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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Arabitol, mannitol and glucose as tracers of primary biogenic organic aerosol: influence of environmental factors on ambient air concentrations and spatial distribution over France" *by* A. Samaké et al.

A. Samaké et al.

abdoulaye.samake2@univ-grenoble-alpes.fr

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ACP-2019-434 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 54âĂŤ59). 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 R2 between arabitol and mannitol (lines 288âĂŤ292), as follows: lines 282-293.

"Based on parallel measurements of spore counts and PM10 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 (R2 = 0.31) to high (R2 = 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 inïňĆuenced 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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Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2019-434/acp-2019-434-AC3-supplement.pdf

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