Response to Reviewers' comments:

Reviewer #2

The authors investigated the association of sugar molecules and the microbial community at taxonomic levels in atmospheric aerosols and found that sugar compounds exhibited significant associations with fungal community structure than bacterial community structure. And this strong associations between fungal community and sugar compounds in rural aerosols may due to the entire fungal community rather than specific fungal taxa. Overall, this study was well-designed, interesting and complete. The results observed could add substantially to the literature, provide a reference for further studies exploring the contribution of bioaerosols to organic carbon in atmospheric aerosols and build links between airborne microorganisms and aerosol chemistry. I would suggest the publication of this manuscript after addressing the following comments.

Response:

Thank the reviewer for the valuable and professional comments. According to the valuable suggestions, we have made extensive corrections to the original manuscript. Please refer to the point-to-point responses to the comments below.

Comment 1

Since the samples were collected in the summer of 2020 (7−13 August). Did the high temperature affect the association of sugar molecules and the microbial community at taxonomic levels in atmospheric aerosols?

Response:

Yes, it's quite possible. Microbial communities and sugar molecules were significantly affected by environmental factors (Zhai et al., 2018;Fu et al., 2012), and the types and amounts of sugar species produced by microorganisms are affected by microbial species and external environmental conditions such as carbon sources, temperature and humidity (Hrynkiewicz et al., 2010). Therefore, we consider that the corresponding relations between microorganisms and sugar compounds vary with the external environmental factors, e.g., temperature, relative humidity and geographical location. To make it clearer, the following sentence was revised as follows:

Page 12, line 364-373: *<u>"In addition, it should be noted that the taxa associated with sugar compounds found in this study were not the same as those found in a survey conducted by Samaké et al. (2020) at a rural site in France, the only study up-to-date investigating the association between microorganisms and sugar compounds. The types and amounts of sugar species produced by microorganisms are affected by microbial species and external environmental conditions such as carbon sources, temperature and humidity (Hrynkiewicz et al., 2010). Here, we performed network construction separately for daytime and nighttime samples. Results showed that the sugar molecules had more associations with microbial taxa at night, particularly fungal taxa (Fig. S7), demonstrating the importance of environmental factors on the relationship between sugar compounds and microbial taxa. More fungal spores could be released at night due to high temperature and high relative humidity (Elbert et al., 2007;Hummel et al., 2015) and the proportions of some fungal genera (e.g., Agaricus) enhanced at night (Table S3), which was a possible explanation for the greater associations between sugar molecules and microbial taxa at night."</u>*

The related analysis was added in Figure S7 as follows:

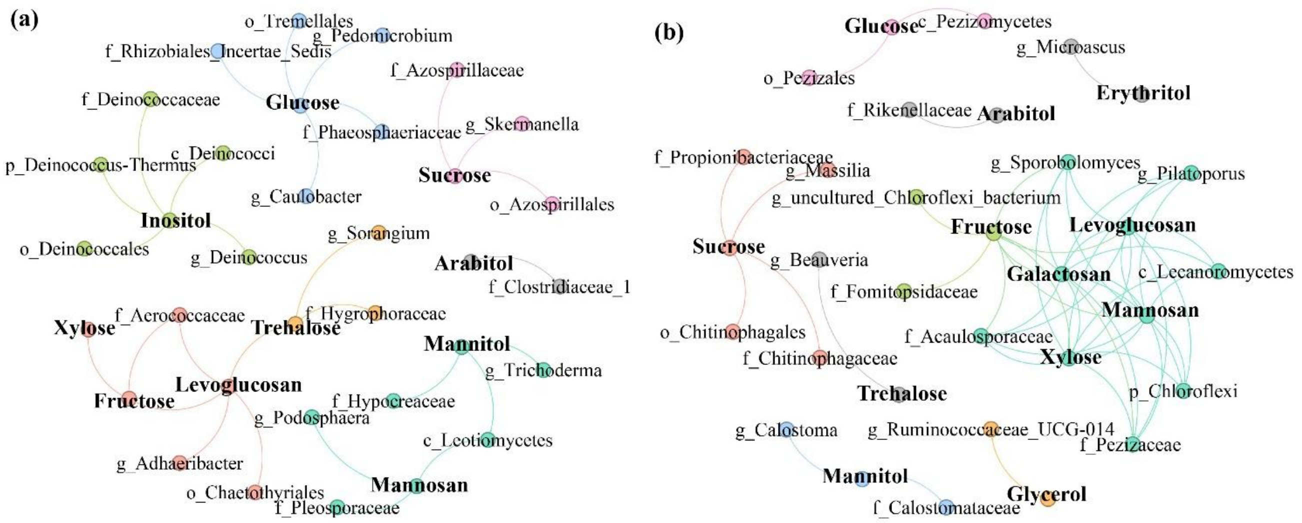

Figure S7. Co-occurrence networks between microbial taxa and sugar components (bold font) were constructed based on Spearman's correlation analysis in the (a) daytime and (b) nighttime samples. The nodes are colored according to different types of modularity classes.

Elbert, W., Taylor, P. E., Andreae, M. O., and PöSchl, U.: Contribution of fungi to primary biogenic aerosols in the atmosphere: Wet and dry discharged spores, carbohydrates, and inorganic ions, Atmos. Chem. Phys., 7, 4569-4588, 2007.

Fu, P., Kawamura, K., Kobayashi, M., and Simoneit, B. R. T.: Seasonal variations of sugars in atmospheric particulate matter from Gosan, Jeju Island: Significant contributions of airborne pollen and Asian dust in spring, Atmos. Environ., 55, 234-239, https://doi.org/10.1016/j.atmosenv.2012.02.061, 2012.

Hrynkiewicz, K., Baum, C., and Leinweber, P.: Density, metabolic activity, and identity of cultivable rhizosphere bacteria on Salix viminalis in disturbed arable and landfill soils, J. Plant Nutr. Soil Sci., 173, 747-756, https://doi.org/10.1002/jpln.200900286, 2010.

Hummel, M., Hoose, C., Gallagher, M., Healy, D. A., Huffman, J. A., O'Connor, D., Pöschl, U., Pöhlker, C., Robinson, N. H., Schnaiter, M., Sodeau, J. R., Stengel, M., Toprak, E., and Vogel, H.: Regional-scale simulations of fungal spore aerosols using an emission parameterization adapted to local measurements of fluorescent biological aerosol particles, Atmos. Chem. Phys., 15, 6127-6146, https://doi.org/10.5194/acp-15-6127-2015, 2015.

Zhai, Y., Li, X., Wang, T., Wang, B., Li, C., and Zeng, G.: A review on airborne microorganisms in particulate matters: Composition, characteristics and influence factors, Environ. Int., 113, 74-90, https://doi.org/10.1016/j.envint.2018.01.007, 2018.

Comment 2

Why the authors chose the TSP samples? If the authors wanted to investigate the contribution of bioaerosols to organic carbon in atmospheric aerosols, it's better to collect PM2.5 or PM10 samples.

Response:

Although the particle sizes of microorganisms are usually smaller than 10 μm, microorganisms tend to attach to the particulate matter for airborne transport (Fröhlich-Nowoisky et al., 2016). Focusing on sugar compounds and microorganisms in TSP samples may offer a broader perspective on the relationship between sugar compounds and microorganisms.

"*Considering the broad size range of airborne microorganisms and their attachment to other coarse particles (Fröhlich-Nowoisky et al., 2016)"* was added in Line 88.

Fröhlich-Nowoisky, J., Kampf, C. J., Weber, B., Huffman, J. A., Pöhlker, C., Andreae, M. O., Lang-Yona, N., Burrows, S. M., Gunthe, S. S., and Elbert, W.: Bioaerosols in the Earth system: Climate, health, and ecosystem interactions, Atmos. Res., 182, 346-376, 2016.

Comment 3

The microbial community may be affected by air quality parameters such as $PM_{2.5}$, PM_{10}, NO_2, O_3, SO_2 and CO. The sugar compounds may be also affected by these pollution factors. I suggest the authors add some analysis about these parameters (also the air temperature).

Response:

The content about the relationship between sugar compounds and environmental factors was added as follows:

Page 9, line 272-273: *"Moreover, the concentrations of anhyrosugars were significantly affected by temperature (Fig. S6), which may also be responsible for the diurnal differences in anhyrosugars."*

Page 10, line 304-306: *"Meanwhile, the temperature significantly affected the sugar alcohols (Fig. S6). Oduber et al. (2021) also reported high correlations between sugar alcohols and air temperature. This phenomenon may be corroborated by the finding that the activities of fungal spores were regulated by temperature (Zhu et al., 2015)."*

The related analysis was added in Figure S6 and revised in Table S5 as follows:

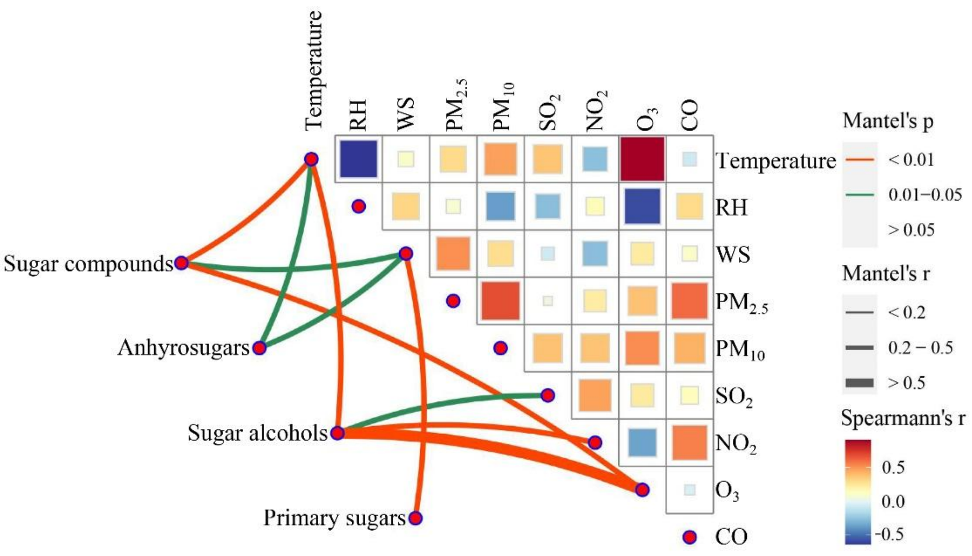

Figure S6. Correlations of sugar compounds with environmental factors based on Bray–Curtis distance. Edge width corresponds to Mantel's r value, and the edge color denotes statistical significance. Pairwise correlations of environmental factors with a heatmap are calculated by Spearman's correlation analysis.

Table S5. Spearman correlation of sugar components and environmental or fungal α-diversity indices.

Factors	Anhyrosugars			Sugar alcohols					Primary saccharides				
	Galactosan	Mannosan	Levoglucosan	Arabitol	Mannitol	Erythritol	Inositol	Glycerol	Glucose	Sucrose	Fructose	Xylose	Trehalose
Galactosan	1												
Mannosan	0.833**	1											
Levoglucosa	0.881**	0.916**	1										
Arabitol	0.547*	0.503	0.398	1									
Mannitol	0.569*	0.560*	0.477	0.864**	1								
Erythritol	0.459	0.411	0.477	0.719**	0.851**	1							
Inositol	0.2	0.516	0.433	-0.086	-0.064	-0.051	1						
Glycerol	0.626*	0.473	0.38	0.688**	0.499	0.284	-0.02	1					
Glucose	0.27	0.516	0.323	0.16	0.073	-0.305	0.503	0.187	1				
Sucrose	-0.169	0.051	0.182	-0.371	-0.473	-0.16	0.429	-0.42	-0.007	1			
Fructose	0.407	0.662**	0.618*	-0.134	-0.13	-0.108	0.754**	-0.024	0.508	0.495	1		
Xylose	0.727**	0.881**	0.921**	0.279	0.367	0.327	0.604*	0.244	0.525	0.292	0.771**	1	
Trehalose	0.31	0.446	0.336	0.745**	0.877**	0.749**	0.09	0.275	0.156	-0.275	-0.103	0.332	1
OC	0.653*	0.855**	0.881**	0.429	0.385	0.433	0.411	0.288	0.341	0.411	0.622*	0.829**	0.345
Ca^{2+}	0.543*	0.534*	0.653*	0.556*	0.415	0.547*	0.218	0.367	0.055	0.358	0.138	0.486	0.415
Cl^{-}	0.829**	0.811**	0.749**	0.798**	0.719**	0.547*	0.116	0.789**	0.292	-0.244	0.191	0.582*	0.49
SO_4^{2-}	0.446	0.705**	0.560*	0.187	0.125	0.055	0.587*	0.178	0.508	0.42	0.829**	0.670**	0.156
Temperature	-0.196	0.033	0.046	-0.644*	-0.670**	-0.604*	0.411	-0.464	0.31	0.679**	0.631*	0.235	-0.604*
RH	0.407	0.244	0.181	0.608*	0.806**	0.670**	-0.297	0.445	-0.207	-0.705**	-0.313	0.053	0.698**
Wind speed	-0.613*	-0.49	-0.675**	-0.407	-0.495	-0.662**	-0.029	-0.244	0.213	-0.046	-0.015	-0.415	-0.376
$PM_{2.5}$	0.152	0.187	-0.007	0.117	0.361	0.442	0.337	0.18	0.09	-0.233	0.037	-0.004	0.429
PM_{10}	0.521	0.732**	0.609*	0.635*	0.53	0.336	0.288	0.389	0.640*	0.174	0.398	0.662**	0.534*
SO_2	0.031	0.277	0.077	0.29	0.242	0.141	0.031	-0.024	0.099	-0.042	0.064	-0.02	-0.053
NO_2	-0.126	0.042	-0.051	-0.267	-0.294	-0.373	-0.289	0.139	0.104	0.241	0.435	-0.038	-0.448
O_3	0.468	0.53	0.464	0.459	0.899**	0.899**	0.815**	-0.011	0.099	-0.16	-0.138	0.354	0.859**
OTU	0.552*	0.503	0.534*	0.679**	0.837**	0.785**	0.051	0.336	-0.011	-0.292	-0.086	0.433	0.851**
Shannon	0.415	0.424	0.257	0.402	0.516	0.095	0.125	0.292	0.604*	-0.635*	0.015	0.279	0.525

Note: *$p < 0.01$; **$p < 0.05$.

Zhu, C., Kawamura, K., and Kunwar, B.: Organic tracers of primary biological aerosol particles at subtropical Okinawa Island in the western North Pacific Rim, J. Geophys. Res. Atmos., 120, 5504-5523, https://doi.org/10.1002/2015JD023611, 2015.

Oduber, F., Calvo, A. I., Castro, A., Alves, C., Blanco-Alegre, C., Fernández-González, D., Barata, J., Calzolai, G., Nava, S., Lucarelli, F., Nunes, T., Rodríguez, A., Vega-Maray, A. M., Valencia-Barrera, R. M., and Fraile, R.: One-year study of airborne sugar compounds: Cross-interpretation with other chemical species and meteorological conditions, Atmos. Res., 251, 105417, https://doi.org/10.1016/j.atmosres.2020.105417, 2021.

Comment 4

The authors used the filters to collect the sugar samples. Did the author test the recovery efficiencies of these sugar compounds from the filter?

Response:

Yes, we tested the recovery efficiencies of these sugar compounds and the recoveries ranged from 80% to 120%. To make it clearer, the following sentence was added:

Page 5, line 131-132: *"The recoveries of sugar compounds ranged from 80% to 120%. Reproducibility of the method was larger than 90%."*

Thank you very much for your comments and suggestions. Your any further comments and suggestions are appreciated.