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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 #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 envi-

ronmental 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 PM10. 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 . Results do not indicate clear source region(s) (Figure R2.1). 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 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.

(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 PM2.5

aerosols measured at Shanghai, China. The distinct spatial behaviors between sulfate (or Ca2+) 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 (Ca2+) and chemically stable species (levoglucosan, Figure R2.1B), 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).

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2019-434/acp-2019-434-AC2-supplement.pdf

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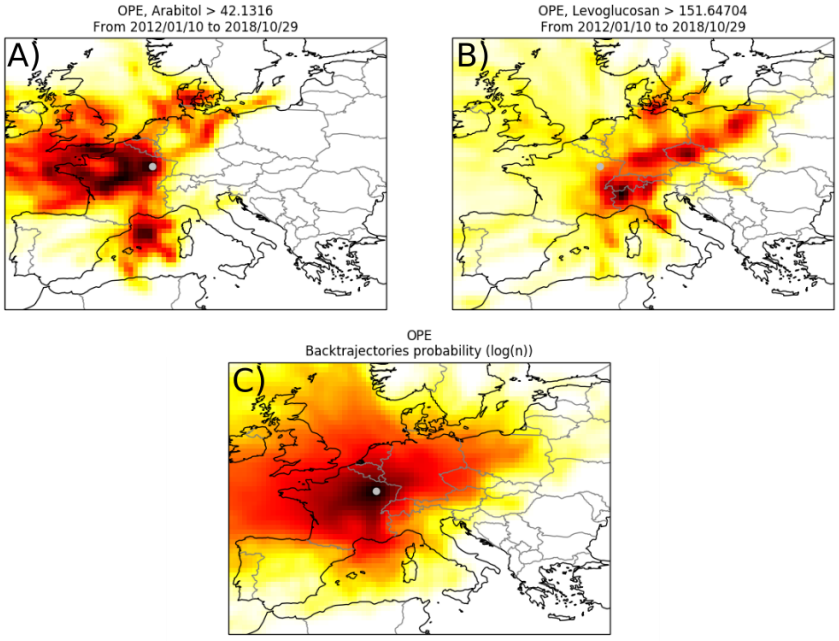

Fig. 1. PSCF analysis for the OPE site (using pyPSCF and HYSPLIT). Back-trajectories associated with arabitol concentrations higher than the 75th percentile divided by the number of back-trajectories.