Anonymous Referee #2

Verma et al. discuss observations of sugars found in the aerosol-phase collected for 1 month in a forested site north of Beijing. The aerosol were collected onto filters and analyzed for the sugars. The authors then describe the pattern of the various sugars throughout the study period and speculate the sources via differences in day- and night-time mass concentration, wind patterns, and PMF. They discuss 5 potential sources, including biomass burning, vegetation, microbial and soil dust, pollen, and fungal.

The results presented here may be of interest to the audience and its scope generally fits within a measurement report. However, along with the concerns discussed by Reviewer #1, the authors nee to address the comments and concerns presented below prior to consideration for publication in ACP.

Major: (1) Statistics: Throughout the text, the authors state that the results are statistically different. However, conducting the t-test with the mean and standard deviation values listed in the table, majority of the observations are statistically similar at the 95% confidence interval and not statistically different. The lack of statistical difference in the observations makes many of the statements the authors use to differentiate day/night and thus sources less substantiated. Further, the correlations shown by the authors in Fig. 5 have very low R values (as stated throughout the text) and suggest that many of the correlations only explain 50% or less of the mass concentration.

Response: We found a positive correlation between sucrose and ambient temperature (r = 0.52), sucrose and solar radiation (r = 0.55), mannitol and RH (r = 0.57), trehalose and arabitol (r=0.58), trehalose and mannitol (r=0.58), with significance levels <0.05. Therefore we mentioned those positive linear correlation values in the text and discussed accordingly.

(2) Contextualization of results: I agree with Reviewer #1 that the listing of numbers from prior results makes it difficult to understand the conclusions in each section and the whole paper. Further, as highlighted with point (1) above, the data not being statistically different makes sections 3.1.1 thru 3.1.3 very long and repetitive. Also, the listing of numbers from prior studies to ascribe sources for the sugars makes the source apportionment very uncertain. This is also relevant for Section 3.5, where they found no differences in the levoglusoan/mannosan ratio and spend 1.5 pages on this. If this is important, it could be summarized in one paragraph at most.

Response: We deleted repeated sentences, several comparisons with previous studies and significantly modified section 3.1.1. to 3.1.3. We also shorten section 3.5. Please see section 3.1.1 - 3.1.3 and 3.5 in the revised MS.

(3) Methods: Reviewer #1 highlighted many of the methods that should be discussed in more detail. Further, PMF needs to be described in more detail to understand how the 5 results were determined (e.g., how many solutions were there allowed to be, how did the time series look, were the results compared against and investigated against external variables, etc.). Also, agree with Reviewer #1 in how were WSOC, OC, Ca2+, etc determined. Response: Authors thank the reviewer's valuable comments and suggestions. We make significant changes in the manuscript especially in the section 2 (Materials and methods), and section 3 (Results and discussions). Please see the revised MS.

We added information about the PMF analysis in the revised MS. Please see lines 187 - 201, 389 - 407, 418 - 429, 442 - 445, 453 - 457, 460 - 462 in the revised MS.

We added Figure S-1 and S-2 as supporting information in the revised MS.

Figure S-1. The scatter plots between observed (input data) and predicted (modeled data) concentrations show statistical parameter (coefficient of determination (r), Intercept, and Slope) with linear equitation of individual sugar compounds. (A blue 1:1 line is provided on this plot for reference (a perfect fit would line up exactly on this line), and the regression line is shown as a dotted red line).

Figure S-2. The time series plots between observed (input data) and predicted (modeled data) concentrations of individual sugar compounds. Blue and red lines are shown for observed (input data) and predicted (modeled data) concentrations, respectively.

(4) PMF: I think this is the more interesting and compelling part of the paper. I highly recommend the authors spend more time expanding on this section while reducing the discussion in the other sections. As highlighted above, there are statistical concerns, thus shortening them while increasing the discussion about PMF, which had lower statistical concern.

Response: We added more sentences in the PMF analysis section. Please see line 386 - 468 in the revised MS.

(5) Figures: The x-axis/date is very hard to read in all figures. It is unclear what the values are shown in different colors in Fig. 3.

Response: We separated Figure 3 as Figures 4a-c, 5a-d and 6a-c. We also modified x-axis in the manuscript and changes are added in the figure captions.

Minor: Please review the grammar throughout the paper, as highlighted by Reviewer #1.

Response: According to the reviewer's suggestion, whole manuscript is properly checked for the grammatical mistakes.

Fig. 4



Fig. 5.



Fig. 6.



Supporting Information



Figure S-1. The Scatter plots between observed (input data) and predicted (modeled data) concentrations show statistical parameter (coefficient of determination (r), Intercept, and Slope) with linear equitation of individual sugar compounds. (A blue 1:1 line is provided on this plot for reference (a perfect fit would line up exactly on this line), and the regression line is shown as a dotted red line).



Figure S-2. The time series plots between observed (input data) and predicted (modeled data) concentrations of individual sugar compounds. (A blue line and redline shown as observed (input data) and predicted (modeled data) concentrations, respectively.

References:

- Akagi, S. K., Yokelson, R. J., Wiedinmyer, C., Alvarado, M. J., Reid, J. S., Karl, T., Crounse, J. D., and Wennberg, P. O.: Emission factors for open and domestic biomass burning for use in atmospheric models, Atmos. Chem. Phys., 11, 4039– 4072, https://doi.org/10.5194/acp-11-4039-2011.
- Claeys, M., Graham, B., Vas, G., Wang, W., Vermeylen, R., Pashynska, V., Cafmeyer, J., Guyon, P., Andreae, M. O., Artaxo, P., and Maenhaut, W.: Formation of secondary organic aerosols through photooxidation of isoprene, Science, 303, 1173–1176, https://doi:10.1126/science.1092805, 2004.
- Davis, J. M., and Loescher, W. H.: [¹⁴C]-Assimilate translocation in the light and dark in celery (*Apium graveokns*) leaves of different ages, Physiol. Plant, 79, 656-662, https://doi.org/10.1111/j.1399-3054.1990.tb00040.x, 1990.
- Davis, J. M., Fellman, J. K., and Loescher, W. H.: Biosynthesis of sucrose and mannitol as a function of leaf age in celery (*Apiulm graveolens* L.), Plant Physio., 186, 129-133, https://doi: 10.1104/pp.86.1.129, 1988.
- Després, V. R., Huffman, J. A., Burrows, S. M., Hoose, C., Safatov, A. S., Buryak, G., Fröhlich-Nowoisky, J., Elbert, W., Andreae, M. O., Pöschl, U., and Jaenicke, R.: Primary biological aerosol particles in the atmosphere: a review, Tellus B, 64, 15598, doi:10.3402/tellusb.v64i0.15598, 2012.
- Graham, B., Mayol-Bracero, O. L., Guyon, P., Roberts, G. C., Decesari, S., Facchini, M. C., Artaxo, P., Maenhaut, W., Koll, P., and Andreae, M. O.: Water-soluble organic compounds in biomass burning aerosols over Amazonia 1. Characterization by NMR and GC-MS, J. Geophys. Res., 107(D20), 8047, doi:10.1029/2001JD000336, 2002.
- Han, F., Kota, S. H., Wang, Y., & Zhang, H.: Source apportionment of PM_{2.5} in Baton Rouge, Louisiana during 2009–2014. Science of the Total Environment, 586, 115– 126. https://doi.org/10.1016/j.scitotenv.2017.01.189, 2017.
- Heald, C. L., and Spracklen, D. V.: Atmospheric budget of primary biological aerosol particles from fungal spores, Geophys. Res. Lett., 36, L09806, doi:10.1029/2009GL037493, 2009.
- Hennigan, C. J., Sullivan, A. P., Collett Jr., J. L., and Robinson, A. L.: Levoglucosan stability in biomass burning particles exposed to hydroxyl radicals, Geophys. Res. Lett., 37, L09806, https://doi.org/10.1029/2010gl043088, 2010.
- Ibrahim, M., Rabah, A. B., Liman, B., and Ibrahim, N. T.: Effect of temperature and relative humidity on the growth of Helminthosporium fulvum, Nigerian J. Basic Appli. Sci., 19, 127–129, 2011.
- Jones, E. B. G., and Mitchell, J. I.: Biodiversity of marine fungi, in biodiversity: international biodiversity seminar, edited by: Cimerman, A. and Gunde-Cimerman, N., 31–42, Ljubljana: National Inst. Chemistry and Slovenia National Commission for UNESCO, 1996.
- Kawamura, K., Tachibana, E., Okuzawa, K., Aggarwal, S. G., Kanaya, Y., Wang, Z. F.: High abundances of water-soluble dicarboxylic acids, ketocarboxylic acids and a-dicarbonyls in the mountaintop aerosols over the north China plain during wheat burning season, Atmos. Chem. Phys. 13 (16), 8285-8302, https://doi.org/10.5194/acp-13-8285-2013, 2013.
- Keller, F., and Matile, P.: Storage of sugars and mannitol in petioles of celery leaves, New Phytol. 113:291–299, https://doi.org/10.1111/j.1469-8137.1989.tb02406.x, 1989.

- Kim, V. K. and Xiao, C. L.: Influence of culture media and environmental factors on mycelial growth and picnidial production of Sphaeropsis pyriputrescens, Mycologia, 97, 25–32, 2005.
- Lelieveld, J., Evans, J.S., Fnais, M., Giannadaki, D., Pozzer, A.: The contribution of outdoor air pollution sources to premature mortality on a global scale, Nature, 525 (7569), 367-371. https://doi.org/10.1038/nature15371, 2015.
- Lewis, D. and Smith, D.: Sugar alcohols (polyols) in fungi and green plants, 1. Distribution, physiology and metabolism, New Phytol., 66, 143–184, https://doi.org/10.2307/2430328, 1967.
- Li, W. J., Shao, L. Y., and Buseck, P. R.: Haze types in Beijing and the influence of agricultural biomass burning, Atmos. Chem. Phys. 10 (17), 8119-8130. https:// doi.org/10.5194/acp-10-8119-2010, 2010.
- Loescher, W. H., Tyson, R. H., Everard, J. D., Redgwell, R. J., and Bieleski, R. L.: Mannitol synthesis in higher plants: evidence for the role and characterization of a NADPH-Dependent mannose 6-Phosphate reductase, Plant Physiol., 98(4), 1396– 1402, https:// doi: 10.1104/pp.98.4.1396, 1992.
- Malik, V. K. and Singh, S.: Effect of temperature and relative humidity on teliospore germination in Ustilago hordei, J. Mycol. Plant Pathol., 34, 410–411, 2004.
- Manninen, H. E., Bäck, J., Sihto Nissilä, S. L., Huffman, J. A., Pessi, A. M., Hiltunen, V., Aalto, P. P., Hidalgo Fernández, P. J., Hari, P., and Saarto, A.: Patterns in airborne pollen and other primary biological aerosol particles (PBAP), and their contribution to aerosol mass and number in a boreal forest, Boreal Environ. Res., 19, 383–405, 2014.
- Miyazaki, Y., Fu., P. Q., Ono, K., Tachibana, E., and Kawamura, K.: Seasonal cycles of water-soluble organic nitrogen aerosols in a deciduous broadleaf forest in northern Japan, J. Geophys. Res. Atmos., 119, 1440–1454, doi:10.1002/2013JD020713, 2014.
- Miyazaki, Y., Jung, J., Fu, P. Q., Mizoguchi, Y., Yamanoi, K., and Kawamura, K.: Evidence of formation of submicrometer water-soluble organic aerosols at a deciduous forest site in northern Japan in summer, J. Geophys. Res., 117, D19213, doi:10.1029/2012JD018250, 2012.
- Pashynska, V., Vermeylen, R., Vas, G., Maenhaut, W., and Claeys, M.: Development of a gas chromatographic/ion trap mass spectrometric method for the determination of levoglucosan and saccharidic compounds in atmospheric aerosols. Application to urban aerosols, J. Mass Spectrom., 37, 1249–1257, https://doi.org/10.1002/jms.391, 2002.
- Rumpho, M. E., Edwards, G. E., and Loescher, W. H.: A pathway for photosynthetic carbon flow to mannitol in celery leaves. Activity and localization of key enzymes, Plant Physiol, 73:869–873, https://doi:10.1104/pp.73.4.869, 1983.
- Sharma, R. and Rajak, R. C.: Keratinophilic fungi: Nature's keratin degrading machines, Resonance, 8, 28–30, 2003.
- Sun, Y., Jiang, Q., Xu, Y., Ma, Y., Zhang, Y., Liu, X., Li, W., Wang, F., Li, J., Wang, P., Li, Z.: Aerosol characterization over the north China plain: haze life cycle and biomass burning impacts in summer, J. Geophys. Res. 121 (5), 2508-2521. https://doi.org/10.1002/2015JD024261, 2016.
- Xu, S., Ren, L., Lang, Y., Hou, S., Ren, H., Wei, L., Wu, L., Deng, J., Hu, W., Pan, X., Sun, Y., Wang, Z., Su, H., Cheng, Y., and Fu, P Q.: Molecular markers of biomass burning and primary biological aerosols in urban Beijing: size distribution and seasonal variation, Atmos. Chem. Phys., 20, 3623–3644,

https://doi.org/10.5194/acp-20-3623-2020, 2020.

- Yan, C., Zheng, M., Sullivan, A. P., Shen, G., Chen, Y., Wang, S., Zhao, B., Cai, S., Desyaterik, Y., Li, X., Zhou, T., Gustafsson, O., and Collett, J. L.: Residential coal combustion as a source of levoglucosan in China, Environ. Sci. Technol. 2018, 52, 3, 1665–1674 http://doi:10.1021/acs.est.7b05858, 2018.
- Zhu, C., Kawamura, K., Fukuda, Y., Mochida, M., and Iwamoto. Y: Fungal spores overwhelm biogenic organic aerosols in a midlatitudinal forest, Atmos. Chem. Phys., 16, 7497–7506, https://doi.org/10.5194/acp-16-7497-2016, 2016.