Characteristics of bacterial and fungal communities and their associations with sugar compounds in atmospheric aerosols at a rural site in North China

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Abstract. Bioaerosols play significant roles in causing health and climate effects. Sugar compounds in air have been widely used to trace the source of bioaerosols. However, knowledge about the association of sugar molecules and the microbial community at taxonomic levels in atmospheric aerosols remains limited. Here, microbial community compositions and sugar molecules in total suspended particles collected from a typical rural site, Gucheng, in the North China Plain were investigated by gas chromatography-mass spectrometry and high-throughput gene sequencing, respectively. Results show that fungal community structure exhibited distinct diurnal variation with largely enhanced contribution of Basidiomycota at night, while bacterial community structure showed no obvious difference between daytime and night. SourceTracker analysis revealed that bacteria and fungi were mainly from plant leaves and unresolved sources (presumably human-related emission and/or long-distance transport), respectively. All the detected anhyrosugars and sugar alcohols, and trehalose showed diurnal variations with lower concentrations in the daytime and higher concentrations at night, which may be affected by enhanced fungal emissions at night, while primary sugars (except trehalose) showed an opposite trend. The Mantel's test showed that more sugar compounds exhibited significant associations with fungal community structure than bacterial community structure. Co-occurrence analysis revealed the strong associations between sugar compounds and a few saprophytic fungal genera with low relative abundances, e.g., *Hannaella*, *Lectera*, *Peniophora*, *Hydnophlebia*, *Sporobolomyces* and *Cyphellophora*. This study suggested that the entire fungal community likely greatly contributed to sugar compounds in rural aerosols, rather than specific fungal taxa, while the contribution of bacteria was limited.

1 Introduction

Bioaerosols, mainly refer to particulate matter with biological origin, including viruses, bacteria, fungi, pollen and their biological debris (Yao, 2018;Fröhlich-Nowoisky et al., 2016;Santl-Temkiv et al., 2020;Shen and Yao, 2022). Bioaerosols have attracted progressively more attention during the past decades due to their adverse health effects. Health problems can arise in living organisms such as humans, animals and plants through respiratory or surface-contact exposure to airborne pathogenic or allergenic microbes (Taipale et al., 2021;Arzt et al., 2011;Hicks et al., 2012), which have caused serious economic losses and even human deaths. Bioaerosols also play potentially important roles in cloud and precipitation formation by acting as cloud condensation nuclei and ice nucleating particles (Hoose et al., 2010;Ariya et al., 2009;Hu et al., 2020;Huang et al., 2021). Certain airborne microorganisms can participate in atmospheric chemical processes by transforming inorganic or organic matter (Liu et al., 2020;Ariya et al., 2002).

To accurately assess the health-related and climatic effects of bioaerosols, and more importantly, to develop effective bioaerosol pollution control approaches, the investigations on the abundance, compositions and emission fluxes of bioaerosols need to be taken seriously. Previous studies have found that in terms of number and mass concentrations, bioaerosols could account for ~30% of total aerosols (size larger than ~1 μm) in urban or rural sites (Després et al., 2012;Huffman et al., 2013;Schumacher et al., 2013); while in the rainforest, bioaerosols account for up to 80% (Huffman et al., 2012;Pöschl et al., 2010). Current estimates of global bioaerosol mass emission fluxes vary widely (about 10–1000 Tg a^{-1}) (Després et al., 2012). In recent decades, molecular biology technology (e.g., fluorescence in situ hybridization techniques, denaturing gradient gel electrophoresis, terminal restriction fragment length polymorphism and sequencing technology) and fluorescence techniques (e.g., ultraviolet aerodynamic particle sizer and wideband integrated bioaerosol sensor) have been applied to study the compositions and abundances of bioaerosols (Prass et al., 2021;Drautz-Moses et al., 2022;Zhang et al., 2017). Whereas, the compositions, sources and emission fluxes of bioaerosols remain less documented and quantified (Yue et al., 2016;Fröhlich-Nowoisky et al., 2016).

Sugar compounds are the main energy storage and cellular structural materials in organisms (Wang et al., 2021), and their presence in the atmosphere is often considered to be closely related to biological sources (Bauer et al., 2008a;Schliemann et al., 2008). Hence sugar compounds are often used as biomarkers to track natural sources of bioaerosols and estimate the contribution of bioaerosols to organic aerosols (Fu et al., 2010;Fu et al., 2013;Fu et al., 2008). For instance, arabitol and mannitol have been commonly used to assess fungal spore emissions (Bauer et al., 2008a;Marynowski et al., 2019), and trehalose was considered a fungal-specific sugar since it is mainly present in mycorrhizal plants (Kaur and Suseela, 2020). Sucrose and glucose, as end products of photosynthesis, were often used to trace the emissions from plant debris and some photosynthetic prokaryotes (Goddijn and van Dun, 1999). Fructose can be used to study the spread of plant debris or pollen in the atmosphere (Fu et al., 2013).

However, most sugar compounds are widely present in living organisms. For example, a variety of bacteria have been found to synthesize mannitol (Monedero et al., 2010). Trehalose participates in osmotic protection and maintains desiccation

tolerance in bacteria, yeast, insects and some plants (Zancan and Sola-Penna, 2005). Glucose, fructose, galactose and mannose were widely found in bacteria (Walmsley et al., 1998;Deutscher et al., 2006). That is, although the sugar compounds used as biomarkers tend to be predominant in organisms they characterize, there are many possible biological sources of these sugar compounds.

A large number of studies using sugar compounds as biomarkers focused on bacteria and fungi at the kingdom level, which may result in inaccurate source identification and emission flux estimation of bioaerosols. To the best of our knowledge, only Samaké et al. (2020) explored the relationship between sugar compounds and microorganisms in PM_{10} collected in an agricultural area in France, and there is still a large lack of understanding of the relationship between microbial communities and sugar compounds at the molecular level. Such information is important for clarifying the sources of bioaerosols and improving the accuracy of bioaerosol emission flux estimates.

In this study, to refine the marker categories of sugar compounds in atmospheric aerosols, airborne total suspended particles (TSP) were collected from a typical rural site, Gucheng, in the North China Plain. Atmospheric microbial communities (bacteria and fungi) and common sugar compounds in the TSP samples were analyzed using high-throughput gene sequencing and gas chromatography-mass spectrometry (GC-MS) techniques, respectively. Statistical analyses were exploited to search for the association between microbial communities and sugar compounds in atmospheric aerosols.

2 Materials and methods

2.1 Observation and sample collection

The observation was carried on at the rooftop of the Gucheng Ecological and Agrometeorological Experiment Station, the Chinese Academy of Meteorological Sciences (39.15°N, 115.73°E; about 5 m above the ground level) in the summer of 2020 (7−13 August). The observation site is located in Dingxing County, Hebei Province, which is about 100 km and 128 km away from two nearby megacities Beijing and Tianjin, respectively (**Fig. S1**). The site is located in a rural area in the north of the North China Plain. It is a high-yield agricultural area with maize as the main summer crop, about 8 km from the nearest river and 178 km from the closest ocean. The site is about 900 m away from the nearest railway and 2.7 km from the expressway. There was no intense agricultural activity observed around the site during the observation period.

Considering the broad size range of airborne microorganisms and their attachment to other coarse particles (Fröhlich-Nowoisky et al., 2016), TSP samples loaded on quartz fiber filters (25 × 20 cm) were collected using a high-volume air sampler (Tisch Environmental, Inc., USA) daily based on daytime (D, 8:00–19:30) and nighttime (N, 20:00–7:30) at a flow rate of 1.05 m^3 min^{-1}. All filters before sampling were combusted at 450°C for 6 h to remove organic matter and microorganisms. The filters after sampling were stored in a refrigerator at −20°C for later analysis.

Meteorological parameters including air temperature, relative humidity, wind speed and direction, and precipitation are real-time monitoring data obtained from a weather station in Xushui, Hebei (about 18 km away from the sampling site). Air quality

 parameters including AQI, PM$_{2.5}$, PM$_{10}$, NO$_2$, O$_3$, SO$_2$ and CO at the Baoding environmental monitoring station (about 36 km away from the sampling site) were obtained from China National Environmental Monitoring Center (http://www.cnemc.cn/).

2.2 Taxonomic identification of airborne microbial community

High-throughput gene sequencing technique was used to analyze airborne bacterial and fungal community structures. A piece of 18 cm^2 filter for each sample was cut and placed in a lysis tube, added to Solution 1 provided by PowerSoil DNA Isolation Kit (MoBio Laboratories, USA) and then placed at 65°C for 20 min to reduce the adsorption of quartz to DNA. Subsequently, the solution was processed according to the manufacturer's instructions. Polymerase chain reaction (PCR) amplification was conducted in a 25 μL reaction solution, including 12.5 μL KAPA 2G Robust Hot Start Ready Mix, 1 μL of 5 μM of each primer, 1.5 μL genome DNA and 9 μL double distilled H$_2$O. The PCR primer pairs are 338F (5'-ACTCCTACGGGAGGCAGCA-3') and 806R (5'-GGACTACHVGGGTWTCTCATAT-3') in bacterial V3-V4 region, and ITS1F (5'-CTTGGTCATTTAGAGGAAGTAA-3') and ITS2 (5'-TGCGTTCTTCATCGATGC-3') in fungal ITS region.

The PCR amplification was performed as follows: denaturation at 94°C for 5 min, 30 cycles for bacteria and 35 cycles for fungi of denaturation, annealing and extension (94°C for 30 s, 50°C for bacteria and 55°C for fungi for 30 s, and 72°C for 60 s), and final extension at 72°C for 7 min. The final PCR products were purified by AMPure XP magnetic beads. Then the purified DNA was used for generating a sequencing library. The library was purified by AMPure XP magnetic beads and qualified with NanoDrop 2000 Spectrophotometers (Thermo Fischer Scientific, Inc., USA). Finally, the library was sequenced using the Illumina Miseq 2500 platform (2×250 paired ends) at Allwegene Company, Beijing, China.

The pair-end reads were quality-controlled using Trimmomatic v0.33, and then merged using FLASH v1.2.11 software to produce the raw tags. Subsequently, the raw tags were identified based on the barcode and primer using Mothur v1.35.1 and were filtered by removing the barcode and primer. Then the clean sequences with ≥ 97% similarity were assigned to one operational taxonomic unit (OTU). Taxonomic annotation of each OTU for bacteria and fungi was performed using the Silva 16S and UNITE databases, respectively.

2.3 Determination of sugar components

A piece of each sampled filter was extracted by sonication for 10 min using a 10 mL dichloromethane and methanol mixture (2:1, v/v). The extracts were filtered through Pasteur pipette filled with quartz wool and concentrated to approximately 100 μL using a rotary evaporator, and then dried under nitrogen. The silylation derivatization was performed by reaction with 60 μL *N, O*-bis-(trimethylsilyl) trifluoroacetamide (BSTFA) including 1% trimethylsilyl chloride and pyridine (5:1, v/v) at 70 °C for 3 h. After the derivatization, hexane solution containing C$_{13}$ n-alkane internal standard (1.43 ng/μL) was added to the derivatives. The samples were stored at −20°C before GC-MS analysis.

Sugar compounds were quantified by a GC-MS system (7890A-5975C, Agilent Technology, USA). The GC was equipped with a split/splitless injection and a fused silica capillary column (DB-5MS, 30 m × 0.25 mm × 0.25 μm, Hewlett-Packard). The GC oven temperature procedure was as follows: initiation at 50°C for 2 min, heating to 120°C at a rate of 15°C/min,

heating to 300°C at a rate of 5°C/min, stabilization for 16 min, and finally decreasing to 50°C. The carrier gas was ultrapure helium (1.3 mL/min). The GC injector temperature was 280°C, the ion source temperature was 230°C, and the quadrupole temperature was 150°C. The MS was operated on Electron Ionization (EI) mode at 70 eV and scanned from 50 to 650 Da.

130 Mass spectral data were processed using the ChemStation software, and sugar compounds were identified and quantified using authentic standards. The recoveries of sugar compounds ranged from 80% to 120%. Reproducibility of the method was larger than 90%.

2.4 Chemical components analyses

Organic (OC) and elemental carbon (EC) were quantified by an OC/EC analyzer (Sunset Laboratory Inc., USA) according to
135 the thermal optical transmission (TOT) method of the NOISH protocol.

A portion of the filters was cut and extracted with 20 mL ultrapure water by ultrasonication for 20 min. Then the extracts were filtered using 0.22 m PTFE filters (MillexGV, Millipore, USA). The filtrate was used for the determination of water-soluble inorganic components including Na^+, K^+, Mg^{2+}, Ca^{2+}, NH_4^+, SO_4^{2-}, NO_3^-, and Cl^- with an ions chromatography system (ICS-5000+, Thermo Fisher, USA).

140 ### 2.5 Statistical analyses

SourceTracker is a Bayesian approach to identifying sources and estimating source proportions for microbial surveys (Knights et al., 2011). The source samples used as source tracking reference databases in this study were soils, plant leaves and seawater. Maize leaves and soil samples were collected around the observation site and the samples were subjected to high-throughput sequencing as described in Section 2.2 to obtain the gene sequences, and the marine microbial sequences were obtained from
145 the NCBI SRA database (https://www.ncbi.nlm.nih.gov/sra/, **Table S1**). The sequences of all source and sink samples were processed together by QIIME 2, and were assigned to OTUs based on the similarity of sequences ≥ 97%. The output data were performed for source tracking analysis based on "SourceTracker2" package in R (https://github.com/caporaso-lab/sourcetracker2).

The one-way ANOVA analysis was performed using SPSS v.22.0 to analyze the diurnal differences in the microbial
150 community. Spearman correlation analysis was used to evaluate the relationship between microbial taxa and environmental or pollution factors. A p-value < 0.05 indicates a significant correlation. Non-metric multidimensional scaling (NMDS), principal coordinate analysis (PCoA), redundancy analysis (RDA), the linear discriminant analysis effect size (LEfSe) analysis (http://huttenhower.sph.harvard.edu/lefse/) and the Mantel's test were conducted using the R scripts. The taxa with linear discriminant analysis (LDA) score > 2 were selected. The infer community assembly mechanisms by phylogenetic bin-based
155 null model analysis (iCAMP) is a general framework to quantitatively estimate community assembly mechanisms (Ning et al., 2020; Chen et al., 2022). The assembly mechanisms of airborne microbes at Gucheng site were investigated by using the iCAMP package in R. For network analysis, Spearman's correlation between microbial taxa with sugar compounds was calculated using "Hmisc" package in R. All p-values were adjusted using the Benjamini and Hochberg false discovery rate

(FDR) controlling procedure (Benjamini et al., 2006). Then, those significant correlations between microbial taxa and sugar

160   molecules (FDR-adjusted p-value < 0.05, correlation coefficients > 0.6) were selected. Network properties were calculated using the "igraph" package in R. Network visualization was conducted using Gephi v0.9.3. Modules were identified using the Louvain method provided by Gephi v0.9.3 (Bastian et al., 2009).

3 Results and Discussion

3.1 Diurnal variation in microbial community

165   **3.1.1 Microbial diversity and richness**

A total of 38565 16S rRNA gene sequences and 56920 ITS sequences were obtained by high-throughput sequencing technology and were assigned to 404 bacterial OTUs and 391 fungal OTUs. The sequencing coverage of all the samples was > 99%, indicating that the sequencing data could represent the microbial communities in the atmosphere.

Alpha diversity indices were used to evaluate the diurnal differences in microbial community structure (**Fig. 1** and **Table S2**).

170   For bacterial communities, OTU and the Chao1 index used to evaluate richness showed no significant diurnal patterns. Except for the samples collected on August 13, the bacterial diversity (Shannon and Simpson's indices) in the daytime samples was slightly lower or comparable to that in the nighttime samples ($p > 0.05$, **Fig. 1a**). In contrast, the diversity and richness of fungal community were significantly higher at night than during the daytime ($p < 0.05$, **Fig. 1b**). Abdel Hameed et al. (2009) found that bacteria and fungi had similar diurnal variation patterns over Cairo City, Egypt. However, the bacterial diversity in

175   aerosols over Guilin, China was lightly abundant during the day than at night (Long et al., 2022), which was similar to the finding over tropical forest aerosols in August (Gusareva et al., 2019). These different results suggest that the diversity and biogeography of airborne microorganisms might be influenced by multiple factors, e.g., geographical locations, airflow patterns, and global atmospheric circulation (Fröhlich-Nowoisky et al., 2012;Martiny et al., 2006;Womack et al., 2010).

3.1.2 Bacterial community structure

180   **Fig. 2** shows the bacterial and fungal community compositions in TSP samples collected at rural Gucheng, North China. At the phylum level, 38 bacterial phyla were detected. Proteobacteria (42.2%), Firmicutes (26.1%), Actinobacteria (9.0%), Cyanobacteria (8.7%) and Bacteroidetes (8.4%) were the most abundant bacterial phyla among all the TSP samples (**Fig. 2a**), which were also observed as the dominant phyla in the air of other regions (Acuna et al., 2022;Chen et al., 2022;Xu et al., 2017). The relative abundance of Cyanobacteria slightly increased in the daytime ($p > 0.05$), which was more abundant in the

185   daytime with warmer temperatures (Singh et al., 2018). Firmicutes had a higher relative abundance in the nighttime ($p < 0.05$, **Fig. 3a**). Gusareva et al. (2019) also found that high temperature had an important impact on the bacterial community structure, and the abundance of Firmicutes was higher at noon when the temperature was the highest. Drautz-Moses et al. (2022) reported significant diurnal differences in the relative abundance of the phylum Firmicutes in aerosols in the lower troposphere.

At the order level, Bacillales (15.1%), Clostridiales (9.6%), Burkholderiales (8.8%), Sphingomonadales (5.8%) and Myxococcales (5.4%) were predominant (**Fig. S2a**) and no significant diurnal differences were found at the order level. At the genus level, *Bacillus* (6.9%), *Curvibacter* (6.3%), *Chroococcidiopsis* (4.2%) and *Tumebacillus* (4.1%) were the most abundant bacterial genera (**Fig. 2b**). *Bacillus* and *Chroococcidiopsis* were the dominant bacterial genera in the air worldwide (Zhao et al., 2022). *Curvibacter* occurring mainly in the aqueous environment (Xu et al., 2021) and spore-forming bacterium *Tumebacillus* occurring in the permafrost (Steven et al., 2008) and soils (Wemheuer et al., 2017) had smaller relative abundances observed in previous studies (Yan et al., 2018). Geographical locations and land use types may shape atmospheric bacterial community structure (Bowers et al., 2011). Shen et al. (2019) found that the relative abundances of *Bacillus* and *Curvibacter* in rural aerosols were about 90 times higher than those in urban aerosols.

3.1.3 Fungal community structure

Fungal community structure exhibited distinct diurnal differences based on the results of NMDS analysis (**Fig. S3**). The Bray-Curtis dissimilarity index of fungal communities was slightly higher at night (0.46) than in the daytime (0.41), suggesting the high β diversity of fungal community at night. Co-occurrence network analysis also revealed that the average degree of fungal communities at night (10.0) was greater than in the daytime (8.9), suggesting that fungal taxa cooperated more closely with each other at night.

From community compositions, fungi were almost always composed of the phyla Ascomycota (63.1%) and Basidiomycota (36.7%) (**Fig. 2c**). Their relative abundances were similar to the results of rural aerosols collected from Backgarden, South China (Fröhlich-Nowoisky et al., 2012). Due to the more abundant Basidiomycota found in natural soils (Hui et al., 2017), Basidiomycota accounted for a relatively higher proportion of rural aerosols compared with urban aerosols (Liu et al., 2019). Significant diurnal differences were found in these two phyla (daytime: Ascomycota (65.3%) vs Basidiomycota (13.6%), nighttime: Ascomycota (28.6%) vs Basidiomycota (34.0%), $p < 0.01$) (**Fig. 3b**). LEfSe analysis likewise shows clear diurnal differences in fungal taxa. The percentage of taxa belonging to Ascomycota increased significantly in the daytime, while taxa belonging to Basidiomycota dominate at night (**Fig. S4**). The release of fungal spores is strongly linked to environmental humidity (Niu et al., 2021). It has been found that Ascomycota prefers to release spores in relatively dry environments, while Basidiomycota prefers wet environments (Almaguer et al., 2014;Elbert et al., 2007). Drautz-Moses et al. (2022) also observed significant diurnal differences in certain fungal phyla, e.g., Basidiomycota had higher proportions at night. At the order level, Pleosporales (67.4%) dominated in the daytime, while Pleosporales (33.5%), Agaricales (34.6%) and Polyporales (13.3%) were the dominant orders in the nighttime (**Fig. S2b**). Polyporales and Agaricales, belonging to Basidiomycota, were observed to enhance in autumn when rainfall is higher (Abrego et al., 2018). These saprotrophic Basidiomycetes could release more spores at night due to higher air moisture (Kramer, 1982).

Significant diurnal variation was also observed for the fungal genera (**Fig. 2d**). More fungal genera showed higher abundances at night, influenced by temperature and humidity (**Table S3**) (Reyes et al., 2016). Among the dominant genera, the abundances of *Coprinopsis*, *Coprinellus* and *Trametes* were significantly higher at night than in the daytime (*Coprinopsis*: 29.9% > 6.9%,

Coprinellus: 11.2% > 8.9% and *Trametes*: 7.7% > 5.8%). The abundances of *Aspergillus* (10.1%), *Periconia* (8.4%) and *Cladosporium* (5.5%) increased significantly in the daytime (**Fig. 3b**). Similar diurnal patterns were found in the studies of airborne fungal spores in Valladolid (Spain) (Reyes et al., 2016) and Oklahoma (USA) (Gillum and Levetin, 2008).

225 Air relative humidity (RH) facilitates the sporulation process, accompanied by a reduction of atmospheric turbulence (Oliveira et al., 2009;Drautz-Moses et al., 2022), so fungal community structure showed more significant diurnal variations than bacteria.

3.2 Possible factors affecting microbial community

Previous studies have shown that airborne microbial community structure may be influenced by the synergistic effects of meteorological and pollution factors (Zhai et al., 2018;Zhao et al., 2022). To analyze the factors affecting the microbial

230 community at the typical rural site, Gucheng, the SourceTracker method and RDA analysis were performed to identify the influences of the surrounding environment on microbial communities.

Results revealed that the airborne fungi at Gucheng site were mainly derived from terrestrial plants (44.1 ± 29.1%), which was similar to the previous view that plant leaves have a greater contribution to local airborne fungi (Qi et al., 2020). The sources of bacteria were mostly unresolved (98.9 ± 1.0%) (**Fig. S5a**). Zhao et al. (2022) found that human-related sources contributed

235 greater to the airborne bacteria than other sources in urban areas. Bacterial communities are more likely to be influenced by human body surface flora than other environments (Jiang et al., 2022). Bacterial community over the Eastern Mediterranean originated likewise mainly from anthropogenic sources (Mazar et al., 2016). However, only the contribution of natural sources including terrestrial plants, soils and ocean to the airborne microbial community was considered in this study, which may partly explain the high unresolved sources of bacteria. It is also noticeable that in this study only local soil and vegetation

240 samples were selected as the source samples, and the impact of long-distance transport was not considered. Therefore, unresolved sources may also come from long-range transported air masses (Gat et al., 2017) and other local sources, e.g., waters, other vegetations and human-related sources.

RDA analysis presented that Ca^{2+} and wind speed (WS) were the main environmental variables affecting the bacterial community structure (**Fig. S5b**). Ca^{2+} is an important tracer of soil dust particles (Mace et al., 2003) and high WS is conducive

245 to the resuspension of soil microorganisms, bringing soil microorganisms into the atmosphere (Savage et al., 2012;Jones and Harrison, 2004). The results implied that the emission from soils played an important role in shaping the bacterial community. For fungi, air temperature and RH were the vital factors affecting the fungal community (**Fig. S5b**). Temperature and RH could affect the growth and release of microorganisms from the sources, especially fungi. A large number of studies have proved that temperature and RH are able to interfere with airborne fungal communities (Wang et al., 2020;Núñez et al., 2021).

250 Different temperatures can activate the formation and release of different types of fungal spores (Pickersgill et al., 2017). High RH could promote the release of many types of basidiospores and ascospores (Elbert et al., 2007), while when the RH decreases to 40–60%, the release of conidia becomes easier (Jones and Harrison, 2004). The oxidation by ozone can effectively inhibit the growth of microorganisms (Zhai et al., 2018). It should be noted that environmental factors explained only 43.4% of the variation in bacterial communities, and it is hypothesized that ecological processes other than environmental filtration had

255   impacts on shaping bacterial communities. Here, iCAMP analysis showed that the relative contributions of deterministic processes (environmental filtration and microbial interactions) and stochastic processes (birth/death, speciation/extinction, and immigration) to shaping bacterial communities were 53.7% and 46.3%, respectively, demonstrating the importance of other ecological processes on bacterial communities.

3.3 Diurnal variation in sugar compounds

260   A total of 13 sugar compounds including three anhyrosugars, five primary sugars and five sugar alcohols were analyzed in this study. The total concentration of sugar compounds in all the TSP samples averaged 289 ± 85.3 ng m^{-3} (**Fig. 4a**), which was comparable to the previously reported concentrations in rural and mountain aerosols (Yan et al., 2019;Li et al., 2012). The daytime concentration of sugar compounds was 247 ± 67.0 ng m^{-3}, which was lower than that (331 ± 84.4 ng m^{-3}) in the nighttime (**Table S4**).

3.3.1 Anhyrosugars

Anhyrosugars (levoglucosan and its isomers, galactosan and mannosan) are the main constituents of cellulose (Simoneit, 2002) and are specific markers for biomass burning (Simoneit et al., 1999). The concentrations of levoglucosan were in the range of 10.3–30.3 ng m^{-3} (average: 18.4 ± 7.43 ng m^{-3}) in the daytime and 2.97–55.8 ng m^{-3} (average: 30.0 ± 20.1 ng m^{-3}) in the nighttime, showing a diurnal variation. The abundance of levoglucosan was about 10–30 times higher than that of galactosan 270   (daytime: 0.7 ± 0.1 ng m^{-3}, nighttime: 1.4 ± 1.0 ng m^{-3}) and mannosan (daytime: 1.3 ± 0.5 ng m^{-3}, nighttime: 2.1 ± 1.6 ng m^{-3}) (**Table S4**). Anhyrosugars concentrations exhibited diurnal variations (**Fig. 4a**), which may be influenced by cooking activities (Verma et al., 2021). Moreover, the concentrations of anhyrosugars were significantly affected by temperature (**Fig. S6**), which may also be responsible for the diurnal differences in anhyrosugars. Yi et al. (2021) also reported that the anhyrosugars concentrations in PM$_{2.5}$ were lower in the daytime (Tai´an: 52.3 ± 36.3 ng m^{-3}, Mt. Tai: 25.4 ± 16.2 ng m^{-3}) than at night 275   (Tai´an: 55.8 ± 44.3 ng m^{-3}, Mt. Tai: 30.0 ± 19.5 ng m^{-3}) in Shandong Province, China.

3.3.2 Primary sugars

Primary sugars including fructose, glucose, sucrose, trehalose and xylose were detected in all the TSP samples. The primary sugars in the daytime and nighttime samples were 152 ± 49.0 ng m^{-3} and 141 ± 47.2 ng m^{-3}, respectively. These primary sugars dominated the total sugar compounds in the daytime samples ($61.3 \pm 0.1\%$), and relatively lower proportions were found at 280   night ($42.1 \pm 0.1\%$, **Table S4**), which was consistent with the results in previous studies (Zhao et al., 2020;Ren et al., 2018). Sucrose was the primary saccharide with the highest concentration in the daytime (67.6 ± 35.8 ng m^{-3}), followed by glucose (42.4 ± 14.7 ng m^{-3}). Sucrose has the potential origins of pollen (Fu et al., 2012) and soil resuspension (Rogge et al., 2007), and can be produced by photosynthesis in plant leaves (Medeiros et al., 2006b). However, no significant correlation was found between sucrose and soil resuspension-related indicators (Ca^{2+} and PM$_{10}$). Sucrose had a significant positive correlation with 285   xylose (**Table S5**), which mainly originates from wood and rice straw (Chen et al., 2013;Sullivan et al., 2011), indicating that

sucrose and xylose may have a similar source. It is speculated that the elevated concentrations of sucrose in the daytime may be caused by the more active metabolic activity of plants.

Trehalose was the dominant primary saccharide at night with an average of 67.6 ± 23.4 ng m^{-3}, followed by glucose (36.6 ± 12.8 ng m^{-3}) (**Fig. 4a**). Trehalose exhibited a diurnal variation with higher concentrations at night and lower concentrations in the daytime, which was in contrast to the variations of the other four detected primary sugars (**Table S3**). Trehalose, as a reserve carbohydrate and pressure protector (Medeiros et al., 2006a), is widely found in various organisms, such as bacteria, fungi, insects, invertebrates, algae and higher plants (Medeiros et al., 2006b). As the most abundant sugar compound in soils, trehalose was used as the tracer for soil dust resuspension (Jia and Fraser, 2011;Rogge et al., 2007). Whereas, in this study, there was no significant correlation between trehalose and soil resuspension-related indicators (Ca^{2+} and PM_{10}). Trehalose was significantly negatively and positively correlated with air temperature and relative humidity, respectively (**Table S5**). Trehalose exhibited an apparent diurnal variation (**Fig. 4a**), which was consistent with the diurnal variation trend of fungal OTUs (**Fig. 1b**). These results indicate that trehalose was possibly affected by fungal emissions.

3.3.3 Sugar alcohols

Arabitol, mannitol, erythritol, glycerol and inositol were the main sugar alcohols detected in the TSP samples. The total concentration of sugar alcohols was averaged 115.4 ± 50.3 ng m^{-3} and their relative abundance was lower in the daytime ($30.6 \pm 0.07\%$) than in the nighttime ($48.4 \pm 0.08\%$) (**Fig. 4a**). Mannitol was the predominant sugar alcohols in all the samples (71.1 ± 34.8 ng m^{-3}), followed by arabitol (23.6 ± 12.5 ng m^{-3}). Mannitol and arabitol were the most predominant organic compounds used to indicate fungal spores and showed significant correlations with fungal community diversity in terms of OTU abundance (**Table S5**). 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 temperatures (Zhu et al., 2015).

The concentrations of mannitol and arabitol are commonly used to evaluate the contribution of fungal spores to ambient OC (Bauer et al., 2008a). The contributions of other primary emission sources such as biomass burning and plant debris to OC could be estimated by levoglucosan and glucose, respectively (Fu et al., 2014;Puxbaum et al., 2007;Puxbaum and Tenze-Kunit, 2003;Andreae and Merlet, 2001). The results estimated based on these tracers show that fungal spore-derived OC dominated (204.3−977.8 ngC m^{-3}), accounting for 3.6–19.7% of OC. Plant debris and biomass burning contributed 0.4–1.6% and 1.1–6.9% to OC, respectively (**Fig. 4b**). Biological sources contributed more to primary OC in rural aerosols, while biomass-burning OC in urban aerosols were dominant in total OC (Wu et al., 2020;Yan et al., 2019). The nighttime fungal spore-derived OC (525.5–977.8 ngC m^{-3}) was significantly higher than that in the daytime (204.3–427.9 ngC m^{-3}) ($p < 0.01$), which was consistent with the observation in other studies (Kang et al., 2018;Zhu et al., 2016). Many fungal spores are wet spores, preferring to release spores at night when the RH is higher.

3.4 Relationship between microbial community and sugar compounds

3.4.1 Correlation between microbial community structure and sugar compounds

To analyze the relationship between different microbial communities and sugar compounds, this study adopted the Mantel's test to determine the possible correlation between microbial community structure and sugar compounds (**Fig. 5a**). The bacterial community structure was closely related to sucrose, while fungal communities were associated with more detected sugar molecules, including all the detectable anhydrosugars, two sugar alcohols mannitol and erythritol and one primary sugar sucrose. These results indicate that the fungal community may have a larger contribution to more sugar molecules than the bacterial community in the rural atmosphere. Compared to bacteria in the air, fungal emissions and concentrations are lower in number, but much (an order of magnitude) higher in mass (Fröhlich-Nowoisky et al., 2016;Hoose et al., 2010).

Anhyrosugars are important tracers of biomass burning (Simoneit et al., 1999) and sucrose is considered mainly from wood and rice straw. Fungi are more common on wood or plant surfaces than bacteria (Lladó et al., 2017;Andrews and Harris, 2000). In this study, three genera *Coprinopsis*, *Coprinellus* and *Trametes* dominant in the fungal community, are typical wood saprophytic fungi (Kamada et al., 2010;Nagy et al., 2012). Minor fungal genera, such as *Periconia* and *Cladosporium*, belong to endophytes (Gunasekaran et al., 2021). As mentioned above, SourceTracker analysis revealed that fungi were mainly derived from plants (**Fig. S5a**). Meanwhile, fungi can use levoglucosan or sucrose as the carbon source to promote growth (Prosen et al., 1993;Van der Nest et al., 2015). It was also found that bacterial community structure may be closely related to sucrose-traced plant origins, although the SourceTracker analysis results did not show the plant origin of bacteria. This inconsistency may be caused by single plant (maize) source samples and/or the negligence of long-distance transport in this study.

The correlations between the fungal community and two sugar alcohols (mannitol and erythritol) were the most significant (correlation coefficients: 0.49 and 0.55) (**Fig. 5a**). Mannitol exists in fungal spores, fruiting bodies and mycelia of fungi (Solomon et al., 2007), and is considered to be the most abundant dissoluble starch inside mycelia and fruiting bodies (Dulermo et al., 2009). In fungi, mannitol can be used to store energy and extinguish reactive oxygen species (Upadhyay et al., 2015). The natural sources of erythritol are mainly yeasts and yeast-like fungi, such as *Moniliella pollinis*, *Trichosporonoides megachiliensis*, *Aureobasidium* sp., *Trigonopsis variabilis*, *Trichosporon* sp., *Torula* sp., and *Candida magnoliae* (Regnat et al., 2018). They mainly produce erythritol when encountering salt or osmotic pressure (Yang et al., 2015). Mannitol has been widely used to trace the origin of fungal spores in aerosols (Bauer et al., 2008b;Bauer et al., 2008a), and erythritol was considered to have similar sources to mannitol (Chen et al., 2013).

3.4.2 Interaction between sugar molecules and microorganisms at taxonomic levels

To elucidate the interaction between sugar molecules and microorganisms at taxonomic levels, a co-occurrence network was constructed based on Spearman's correlation between different microbial taxa and sugar molecules. Based on correlation analysis, 27 edges, which depict significant correlations between sugar molecules and taxa (FDR-adjusted p-value < 0.05,

correlation coefficients > 0.6), were captured between 28 nodes (**Fig. 5b**). All microbial taxa related to the sugar molecules belonged to fungi, suggesting the interaction between sugar molecules and fungal taxa. Modularity analysis revealed that the entire network was divided into seven modules, only three of which were comprised of sugar molecules nodes and more than two OTUs nodes (Modules I, II and IV). In general, nodes in one module interconnected more frequently among themselves than with nodes in other modules. However, the nodes of these three modules were mixed, suggesting that certain taxa (e.g., *Sporobolomyces*) may contribute to multiple sugar molecules at the same time. A previous study has shown that there is a significant positive correlation between *Sporobolomyces* and sugar compounds (Samaké et al., 2020).

Meanwhile, we found that only three sugar alcohols (mannitol, arabitol and erythritol) used to trace fungal spores were associated with a few fungal genera (*Peniophora*, *Sporobolomyces*, *Hydnophlebia*, *Hannaella*, *Lectera* and *Cyphellophora*) (**Fig. 5b**), and other sugar molecules had limited links with microbial taxa. The three sugar alcohols had significant positive correlations ($p < 0.05$) with fungal OTU abundances (**Table S5**), possibly implying the important roles of sugar alcohols playing in tracing fungal spores. In contrast, no bacterial taxon was significantly related to sugar molecules. Combined with the results of the Mantel's test and network analysis, it is considered that sugar compounds in rural aerosols had a strong association with overall fungal community structure, while bacteria have a weak effect on sugar compounds possibly due to their limited mass.

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.

4 Conclusions

In this study, the airborne microbial taxonomy and the compositions of sugar molecules in atmospheric aerosols collected in rural Gucheng, North China Plain were obtained. Results confirm that fungal communities have more pronounced diurnal differences than bacteria. Airborne fungi mainly originated from plant leaves, while the sources of airborne bacteria were mostly unresolved, which may be human-related and/or long-distance transport sources. Among sugar compounds, sucrose dominated in the daytime samples ($27.2 \pm 12.7\%$), while mannitol dominated at night ($31.1 \pm 5.9\%$). From the perspective of the overall microbial community, most sugar molecules were strongly associated with the fungal community structure, while

the bacteria community structure was weakly related to sucrose only. At the microbial taxon level, only a few saprophytic fungal taxa were closely associated with sugar compounds. The results suggest that the entire fungal community may largely contribute to sugar compounds, while bacterial community contributed little to them. The presented results in this study will help to understand the contribution of bioaerosols to organic carbon in atmospheric aerosols and build links between airborne

385 microorganisms and aerosol chemistry. Importantly, it should be noted that the uncertainties in estimating the concentrations and emission fluxes of bioaerosols using certain sugar compounds need to be seriously considered. For example, arabitol, usually used as a fungal spore tracer, showed no significant correlation with fungi in this study. More likely, different seasons, climates and locations may lead to differences in the microbial community structure as well as corresponding sugar composition and metabolites.

390 Moreover, the airborne microbiota is highly complex, and biomolecules such as carbohydrates, proteins and lipids are major chemical compositions and metabolites of airborne microorganisms. In this study, the measured carbohydrate species are limited to small molecule compounds. In the future, it is necessary to detect more carbohydrates and other compounds in aerosols from different typical regions by using high-resolution mass spectrometry to further study the relationship between microorganisms and atmospheric aerosol chemistry.

395 **Data availability**

The sequence data is available in the NCBI Sequence Read Archive under accession numbers SUB12879673 and SUB12879615.

Author contributions

MTN analyzed data and prepared all figures. SH and WW conducted the observation. SH, YJW, RJ, QZ, ZHW and DHZ
400 performed experiments. MTN and WH wrote the manuscript. WYX, LBW, JJD, FXS and PQF revised the original manuscript draft. WH and PQF supervised the project. All authors were involved in helpful discussions and contributed to the manuscript.

Competing interests

The contact authors have declared that none of the authors has any competing interests.

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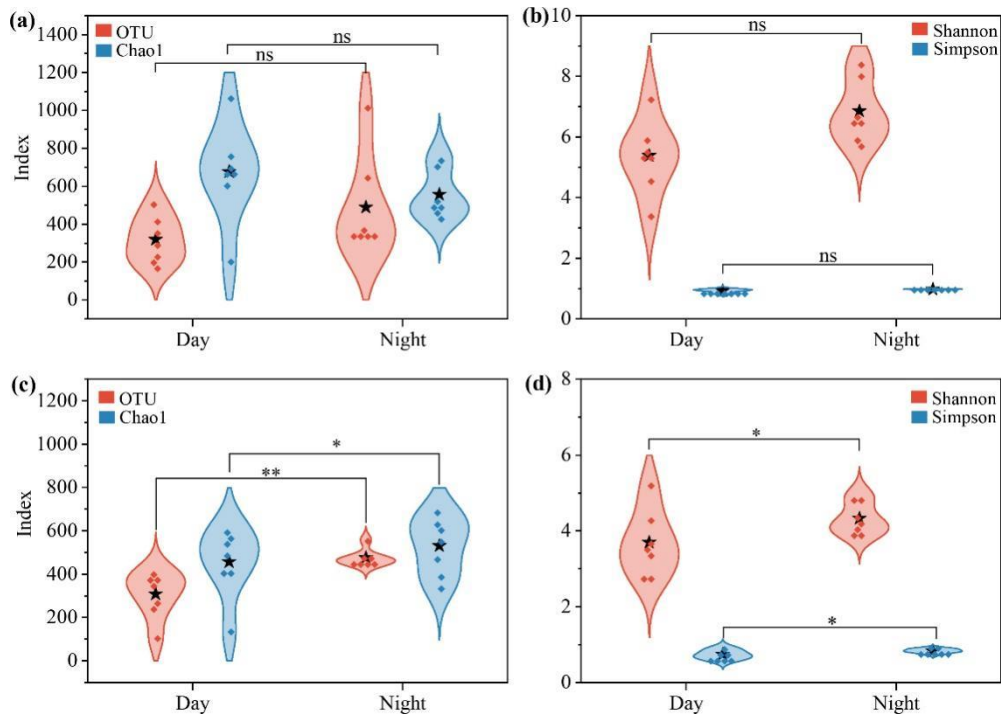
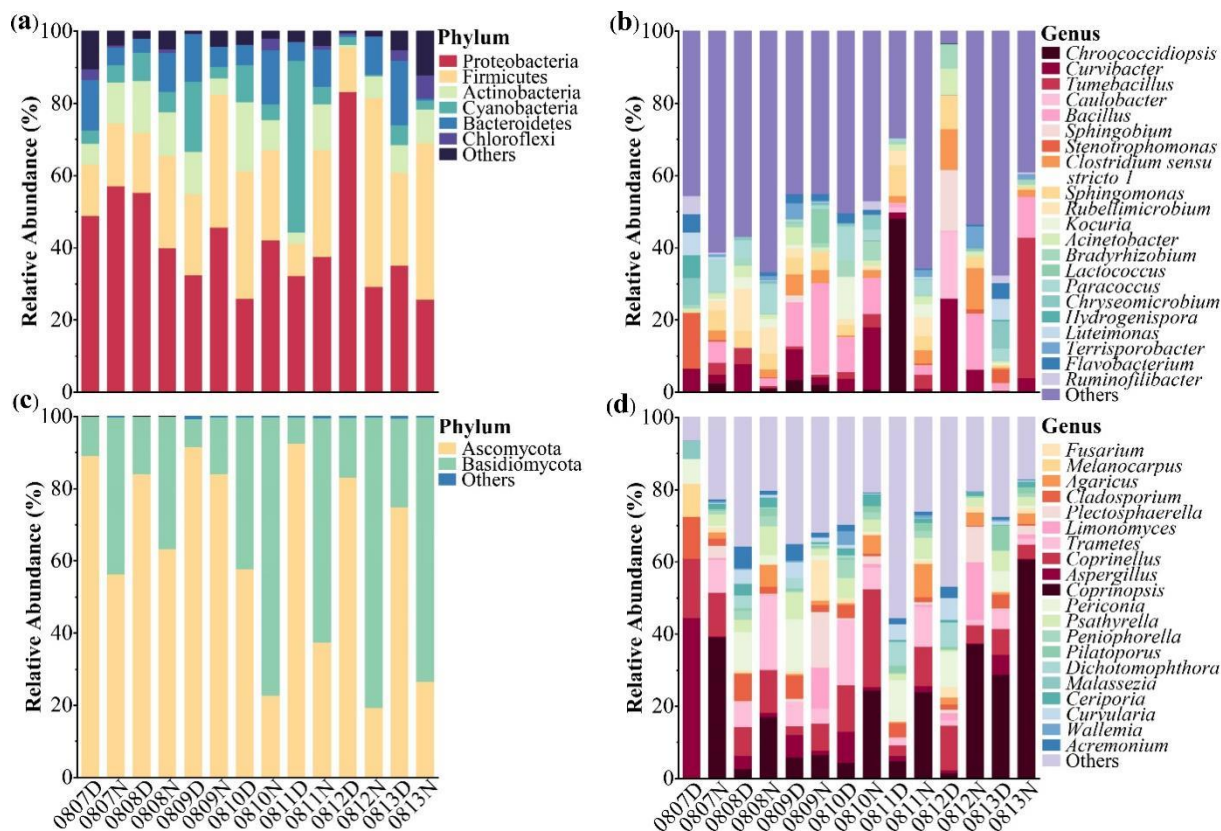

740 **Figure 1. The α diversity of airborne microbial community at rural Gucheng site.** (**a-b**) bacterial and (**c-d**) fungal diversity and richness in the daytime and nighttime samples were compared based on the one-way ANOVA analysis. The dots in the violin shapes represent all data points. Stars represent the average value of all data. * and ** represent that the p-value was less than 0.05 and 0.01, respectively; ns indicates that there is no significant difference.

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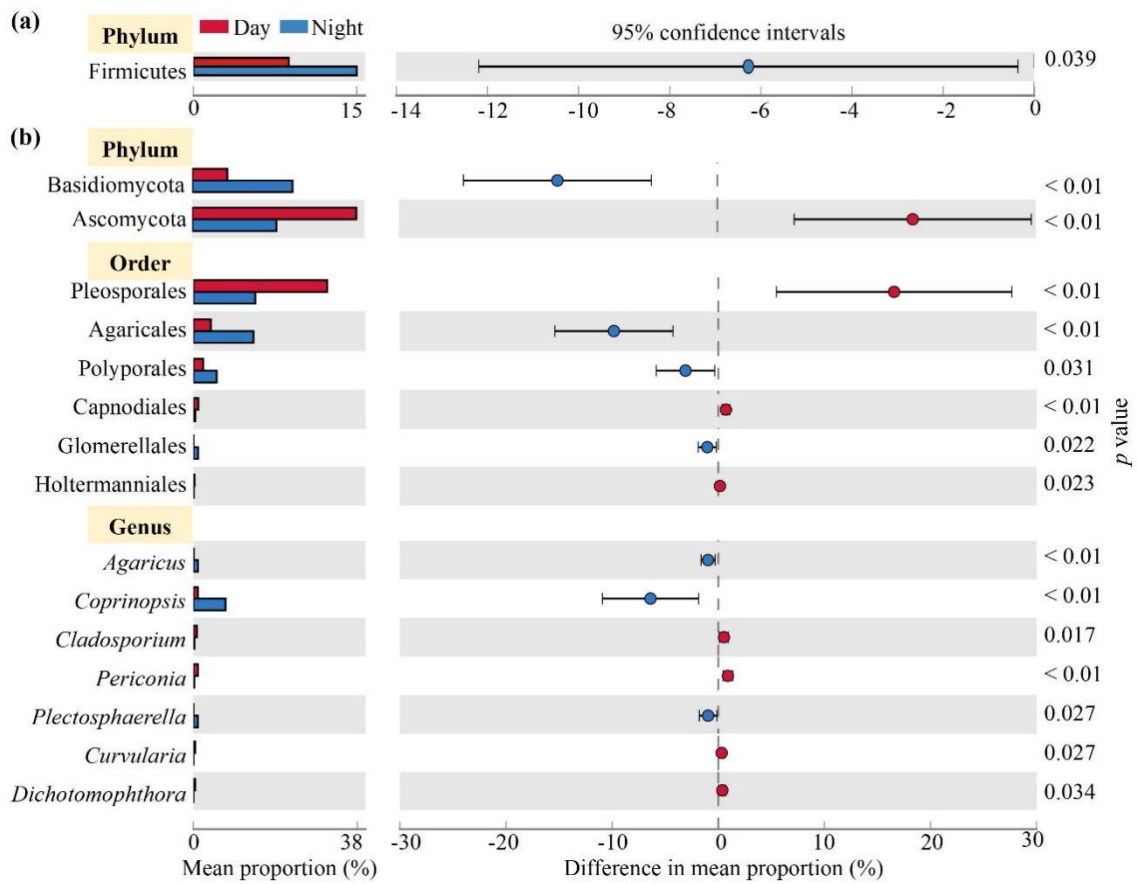
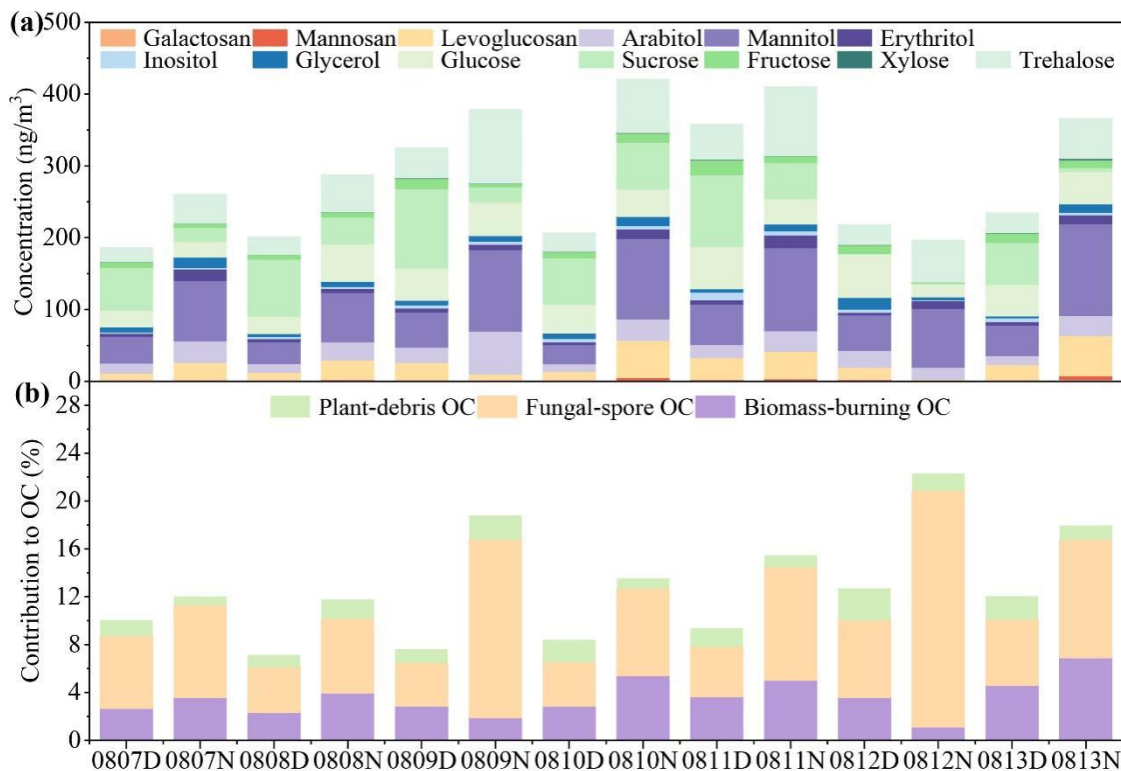
Figure 2. Taxonomic compositions of microorganisms in TSP samples collected at rural Gucheng site. Relative abundances of dominant bacterial (**a**–**b**) and fungal (**c**–**d**) taxa at the phylum (left panel) and genus (right panel) levels were shown. D and N in the sample IDs mean daytime and nighttime, respectively.

 Figure 3. Diurnal variations in the proportions of microbial taxa. The relative abundances of (**a**) bacterial and (**b**) fungal phyla, orders and genera with significant diurnal differences ($p < 0.05$) based on the one-way ANOVA analysis were shown in the left part of the figure. The right part of the figure shows the differences between the average abundances of the microbial taxa in the daytime and nighttime samples.

755 **Figure 4. Diurnal variations of sugar molecules in TSP samples from rural Gucheng site.** The concentrations of 13 sugar compounds including three anhyrosugars, five primary sugars and five sugar alcohols are shown in (**a**). (**b**) presents the contributions of plant-debris OC, fungal spore OC and biomass-burning OC to total OC. The concentrations of plant-debris OC, fungal spore OC and biomass-burning OC were estimated according to the previous studies (Bauer et al., 2008a;Fu et al., 2014;Puxbaum et al., 2007). D and N in the sample IDs mean daytime and nighttime, respectively.

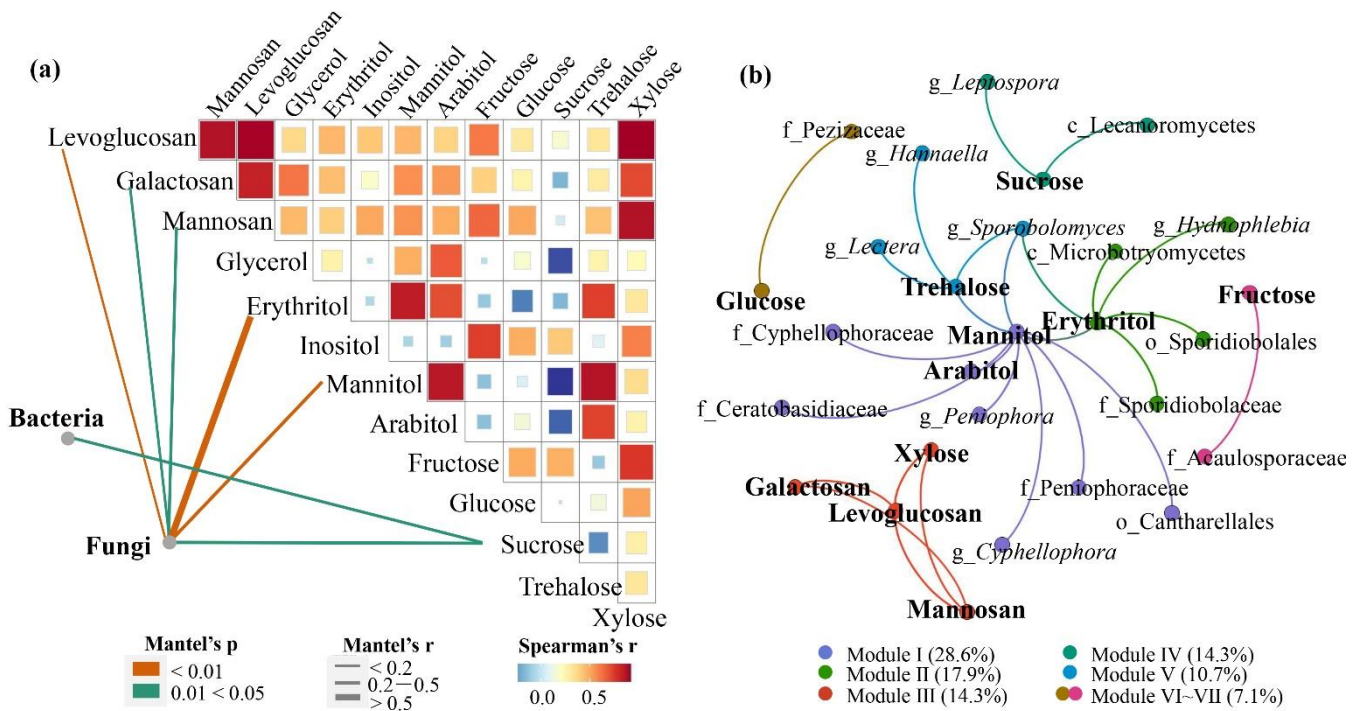



Figure 5. Association between microbial community and sugar compounds. (**a**) presents the correlations of the microbial community structures with sugar compounds based on the Bray–Curtis distance. Edge width corresponds to Mantel's r value, and the edge color denotes statistical significance. Pairwise correlations of sugar compounds with a heatmap are calculated by Spearman's correlation analysis. (**b**) Co-occurrence networks between microbial taxa and sugar components were constructed based on Spearman's correlation analysis. The nodes are colored according to different types of modularity classes.