

General comments:

In this study the authors reported measurement of PM_{2.5} component over 3 different sites in China during a sampling period of 1 month, during spring 2019. Different saccharides were measured, including biomass burning proxy such as levoglucosan, manosan and galactosan, as well as more uncommon mono(di)saccharide, aiming at tracing the primary biogenic and possibly secondary biogenic sources. After a discussion on the potential link between emissions sources based on correlation and ratio of species, the authors attempt a source-apportionment of the different saccharide using a Non-Negative matrix Factorization (NMF) method and successfully identify 5 different factors of saccharides.

This interesting study reports a comprehensive observational dataset (although not covering the full year) and gives useful insight concerning the sources of organic components thanks to the use of proxy species not-usually used in the literature.

Reply:

Dear Prof. Samuel Weber,

We appreciate the positive comments and suggestions about the manuscript. We agree with the reviewer's comments, and have updated the manuscript on the basis of these suggestions.

Specific comments:

- Samake et al. (2019) highlight that the different polyols are mostly in the coarse fraction of the PM. Also, it has been hypothesis that the different size distribution of polyols may be a proxy of the different microbiota. Did the authors have also sampled the PM₁₀ fraction and could provide the size distribution of the different saccharides?

Reply: Thank for the reviewer's suggestion. Indeed, previous results have indicated that polyols (especially mannitol and arabitol) and glucose were prevalent existed in the coarse fraction (Fu et al., 2012; Fuzzi et al., 2007; Pio et al., 2008; Yttri et al., 2007), and were mainly associated with the coarse PM fraction (Samaké et al., 2019). But PM₁₀ fraction was not collected due to some practical difficulties, we can't provide the size distribution of the saccharides in this study.

We've cited a reference and rephrased the sentence in line 428-430. "The contribution of fungal spores might be underestimated because previous results had indicated that mannitol and arabitol were mainly associated with the coarse PM fraction (Samaké et al., 2019)."

- The source apportionment (SA) is a very interesting part, although it lacks of important information that should be reported: Why didn't you included the whole species available in the SA? It could help identify more robustly BB, but also

saccharides from soil resuspension (with Ca^{2+}), and moreover quantify the apportionment of the different factors to the total $\text{PM}_{2.5}$ mass.

Reply: The source apportionment including the other species could quantify the apportionment of the different factors to the total $\text{PM}_{2.5}$ mass. We have tried to include the whole species available in the source apportionment. To make the result be better correlate with the five sources of saccharides, we ran a five-factor NMF. The result is shown as below.

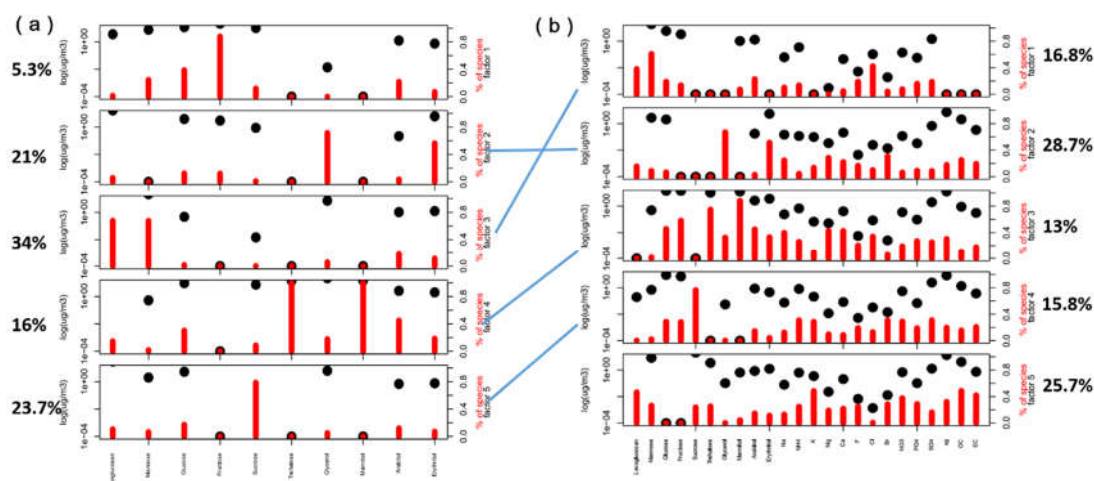


Figure 1. The factor profile obtained by NMF analysis based on the saccharide components (a) and the factor profile based on all the species (b).

In Figure 1a, the sources of plant detritus (factor 1), plant senescence (factor 2), biomass burning (factor 3), soil microbiota (factor 4) and airborne pollen (factor 5) respectively contributed 5.3%, 21.0%, 34%, 16.0% and 23.7% to the total saccharides. We matched the factors one-to-one in the two figures according to the characteristic saccharide species. The other various species showed decentralized load on these factors. Based on the compositional data of saccharides, five factors associated to the total $\text{PM}_{2.5}$ mass were correspond one-to-one to the factors associated to the total saccharides. Factor 1-4 were correspond to the sources of biomass burning, soil microbiota, plant senescence and airborne pollen, respectively. Factor 5 was more appropriate to be thought as a mixed source.

Thus, in Figure 1b, the sources of biomass burning (factor 1), plant senescence (factor 2), soil microbiota (factor 3), airborne pollen (factor 4) and mix sources (factor 5) respectively contributed 16.8%, 28.7%, 13%, 15.8% and 25.7% to the total $\text{PM}_{2.5}$ mass. However, we think the naming of these factors associated to the total $\text{PM}_{2.5}$ mass are not accurate and comprehensive. In order to get more clear information about the sources and their contribution to the total saccharides, we decided to only report the source apportionment of saccharides.

- It is stated that the SA is still uncertain, but no estimation of the uncertainties is given. It would be of great interest to report the species uncertainties, for instance with bootstrapping your input data.

Reply: We only have 91 samples in total, so we cannot carry out resampled runs for many times. The analytical uncertainty was high in present study due to the limited sample number by using the currently used formula in PMF model. We used 0.3 plus the analytical detection limit for estimating uncertainty according to the method of Xie et al. (1999). The constant 0.3 corresponding to the log(Geometric Standard Deviation, GSD) was calculated from the normalized concentrations for all measured species, and was used to represent the variation of measurements. The use of GSD was suitable for our measurement set in a small sample size.

- The timeserie contribution would also be of great interest. Even if the authors did not include a total variable (namely, PM_{2.5}), the timeserie of the total saccharide for the 5 factors would be informative.

Reply: We agree with the reviewer's view of the importance on the timeserie contribution. The timeserie of the total saccharide for the 5 factors are shown in Figure S5. We've rewritten the relevant content from Line 525. "During the sampling periods, daily variations on proportion of the five factors are shown in Figure S5. Factor 2 soil microbiota emissions could be associated to soil reclamation and cultivation of farming periods, and factors 3 plant senescence and factor 5 plant detritus could be associated to harvesting of vegetation or crop. During the observation period of a month, along with the weather warming as sunshine enhanced, human left two obvious traces of cultivated soil during 9-17 March and 27 March-8 April and a trace of vegetation or crop harvest during 17-30 March. The stronger pollen discharge occurred in March, probably due to the flowering of certain plants. The BB emissions peaked on 9, 16 March, and 1 April were more prone to be open burnings."

- The "Soil microbiota" factor, identified mainly by the presence of Trehalose and Mannitol (and Arabitol) denotes with the finding of Samake et al. (2020) that found that Arabitol and Mannitol are associated with fungi and bacteria from the leaves and not with the soil (even if some mixing are probable). I would suggest naming it "Soil and leave microbiota".

Reply: We agree with the reviewer's suggestion, "Soil and leaves microbiota" is more specific. We've named it "Soil and leave microbiota" and gave an explanation in line 502-507. "These saccharide compounds had all been detected in the suspended soil particles and associated microbiota (e.g., fungi, bacteria and algae) (Simoneit et al., 2004; Rogge et al., 2007). A recent study found that leaves were a major source

of saccharides-associated microbial taxa in a rural area of France (Samaké et al., 2020). Hence, this factor was attributed to soil and leaves microbiota.”

- Overall, the naming of the different factors identified is too rapidly explained, and more detailed could be written to ease the interpretation of the different factors.

Reply: Since each type of sugar has been described in the text, the factors were resolved in a little brief way. In the new version, the naming of the different factors have been more detailed explained from Line 497.

“As shown in Figure 6a, factor 1 was characterized by high level of levoglucosan (71.8%) and mannosan (78.7%), suggesting the source of BB (Simoneit et al., 1999; Nolte et al., 2001). Factor 2 was characterized by trehalose (99.9%) and mannitol (100.0%), and was enriched in the other saccharides components, i.e., arabitol (44.1%), glucose (29.6%), erythritol (18.2%), glycerol (17.8%), levoglucosan (14.7%), and sucrose (8.6%). These saccharide compounds had all been detected in the suspended soil particles and associated microbiota (e.g., fungi, bacteria and algae) (Simoneit et al., 2004; Rogge et al., 2007). A recent study found that leaves were a major source of saccharides-associated microbial taxa in a rural area of France (Samaké et al., 2020). Hence, this factor was attributed to soil and leaves microbiota. Factor 3 has high levels of glycerol (71.4%) and erythritol (58.2%), and showed loadings of glucose (12.8%) and fructose (11.8%). Kang et al. (2018) reported that glycerol and erythritol presented larger amounts in winter and autumn, when the vegetation decomposed. This factor was thought as the sources from plant senescence and decay by microorganisms. Factor 4 exhibited a predominance of sucrose (78.7%), and showed loadings of glucose (17.2%), arabitol (11.8%). This factor was regarded as the source of airborne pollen, because pollen is the reproductive unit of plants and contains these saccharides and saccharide alcohols as nutritional components (Bielecki, 1995; Miguel et al., 2006; Fu et al., 2012). Factor 5 characterized by the dominance of fructose (88.2%) was resolved, and was enriched in glucose (38.2%) and arabitol (21.2%), thus it could be regarded as the source of plant detritus.”

Minor comment:

- Please provide the pie chart of Figure 6b in a non-3D way, as the relative proportion is much harder to see in 3D compare to regular 2D graph.

Reply: We agree with the reviewer’s comment. We’ve provided the pie chart of Figure 6b in a 2D way in the new version of manuscript.

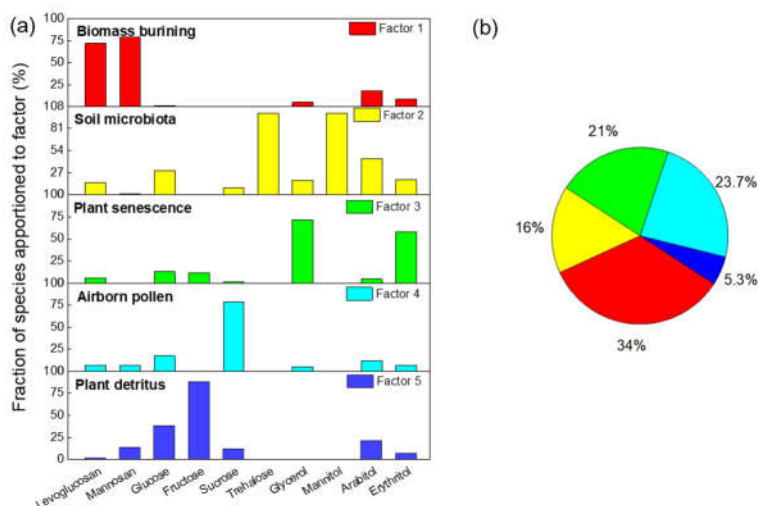


Figure 6. Factor profile obtained by NMF analysis (a). Source contribution of the five factors to the total saccharides in PM_{2.5} samples (b).

References:

- Fuzzi, S., Decesari, S., Facchini, M. C., Cavalli, F., Emblico, L., Mircea, M., Andreae, M. O., Trebs, I., Hoffer, A., Guyon, P., Artaxo, P., Rizzo, L. V., Lara, L. L., Pauliquevis, T., Maenhaut, W., Raes, N., Chi, X., Mayol-Bracero, O. L., Soto-García, L. L., Claeys, M., Kourtchev, I., Rissler, J., Swietlicki, E., Tagliavini, E., Schkolnik, G., Falkovich, A. H., Rudich, Y., Fisch, G., and Gatti, L. V.: Overview of the inorganic and organic composition of size-segregated aerosol in Rondônia, Brazil, from the biomassburning period to the onset of the wet season, *J. Geophys. Res.*, 112, D01201, <https://doi.org/10.1029/2005JD006741>, 2007.
- Pio, C. A., Legrand, M., Alves, C. A., Oliveira, T., Afonso, J., Caseiro, A., Puxbaum, H., Sanchez-Ochoa, A., and Gelencsér, A.: Chemical composition of atmospheric aerosols during the 2003 summer intense forest fire period, *Atmos. Environ.*, 42, 7530–7543, <https://doi.org/10.1016/j.atmosenv.2008.05.032>, 2008.
- Samaké, A., Jaffrezo, J.-L., Favez, O., Weber, S., Jacob, V., Albinet, A., Riffault, V., Perdrix, E., Waked, A., Golly, B., Salameh, D., Chevrier, F., Oliveira, D. M., Bonnaire, N., Besombes, J.-L., Martins, J. M. F., Conil, S., Guillaud, G., Mesbah, B., Rocq, B., Robic, P.-Y., Hulin, A., Meur, S. L., Descheemaeker, M., Chretien, E., Marchand, N., and Uzu, G.: Polyols and glucose particulate species as tracers of primary biogenic organic aerosols at 28 French sites, 19, 3357–3374, <https://doi.org/10.5194/acp-19-3357-2019>, 2019.
- Samaké, A., Bonin, A., Jaffrezo, J.-L., Taberlet, P., Weber, S., Uzu, G., Jacob, V., Conil, S., and Martins, J. M. F.: High levels of primary biogenic organic aerosols are driven by only a few plant-associated microbial taxa, 20, 5609–5628, <https://doi.org/10.5194/acp-20-5609-2020>, 2020.