ACP-2019-434

Answer to Anonymous Referee #1 comments

This manuscript provides detailed insights into the biogenic primary organic aerosol emission sources of the primary sugar compounds (SC), i.e., glucose, arabitol and mannitol. The study has been carefully designed and the results have been interpreted in detail. The study covers 16 nation-wide sites all over France and contains a very comprehensive data set. It is clearly shown that the main drivers of SC atmospheric concentrations are ambient air temperature, relative humidity and vegetation density.

We thank the reviewer for his/her review. We have studied the comments and we have made revisions point by point. The detailed responses to the comments are given below, point by point, *in blue color*, including changes directly made to the manuscript, *in red color*.

Specific comments:

1. Introduction: glucose is recognized as a tracer for plant pollen but also for biomass burning. I miss some discussion about this issue in the introduction.

The reviewer is right that glucose can also originate from the thermal degradation of the plant materials (e.g., cellulose, a polymer of glucose). However, as evidenced in Figure R1.1, the concentrations of levoglucosan (a well-established tracer of biomass burning source) and those of glucose clearly display very different annual atmospheric evolution cycles: higher concentrations of levoglucosan in France are observed in the coldest season (winter) due to the increased biomass burning while those of glucose are observed in in warm seasons and coinciding with negligible ambient concentrations of levoglucosan. Such different temporal patterns indicate that the biomass burning is not an important source of atmospheric glucose.

Glucose can have a broad primary biogenic sources, e.g. from terrestrial plant pollen, fruits, and detritus, or from the degradation of the soil microorganisms (Xiao et al., 2018; Zhu et al., 2015) or even possibly from bubble bursting processes in remote oceans (Fu et al., 2013; Gao et al., 2011; Leck and Bigg, 2005).

For these reasons, we have removed the term "specific" in lines 54—59.

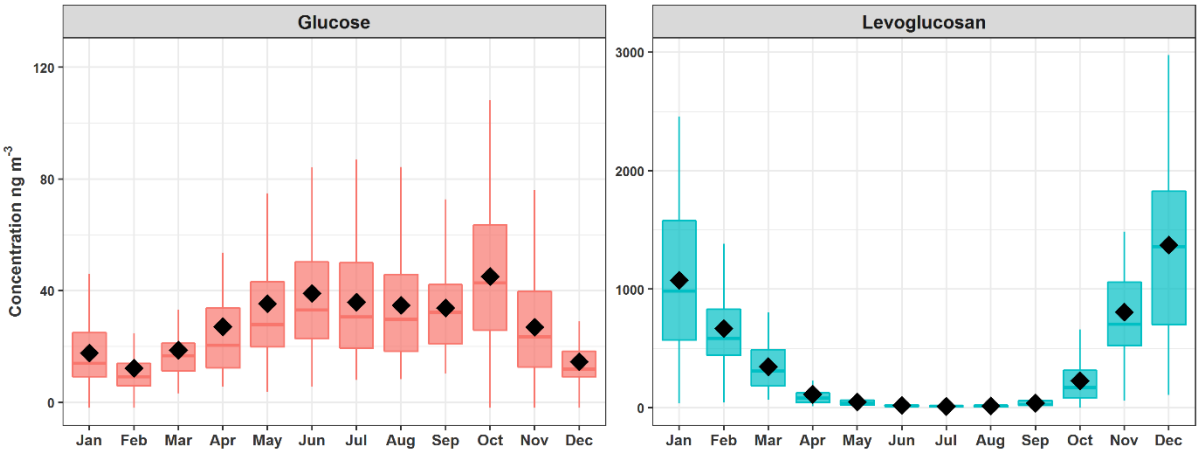

Figure R1.1: Annual evolution cycles of the glucose (left) and levoglucosan (right) concentrations in PM_{10} measured at the urban site of Grenoble Les Frênes, from the years 2012 to 2018. The black marker inside each boxplot indicates the average value, while the top, middle and bottom of the box represent 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.

2. In several parts of the text, figures and tables, mention is made of "glucose" but in fact "free cellulose" is meant. In order to avoid confusion, I suggest to make this more clear and replace "glucose" by "free cellulose".

In fact, both glucose and free cellulose are measured and analyzed separately in the present work. We used glucose when the monosaccharide "glucose" is meant and free cellulose we considered the cellulose ambient cellulose.

Technical corrections: References: should be ordered chronologically.

The references are now ordered chronologically, as suggested by the reviewer.

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Fu, P. Q., Kawamura, K., Chen, J., Charrière, B., and Sempéré, R.: Organic molecular composition of marine aerosols over the Arctic Ocean in summer: contributions of primary emission and secondary aerosol formation, Biogeosciences, 10(2), 653–667, doi:10.5194/bg-10-653-2013, 2013.

Gao, Q., Nilsson, U., Ilag, L. L., and Leck, C.: Monosaccharide compositional analysis of marine polysaccharides by hydrophilic interaction liquid chromatography-tandem mass spectrometry, Anal. Bioanal. Chem., 399(7), 2517–2529, doi:10.1007/s00216-010-4638-z, 2011.

Leck, C. and Bigg, E. K.: Biogenic particles in the surface microlayer and overlaying atmosphere in the central Arctic Ocean during summer, Tellus B, 57(4), 305–316, doi:10.1111/j.1600-0889.2005.00148.x, 2005.

Xiao, M., Wang, Q., Qin, X., Yu, G., and Deng, C.: Composition, Sources, and Distribution of PM2.5 Saccharides in a Coastal Urban Site of China, Atmosphere, 9(7), 274, doi:10.3390/atmos9070274, 2018.

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

Answer to Anonymous Referee #2 comments

The manuscript "Arabitol, mannitol and glucose as tracers of primary biogenic organic aerosol: influence of environmental factors on ambient air concentrations and spatial distribution over France" describes the primary sugar compounds (SC, defined as glucose, arabitol and mannitol) concentrations in PM10 for 16 increasing space scale sites (local to nation-wide), distributed in several French geographic areas of different environmental conditions. This paper first time investigates the spatial behavior of these chemicals and evidencing their major effective environmental drivers.

We thank the reviewer for his/her attention to our manuscript that greatly contribute to improve the quality of this research paper. All comments have been considered and answered. The detailed responses to the comments are given below, point by point, *in blue color*, including changes directly made to the manuscript, *in red color*.

Major comments:

(1) Updating the references used in this manuscript to more current state is suggested.
We do agree with the reviewer and we have updated the references with several works recently published, including those in 2019. However, very few scientific papers have been published recently on the short term (daily) and the spatial characterization of polyols and glucose in PM_{10}. This is why older pioneering works are also cited in the present work.

(2) LOD (limit of detection) of the detected chemicals should be included in the experimental section.
As suggested by reviewer, the information about the quantification limits have been included in the experimental section (lines 176—177).

(3) The regional transport is also very important for the spatial behavior and distribution of the chemical species in the ambient. In addition, only temporal variations and tracer ratios were shown and discussed in the results and discussion section. More deep analysis (i.e., the influences from nearby regions/sources, combine the chemical analysis results with synoptic data,. . .) are recommended to make this paper more interesting and innovative. At least, choose one or two cases to explain the contribution from regional transport by backward trajectory analysis.

We agree that regional transport may impact PM polyol concentrations. However, we do not think that it explains the main temporal signals observed in this work. Since the correlation matrix corresponds to averaged values of composite data, i.e. aggregate on consecutive three days or six days intervals, it already account for potential regional transport between sites, and a decreased of correlation with inter-site distance is observed and is so probably indicative of local source contribution rather than transportation.

However, as suggested by the reviewer, we achieved additional back trajectory analyses. , This was done for arabitol concentrations at the remote OPE-ANDRA site for the period 2012−2018, applying Potential Source Contribution Function (PSCF) to HYSPLIT data and using the pyPSCF python package[1]. Results do not indicate clear source region(s) (Figure R2.2). Indeed, even if it seems that air-masses associated with high arabitol loading (>75th concentration percentile) never come from the East, it is in fact explained by the climatic wind condition in this region where no easterlies wind are observed during summer (anticyclonic condition). Finally, since no specific region is pointed out by the PSCF analysis, it may be explain either if the arabitol is emitted everywhere, or by a local (<few grid cells, within around 100 km from the station) source since all back-trajectories will then be associated with high concentration. Since correlations between sites decrease with the distance, the first hypothesis is

[1] https://gricad-gitlab.univ-grenoble-alpes.fr/webersa/pyPSCF.git
(documentation: https://pypscf.readthedocs.io/en/lastest)

most probably not valid. These two arguments are in favor of -local sources being predominant for the polyols, as opposed to regional (> 100 km) transport.

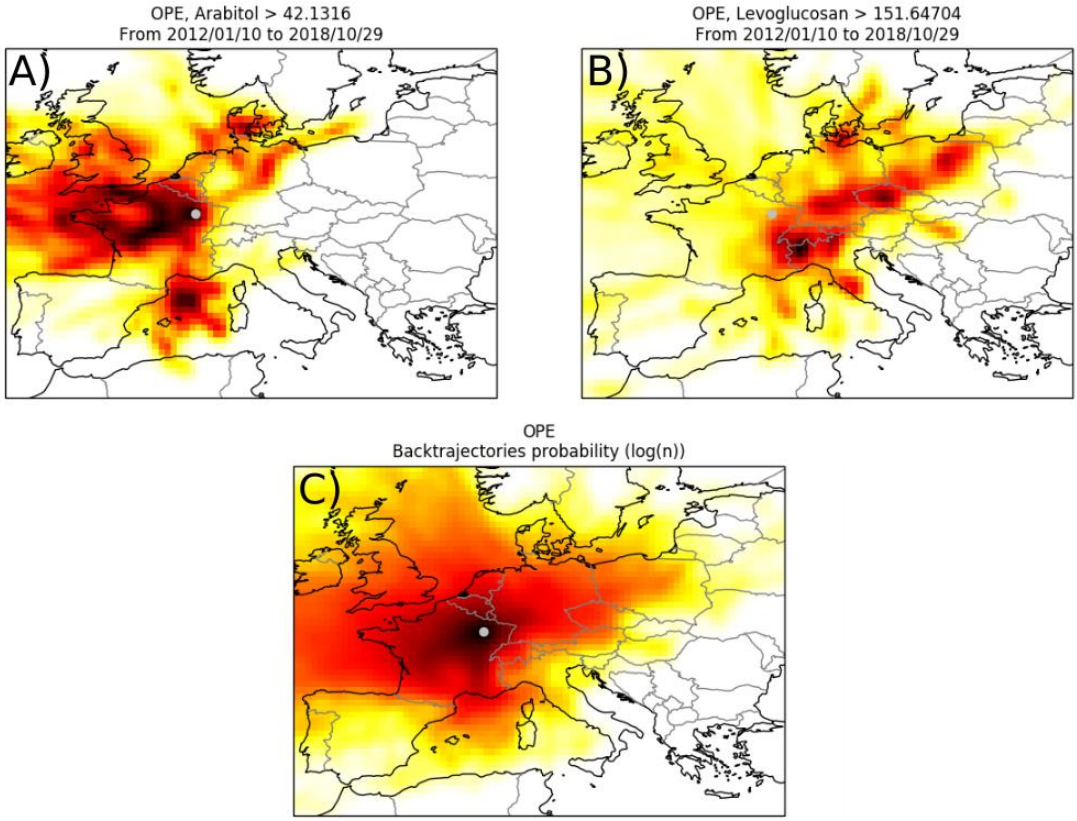

Figure R2.2:A) PSCF analysis for the OPE site (using pyPSCF and HYSPLIT). Back-trajectories associated with arabitol concentrations higher than the 75th percentile divided by the number of back-trajectories. B) displays all the back-trajectories. The colorscale indicated the probability that the specie comes from this cell (the darker color indicate higher probability).

(4) Page 6, Line 174-180. The normalized cross-correlation (NCC) test was chosen in this manuscript, and author mentioned a thorough discussion on the normalized cross-correlation method can be found elsewhere (Kaso, 2018; Yoo and Han, 2009). However, there was no related applied reference of NCC method was given, more field observation references used this methods are suggested to add.

The reviewer is right. The references related to the NCC method described only the concept and theory of NCC method. Sorry about it. Additional references (Bardal and Sætran, 2016; Dai and Zhou, 2017; Eisner et al., 2009; Lainer et al., 2016; Le Pichon et al., 2019) are now given to illustrate NCC applications in atmospheric sciences (lines 206—207).

(5) The lines in the figures are too thick to find the points, especially for Fig. (2a), Fig. (2b) and Fig. 5. It is difficult to separate the different color lines. Moreover, the thickness of the lines seems not consistent, i.e., the blue lines seem thicker than other color lines.

These figures have been modified accordingly. Thanks for suggestion.

(6) Figure S2 is suggested to add in Figure 3. Discuss the Normalized cross-correlation values for the daily evolution of particulate for glucose, polyols, calcium and sulfate together. It can exhibit the differences of NCC between these chemicals more directly. Moreover, how about NCC of other

inorganic ions, i.e., NH4+, NO3- (similar as SO42- , are the main components of secondary inorganic aerosols), K+ (biomass burning tracer) and Cl-.

The present work do not aim at discussing these species. Nevertheless, since they may act as a negative control for the local emission of polyols, we initially presented some of them in the submitted manuscript and SI. As suggested by the reviewer, former Fig S2 has also added in the main text together with fig 3. and we are now presenting some major secondary inorganics (ammonium) and biomass burning proxy (levoglucosan) as follows:

Lines 266-279 "Unlike SC, ambient air concentrations of sulfate (Fig. 3C) and ammonium (Fig. 3D), associated with long-range aerosol transport (Abdalmogith and Harrison, 2005; Amato et al., 2016; Coulibaly et al., 2015; Pindado and Perez, 2011; Waked et al., 2014) and levoglucosan ((Fig. 3E), associated with biomass burning in cold season (Weber et al., 2019; Xiao et al., 2018), display stronger positive correlations ($R > 0.72$-0.98, $p < 0.01$) at all pairs of sites considered in the present work. Moreover, ambient concentrations of calcium (Fig. 3F), associated with local fugitive dust sources or/and long-range aerosol transport (Ram et al., 2010; Wan et al., 2019) display random correlation patterns. These results are in agreement with Zhu et al. (2018) who also reported non-significant correlations between SC and sulfate in $PM_{2.5}$ aerosols measured at Shanghai, China. The distinct spatial behaviors between sulfate (or Ca^{2+}) and SC in the present work further suggest a dominant regional influence for atmospheric SC, as opposed to processes associated with either local sources for calcium or long-range transport for sulfate".

For secondary species (sulfate and ammonium), potential long range transport (Ca^{2+}) and chemically stable species (levoglucosan, Figure R2.2B), the correlation are still high ($r>0.7$) even after hundreds of kilometers. For these species, we can make the hypothesis that the regional transport play a major role in concentrations seen at a given site. However, an in-depth analysis of the sources and evolution of the concentrations of these species is beyond the scope of this study and would require a dedicated future work.

Specific comments:

(1) Line 190: The linear regression (lm) package in R was employed for multiple regression analyses. What does "lm" in the bracket means??

The linear model aka "lm" in the brackets is the name of the statistical package employed for multiple regression analyses. Definition of "lm" is now added in the main text (line 217).

(2) Line 320: these findings highlight that particulate SC PM10 and cellulose in both urban background and rural agricultural areas. . ., should be changed to "these findings highlight that SC in PM10 and cellulose in both urban background and rural agricultural areas

Thank you for your attentive review, this sentence has been corrected (lines 367—368).

Answer to Anonymous Referee #3 comments

This paper describes the evolutions of glucose, mannitol and arabitol in the aerosol covering 16 sites all over France. The study consists in a huge and precious dataset. For the first time, the distance-dependent correlation is demonstrated, investigating also the main drivers of atmospheric sugar concentrations.

We thank the reviewer for his/her attention to our manuscript that greatly contribute to improve the quality of this research paper. We have considered each of the comments and we have made revisions point by point. The detailed responses to the comments are given below, point by point, *in blue color*, including changes directly made to the manuscript, *in red color*.

General comment: please check all manuscript, including figures and tables, and modify the term "polyols" with "mannitol and arabitol", as necessary, to avoid confusion., as suggested in the initial revision.

We do agree with the reviewer that we are not analyzing all the sugar alcohol species. However, we clearly specify that the term polyols is to refer to the sum of arabitol and mannitol concentrations (lines 178—179). This has been added to the main text:

"Hereafter, the term "Polyols" is used to refer uniquely to the sum of arabitol and mannitol concentrations".

Line 41-43. The authors affirmed that "sugar alcohols . . .- including arabitol and mannitol. . .- have been recognized as tracers for airborne fungi". One of the main objectives of my recent research is the source investigation of water soluble organic compounds, such as for example sugars, and I quite sure that some sugars alcohols have another source. For examples I saw that sorbitol have some correlation with biomass burning tracers, while arabitol and mannitol, mainly distributed in the coarse fraction of aerosol, plausibly originate from fungal spores. So, I suggest to focus your affirmation only on the arabitol and mannitol.

We do agree with the reviewer and we have focused our affirmation only on the arabitol and mannitol.

Line 45. The authors define glucose "a specific tracer for plant materials" but I think that the authors should remove "specific" because glucose can have different sources: plant materials, soil emissions (as suggested by the authors) and also marine biogenic material derived from degradation of polysaccharides present in the marine microlayer. I suggest to read some papers of Prof. Leck because she investigated the organic compounds (such as saccharides) in the marine aerosol. I know that the paper is focused on the aerosol samples collected in the areas far from the coast but in the introduction I think that the authors should consider all sources.

Indeed, glucose can have a broad of biogenic sources, e.g. from terrestrial plant pollen, fruits, and detritus, or from the degradation of the soil microorganisms (Kang et al., 2018; Li et al., 2018; Xiao et al., 2018) or even possibly from bubble bursting processes in remote oceans (Fu et al., 2013; Gao et al., 2011; Leck and Bigg, 2005); we have removed the term "specific" (see lines 54—59). This point is also further discussed in the response to the comment 1 of anonymous referee # 1.

Line 174. Can you specify some details about the dataset matrix using to perform the normalized cross correlation.

The raw data used in the present study consisted in to daily (24 hours) aerosols collected at 16 sites in different geographic regions in France. For pairwise normalized cross-correlation analyses, original daily series were first converted as follows: starting on identical days (for each pairs of sites), arrangement on the original daily data into consecutive 3-day intervals (or 6-day intervals in the case of

OPE-ANDRA) and calculation of the average concentration values for the middle-day were performed. We directly used this resultant data for the correlation analysis between site pairs (lines 207—212).

In this respect, the manuscript has been revised as follows: lines 207-212

"To achieve pair-wise correlation analysis between the sampling sites collected during the same periods, the original raw daily measurements were processed as follows: starting on identical days for each pairs of sites, arrangement on the original daily data into consecutive 3-day intervals (or 6-day intervals in the case of OPE-ANDRA) and calculation of the average concentration values for the middle-day were performed. The resultant data were used for correlation analysis between site pairs (Table S3)".

Line 257. You correctly affirmed that mannitol-to arabitol ratio can suggest the temporal and spatial evolution of their emission processes, using this reference: Gosselin et al. 2016. This paper demonstrated also that, in some cases, mannitol and arabitol can have different sources: "mannitol is a common polyol in higher plants while arabitol is only found in fungal spores and lichen". I suggest to insert this concept in the manuscript and to consider the R2 between two polyols in the discussion because maybe either conclusion can be also obtained (this is just a suggestion).

Thank you for this interesting suggestion. We added discussion about R^2 between arabitol and mannitol (lines 288—292), as follows: lines 282-293.

"Based on parallel measurements of spore counts and PM_{10} polyol concentrations at three sites within the area of Vienna (Austria), Bauer et al. (2008a) found an average arabitol and mannitol content per fungal spores of respectively 1.2 pg spore-1 (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 often identified in the green algae and lower plants (Buiarelli et al., 2013; Di Filippo et al., 2013; Vélëz et al., 2007; Xu et al., 2018; Zhang et al., 2010). Gosselin et al., 2016 observed a relatively low (R^2 = 0.31) to high (R^2 = 0.84) coefficient of determination between mannitol and arabitol for total suspended particles (TSP) collected at a pine-forested area during dry and rainy periods, respectively. High correlation in rainy periods possibly suggested that both chemical species in the TSP fraction in this pine-forested area could have been derived mainly from the same sources, i.e., actively wet-discharged ascospores and basidiospores, while the poor correlation in dry periods could have been likely due to more complex sources, i.e., dry discharged spores, plants, algae, etc."

Note that the study by Gosselin et al., 2016 has been conducted on total suspended particles (TSP) and in a specific pine-forested area of North America. The high coefficient of determination reported by Gosselin et al., 2016 during the rainy periods possibly suggest that both chemical species in the TSP fraction in this pine-forested area could have been derived mainly from the same sources, i.e., actively wet-discharged ascospores and basidiospores. However, the relatively poor correlation in dry periods could have been likely due to more complex sources, i.e., a mixture of actively wet and/or dry discharged spores, or influence of additional biogenic sources such as plants, algae, etc. Indeed, active release of wet discharged ascospores and basidiospores occurs in most ascomycetes and in basidiomycetes (Ingold and Hudson, 1993; Zhang et al., 2010) which is influenced by ambient humidity and rainfall (Elbert et al., 2007; Zhang et al., 2015). In contrast, dry discharged spores are preferentially emitted under dry and warm conditions. Thus, these correlation patterns could be at least partially explained by the different fungal habitats and/or different emission processes during rainy and dry periods.

Section 3.2. The distance-dependent correlations and the SC evolution synchronous at an urban city scale and throughout the same geographical regions are the very interesting topics in the manuscript and I appreciate this work because it was a lack of the sugars knowledge. The distance-dependent correlations is very clear using your approach but I suggest to clarify the main reasons for the decrease of NCC when the distance was above 200 km. You report some explanations but I suggest to deeply discuss the reasons or the suggestion of this behavior.

Thank for this positive comment. We also believe that this point is quite innovative in the current literature and that it can considerably improve our knowledge about primary sugars in the atmosphere. We believe that the main reasons of such distance correlation patterns are most probably associated with different airborne microbial community assemblies that are shaped by different regional environmental factors (e.g. meteorological conditions, vegetation types and cover, etc.). Indeed, our recent interdisciplinary work (submitted for publication in "Science advances") has shown that the atmospheric concentration dynamics of polyols and some major saccharides (trehalose, glucose) are driven by only a few specific airborne fungal and bacterial genera. Further analyses for sites located in different climatic regions of France have also shown that airborne microbial assemblies associated with these chemical species vary regionally (unpublished data). This makes sense since different biotopes (meteorological conditions, vegetation types and cover) harbor distinct microbial communities (Bowers et al., 2012; Liu et al., 2019).

Line 301. Please remove "s" from "corresponds".

Thank you for your careful review, we have removed this "s".

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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)~~(Amato et al., 2017; Després et al., 2012; Elbert et al., 2007; Fang et al., 2018; Fröhlich-Nowoisky et al., 2016; Morris et al., 2011; 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)~~(Fröhlich-Nowoisky et al., 2009, 2016; Humbal et al., 2018; Lecours et al., 2017)~~, 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)~~(Bozzetti et al., 2016; China et al., 2018; Di Filippo et al., 2013; Heald and Spracklen, 2009; Jia et al., 2010)~~. 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)~~(Després et al., 2012; Elbert et al., 2007; Jacobson and Streets, 2009; Sesartic and Dallafior, 2011)~~.

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)~~(Bauer et al., 2008a; Di Filippo et al., 2013; Gosselin et al., 2016; Zhang et al., 2010,~~

2015). 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)(Bauer et al., 2008a; Gosselin et al., 2016; Holden et al., 2011; Jia and Fraser, 2011; Medeiros et al., 2006b). For instance, among sugar alcohols, (aka polyols) including 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 et al., 2019; Srivastava et al., 2018; Verma et al., 2018; Weber et al., 2018, 2019)(Amato et al., 2017; Bauer et al., 2008a, 2008b; Golly et al., 2018; Medeiros et al., 2006b; Samaké et al., 2019; Verma et al., 2018; Weber et al., 2018; Zhang et al., 2010; Zhu et al., 2015, 2016). Similarly, glucose has also been used as a specific 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)(Chen et al., 2013; Fu et al., 2013; Liang et al., 2016; Medeiros et al., 2006b; Pietrogrande et al., 2014; Rathnayake et al., 2017; Rogge et al., 2007; Simoneit et al., 2004b; Wan and Yu, 2007; Wan et al., 2019).

In this context, atmospheric concentrations of specific polyols 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)(Barbaro et al., 2015; Chen et al., 2013; Fu et al., 2012; Golly et al., 2018; Graham et al., 2003; Jia et al., 2010; Liang et al., 2016; Pietrogrande et al., 2014; Rogge et al., 2007; Simoneit et al., 2004a; Verma et al., 2018; Yttri et al., 2007; Zhu et al., 2018). 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)(Heald and Spracklen, 2009; 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)(Di Filippo et al., 2013; Golly et al., 2018; Gosselin et al., 2016; Graham et al., 2003; Holden et al., 2011; Medeiros et al., 2006b; Simoneit et al., 2004b; Wan et al., 2019; Yan et al., 2019; Yttri et al., 2007, 2011a; Zhu et al., 2015). 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)(Medeiros et al., 2006a; Rogge et al., 2007; Simoneit et al., 2004b; 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)~~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)~~(Ramoni and Seiboth, 2016)~~. Plant materials contain cellulose which has been reported as a suitable proxy to evaluate the vegetative debris contribution to OM mass (Bozzetti et al., 2016; Daellenbach et al., 2017; Glasius et al., 2018; Hiranuma et al., 2019; Puxbaum and Tenze-Kunit, 2003; Sánchez-Ochoa et al., 2007; Yttri et al., 2011b)~~(Bozzetti et al., 2016; Glasius et al., 2018; Puxbaum and Tenze-Kunit, 2003; Sánchez-Ochoa et al., 2007; Yttri et al., 2011b)~~. The ambient PM_{10} cellulose has been shown to be abundant in the European semi-rural or background environments (accounting for 2 to 10 % of OM mass) (Glasius et al., 2018; Sánchez-Ochoa et al., 2007)~~(Glasius et al., 2018; Sánchez-Ochoa et al., 2007)~~ and Nordic rural environments in Norway (contributing to 12 to 18 % of total carbon mass) (Yttri et al., 2011b) ~~(Yttri et al., 2011b)~~. Thus, simultaneous concentration measurements of cellulose and SC can provide essential information into their emission source dynamics.

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

2. Material and methods

2.1 Sampling sites

Daily PM_{10} concentrations reported in the present work were obtained from different research and monitoring programs conducted over the last six years in France. Within the framework of the present study, we carefully

selected sites sharing at least one complete year of concurrent monitoring with another one, to be representative of the annual variation cycles. The final dataset includes data from 16 sites, which are distributed in different regions of France (Fig. ~~ure~~ 1) and cover several main types of environmental conditions in terms of site topography, local vegetation, and climate. The characteristics and data available at each sampling site are listed in Table S1 of the supplementary material (SM), together with the information on the annual average concentrations of aerosol chemical composition (Table S2). Detailed information on the sampling conditions can be found in Samaké et al. (2019), such as the campaign periods, number of collected PM samples, sampling flow rates, sample storage and handling, etc. Note that, the previous database (Samaké et al., 2019)~~(Samaké et al., 2019)~~ has been updated here with arabitol and mannitol in PM_{10} collected at the suburban site of Nogent-sur-Oise for a series covering the years 2013 to 2017.

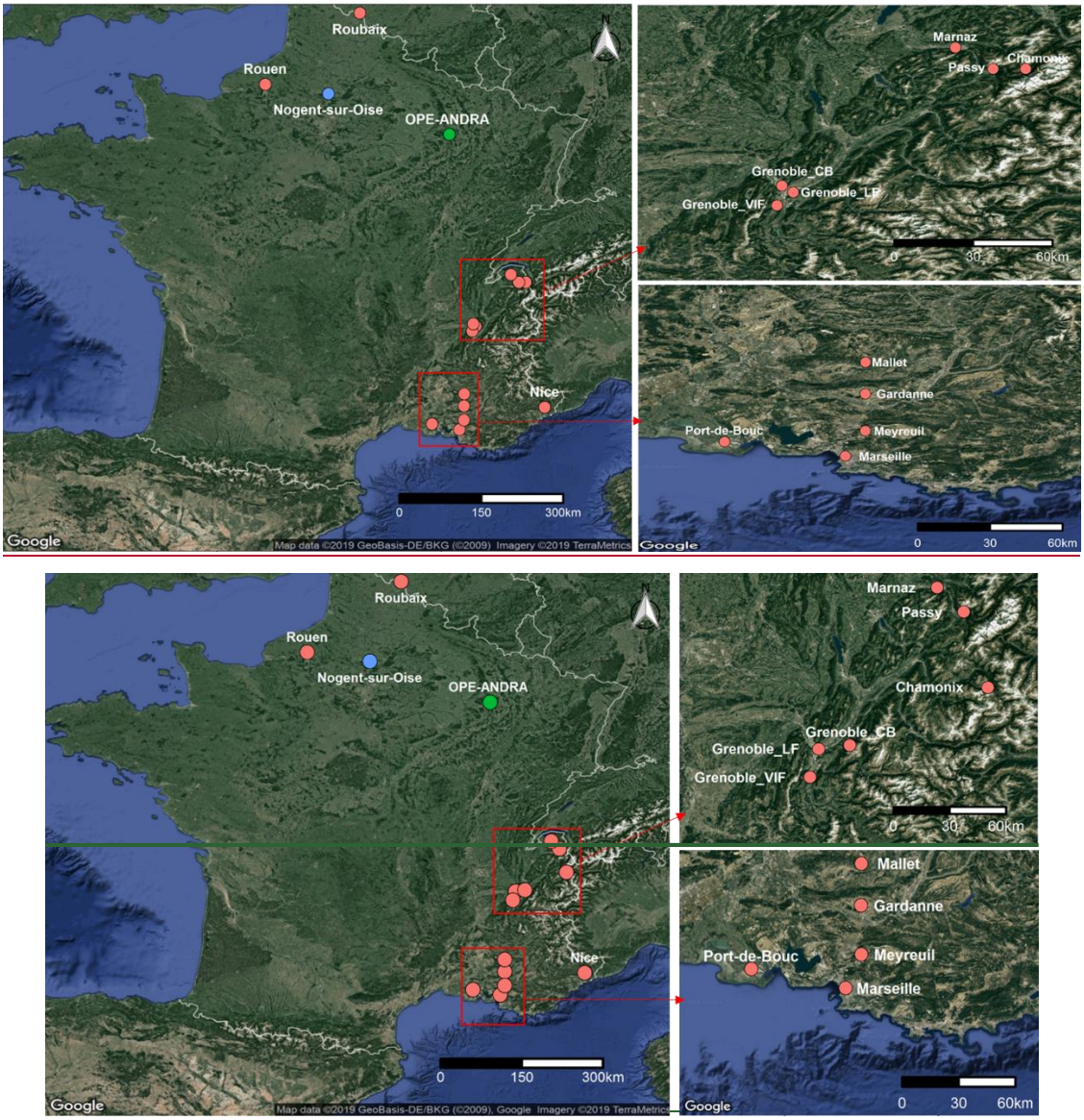


Figure 3: Geographical location of the selected sampling sites. The red and blue dots indicate respectively urban and suburban sites while the green one corresponds to a rural site, surrounded by field crop areas.

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)~~(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 cm^2) 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)~~(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)~~(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)~~(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 the range of 10-15 %. The quantification limits primary sugar compounds and cellulose ranged from 0.63 to 0.89 ng m^{-3}. About 2 800 samples are considered in this work for the polyols and glucose series, while 290 samples

(from the sites of Grenoble_LF and OPE-ANDRA) are considered for the cellulose series. Hereafter, the term "Polyols" is used to refer uniquely to the sum of arabitol and mannitol concentrations.

2.3 Meteorological data and LAI measurements

Ambient weather data were not available at all monitoring sites (see Table S1). In this study, data including daily relative humidity (%), night-time temperature (°C), average and maximum temperatures (°C), wind speed (m s^{-1}), solar radiation (W m^{-2}), and rainfall level (mm) for the sites of Marnaz and OPE-ANDRA (Fig ~~ure~~ 1), representing different climatic regions and environmental conditions, were obtained from the French meteorological data sharing service system (Météo-France) and ANDRA (French national radioprotective agency, in charge of the OPE-ANDRA site), respectively.

The leaf area index (LAI), which is defined as the projected area of leaves over a unit of land, is an important measure of the local vegetation density variation (Heald and Spracklen, 2009; Yan et al., 2016a, 2016b)~~(Heald and Spracklen, 2009; Yan et al., 2016a, 2016b)~~. For this study, we used the MODIS Collection 6 LAI product because it is considered to have the highest quality among all the MODIS LAI products (Yan et al., 2016a, 2016b)~~(Yan et al., 2016a, 2016b)~~. The MCD15A3H product uses both Terra and Aqua reflectance observations as inputs to estimate daily LAI at 500 m spatial resolution, and a 4-day composite is calculated to reduce the noise from abiotic factors. Using a 2 × 2 km grid box around the monitoring site, the local vegetation density variation was retrieved from LP DAAC (https://lpdaac.usgs.gov/, last accessed: 15 March 2019) for the sites of Marnaz, OPE-ANDRA, and Grenoble_LF.

2.4 Data analyses

All the statistical analyses were carried out using the open-source R software (R studio interface, version 3.4.1). Several statistical analyses were performed on the concentrations to identify the spatial patterns of emission sources and the potential parameters of influence as explained below.

The normalized cross-correlation (NCC) test was chosen to examine the potential similarities among the monitoring sites for particulate SC concentrations, in terms of short-term temporal trends (e.g. synchronized periods of increase or decrease, simultaneous fluctuations during specific episodes). The main advantage of NCC over the traditional correlation tests is that it is less sensitive to linear changes in the amplitudes of the two-time series compared. Therefore, to reduce the possibility of spurious "anti-correlation" due to highly variable concentration ranges, data were amplitude-normalized prior to correlation analysis. A thorough discussion on the normalized cross-correlation method can be found elsewhere (Bardal and Sætran, 2016; Dai and Zhou, 2017; Eisner et al., 2009; Kaso, 2018; Lainer et al., 2016; Le Pichon et al., 2019)~~(Kaso, 2018; Yoo and Han, 2009)~~. To achieve pair-wise correlation analysis between the sampling sites collected during the same periods, the original raw daily measurements were processed as follows: starting on identical days for each pairs of sites, arrangement on the original daily data into consecutive 3-day intervals (or 6-day intervals in the case of OPE-ANDRA) and calculation of the average concentration values for the middle-day were performed. The resultant data were used for correlation analysis between site pairs (Table S3).

Multiple linear regression (MLR) was used to assess the strength of the relationships between atmospheric concentrations of particulate SC and local environmental factors including the daily mean relative humidity, night-time temperature, average and maximum temperature, wind speed, solar radiation, rain levels and LAI. Because the LAI is a 4-day composite, daily values of the other variables were re-scaled into consecutive 4-day averaged values. The linear regression (linear model or lm) package in R was employed for multiple regression analyses.

The concentration data were log-transformed to obtain regression residual distributions as close as possible to the normal Gaussian one (Fig.ure S1). Stepwise forward selection was used to select the predictors that explain well the temporal variation of SC concentrations at the site of Marnaz.

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

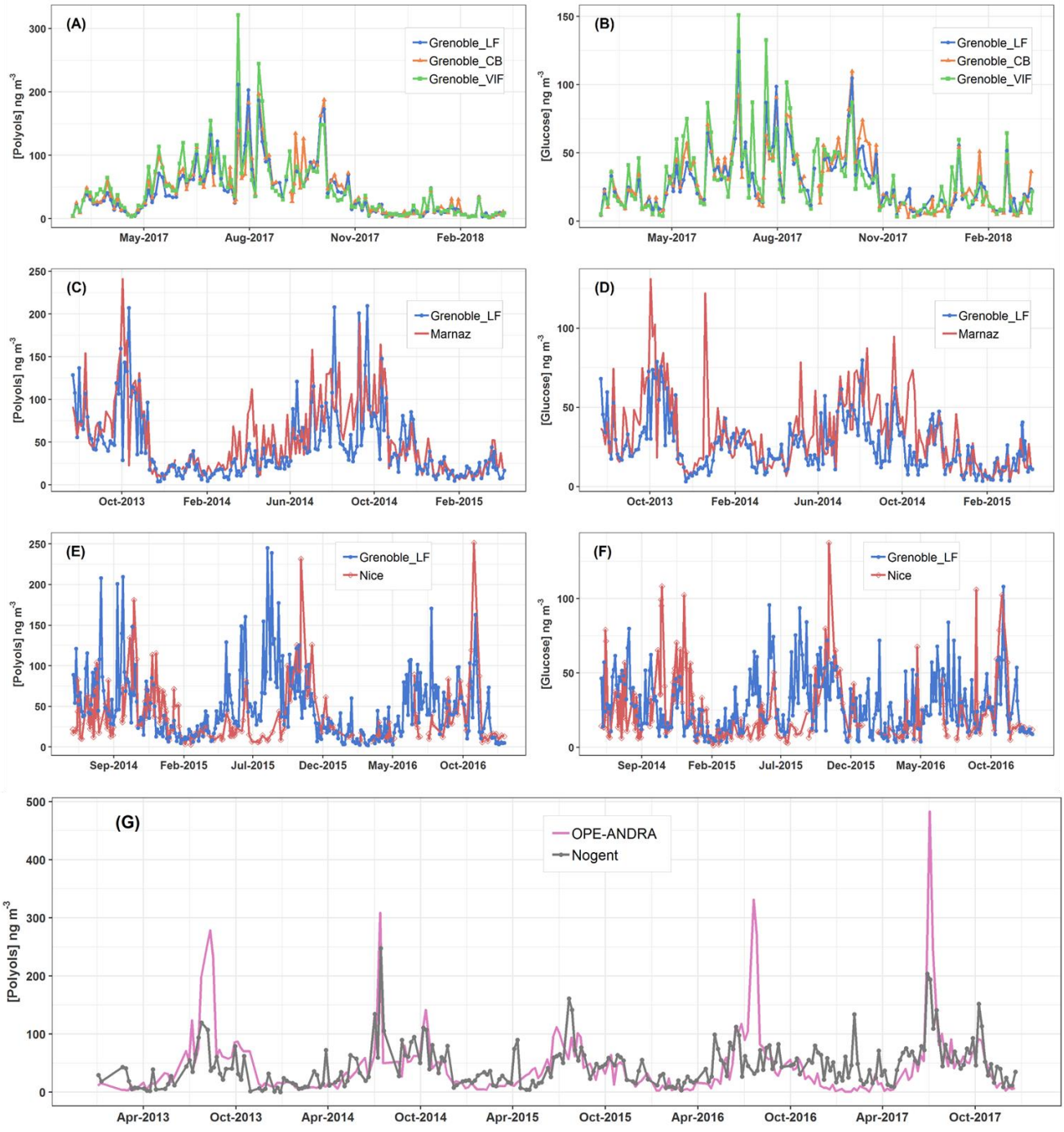
3. Results and discussion

3.1 Example of spatial coherence of the concentrations at different scales

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

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

The following sections are dedicated to the investigation of the processes that can lead to these similarities and differences according to these spatial scales.


(A)
[Polyols] ng m-3
Grenoble_LF
Grenoble_CB
Grenoble_VIF
May-2017
Aug-2017
Nov-2017
Feb-2018
(B)
[Glucose] ng m-3
(C)
Marnaz
Oct-2013
Feb-2014
Jun-2014
Oct-2014
Feb-2015
(D)
(E)
Nice
Sep-2014
Feb-2015
Jul-2015
Dec-2015
May-2016
Oct-2016
(F)
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OPE-ANDRA
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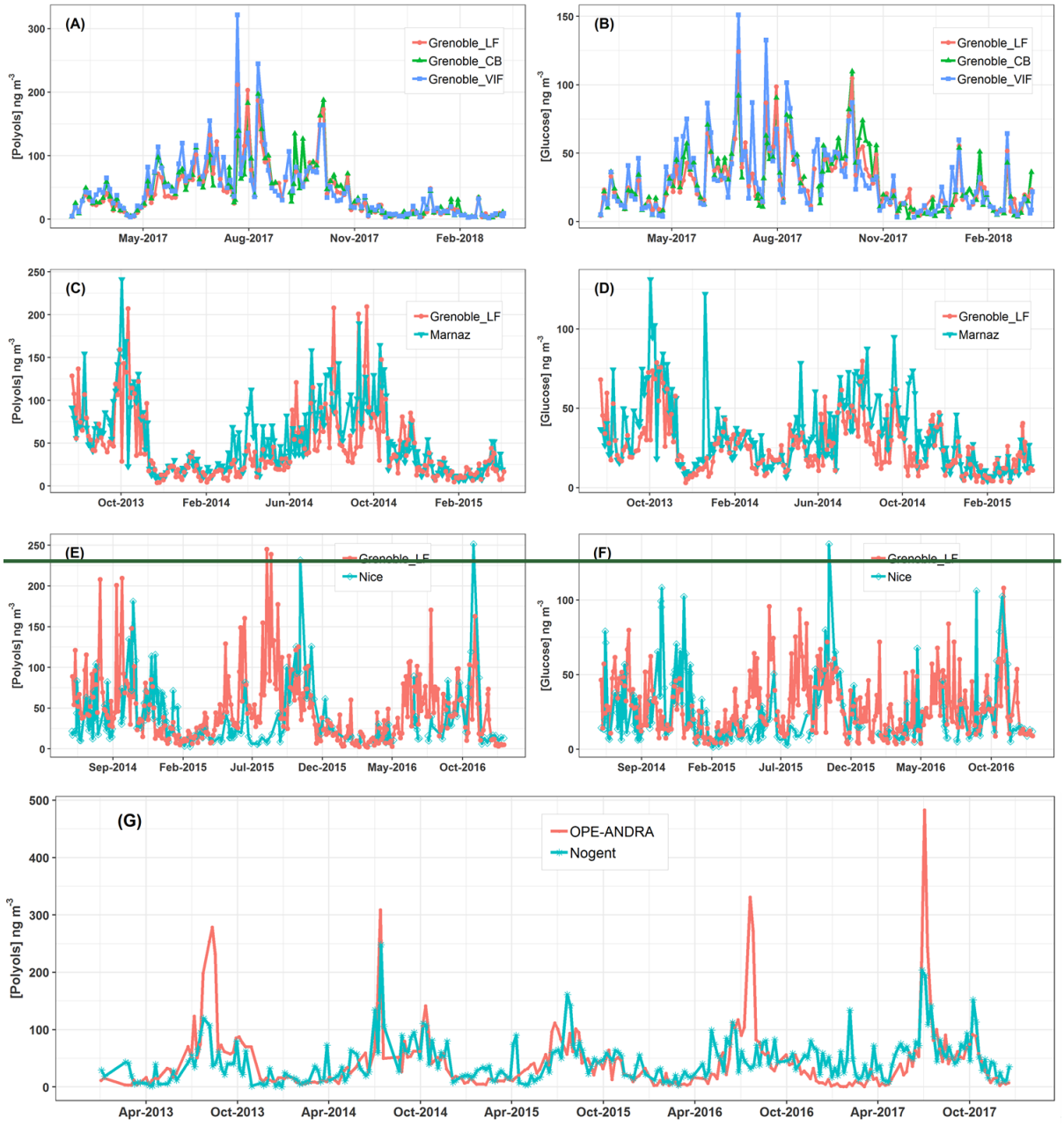
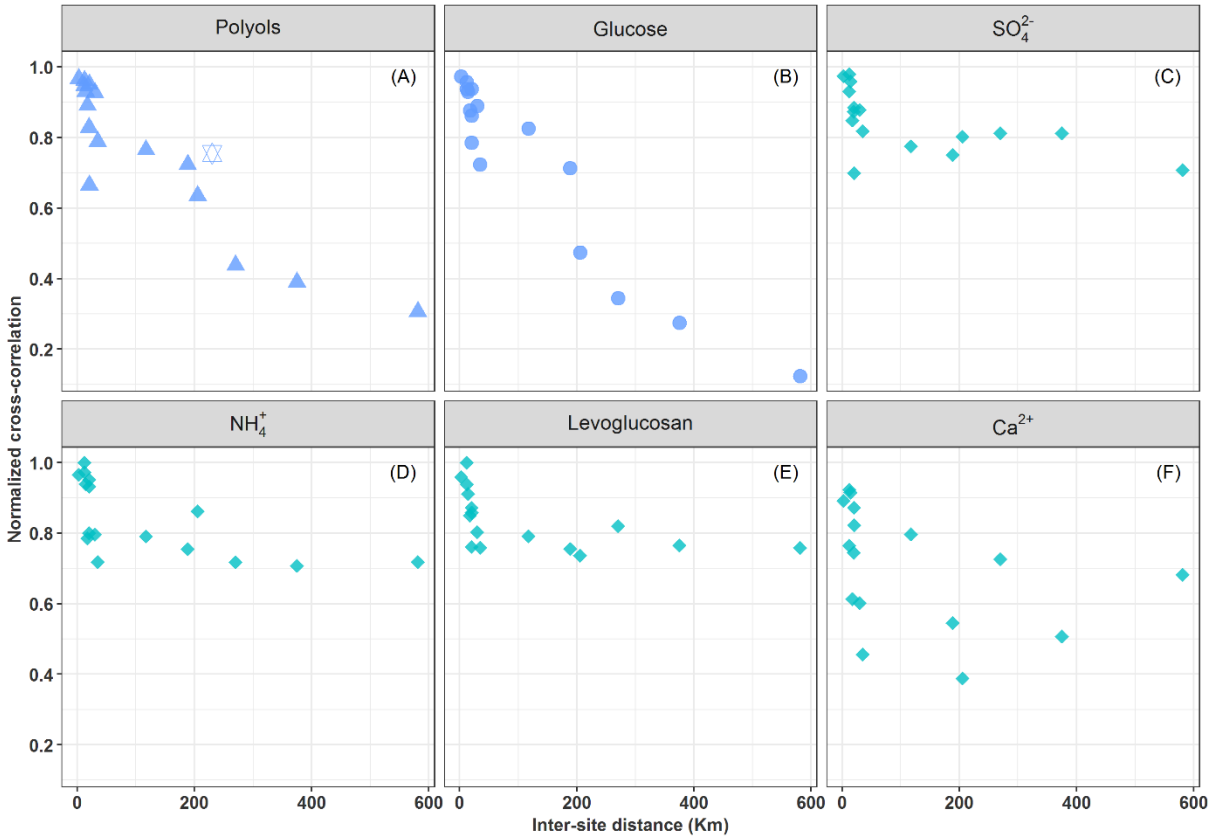


Figure 4: Concentrations (in ng m^{-3}) of (left) ambient particulate polyols (defined as the sum of arabitol and mannitol) and glucose (right) over different monitoring sites in France. Since PM_{10} were collected every 3-days at Nogent-sur-Oise and 6-days at OPE-ANDRA, the original data sets are averaged 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 ~~glucose~~ and glucose ~~polyols~~ (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-Oise (high correlation for a distance above 230 km), both sites being located in highly-impacted agricultural areas. This overall pattern suggests that the processes responsible for the atmospheric concentrations of SC present a spatial homogeneity over typical areas of at least several tens of km.



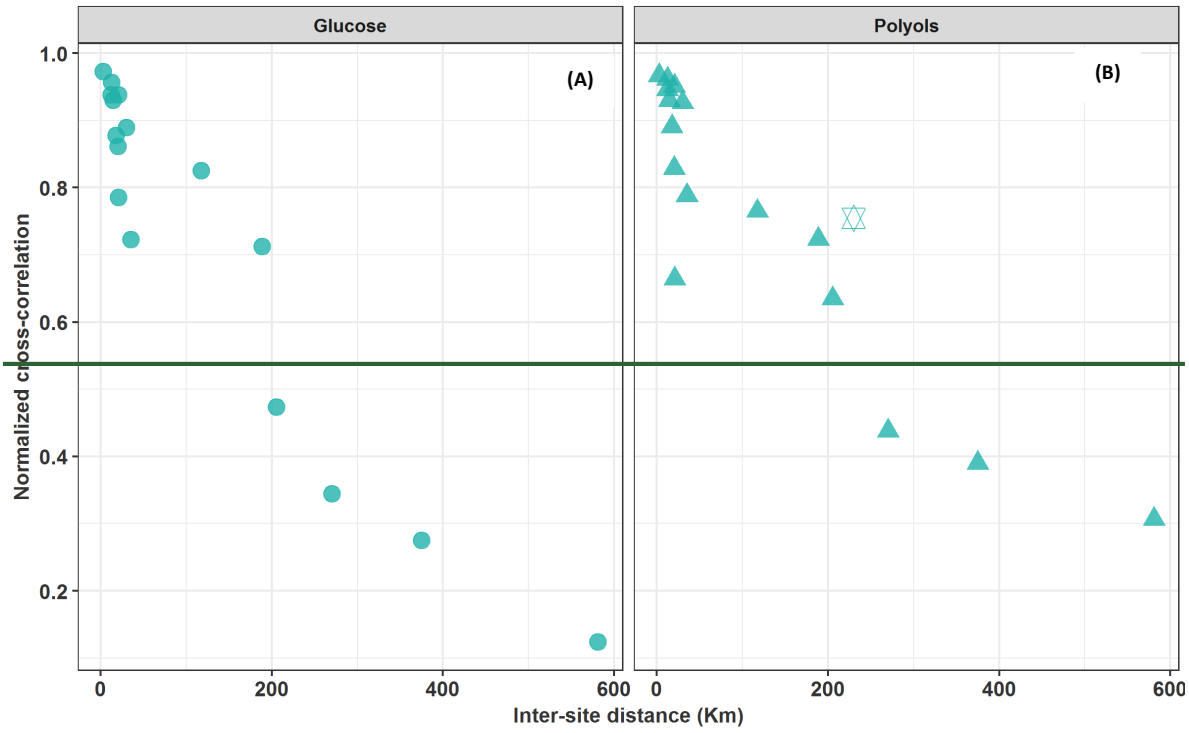


Figure 5: Normalized cross-correlation values for the daily evolution of particulate ~~glucose~~ ~~polyols~~ (A), ~~glucose~~ ~~and polyols~~ (B), sulfate (C), ammonium (D), levoglucosan (E) and calcium (F) concentrations over pairs of sites located at multiple increasing space scales across France. The hexagram corresponds to the correlation between the sites of OPE-ANDRA and Nogent-sur-Oise, both sites being surrounded by crop field areas.

Unlike SC, ambient air concentrations of sulfate (Fig. 3C) and ammonium (Fig. 3D), associated with long-range aerosol transport (Abdalmogith and Harrison, 2005; Amato et al., 2016; Coulibaly et al., 2015; Pindado and Perez,

2011; Waked et al., 2014)~~(Abdalmogith and Harrison, 2005; Amato et al., 2016; Coulibaly et al., 2015; Pindado and Perez, 2011; Waked et al., 2014)~~ and levoglucosan ((Fig. 3E), associated with biomass burning in cold season (Weber et al., 2019; Xiao et al., 2018), display stronger positive correlations ($R > 0.72$-0.98, $p < 0.01$) at all pairs of sites considered in the present work~~ (Figure S2)~~. Moreover, ambient concentrations of calcium (Fig. 3F), associated with local fugitive dust sources or/and long-range aerosol transport (Ram et al., 2010; Wan et al., 2019) display random correlation patterns ~~(Figure S2)~~. These results are in agreement with Zhu et al. (2018) who also reported non-significant correlations between SC and sulfate in $PM_{2.5}$ aerosols measured at Shanghai, China. The distinct spatial behaviors between sulfate (or Ca^{2+}) and SC in the present work further suggest a dominant regional influence for atmospheric SC, as opposed to processes associated with either local sources for calcium or long-range transport for sulfate.

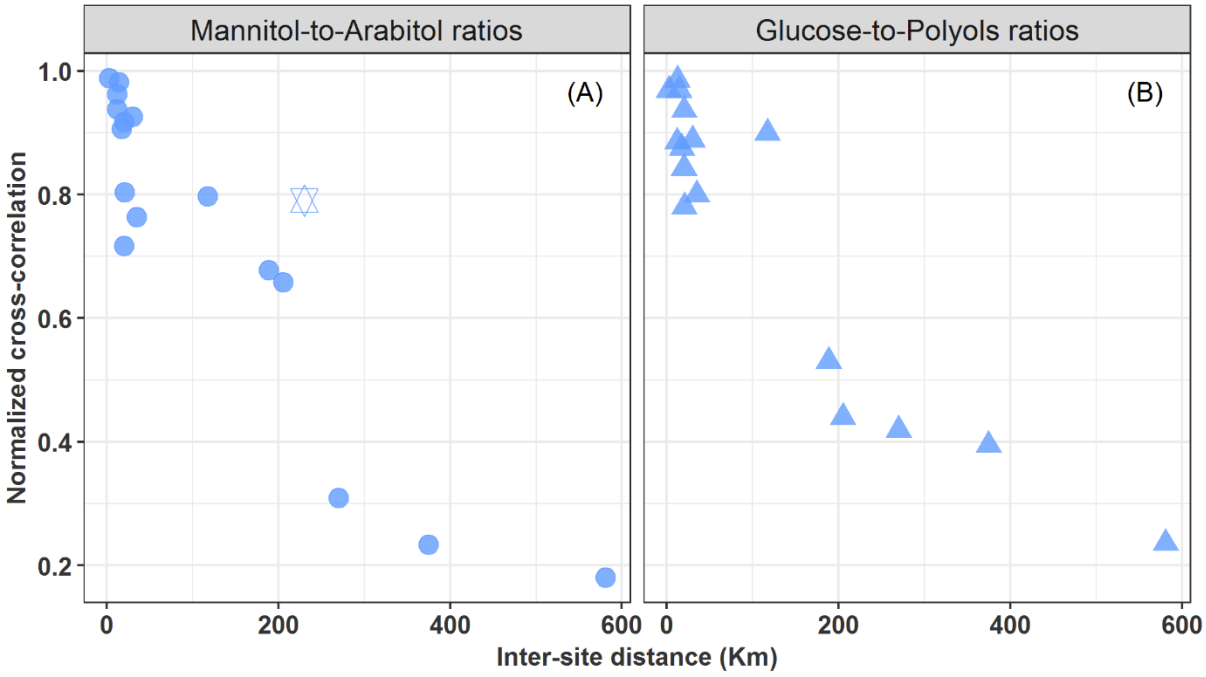
Mannitol and arabitol are well-known materials of fungal spores, serving as osmo-regulatory solutes (Medeiros et al., 2006b; Simoneit et al., 2004; Verma et al., 2018; Xiao et al., 2018; Zhang et al., 2015)~~(Medeiros et al., 2006b; Simoneit et al., 2004b; Verma et al., 2018; Zhang et al., 2010, 2015)~~. Based on parallel measurements of spore counts and PM_{10} polyol concentrations at three sites within the area of Vienna (Austria), Bauer et al. (2008a) found an average arabitol and mannitol content per fungal spores of respectively 1.2 pg spore^{-1} (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 often identified in the green algae and lower plants (Buiarelli et al., 2013; Di Filippo et al., 2013; Gosselin et al., 2016; Vélëz et al., 2007; Xu et al., 2018; Zhang et al., 2010)~~(Buiarelli et al., 2013; Di Filippo et al., 2013; Vélëz et al., 2007; Xu et al., 2018; Zhang et al., 2010)~~. Gosselin et al., 2016 observed a relatively low ($R^2 = 0.31$) to high ($R^2 = 0.84$) coefficient of determination between mannitol and arabitol for total suspended particles (TSP) collected at a pine-forested area during dry and rainy periods, respectively. High correlation in rainy periods possibly suggested that both chemical species in the TSP fraction in this pine-forested area could have been derived mainly from the same sources, i.e., actively wet-discharged ascospores and basidiospores, while the relatively poor correlation in dry periods could have been likely due to more complex sources, i.e., dry discharged spores, plants, algae, etc. Being important chemical species for the metabolism of the~~se~~ microorganisms (Shcherbakova, 2007), it may well be that the concentration ratio of mannitol-to-arabitol could deliver some information on the spatial or temporal evolution of their emission processes (Gosselin et al., 2016)~~(Gosselin et al., 2016)~~. The annual average mannitol-to-arabitol 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 those in the cold period (Dec-May) (Table S1). These ratios are within the range of those previously reported for PM_{10} aerosols collected at various urban and rural background sites in Europe (Bauer et al., 2008; Yttri et al., 2011b)~~(Bauer et al., 2008a; Yttri et al., 2011b)~~. Similarly, Burshtein et al., (2011) also reported comparable ratios for PM_{10} aerosols collected during autumn and winter from a Mediterranean region in Israel.

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

However, as evidenced in Fig.~~ure~~ 4, both mannitol-to-arabitol and glucose-to-polyols ratios show a clear distance-dependent correlation, with higher correlations ($R = 0.64$ to 0.98, $p < 0.01$) observed for pairs of sites within 150-190 km distance. This spatial consistency highlights once again that the dominant emission processes should be effective regionally, rather than being specific local input processes, and that atmospheric dynamics of the concentration levels (i.e., driven by the interplay of emission and removal processes) are determined by quite

similar environmental factors (e.g. meteorological conditions, vegetation, land use, etc.) at such a regional scale. This implies that local events and phenomena, such as the mechanical resuspension of topsoil and associated biota (like bacteria, fungi, plant materials, etc.) might not be their major atmospheric input processes, particularly in urban background areas typically characterized by less bare soil, and with a variable nature of the unpaved topsoil at the regional scale (Karimi et al., 2018)~~(Karimi et al., 2018)~~. Furthermore, Karimi et al. (2018)~~Karimi et al. (2018)~~ also recently reported heterogeneous topsoil microbial structure within patches of 43 to 260 km across different regions of France. It follows that the hypotheses of emissions related to mechanical resuspension of topsoil particles and associated biota, or microbiota emitted actively from surface soil into the air generally assumed in most pioneering reports (Medeiros et al., 2006b; Rogge et al., 2007; Simoneit et al., 2004; Wan and Yu, 2007)~~(Medeiros et al., 2006b; Rogge et al., 2007; Simoneit et al., 2004b; Wan and Yu, 2007)~~ are most probably not valid.

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



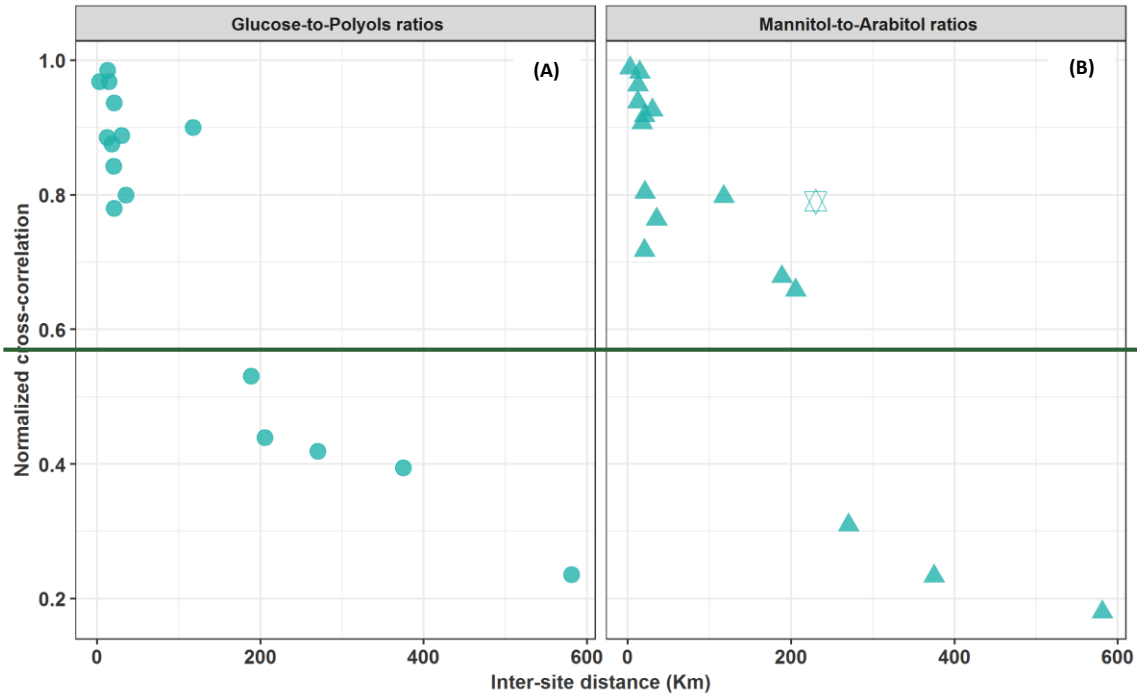


Figure 6: Normalized cross-correlation values for daily evolution of particulate mannitol-to-arabitol (A) glucose-to-polyols (BA) and ~~mannitol-to-arabitol (B)~~ ratios over pairs of sites located at multiple increasing space scales across France. The hexagram corresponds to the correlation between 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 corresponds

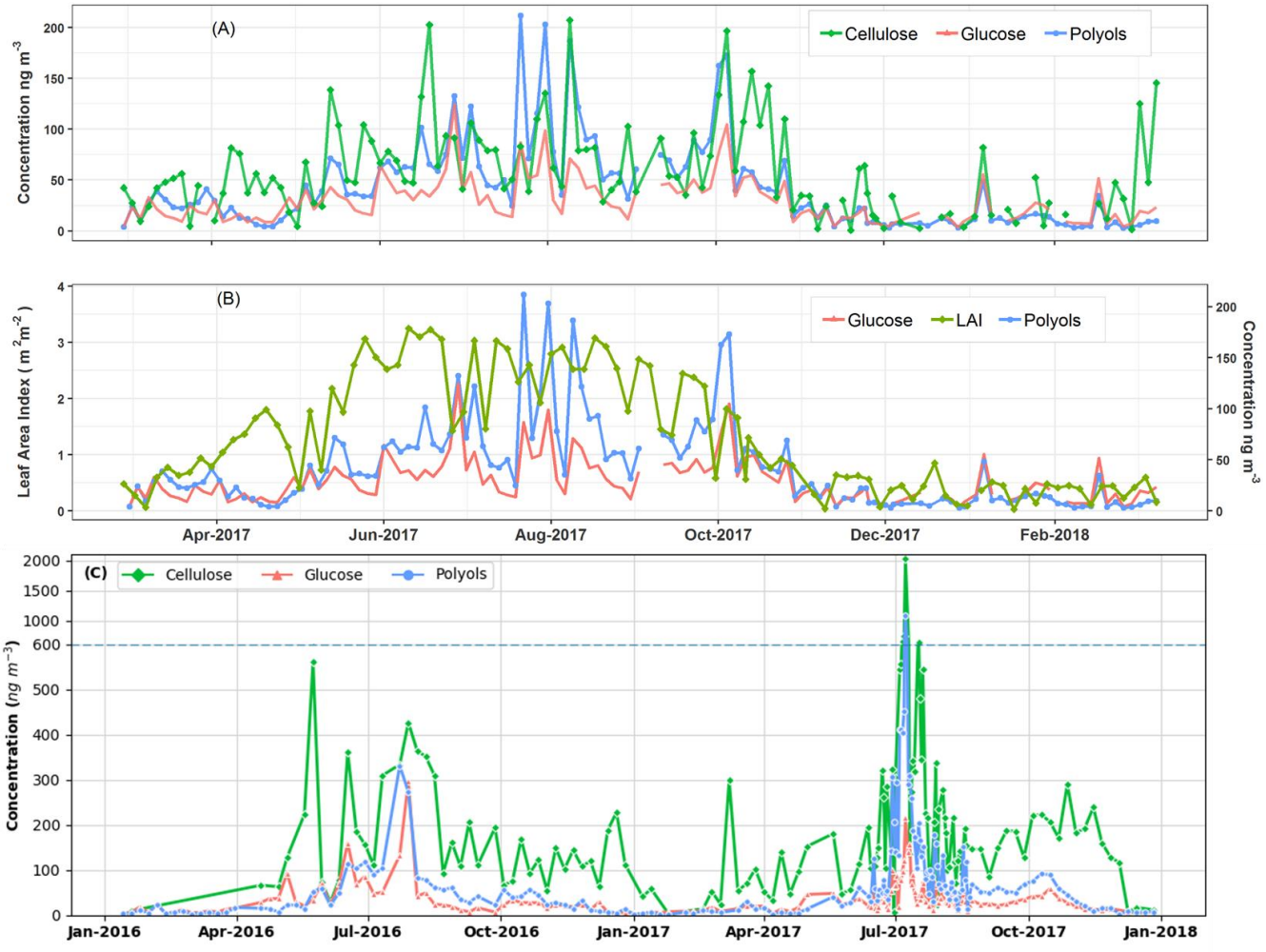
to respectively to 0.1 to 6.6 % and 0.01 to 5.3 % of total organic matter (OM) mass in PM_{10}. These values are comparable to those previously reported for various sites in Europe (Daellenbach et al., 2017; Sánchez-Ochoa et al., 2007; Vlachou et al., 2018; Yttri et al., 2011b)~~(Daellenbach et al., 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.~~ure~~ 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 (F~~f~~ig.~~ure~~ 5A). Moreover, the maximum atmospheric concentrations of polyols also mainly occur when the vegetation density (LAI) is at its highest in late summer (Fig.~~ure~~ 5B). Similar global behaviors are also observed for atmospheric particulate glucose 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 ~~these findings highlight that particulate SC 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)~~(Bozzetti et al., 2016; Burshtein et al., 2011; Daellenbach et al., 2017; Pashynska et al., 2002; Verma et al., 2018; Vlachou et al., 2018; 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)~~(Bozzetti et al., 2016)~~.

As also evidenced in Fig.~~ure~~ 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)~~(Bodenhausen et al., 2014; Bringel and Couée, 2015; Lindow and Brandl, 2003; Moricca and Ragazzi, 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)~~(Bozzetti et al., 2016; Burshtein et al., 2011; Pashynska et al., 2002; Yttri et al., 2007)~~.



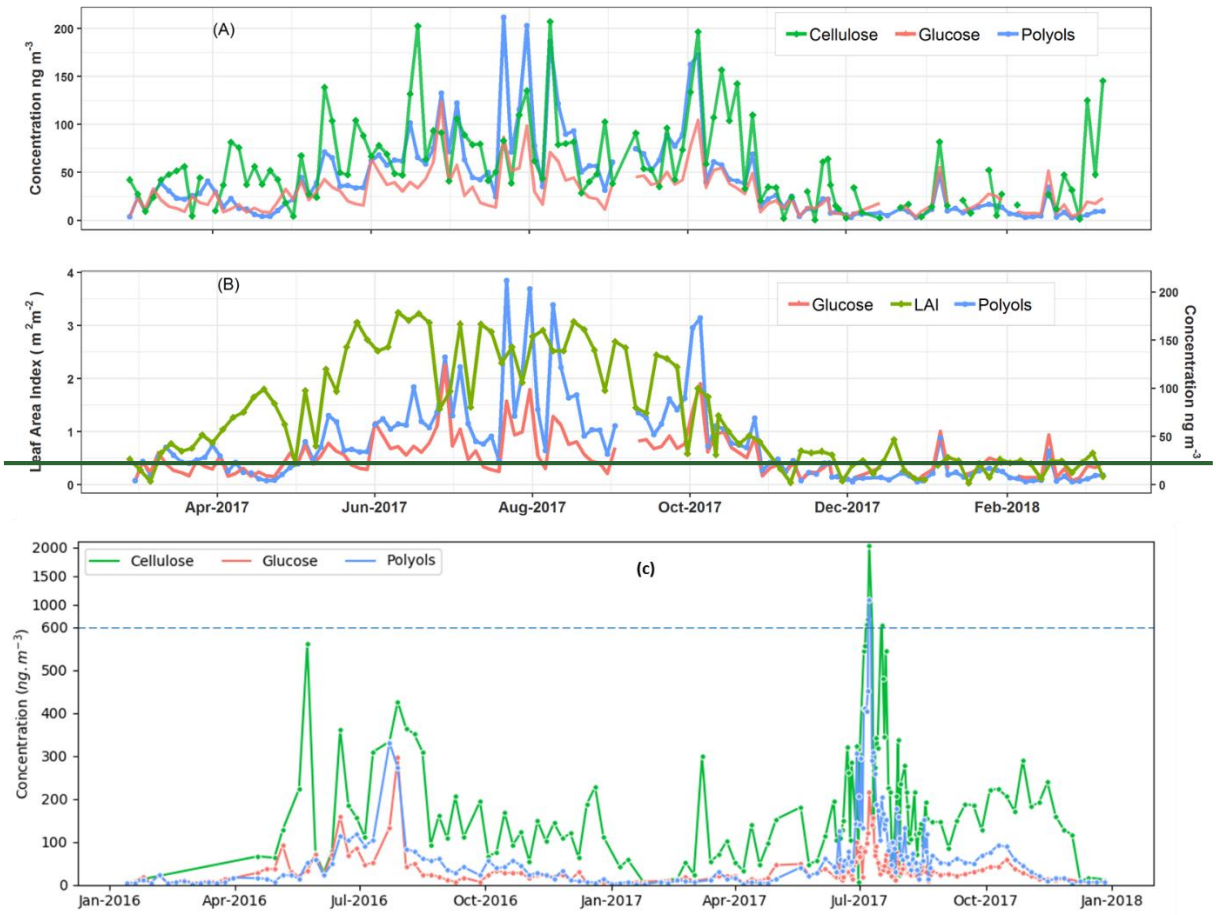


Figure 7: Temporal covariation cycles of the daily particulate polyols and glucose concentrations along with vegetation indicators at the urban background site of Grenoble (A and B) and the rural agricultural background site of OPE-ANDRA (C), respectively. Note that PM_{10} aerosols are intensively collected 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.~~ure~~ 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)~~(Claeys et al., 2004; Graham et al., 2003)~~.

Overall, the environmental factors including the mean night-time temperature, relative humidity, wind speed and the leaf area index explain up to 82 % (adjusted R^2 = 0.82, see Table 1) of the annual temporal variation cycles of SC concentrations. The mean night-time temperature and LAI contribute respectively to 54 % and 37 % of the observed annual variabilities of SC concentrations. The atmospheric humidity is also a driver for these chemical

species (3 % of the explained variation). These results are consistent with previous studies showing that concentrations of mannitol (in both PM_{10} and $PM_{2.5}$ size fractions) linearly correlate best with the LAI, atmospheric water vapor and temperature (Heald and Spracklen, 2009; Hummel et al., 2015; Myriokefalitakis et al., 2017)~~(Heald and Spracklen, 2009; Hummel et al., 2015)~~. All of these drivers have been previously shown to induce the initial release and influence the long-term airborne microbial (i.e. bacteria, fungi) concentrations (China et al., 2016; Elbert et al., 2007; Grinn-Gofroń et al., 2019; Jones and Harrison, 2004; Rathnayake et al., 2017; Zhang et al., 2015).

Besides, the wind speed (range of 0.2 to 5.6 m s^{-1}) seems an additional effective driver affecting the contribution of the local vegetation to SC concentrations in the atmosphere. Albeit enough air movement is required to passively release microorganisms along with plant debris into the atmosphere, strong air motions induce higher dispersion. These observations are in good agreement with those previously reported (Jones and Harrison, 2004; Liang et al., 2013; Zhang et al., 2010, 2015; Zhu et al., 2018b)~~(Jones and Harrison, 2004; Liang et al., 2013; Zhang et al., 2010, 2015; Zhu et al., 2018)~~. For instance Liang et al. (2013)~~Liang et al. (2013)~~ have found a negative correlation between wind speed and polyols concentrations, and the highest atmospheric fungal spores concentrations were observed for a wind speed range of 0.6 to 1.0 m s^{-1}.

Table 1: Multiple linear regression for ambient polyols and glucose concentrations and their effective environmental factors at the Marnaz site. Contributions of predictor are normalized to sum 1. "Relaimpo package under R" was 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

.

One of the limitations of this study is that 4-day averaged observations do not allow to evaluate the driver contributions that might explain some short term events for which the influence of meteorological parameters such as rainfall or solar radiation could also be significant (Grinn-Gofroń et al., 2019; Heald and Spracklen, 2009; Jones and Harrison, 2004)~~(Grinn-Gofroń et al., 2019; Heald and Spracklen, 2009; Jones and Harrison, 2004)~~. However, such simple parameterizations could be a first step in considering SC concentrations in CTM models, and further work is required in this direction in order to generate a robust parametrization of the emissions.

3.5 Specific case of a highly-impacted agricultural area

This section focuses on evidencing the environmental drivers of PM_{10} SC concentrations specific to agricultural 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 ~~29~~3923~~3~~ 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.~~ure~~ 6). Year 2015 has been found to be particularly hot and dry at OPE-ANDRA (Fig.~~ure~~ 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)~~(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 ~~OPE~~ site (Fig.~~ure~~ 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)~~(Golly et al., 2018; Rogge et al., 2007; Simoneit et al., 2004b)~~. 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.

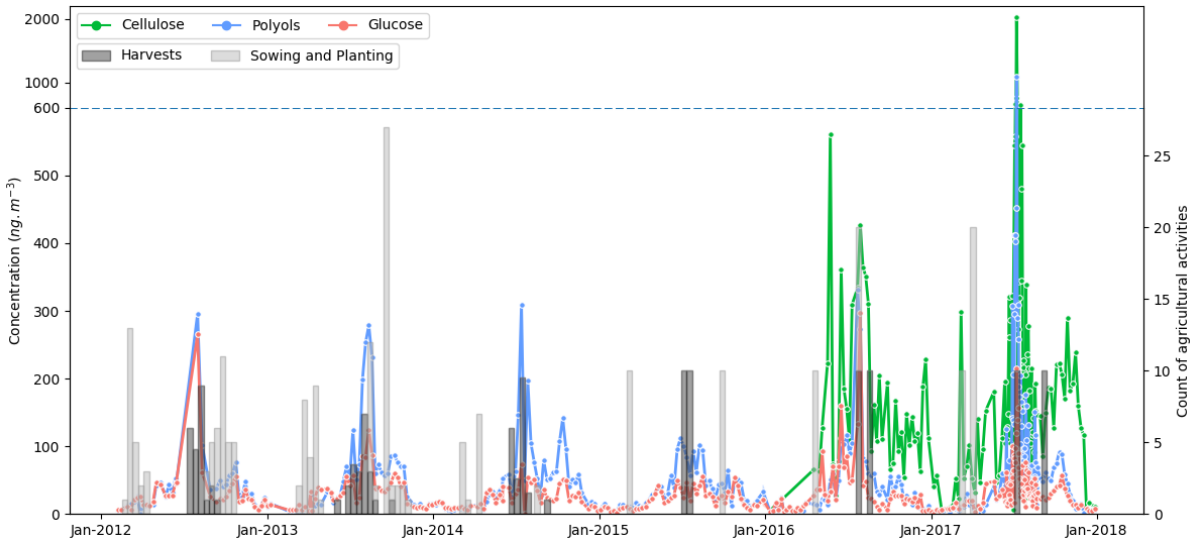


Figure 8: Daily evolution cycles of polyols and glucose concentrations in aerosols collected from the OPE-ANDRA monitoring site, from 2012 to 2018. Cellulose concentrations have been measured from January 2016 to January 2018. Colored bars correspond to the sum of the various agricultural practices performed (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.

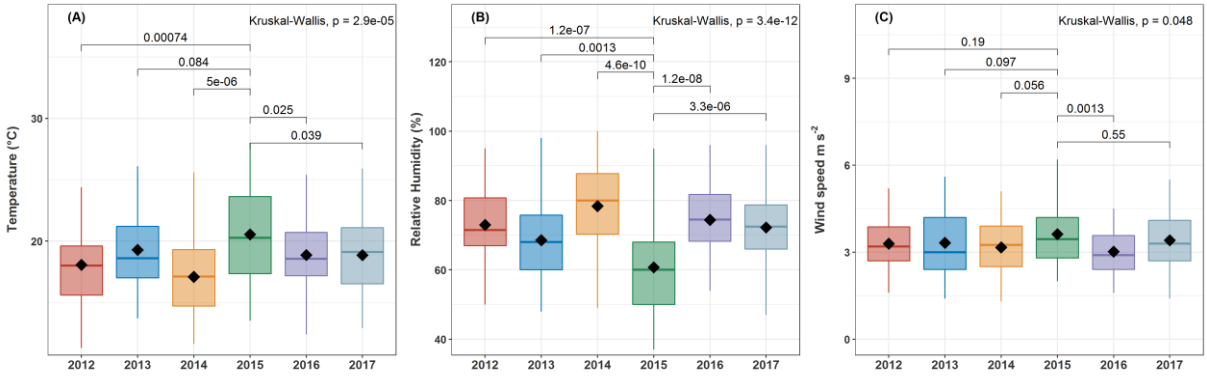


Figure 9: Boxplots of (A) maximum ambient temperature, (B) relative humidity and (C) wind speed at OPE-ANDRA from 2012 to 2017. Analyses are performed for warmer periods (June to November). Only statistically different meteorological factors are presented. The black marker inside each boxplot indicates 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).

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

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

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Author contributions: JLJ was the (co-)supervisor for the PhD for AS, FC, SW, and for the post-doc of DS, BG, and AW. He directed all the personnel who performed the analysis at IGE. He is the coordinator for the CNRS LEFE-EC2CO CAREMBIOS program that is funding the work of AS. GU and JMF-M were the co-supervisor for the PhD of AS or SW. EP, OF, and VR supervised the PhD of DMO who investigated the sites in northern France. OF, JL-J, JL-B, AA and NM were coordinating and partners of the different initial programs for the collection and chemical analysis of the samples. VJ developed the analytical techniques for polyols and cellulose measurements. TC performed the cellulose measurements. Samples analyses at LSCE were performed by NB. AC gave advices for the statistical aspects of the data processing. AS and JLJ processed the data and wrote up the manuscript. SW participated to the visualization of the results. SC is supervising the OPE station and collected the agricultural activities records. All authors from AASQA (author affiliation nos. 9~~7~~ to 16~~4~~) are representatives for each network that conducted the sample collection and the general supervision of the sampling sites. All authors reviewed and commented on the manuscript.

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

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