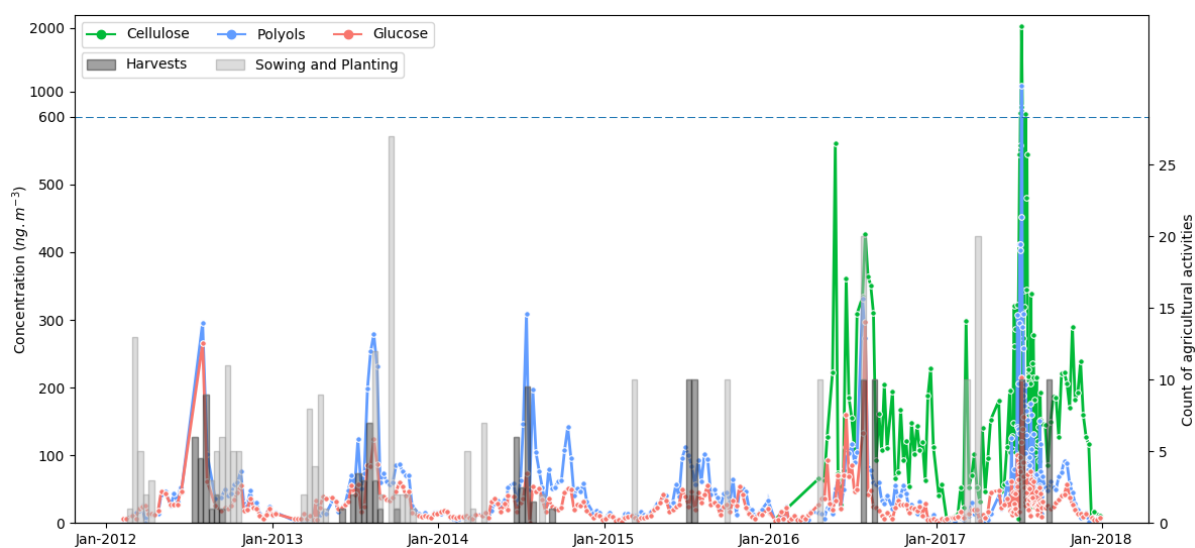
Figure 6: Daily evolution cycles of polyols and glucose concentrations in aerosols collected from the OPE-ANDRA monitoring site, from 2012 to 2018. Cellulose concentrations have been measured from January 2016 to January 2018.

Colored bars correspond to the sum of the various agricultural practices performed (data for 69 parcels are averaged over 12 days for better clarity). Records of agricultural activities after October 2014 were available for only two parcels within the immediate vicinity of the PM_{10} sampler. Records are multiplied by 10 for this period.
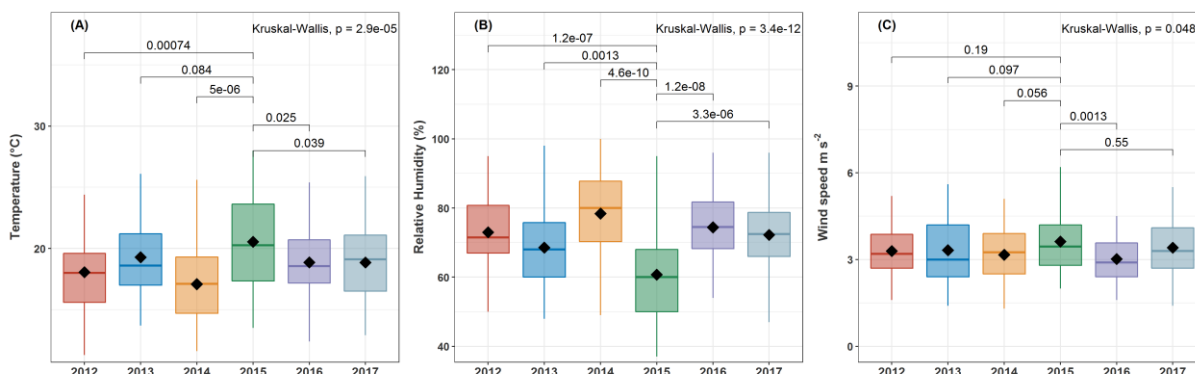
Figure 7: Boxplots of (A) maximum ambient temperature, (B) relative humidity and (C) wind speed at OPE-ANDRA from 2012 to 2017. Analyses are performed for warmer periods (June to November). Only statistically different meteorological factors are presented. The black marker inside each boxplot indicates the average value, while the top, middle and bottom of the box represent the 75th, median and 25th percentiles, respectively. The whiskers at the top and bottom of the box extend from the 95th to the 5th percentiles. Statistical differences between average values were assessed with the Kruskall-Wallis method ($p < 0.05$).

4. Conclusions

The short-term temporal (daily) and spatial (local to nation-wide) evolutions of particulate polyols (defined here as the sum of arabitol and mannitol) and glucose concentrations are rarely discussed in the current literature. The present work aimed at investigating the spatial behavior of these chemicals and evidencing their major effective environmental drivers. The major results mainly showed that:

- The short-term evolution of ambient polyols and glucose concentrations is highly synchronous across an urban city-scale and remains very well correlated throughout the same geographic areas of France, even if the monitoring sites are situated in different cities at about 150-190 km. However, sampling sites located in two distinct geographic areas are poorly correlated. This indicates that emission sources of these chemicals are uniformly distributed, and their accumulation and removal processes are driven by quite similar environmental parameters at the regional scale. Therefore, local phenomena such as atmospheric resuspension of topsoil particles and associated microbiota, microbial direct emissions (e.g. sporulation), cannot be the main emission processes of particulate polyols and glucose in urban areas not directly influenced by agricultural activities.

- The atmospheric concentrations of polyols (or glucose) and cellulose display remarkably synchronous temporal evolution cycles at the background urban site of Grenoble, indicating a common source related to plant debris.

- Higher ambient concentrations of polyols and glucose at the rural site of OPE-ANDRA occur during each harvest period, pointing out resuspension processes of plant materials (crop detritus, leaves debris) and associated microbiota for agricultural and nearby urbanized areas. This is associated with higher PM_{10} cellulose concentration levels, as high as 0.4 to 2.0 µg m^{-3} on a daily basis (accounting up to 7.5 to 32.4 % of the OM mass).

462　　　• Multiple linear regression analysis of the yearly series from the site of Marnaz gave insightful information
463　　　　on which parameter controls the ambient concentrations of polyols and glucose. Ambient air night-time
464　　　　temperature, relative humidity and vegetation density are the most important drivers, whilst wind speed
465　　　　conditions tend to affect the contribution of local vegetation.

466 Altogether, these results improve our understanding of the spatial behavior tracers of PM_{10} PBOA emission sources
467 in France, and in general, which is imperative for further implementation of this important mass fraction of OM
468 into chemical transport models. Further investigations of airborne microbial fingerprint (bacteria and fungi) are
469 ongoing, which may deepen our understanding of the PBOA source profile.

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485 BG, and AW. He directed all the personnel who performed the analysis at IGE. He is the coordinator for the CNRS
486 LEFE-EC2CO CAREMBIOS program that is funding the work of AS. GU and JMF-M were the co-supervisor for
487 the PhD of AS or SW. EP, OF, and VR supervised the PhD of DMO who investigated the sites in northern France.
488 OF, JL-J, JL-B, AA and NM were coordinating and partners of the different initial programs for the collection and
489 chemical analysis of the samples. VJ developed the analytical techniques for polyols and cellulose measurements.
490 TC performed the cellulose measurements. Samples analyses at LSCE were performed by NB. AC gave advices
491 for the statistical aspects of the data processing. AS and JLJ processed the data and wrote up the manuscript. SW
492 participated to the visualization of the results. SC is supervising the OPE station and collected the agricultural
493 activities records. All authors from AASQA (author affiliation nos. 9 to 16) are representatives for each network
494 that conducted the sample collection and the general supervision of the sampling sites. All authors reviewed and
495 commented on the manuscript.

496 **Competing interests:** The authors declare that they have no conflict of interest.

497 **References**

498 Abdalmogith, S. S. and Harrison, R. M.: The use of trajectory cluster analysis to examine the long-range transport
499 of secondary inorganic aerosol in the UK, Atmos. Environ., 39(35), 6686–6695,
500 doi:10.1016/j.atmosenv.2005.07.059, 2005.

501 Amato, F., Alastuey, A., Karanasiou, A., Lucarelli, F., Nava, S., Calzolai, G., Severi, M., Becagli, S., Gianelle, V.
502 L., Colombi, C., Alves, C., Custódio, D., Nunes, T., Cerqueira, M., Pio, C., Eleftheriadis, K., Diapouli, E., Reche,
503 C., Minguillón, M. C., Manousakas, M.-I., Maggos, T., Vratolis, S., Harrison, R. M., and Querol, X.: Airuse-life+:
504 a harmonized PM speciation and source apportionment in five southern European cities, Atmos. Chem. Phys.,
505 16(5), 3289–3309, doi:10.5194/acp-16-3289-2016, 2016.

506 Amato, P., Brisebois, E., Draghi, M., Duchaine, C., Fröhlich-Nowoisky, J., Huffman, J. A., Mainelis, G., Robine,
507 E., and Thibaudon, M.: Main biological aerosols, specificities, abundance, and diversity, in Microbiology of
508 Aerosols, pp. 1–21, John Wiley & Sons, Ltd., doi:10.1002/9781119132318, 2017.

509 Ariya, P. A., Sun, J., Eltouny, N. A., Hudson, E. D., Hayes, C. T., and Kos, G.: Physical and chemical
510 characterization of bioaerosols – implications for nucleation processes, Int. Rev. Phys. Chem., 28(1), 1–32,
511 doi:10.1080/01442350802597438, 2009.

512 Barbaro, E., Kirchgeorg, T., Zangrando, R., Vecchiato, M., Piazza, R., Barbante, C., and Gambaro, A.: Sugars in
513 Antarctic aerosol, Atmos. Environ., 118, 135–144, doi:10.1016/j.atmosenv.2015.07.047, 2015.

514 Bardal, L. M. and Sætran, L. R.: Spatial correlation of atmospheric wind at scales relevant for large scale wind
515 turbines, J. Phys. Conf. Ser., 753, 032033, doi:10.1088/1742-6596/753/3/032033, 2016.

516 Bauer, H., Claeys, M., Vermeylen, R., Schueller, E., Weinke, G., Berger, A., and Puxbaum, H.: Arabitol and
517 mannitol as tracers for the quantification of airborne fungal spores, Atmos. Environ., 42(3), 588–593,
518 doi:10.1016/j.atmosenv.2007.10.013, 2008.

519 Bodenhausen, N., Bortfeld-Miller, M., Ackermann, M., and Vorholt, J. A.: A synthetic community approach
520 reveals plant genotypes affecting the phyllosphere microbiota, PLoS Genet., 10(4), doi:
521 10.1371/journal.pgen.1004283, 2014.

522 Bowers, R. M., Sullivan, A. P., Costello, E. K., Collett, J. L., Knight, R., and Fiereri, N.: Sources of bacteria in
523 outdoor air across cities in the midwestern United States., Appl. Environ. Microbiol., 77(18), 6350–6356,
524 doi:10.1128/AEM.05498-11, 2011.

525 Bozzetti, C., Daellenbach, K. R., Hueglin, C., Fermo, P., Sciare, J., Kasper-Giebl, A., Mazar, Y., Abbaszade, G.,
526 El Kazzi, M., Gonzalez, R., Shuster-Meiseles, T., Flasch, M., Wolf, R., Křepelová, A., Canonaco, F., Schnelle-
527 Kreis, J., Slowik, J. G., Zimmermann, R., Rudich, Y., Baltensperger, U., El Haddad, I., and Prévôt, A. S. H.: Size-
528 resolved identification, characterization, and quantification of primary biological organic aerosol at a European
529 rural site, Environ. Sci. Technol., 50(7), 3425–3434, doi:10.1021/acs.est.5b05960, 2016.

530 Bringel, F. and Couée, I.: Pivotal roles of phyllosphere microorganisms at the interface between plant functioning
531 and atmospheric trace gas dynamics, Front. Microbiol., 6, 486, doi:10.3389/fmicb.2015.00486, 2015.

532 Buiarelli, F., Canepari, S., Di Filippo, P., Perrino, C., Pomata, D., Riccardi, C., and Speziale, R.: Extraction and
533 analysis of fungal spore biomarkers in atmospheric bioaerosol by HPLC–MS–MS and GC–MS, Talanta, 105, 142–
534 151, doi:10.1016/j.talanta.2012.11.006, 2013.

535 Burshtein, N., Lang-Yona, N., and Rudich, Y.: Ergosterol, arabitol and mannitol as tracers for biogenic aerosols
536 in the eastern Mediterranean, Atmospheric Chem. Phys., 11(2), 829–839, doi:10.5194/acp-11-829-2011, 2011.

537 Chen, J., Kawamura, K., Liu, C.-Q., and Fu, P.: Long-term observations of saccharides in remote marine aerosols
538 from the western North Pacific: A comparison between 1990–1993 and 2006–2009 periods, Atmos. Environ., 67,
539 448–458, doi:10.1016/j.atmosenv.2012.11.014, 2013.

540 China, S., Wang, B., Weis, J., Rizzo, L., Brito, J., Cirino, G. G., Kovarik, L., Artaxo, P., Gilles, M. K., and Laskin,
541 A.: Rupturing of biological spores as a source of secondary particles in Amazonia, Environ. Sci. Technol., 50(22),
542 12179–12186, 2016.

543 China, S., Burrows, S. M., Wang, B., Harder, T. H., Weis, J., Tanarhte, M., Rizzo, L. V., Brito, J., Cirino, G. G.,
544 Ma, P.-L., Cliff, J., Artaxo, P., Gilles, M. K. and Laskin, A.: Fungal spores as a source of sodium salt particles in
545 the Amazon basin, Nat. Commun., 9(1), doi:10.1038/s41467-018-07066-4, 2018.

546 Claeys, M., Graham, B., Vas, G., Wang, W., Vermeylen, R., Pashynska, V., Cafmeyer, J., Guyon, P., Andreae, M.
547 O., Artaxo, P., and Maenhaut, W.: Formation of secondary organic aerosols through photooxidation of isoprene,
548 Science, 303(5661), 1173, doi:10.1126/science.1092805, 2004.

549 Coulibaly, S., Minami, H., Abe, M., Hasei, T., Sera, N., Yamamoto, S., Funasaka, K., Asakawa, D., Watanabe,
550 M., Honda, N., Wakabayashi, K., and Watanabe, T.: Seasonal fluctuations in air pollution in Dazaifu, Japan, and
551 effect of long-range transport from mainland east Asia, Biol. Pharm. Bull., 38(9), 1395–1403,
552 doi:10.1248/bpb.b15-00443, 2015.

553   Coz, E., Artíñano, B., Clark, L. M., Hernandez, M., Robinson, A. L., Casuccio, G. S., Lersch, T. L., and Pandis,
554   S. N.: Characterization of fine primary biogenic organic aerosol in an urban area in the northeastern United States,
555   Atmos. Environ., 44(32), 3952–3962, 2010.

556   Daellenbach, K. R., Stefenelli, G., Bozzetti, C., Vlachou, A., Fermo, P., Gonzalez, R., Piazzalunga, A., Colombi,
557   C., Canonaco, F., Hueglin, C., Kasper-Giebl, A., Jaffrezo, J.-L., Bianchi, F., Slowik, J. G., Baltensperger, U., El-
558   Haddad, I., and Prévôt, A. S. H.: Long-term chemical analysis and organic aerosol source apportionment at nine
559   sites in central Europe: source identification and uncertainty assessment, Atmos. Chem. Phys., 17(21), 13265–
560   13282, doi:10.5194/acp-17-13265-2017, 2017.

561   Dai, Y.-H. and Zhou, W.-X.: Temporal and spatial correlation patterns of air pollutants in Chinese cities, PLoS
562   ONE, 12(8), doi:10.1371/journal.pone.0182724, 2017.

563   Després, V. R., Alex Huffman, J., Burrows, S. M., Hoose, C., Safatov, A. S., Buryak, G., Fröhlich-Nowoisky, J.,
564   Elbert, W., Andreae, M. O., Pöschl, U., and Jaenicke, R.: Primary biological aerosol particles in the atmosphere:
565   a review, Tellus B., 64(1), 15598, doi:10.3402/ tellusb.v64i0.15598, 2012.

566   Di Filippo, P., Pomata, D., Riccardi, C., Buiarelli, F., and Perrino, C.: Fungal contribution to size-segregated
567   aerosol measured through biomarkers, Atmos. Environ., 64, 132–140, 2013.

568   Eisner, A. D., Richmond-Bryant, J., Hahn, I., Drake-Richman, Z. E., Brixey, L. A., Wiener, R. W., and Ellenson,
569   W. D.: Analysis of indoor air pollution trends and characterization of infiltration delay time using a cross-
570   correlation method, J. Environ. Monit., 11(12), 2201, doi:10.1039/b907144j, 2009.

571   Elbert, W., Taylor, P. E., Andreae, M. O., and Pöschl, U.: Contribution of fungi to primary biogenic aerosols in
572   the atmosphere: wet and dry discharged spores, carbohydrates, and inorganic ions, Atmos. Chem. Phys., 7(17),
573   4569–4588, doi:10.5194/acp-7-4569-2007, 2007.

574   Fang, Z., Guo, W., Zhang, J., and Lou, X.: Influence of heat Events on the composition of airborne bacterial
575   communities in urban ecosystems, Int. J. Environ. Res. Public. Health, 15(10), 2295, doi:10.3390/ijerph15102295,
576   2018.

577   Fröhlich-Nowoisky, J., Pickersgill, D. A., Després, V. R. and Pöschl, U.: High diversity of fungi in air particulate
578   matter, Proc. Natl. Acad. Sci. U. S. A., 106(31), 12814–12819, doi:10.1073/pnas.0811003106, 2009.

579   Fröhlich-Nowoisky, J., Kampf, C. J., Weber, B., Huffman, J. A., Pöhlker, C., Andreae, M. O., Lang-Yona, N.,
580   Burrows, S. M., Gunthe, S. S., Elbert, W., Su, H., Hoor, P., Thines, E., Hoffmann, T., Després, V. R., and Pöschl,
581   U.: Bioaerosols in the earth system: climate, health, and ecosystem interactions, Atmos. Res., 182, 346–376,
582   doi:10.1016/j.atmosres.2016.07.018, 2016.

583   Fröhlich-Nowoisky, J., Amato, P., Renard, P., Brisebois, E., and Duchaine, C.: Quantification and characterization
584   of bioaerosols (offline techniques), in Microbiology of Aerosols, pp. 49–82, John Wiley & Sons, Ltd., doi:
585   10.1002/9781119132318, 2017.

586   Glasius, M., Hansen, A. M. K., Claeys, M., Henzing, J. S., Jedynska, A. D., Kasper-Giebl, A., Kistler, M.,
587   Kristensen, K., Martinsson, J., Maenhaut, W., Nøjgaard, J. K., Spindler, G., Stenström, K. E., Swietlicki, E., Szidat,
588   S., Simpson, D., and Yttri, K. E.: Composition and sources of carbonaceous aerosols in northern Europe during
589   winter, Atmos. Environ., 173, 127–141, doi:10.1016/j.atmosenv.2017.11.005, 2018.

590   Golly, B., Waked, A., Weber, S., Samaké, A., Jacob, V., Conil, S., Rangognio, J., Chrétien, E., Vagnot, M.-P.,
591   Robic, P.-Y., Besombes, J.-L., and Jaffrezo, J.-L.: Organic markers and OC source apportionment for seasonal
592   variations of $PM_{2.5}$ at 5 rural sites in France, Atmos. Environ., 198, 142–157, doi:10.1016/j.atmosenv.2018.10.027,
593   2018.

594   Gosselin, M. I., Rathnayake, C. M., Crawford, I., Pöhlker, C., Fröhlich-Nowoisky, J., Schmer, B., Després, V. R.,
595   Engling, G., Gallagher, M., Stone, E., Pöschl, U., and Huffman, J. A.: Fluorescent bioaerosol particle, molecular
596   tracer, and fungal spore concentrations during dry and rainy periods in a semi-arid forest, Atmos. Chem. Phys.,
597   16(23), 15165–15184, doi:10.5194/acp-16-15165-2016, 2016.

598 Graham, B., Guyon, P., Taylor, P. E., Artaxo, P., Maenhaut, W., Glovsky, M. M., Flagan, R. C., and Andreae, M.
599 O.: Organic compounds present in the natural Amazonian aerosol: Characterization by gas chromatography-mass
600 spectrometry: organic compounds in Amazonian aerosols., J. Geophys. Res. Atmospheres, 108(D24), 4766,
601 doi:10.1029/2003JD003990, 2003.

602 Grinn-Gofroń, A., Nowosad, J., Bosiacka, B., Camacho, I., Pashley, C., Belmonte, J., De Linares, C., Ianovici, N.,
603 Manzano, J. M. M., Sadyś, M., Skjøth, C., Rodinkova, V., Tormo-Molina, R., Vokou, D., Fernández-Rodríguez,
604 S., and Damialis, A.: Airborne alternaria and Cladosporium fungal spores in Europe: forecasting possibilities and
605 relationships with meteorological parameters, Sci. Total Environ., 653, 938–946,
606 doi:10.1016/j.scitotenv.2018.10.419, 2019.

607 Heald, C. L. and Spracklen, D. V.: Atmospheric budget of primary biological aerosol particles from fungal spores,
608 Geophys. Res. Lett., 36(9), doi:10.1029/2009GL037493, 2009.

609 Hill, T. C. J., DeMott, P. J., Conen, F., and Möhler, O.: Impacts of bioaerosols on atmospheric ice nucleation
610 processes, in Microbiology of Aerosols, pp. 195–219, John Wiley & Sons, Ltd., doi:10.1002/9781119132318,
611 2017.

612 Hiranuma, N., Adachi, K., Bell, D. M., Belosi, F., Beydoun, H., Bhaduri, B., Bingemer, H., Budke, C., Clemen,
613 H.-C., Conen, F., Cory, K. M., Curtius, J., DeMott, P. J., Eppers, O., Grawe, S., Hartmann, S., Hoffmann, N.,
614 Höhler, K., Jantsch, E., Kiselev, A., Koop, T., Kulkarni, G., Mayer, A., Murakami, M., Murray, B. J., Nicosia, A.,
615 Petters, M. D., Piazza, M., Polen, M., Reicher, N., Rudich, Y., Saito, A., Santachiara, G., Schiebel, T., Schill, G.
616 P., Schneider, J., Segev, L., Stopelli, E., Sullivan, R. C., Suski, K., Szakáll, M., Tajiri, T., Taylor, H., Tobo, Y.,
617 Ullrich, R., Weber, D., Wex, H., Whale, T. F., Whiteside, C. L., Yamashita, K., Zelenyuk, A., and Möhler, O.: A
618 comprehensive characterization of ice nucleation by three different types of cellulose particles immersed in water,
619 Atmos. Chem. Phys., 19(7), 4823–4849, doi:10.5194/acp-19-4823-2019, 2019.

620 Holden, A. S., Sullivan, A. P., Munchak, L. A., Kreidenweis, S. M., Schichtel, B. A., Malm, W. C., and Collett, J.
621 L.: Determining contributions of biomass burning and other sources to fine particle contemporary carbon in the
622 western United States, Atmos. Environ., 45(11), 1986–1993, doi:10.1016/j.atmosenv.2011.01.021, 2011.

623 Huffman, J. A. and Santarpia, J.: Online Techniques for quantification and characterization of biological aerosols,
624 in Microbiology of Aerosols, pp. 83–114, John Wiley & Sons, Ltd., doi:10.1002/9781119132318, 2017.

625 Humbal, C., Gautam, S. and Trivedi, U.: A review on recent progress in observations, and health effects of
626 bioaerosols, Environ. Int., 118, 189–193, doi:10.1016/j.envint.2018.05.053, 2018.

627 Hummel, M., Hoose, C., Gallagher, M., Healy, D. A., Huffman, J. A., O'Connor, D., Pöschl, U., Pöhlker, C.,
628 Robinson, N. H., Schnaiter, M., Sodeau, J. R., Stengel, M., Toprak, E., and Vogel, H.: Regional-scale simulations
629 of fungal spore aerosols using an emission parameterization adapted to local measurements of fluorescent
630 biological aerosol particles, Atmos. Chem. Phys., 15(11), 6127–6146, doi:10.5194/acp-15-6127-2015, 2015.

631 Jacobson, M. Z. and Streets, D. G.: Influence of future anthropogenic emissions on climate, natural emissions, and
632 air quality, J. Geophys. Res., 114(D8), D08118, doi:10.1029/2008JD011476, 2009.

633 Jaenicke, R.: Abundance of cellular material and proteins in the atmosphere, Science, 308(5718), 73–73,
634 doi:10.1126/science.1106335, 2005.

635 Jia, Y., Bhat, S., and Fraser, M. P.: Characterization of saccharides and other organic compounds in fine particles
636 and the use of saccharides to track primary biologically derived carbon sources, Atmos. Environ., 44(5), 724–732,
637 doi:10.1021/es103104e, 2010.

638 Jones, A. M. and Harrison, R. M.: The effects of meteorological factors on atmospheric bioaerosol
639 concentrations—a review, Sci. Total Environ., 326(1), 151–180, doi:10.1016/j.scitotenv.2003.11.021, 2004.

640 Kang, M., Ren, L., Ren, H., Zhao, Y., Kawamura, K., Zhang, H., Wei, L., Sun, Y., Wang, Z., and Fu, P.: Primary
641 biogenic and anthropogenic sources of organic aerosols in Beijing, China: Insights from saccharides and n-alkanes,
642 Environ. Pollut., doi:10.1016/j.envpol.2018.09.118, 2018.

Karimi, B., Terrat, S., Dequiedt, S., Saby, N. P. A., Horrigue, W., Lelièvre, M., Nowak, V., Jolivet, C., Arrouays, D., Wincker, P., Cruaud, C., Bispo, A., Maron, P.-A., Bouré, N. C. P., and Ranjard, L.: Biogeography of soil bacteria and archaea across France, Sci. Adv., 4(7), eaat1808, doi:10.1126/sciadv.aat1808, 2018.

Kaso, A.: Computation of the normalized cross-correlation by fast Fourier transform, PLOS ONE, 13(9), e0203434, doi:10.1371/journal.pone.0203434, 2018.

Kembel, S. W. and Mueller, R. C.: Plant traits and taxonomy drive host associations in tropical phyllosphere fungal communities, Botany, 92(4), 303–311, doi:10.1139/cjb-2013-0194, 2014.

Kunit, M. and Puxbaum, H.: Enzymatic determination of the cellulose content of atmospheric aerosols, Atmos. Environ., 30(8), 1233–1236, doi:10.1016/1352-2310(95)00429-7, 1996.

Laforest-Lapointe, I., Messier, C., and Kembel, S. W.: Tree leaf bacterial community structure and diversity differ along a gradient of urban intensity, mSystems, 2(6), e00087-17, doi:10.1128/mSystems.00087-17, 2017.

Lainer, M., Hocke, K., and Kämpfer, N.: Variability of mesospheric water vapor above Bern in relation to the 27-day solar rotation cycle, J. Atmos. Sol.-Terr. Phys., 143–144, 71–87, doi:10.1016/j.jastp.2016.03.008, 2016.

Le Pichon, A., Blanc, E., and Hauchecorne, A.: Infrasound monitoring for atmospheric studies, 2nd ed., Springer International Publishing, Switzerland., doi:10.1007/978-3-319-75140-5,2019.

Lecours, P. B., Duchaine, C., Thibaudon, M., and Marsolais, D.: Health impacts of bioaerosol exposure, in Microbiology of Aerosols, pp. 249–268, John Wiley & Sons, Ltd., doi:10.1002/9781119132318, 2017.

Li, L., Ren, L., Ren, H., Yue, S., Xie, Q., Zhao, W., Kang, M., Li, J., Wang, Z., Sun, Y., and Fu, P.: Molecular characterization and seasonal variation in primary and secondary organic aerosols in Beijing, China, J. Geophys. Res. Atmospheres, 123(21), 12,394-12,412, doi:10.1029/2018JD028527, 2018.

Liang, L., Engling, G., He, K., Du, Z., Cheng, Y., and Duan, F.: Evaluation of fungal spore characteristics in Beijing, China, based on molecular tracer measurements, Environ. Res. Lett., 8(1), 014005, doi:10.1088/1748-9326/8/1/014005, 2013.

Lindow, S. E. and Brandl, M. T.: Microbiology of the Phyllosphere, Appl. Environ. Microbiol., 69(4), 1875–1883, doi:10.1128/AEM.69.4.1875-1883.2003, 2003.

Lymperopoulou, D. S., Adams, R. I., and Lindow, S. E.: Contribution of vegetation to the microbial composition of nearby outdoor air, Appl. Environ. Microbiol., 82(13), 3822–3833, doi:10.1128/AEM.00610-16, 2016.

Manninen, H. E., Bäck, J., Sihto-Nissilä, S.-L., Huffman, J. A., Pessi, A.-M., Hiltunen, V., Aalto, P. P., Hidalgo Fernández, P. J., Hari, P., Saarto, A., Kulmala, M., and Petäjä, T.: Patterns in airborne pollen and other primary biological aerosol particles (PBAP), and their contribution to aerosol mass and number in a boreal forest, Boreal Environ. Res., 383–405, doi:hdl.handle.net/10138/165208, 2014.

Martin, S. T., Andreae, M. O., Artaxo, P., Baumgardner, D., Chen, Q., Goldstein, A. H., Guenther, A., Heald, C. L., Mayol-Bracero, O. L., McMurry, P. H., Pauliquevis, T., Pöschl, U., Prather, K. A., Roberts, G. C., Saleska, S. R., Silva Dias, M. A., Spracklen, D. V., Swietlicki, E., and Trebs, I.: Sources and properties of Amazonian aerosol particles, Rev. Geophys., 48(2), RG2002, doi:10.1029/2008RG000280, 2010.

Medeiros, P. M., Fernandes, M. F., Dick, R. P., and Simoneit, B. R. T.: Seasonal variations in sugar contents and microbial community in a ryegrass soil, Chemosphere, 65(5), 832–839, doi:10.1016/j.chemosphere.2006.03.025, 2006a.

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

Meisner, A., Jacquiod, S., Snoek, B. L., ten Hooven, F. C., and van der Putten, W. H.: Drought legacy effects on the composition of soil fungal and prokaryote communities, Front. Microbiol., 9, doi:10.3389/fmicb.2018.00294, 2018.

687 Mhuireach, G., Johnson, B. R., Altrichter, A. E., Ladau, J., Meadow, J. F., Pollard, K. S., and Green, J. L.: Urban
688 greenness influences airborne bacterial community composition, Sci. Total Environ., 571, 680–687,
689 doi:10.1016/j.scitotenv.2016.07.037, 2016.

690 Morris, C. E., Sands, D. C., Bardin, M., Jaenicke, R., Vogel, B., Leyronas, C., Ariya, P. A., and Psenner, R.:
691 Microbiology and atmospheric processes: research challenges concerning the impact of airborne micro-organisms
692 on the atmosphere and climate, Biogeosciences, 8(1), 17–25, doi:10.5194/bg-8-17-2011, 2011.

693 Morris, C. E., Conen, F., Alex Huffman, J., Phillips, V., Pöschl, U., and Sands, D. C.: Bioprecipitation: a feedback
694 cycle linking Earth history, ecosystem dynamics and land use through biological ice nucleators in the atmosphere,
695 Glob. Change Biol., 20(2), 341–351, doi:10.1111/gcb.12447, 2014.

696 Myriokefalitakis, S., Fanourgakis, G., and Kanakidou, M.: The Contribution of bioaerosols to the organic carbon
697 budget of the atmosphere, in Perspectives on Atmospheric Sciences, pp. 845–851, Springer International
698 Publishing., doi:10.1007/978-3-319-35095-0, 2017.

699 Nirmalkar, J., Deshmukh, D. K., Deb, M. K., Tsai, Y. I., and Pervez, S.: Characteristics of aerosol during major
700 biomass burning events over eastern central India in winter: A tracer-based approach, Atmos. Pollut. Res.,
701 doi:10.1016/j.apr.2018.12.010, 2018.

702 Pashynska, V., Vermeylen, R., Vas, G., Maenhaut, W., and Claeys, M.: Development of a gas chromatographic/ion
703 trap mass spectrometric method for the determination of levoglucosan and saccharidic compounds in atmospheric
704 aerosols. Application to urban aerosols, J. Mass Spectrom., 37(12), 1249–1257, doi:10.1002/jms.391, 2002.

705 Perrino, C. and Marcovecchio, F.: A new method for assessing the contribution of primary biological atmospheric
706 particles to the mass concentration of the atmospheric aerosol, Environ. Int., 87, 108–115,
707 doi:10.1016/j.envint.2015.11.015, 2016.

708 Pietrogrande, M. C., Bacco, D., Visentin, M., Ferrari, S., and Casali, P.: Polar organic marker compounds in
709 atmospheric aerosol in the Po valley during the supersito campaigns — part 2: seasonal variations of sugars,
710 Atmos. Environ., 97, 215–225, doi:0.1016/j.atmosenv.2014.07.056, 2014.

711 Pindado, O. and Perez, R. M.: Source apportionment of particulate organic compounds in a rural area of Spain by
712 positive matrix factorization, Atmos. Pollut. Res., 2(4), 492–505, doi:10.5094/APR.2011.056, 2011.

713 Pirttilä, A. M. and Frank, A. C., Eds.: Endophytes of forest trees, Springer Netherlands, Dordrecht.,
714 doi:10.1007/978-3-319-89833-9, 2011.

715 Puxbaum, H. and Tenze-Kunit, M.: Size distribution and seasonal variation of atmospheric cellulose, Atmos.
716 Environ., 37(26), 3693–3699, doi:10.1016/S1352-2310(03)00451-5, 2003.

717 Rajput, P., Chauhan, A. S., and Gupta, T.: Bioaerosols over the indo-gangetic plain: influence of biomass burning
718 emission and ambient meteorology, in Environmental Contaminants: measurement, modelling and control, pp.
719 93–121, Springer Singapore., doi:10.1007/978-981-10-7332-8, 2018.

720 Ram, K., Sarin, M. M., and Hegde, P.: Long-term record of aerosol optical properties and chemical composition
721 from a high-altitude site (Manora Peak) in central Himalaya, Atmos. Chem. Phys., 13, doi:10.5194/acp-10-11791-
722 2010, 2010.

723 Ramoni, J. and Seiboth, B.: Degradation of plant cell wall polymers by fungi, in Environmental and microbial
724 relationships, vol. IV, pp. 127–148, Springer International Publishing, Cham., doi: 10.1007/978-3-540-71840-6
725 2016.

726 Rathnayake, C. M., Metwali, N., Jayarathne, T., Kettler, J., Huang, Y., Thorne, P. S., O'Shaughnessy, P. T., and
727 Stone, E. A.: Influence of rain on the abundance of bioaerosols in fine and coarse particles, Atmos. Chem. Phys.,
728 17(3), 2459–2475, doi:10.5194/acp-17-2459-2017, 2017.

729 Reddy, S. M., Girisham, S., and Babu, G. N.: Applied Microbiology (agriculture, environmental, food and
730 industrial microbiology), Scientific Publishers, doi:9789387307407, 2017.

731   Rogge, W. F., Medeiros, P. M., and Simoneit, B. R. T.: Organic marker compounds in surface soils of crop fields
732   from the San Joaquin Valley fugitive dust characterization study, Atmos. Environ., 41(37), 8183–8204,
733   doi:10.1016/j.atmosenv.2007.06.030, 2007.

734   Samaké, A., Jaffrezo, J.-L., Favez, O., Weber, S., Jacob, V., Albinet, A., Riffault, V., Perdrix, E., Waked, A.,
735   Golly, B., Salameh, D., Chevrier, F., Oliveira, D. M., Bonnaire, N., Besombes, J.-L., Martins, J. M. F., Conil, S.,
736   Guillaud, G., Mesbah, B., Rocq, B., Robic, P.-Y., Hulin, A., Meur, S. L., Descheemaecker, M., Chretien, E.,
737   Marchand, N., and Uzu, G.: Polyols and glucose particulate species as tracers of primary biogenic organic aerosols
738   at 28 French sites, Atmos. Chem. Phys., 19(5), 3357–3374, doi:10.5194/acp-19-3357-2019, 2019.

739   Sánchez-Ochoa, A., Kasper-Giebl, A., Puxbaum, H., Gelencser, A., Legrand, M., and Pio, C.: Concentration of
740   atmospheric cellulose: a proxy for plant debris across a west-east transect over Europe, J. Geophys. Res.,
741   112(D23), doi:10.1029/2006JD008180, 2007.

742   Sesartic, A. and Dallafior, T. N.: Global fungal spore emissions, review and synthesis of literature data,
743   Biogeosciences, 8(5), 1181–1192, doi:10.5194/bg-8-1181-2011, 2011.

744   Shcherbakova, L. A.: Advanced methods of plant pathogen diagnostics, in Comprehensive and molecular
745   phytopathology, pp. 75–116, Elsevier, Amsterdam, doi:9780080469331, 2007.

746   Simoneit, B. R. T., Elias, V. O., Kobayashi, M., Kawamura, K., Rushdi, A. I., Medeiros, P. M., Rogge, W. F., and
747   Didyk, B. M.: Sugars dominant water-soluble organic compounds in soils and characterization as tracers in
748   atmospheric particulate matter, Environ. Sci. Technol., 38(22), 5939–5949, doi:10.1029/2004JD004598, 2004.

749   Srivastava, D., Favez, O., Bonnaire, N., Lucarelli, F., Haeffelin, M., Perraudin, E., Gros, V., Villenave, E., and
750   Albinet, A.: Speciation of organic fractions does matter for aerosol source apportionment: part 2: intensive short-
751   term campaign in the Paris area (France), Sci. Total Environ., 634, 267–278, doi:10.1016/j.scitotenv.2018.03.296,
752   2018.

753   Sullivan, A. P., Frank, N., Kenski, D. M., and Collett, J. L.: Application of high-performance anion-exchange
754   chromatography–pulsed amperometric detection for measuring carbohydrates in routine daily filter samples
755   collected by a national network 2: examination of sugar alcohols/polyols, sugars, and anhydrosugars in the upper
756   Midwest, J. Geophys. Res. Atmospheres, 116(D8), D08303, doi:10.1029/2010JD014169, 2011.

757   Tanarhte, M., Bacer, S., Burrows, S. M., Huffman, J. A., Pierce, K. M., Pozzer, A., Sarda-Estève, R., Savage, N.
758   J., and Lelieveld, J.: Global modeling of fungal spores with the EMAC chemistry climate model: uncertainties in
759   emission parametrizations and observations, Atmospheric Chem. Phys. Discuss., 1–31, doi:10.5194/acp-2019-
760   251, 2019.

761   Vélëz, H., Glassbrook, N. J., and Daub, M. E.: Mannitol metabolism in the phytopathogenic fungus alternaria
762   alternata, Fungal Genet. Biol., 44(4), 258–268, doi:10.1016/j.fgb.2006.09.008, 2007.

763   Verma, S. K., Kawamura, K., Chen, J., and Fu, P.: Thirteen years of observations on primary sugars and sugar
764   alcohols over remote Chichijima Island in the western north Pacific, Atmos. Chem. Phys., 18(1), 81–101,
765   doi:https://doi.org/10.5194/acp-18-81-2018, 2018.

766   Vlachou, A., Daellenbach, K. R., Bozzetti, C., Chazeau, B., Salazar, G. A., Szidat, S., Jaffrezo, J.-L., Hueglin, C.,
767   Baltensperger, U., Haddad, I. E., and Prévôt, A. S. H.: Advanced source apportionment of carbonaceous aerosols
768   by coupling offline AMS and radiocarbon size-segregated measurements over a nearly 2-year period, Atmos.
769   Chem. Phys., 18(9), 6187–6206, doi:10.5194/acp-18-6187-2018, 2018.

770   Waked, A., Favez, O., Alleman, L. Y., Piot, C., Petit, J.-E., Delaunay, T., Verlinden, E., Golly, B., Besombes, J.-
771   L., Jaffrezo, J.-L., and Leoz-Garziandia, E.: Source apportionment of PM_{10} in a north-western Europe regional
772   urban background site (Lens, France) using positive matrix factorization and including primary biogenic
773   emissions, Atmos. Chem. Phys., 14(7), 3325–3346, doi:10.5194/acp-14-3325-2014, 2014.

774   Wan, E. C. H. and Yu, J. Z.: Analysis of sugars and sugar polyols in atmospheric aerosols by chloride attachment
775   in liquid chromatography/negative ion electrospray mass spectrometry, Environ. Sci. Technol., 41(7), 2459–2466,
776   doi:10.1021/es062390g, 2007.

777 Wang, X., Shen, Z., Liu, F., Lu, D., Tao, J., Lei, Y., Zhang, Q., Zeng, Y., Xu, H., Wu, Y., Zhang, R., and Cao, J.:
778 saccharides in summer and winter $PM_{2.5}$ over Xi'an, northwestern China: sources, and yearly variations of biomass
779 burning contribution to $PM_{2.5}$, Atmos. Res., 214, 410–417, doi:10.1016/j.atmosres.2018.08.024, 2018.

780 Wan, X., Kang, S., Rupakheti, M., Zhang, Q., Tripathee, L., Guo, J., Chen, P., Rupakheti, D., Panday, A. K.,
781 Lawrence, M. G., Kawamura, K., and Cong, Z.: Molecular characterization of organic aerosols in the Kathmandu
782 valley, Nepal: insights into primary and secondary sources, Atmos. Chem. Phys., 19(5), 2725–2747,
783 doi:10.5194/acp-19-2725-2019, 2019.

784 Weber, S., Uzu, G., Calas, A., Chevrier, F., Besombes, J.-L., Charron, A., Salameh, D., Ježek, I., Močnik, G., and
785 Jaffrezo, J.-L.: An apportionment method for the oxidative potential of atmospheric particulate matter sources:
786 application to a one-year study in Chamonix, France, Atmos. Chem. Phys., 18(13), 9617–9629, doi:10.5194/acp-
787 18-9617-2018, 2018.

788 Weber, S., Salameh, D., Albinet, A., Alleman, L. Y., Waked, A., Besombes, J.-L., Jacob, V., Guillaud, G.,
789 Meshbah, B., Rocq, B., Hulin, A., Dominik-Sègue, M., Chrétien, E., Jaffrezo, J.-L., and Favez, O.: Comparison
790 of PM_{10} Sources profiles at 15 French sites using a harmonized constrained positive matrix factorization approach,
791 Atmosphere, 10(6), 310, doi:10.3390/atmos10060310, 2019.

792 Wéry, N., Galès, A., and Brunet, Y.: Bioaerosol sources, in Microbiology of Aerosols, pp. 115–135, John Wiley
793 & Sons, Ltd., doi:10.1002/9781119132318, 2017.

794 Whipps, J. M., Hand, P., Pink, D., and Bending, G. D.: Phyllosphere microbiology with special reference to
795 diversity and plant genotype, J. Appl. Microbiol., 105(6), 1744–1755, doi:10.1111/j.1365-2672.2008.03906.x,
796 2008.

797 Xiao, M., Wang, Q., Qin, X., Yu, G., and Deng, C.: Composition, sources, and distribution of $PM_{2.5}$ saccharides
798 in a coastal urban site of China, Atmosphere, 9(7), 274, doi:10.3390/atmos9070274, 2018.

799 Xu, J., He, J., Xu, H., Ji, D., Snape, C., Yu, H., Jia, C., Wang, C., and Gao, J.: Simultaneous measurement of
800 multiple organic tracers in fine aerosols from biomass burning and fungal spores by HPLC-MS/MS, RSC. Adv.,
801 8(59), 34136–34150, doi:10.1039/C8RA04991B, 2018.

802 Yan, C., Sullivan, A. P., Cheng, Y., Zheng, M., Zhang, Y., Zhu, T., and Collett, J. L.: Characterization of
803 saccharides and associated usage in determining biogenic and biomass burning aerosols in atmospheric fine
804 particulate matter in the north China Plain, Sci. Total Environ., 650, 2939–2950,
805 doi:10.1016/j.scitotenv.2018.09.325, 2019.

806 Yan, K., Park, T., Yan, G., Chen, C., Yang, B., Liu, Z., Nemani, R., Knyazikhin, Y., and Myneni, R.: Evaluation
807 of MODIS LAI/FPAR product collection 6: part 1: consistency and improvements, Remote Sens., 8(5), 359,
808 doi:10.3390/rs8050359, 2016a.

809 Yan, K., Park, T., Yan, G., Liu, Z., Yang, B., Chen, C., Nemani, R., Knyazikhin, Y., and Myneni, R.: Evaluation
810 of MODIS LAI/FPAR product collection 6: part 2: validation and intercomparison, Remote Sens., 8(6), 460,
811 doi:10.3390/rs8060460, 2016b.

812 Yttri, K. E., Dye, C., and Kiss, G.: Ambient aerosol concentrations of sugars and sugar-alcohols at four different
813 sites in Norway, Atmos. Chem. Phys., 7(16), 4267–4279, doi:10.5194/acp-7-4267-2007, 2007.

814 Yttri, K. E., Simpson, D., Stenström, K., Puxbaum, H., and Svendby, T.: Source apportionment of the
815 carbonaceous aerosol in Norway – quantitative estimates based on 14C, thermal-optical and organic tracer
816 analysis, Atmos. Chem. Phys., 11, 9375-9394, doi:10.5194/acpd-11-7375-2011, 2011a.

817 Yttri, K. E., Simpson, D., Nøjgaard, J. K., Kristensen, K., Genberg, J., Stenström, K., Swietlicki, E., Hillamo, R.,
818 Aurela, M., Bauer, H., Offenberg, J. H., Jaoui, M., Dye, C., Eckhardt, S., Burkhart, J. F., Stohl, A., and Glasius,
819 M.: Source apportionment of the summer time carbonaceous aerosol at Nordic rural background sites, Atmos.
820 Chem. Phys., 11(24), 13339–13357, doi:10.5194/acp-11-13339-2011, 2011b.

821   Yue, S., Ren, H., Fan, S., Wei, L., Zhao, J., Bao, M., Hou, S., Zhan, J., Zhao, W., Ren, L., Kang, M., Li, L., Zhang,
822   Y., Sun, Y., Wang, Z., and Fu, P.: High abundance of fluorescent biological aerosol particles in winter in Beijing,
823   China, ACS Earth Space Chem., 1(8), 493–502, doi:10.1021/acsearthspacechem.7b00062, 2017.

824   Zamfir, M., Gerstner, D. G., Walser, S. M., Bünger, J., Eikmann, T., Heinze, S., Kolk, A., Nowak, D., Raulf, M.,
825   Sagunski, H., Sedlmaier, N., Suchenwirth, R., Wiesmüller, G. A., Wollin, K.-M., Tesseraux, I., and Herr, C. E.
826   W.: A systematic review of experimental animal studies on microbial bioaerosols: Dose-response data for the
827   derivation of exposure limits, Int. J. Hyg. Environ. Health, 222(2), 249–259, doi:10.1016/j.ijheh.2018.11.004,
828   2019.

829   Zhang, T., Engling, G., Chan, C.-Y., Zhang, Y.-N., Zhang, Z.-S., Lin, M., Sang, X.-F., Li, Y. D., and Li, Y.-S.:
830   Contribution of fungal spores to particulate matter in a tropical rainforest, Environ. Res. Lett., 5(2), 024010,
831   doi:10.1088/1748-9326/5/2/024010, 2010.

832   Zhang, Z., Engling, G., Zhang, L., Kawamura, K., Yang, Y., Tao, J., Zhang, R., Chan, C., and Li, Y.: Significant
833   influence of fungi on coarse carbonaceous and potassium aerosols in a tropical rainforest, Environ. Res. Lett.,
834   10(3), 034015, doi:10.1088/1748-9326/10/3/034015, 2015.

835   Zhu, C., Kawamura, K., and Kunwar, B.: Organic tracers of primary biological aerosol particles at subtropical
836   Okinawa Island in the western North Pacific Rim: organic biomarkers in the north pacific, J. Geophys. Res.
837   Atmospheres, 120(11), 5504–5523, 2015.

838   Zhu, W., Luo, L., Cheng, Z., Yan, N., Lou, S., and Ma, Y.: Characteristics and contributions of biogenic secondary
839   organic aerosol tracers to $PM_{2.5}$ in Shanghai, China, Atmospheric Pollut. Res., 9(2), 179–188,
840   doi:10.1016/j.apr.2017.09.001, 2018a.

841   Zhu, W., Cheng, Z., Luo, L., Lou, S., Ma, Y., and Yan, N.: Investigation of fungal spore characteristics in $PM_{2.5}$
842   through organic tracers in Shanghai, China, Atmospheric Pollut. Res., 9(5), 894–900,
843   doi:10.1016/j.apr.2018.01.009, 2018b.

844