Measurement Report:Characterization of sugars and amino acids in atmospheric fine particulates: identify local primary sources

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Abstract. Sugars and amino acids are major classes of organic components in atmospheric fine particles and play important roles in atmospheric processes. However, the identification of their sources in different regions is less explored. To characterize local primary sources (biomass burning, plant and soil sources) and evaluate their contributions to the total sugar compounds and amino acids (AAs) pool in different regions, fine particulate matter samples were collected from the urban, rural and forest areas in Nanchang, China. The concentrations and compositions of sugar compounds (anhydrosugars, primary sugars and sugar alcohols), free amino acids (FAAs) and combined amino acids (CAAs) were analyzed by gas chromatography–mass spectrometry (GC-MS) after silylation derivatization. Urban area had significantly higher average Σsugars concentration (317 ± 139 ng m^{-3}) than that of the rural (181 ± 72 ng m^{-3}) and forest (275 ± 154 ng m^{-3}) areas ($p < 0.05$). Overall, the distribution pattern of sugar compounds and CAAs in PM$_{2.5}$ was generally similar in three areas. Levoglucosan accounted for 24.4%, 22.0% and 21.7%, respectively of the total sugars pool in the urban, rural and forest areas. These suggest besides biomass burning (BB), plant and soil sources have important contributions to aerosol sugars and CAAs in three areas. In the urban area, the concentrations of anhydrosugars showed a positive correlation with combined Gly concentrations, but no correlation was found between these two compounds in the rural and forest areas, indicating that the urban area is mainly affected by local combustion sources. This conclusion was also supported by the positive correlation between levoglucosan and non-sea-salt potassium only observed in the urban area (Lev $= 0.07$ K$^+$$+37.7$, r$=0.6$, p$<0.05$). In addition, the average levoglucosan and mannosan (L/M) ratio in the urban area (59.9) was much higher than those in the rural (6.9) and forest areas (7.2), implying BB aerosols collected in the urban area originated from lignite burning while the type of biofuels used in the rural and forest areas is mainly softwood. The concentrations of sugar alcohols in the rural and forest areas were positively correlated with that of CAAs, which are abundant in the topsoil (r $= 0.53 \sim 0.62$, $p < 0.05$), suggesting that the contribution of local topsoil sources is large in these two areas. In the rural and forest areas, the concentrations of primary sugars were positively correlated with that of combined CAA species abundant in local dominant vegetation. Our findings suggest that combining specific sugar tracers and chemical profiles CAAs in

local emission sources can provide insight to primary sources characteristics including the types of
biofuels burned, the contribution of topsoil sources and local vegetation types.

1 Introduction

Organic aerosols (OAs), are recently highlighted because they account for a significant fraction (20%–90%) of fine aerosols (Verma et al., 2021; Wu et al., 2018; Xu et al., 2020a). Furthermore, they may influence the global climate as condensation nuclei (CCN) and ice nuclei (IN) (Ng et al., 2010; Steinfeld, 1998), contribute a large amount of terrestrial organic carbon to the remote oceans (Zhu et al., 2015), and cause human health problems (Nel, 2005; Fu et al., 2008). Thus, a profound understanding of the abundance, composition and sources of OAs in fine aerosols is of great significance to reduce the adverse effects of OAs on the atmospheric environments and human being health (Fan et al., 2020).

As the vital fraction of water-soluble organic compounds (WSOC), sugars and AAs are ubiquitous in the atmosphere from different geographical locations, including urban (Wang et al., 2019), forests (Mace et al., 2003), mountains (Barbaro et al., 2020; Fu et al., 2008), marine (Fu et al., 2013; Matsumoto and Uematsu, 2005) and polar regions (Feltracco et al., 2020; Feltracco et al., 2019). Moreover, chemical fingerprints of these two major classes of organic components in atmospheric particles have been used to elucidate relative contribution of different sources to aerosols (Jia et al., 2010; Matos et al., 2016; Yan et al., 2019). Specifically, levoglucosan and related anhydrosugar isomers (mannosan and galactosan), produced from pyrolysis of cellulose and hemicellulose at temperatures higher than 300°C and ranking as the most abundant compounds in biomass burning emissions, have been recognized as specific molecular markers for biomass burning (BB) sources (Simoneit, 2002). Although there is still lack of knowledge about BB and amino acids, the potential correlation between the free glycine (Gly) concentrations and BB aerosols has been suggested in past studies (Zhu et al., 2020a; Samy et al., 2013; Violaki and Mihalopoulos, 2010).

Primary sugars and sugar alcohols are frequently utilized in tracking and estimating the contribution of soil and associated biogenic aerosols, including fungi, viruses, bacteria, pollen, and plant and animal debris (Liang et al., 2016). For instance, plant-debris-derived aerosol organic carbon was estimated based on the relationship of glucose and plant debris (Fan et al., 2020). Sucrose have been widely employed to indicate airborne pollen grains (Wang et al., 2021; Fu et al., 2012; Yan et al., 2019). The sugar alcohols arabitol and mannitol, are found to be produced by fungi, lichens, soil biota, and algae to overcome environmental stress (Medeiros et al., 2006). These sugar alcohols have been recently proposed as molecular tracers for fungal spores (Elbert et al., 2007). In addition, algae and higher plants are also proved to be an important source of mannitol (Feltracco et al., 2020). Similarly, the level of combined amino acids (CAAs) in aerosols has been used as marker for primary biological aerosol particles (PABPs) and its composition can distinguish different types PABPs based on chemical composition of CAAs in corresponding biological sources (Axelrod et al., 2021; Abe et al., 2016).

Obviously, sugars and AAs in aerosols may share similar source to some extent. However, the concentrations, compositions and source apportionment of sugars and AAs in aerosols have been studied separately in previous studies. Till now, few studies have focused on sugars and amino acids in aerosols simultaneously, and the concentration and composition profiles of sugar compounds and AAs in different

areas impacted by different local primary sources is still poorly understood (Barbaro et al., 2019; Ruiz-Jimenez et al., 2021). It is expected that coupling source specific sugars with most abundant AA species in local primary sources may provide insight into more source signatures.

In recent years, we investigated the source and possible atmospheric transformation process of amino acids and proteinaceous material in different size-segregated particles collected from the forest, rural, suburban and urban sites under different ambient oxidation levels (Zhu et al., 2020a; Zhu et al., 2020b; Zhu et al., 2021b; Zhu et al., 2021a; Wen et al., 2022). The nitrogen isotope composition of free glycine ($\delta^{15}N_{F\text{-}Gly}$) in potentially emission sources were analysed and $\delta^{15}N_{F\text{-}Gly}$ values released by BB sources are significantly higher than those from natural primary sources (Zhu et al., 2020a). In the study of Zhu et al. (2020b), the origin of FAAs and CAAs and their atmospheric transformation processes in $PM_{2.5}$ were explored by analysing the concentrations of FAAs and CAAs, and $\delta^{15}N$ values of free and combined Gly in $PM_{2.5}$ and main primary emission sources (plant, soil, and aerosol from BB). Combined compound-specific $\delta^{15}N$ patterns of hydrolyzed amino acids (HAAs), degradation index with the variance within trophic HAAs in fine and coarse particles, we demonstrated that AAs in coarse particles have advanced bacterial degradation states compared to fine particles (Zhu et al., 2021b). Correlations between the concentrations of FAAs, CAAs and oxidants under different ambient NO_2 and O_3 levels were discussed to elucidate the mechanism of FAAs formation from the protein degradation in relation to ambient NO_2 and O_3 concentrations (Zhu et al., 2021a). Although primary sources are proved to be important sources of AAs in the atmosphere in our previous study, further information of local primary sources characteristics including the type of biofuels burned, local vegetation types and the contribution of topsoil sources are missing.

To further clarify local primary source information in different locations, the concentration and composition of sugars, FAAs and CAAs in $PM_{2.5}$ samples collected from the urban, rural and forest areas in Nanchang, China were analysed. In order to obtain a sufficient sample size for analysis of sugar compounds (anhydrosugars, primary sugars and sugar alcohols), FAAs and CAAs in one sample simultaneously, in this study different filter samples were collected during the same sampling period as our previous study (Zhu et al., 2020b). Given the documented roles of specific sugars as tracers for BB and PBAPs sources and the distribution of CAAs in primary sources, the variation pattern of the concentration and composition of specific CAAs species predominant in local primary source and the corresponding sugar tracers were compared. The correlations of source specific sugars with free amino acids (FAAs) and with major CAA species in local primary sources were evaluated (1) to character the concentration and composition profiles of sugar compounds, FAAs and CAAs in urban, rural and forest areas, (2) to obtain the information of the type of biofuels, local vegetation types and the contribution of topsoil sources in different sampling areas.

2 Materials and Methods

2.1 Sample Collection

$PM_{2.5}$ samples were collected at three different areas (urban, rural and forest) in Nanchang, China from April 30, 2019 to May 13, 2019 (Figure S1). More details of sampling locations are available in

supplement material (Table S1). Forty-two filter samples were collected daily with a duration of 23.5 h on prebaked quartz fiber filters (450 °C, 10h) using high volume air samplers (KC-1000, Qingdao Laoshan Electronic Instrument company, China) at a flow rate of 1.05 ± 0.03 m^3 min^{-1}. Sampling was performed on the roof of buildings (15 m height) at each site. After the sampling the filter was recovered into a pre-combusted glass jar with a Teflon-lined screw cap, immediately placed in the freezer (the temperature is set to -20℃) and then transported to the laboratory within 1 hour and stored at -20℃ before analysis, to prevent loss of semi-volatile/volatile organic compounds from the samples.

The field blank samples were collected by using a modified version of (Chow et al., 2010). After treated at 450 °C for 10 hours, field blank filters were loaded into filter holders and accompany the sampled filters to each sampling site. Field blanks accompanied sample shipments and were placed in the high-volume air samplers along with the sampled filters. The passive period for field blank filters was consistent with the samples. The only difference between samples and field blanks is that air is not drawn through field blanks. After the sampling, field blank filters were also recovered into filter holders and accompanied with the samples back to the laboratory. Three field blank samples were collected in this study, accounting for 7% of total sample number.

2.2 Analysis of sugar concentrations

For sugars analysis, samples were prepared using a modified version of (Fu et al., 2008). In brief, a filter aliquot (~160 m^3 of air) of each aerosol sample was extracted three times with dichloromethane/methanol (2:1; v/v) under ultrasonication for 10 min. The solvent extract was filtered through quartz wool packed in a Pasteur pipette and then dried with pure nitrogen gas. The dried extracts were reacted with 50 ml of N,O-bis-(trimethylsilyl) trifluoroacetamide (BSTFA) containing 1% trimethylsilyl chloride (Sigma-Aldrich, St Louis, MO, USA) and 10 μl of pyridine (Sigma-Aldrich, St Louis, MO, USA) at 70℃ for 3 h. After the reaction, derivatives were diluted by the addition of 140 μl of n-hexane containing the internal standard (C13 n-alkane, 1.43 ng μl^{-1}) prior to analysis by gas chromatography-mass spectrometry (GC-MS).

GC-MS analyses were performed on a Thermo Scientific TRACE GC (Thermo Scientific, Bremen, Germany) connected into a Thermo Scientific ISQ QD single quadrupole MS. The GC separation was achieved on a DB-5MS fused silica capillary column (30m × 0.25mm i.d., 0.25 μm film thickness) with the same GC oven temperature program: temperature hold at 90℃ for 1 min, increase from 90 to 120℃ at a rate of 12℃ min^{-1}, then further increase from 120 to 285℃ at a rate of 6℃ min^{-1} with a final isotherm hold at 285℃ for 18 min. Helium was used as the carrier gas at a constant flow rate of 1.2 ml min^{-1}. The temperatures of GC injector and MS ion source were 280 °C and 250 °C, respectively. The mass spectrometer was operated in the electron impact (EI) mode at 70 eV and scanned from 50 to 650 Da. The individual compounds (TMS derivatives) were identified by comparing mass spectra with those reported in the literature and library data and authentic standards (Medeiros and Simoneit, 2007). Compounds were quantified using total ion current (TIC) peak area of each authentic standard relative to that of internal standard (C13 n-alkane), and converted to compound mass using calibration curves of internal standards. In general, the obtained calibration curves show good linearity ($R^2 > 0.99$) in the exploited range (Table S2). The field blank filters were also analyzed by the procedure mentioned above

155 in this section and then were run in parallel with the samples for all analyses in order to monitor significant background interferences (Urban et al., 2012). No signal different than that of the base line was obtained for any of the studied sugars. The concentrations of sugar reported here are corrected for the field blanks. The detailed method validation was provided in supporting information.

2.2 Analysis of FAAs and CAAs concentrations

160 The concentrations of FAAs and CAAs were prepared following the method described by our previous study and analyzed by gas chromatograph-mass spectrometer (Thermo Scientific, Bremen, Germany) (Zhu et al., 2020b). For more details of their concentrations analysis method are provided in the supplementary information.

2.3 Analysis of water-soluble ions

165 Water-soluble ions (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^- and SO_4^{2-}) were measured by ion chromatography (Dionex Aquion (AQ)™, ICS-90, Thermo Scientific™, USA) (Guo et al., 2020). Briefly, one eighth quartz filters were cut into pieces and transferred to a Nalgene tube. After added 50 mL Milli-Q water, the tube was ultrasonic vibration (30min), shaking (30min), and centrifugation (10min). Then, the supernatant was filtered by using pinhole filter with a 0.22μm microporous membrane. The extract

170 was stored in a refrigerator at -20°C for analyses. The anions (Cl^-, NO_3^- and SO_4^{2-}) were determined by the ion chromatography system with an AS23 4 × 250 mm analytical column, while the cations (Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}) were determined using the same IC system with a CS12A 4 × 250 mm column. A sample volume was 100 μL for both anion and cation analyses. Strict quality control was performed to avoid any possible contamination of the samples. In order to ensure the reliability of data, standard

175 samples were re-measured every twenty samples. The standard solutions were bought from Merck, Germany. More details on quality assurance and control of water-soluble ions are available in the supporting information.

The mass concentrations of non-sea salt K^+ (nss-K^+) and non-sea salt Ca^+ (nss-Ca^+) were calculated based on the measured Na^+ was assumed to be derived from sea salt as follows Ren et al. (2018) and Zhang et

180 al. (2018):

nss-K^+= K^+ -Na^+×0.037

nss-Ca^+= Ca^+ -Na^+×0.0385

2.4 Statistical Analyses

Statistical analysis and graphs were mainly was conducted by R (version 4.0.2) and Origin 2018

185 (OriginLab Corporation, USA). Normality tests of Shapiro-Wilk were performed. The concentrations sugar compounds (anhydrosugars, primary sugars and sugar alcohols), FAAs and CAAs are normally distributed. Pearson's correlations were conducted to examine the relationship between target sugar category and TFAA, between target sugar category and TCAA, between specific sugar tracers and individual CAA and between specific sugar tracers and individual FAA. Significant differences in the

190 sugars concentration among three sampling sites were tested using the one-way analysis of variance

(ANOVA) procedure, and compared using the LSD test; differences were considered significant at the level of $p < 0.05$.

3 Results and discussion

3.1 Sugars in the urban, rural and forest locations

3.1.1 Sugars concentrations in PM$_{2.5}$

The temporal variations of total sugars concentrations in PM$_{2.5}$ measured at the urban, rural and forest locations during the sampling campaign were shown in Figure 1. The total sixteen sugars (Σsugars) were quantified in the atmospheric fine particulates. Average Σsugars concentration measured in the urban, rural and forest location was 317±139, 181±72, 275±154 ng m^{-3}, respectively. Urban location had significantly higher average Σsugars concentration than that observed in the rural location ($p < 0.05$), but the differences in average Σsugars concentration between rural and forest locations are not significant ($p > 0.05$) (Figure S2).
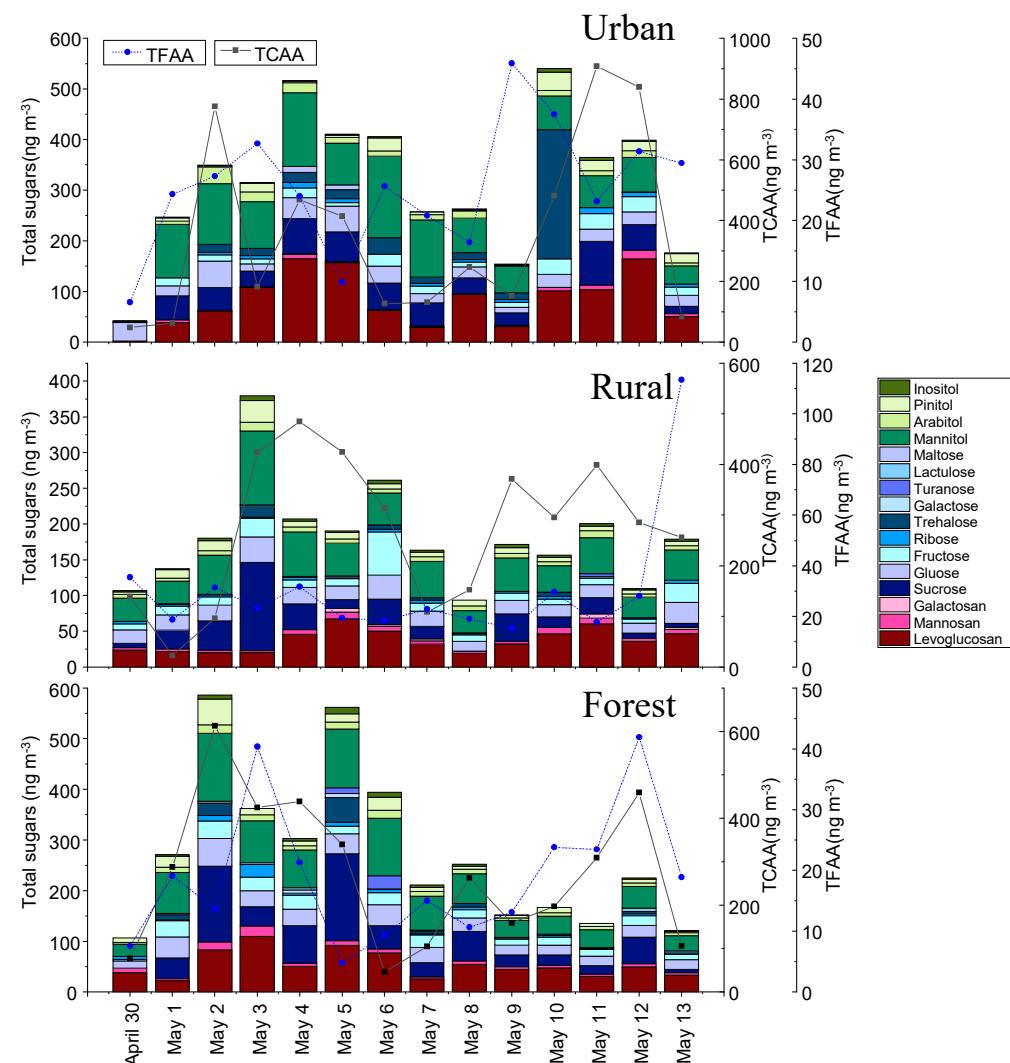
Figure 1. Temporal variations in the concentrations of total sugars and total combined amino acids measured in PM2.5 samples collected at the urban, rural and forest sites in Nanchang, China.

Three target sugar categories (anhydrosugars, primary sugars, alcohol-sugar) were compared between the three sampling locations. The average concentrations of anhydrosugars in the urban location (88 ± 56 ng m^{-3}) was higher than those in the rural (44 ± 20 ng m^{-3}) and forest locations (62 ± 31 ng m^{-3}) (Figure S2). Similarly, the highest average concentration of levoglucosan, a good tracer of BB (Sheesley et al., 2003), was found in the urban location (83 ± 53 ng m^{-3}), followed by that in the rural (37 ± 16 ng m^{-3}) and forest locations (54 ± 27 ng m^{-3}) (Figure 2), indicating enhanced biomass burning activities in the urban location.
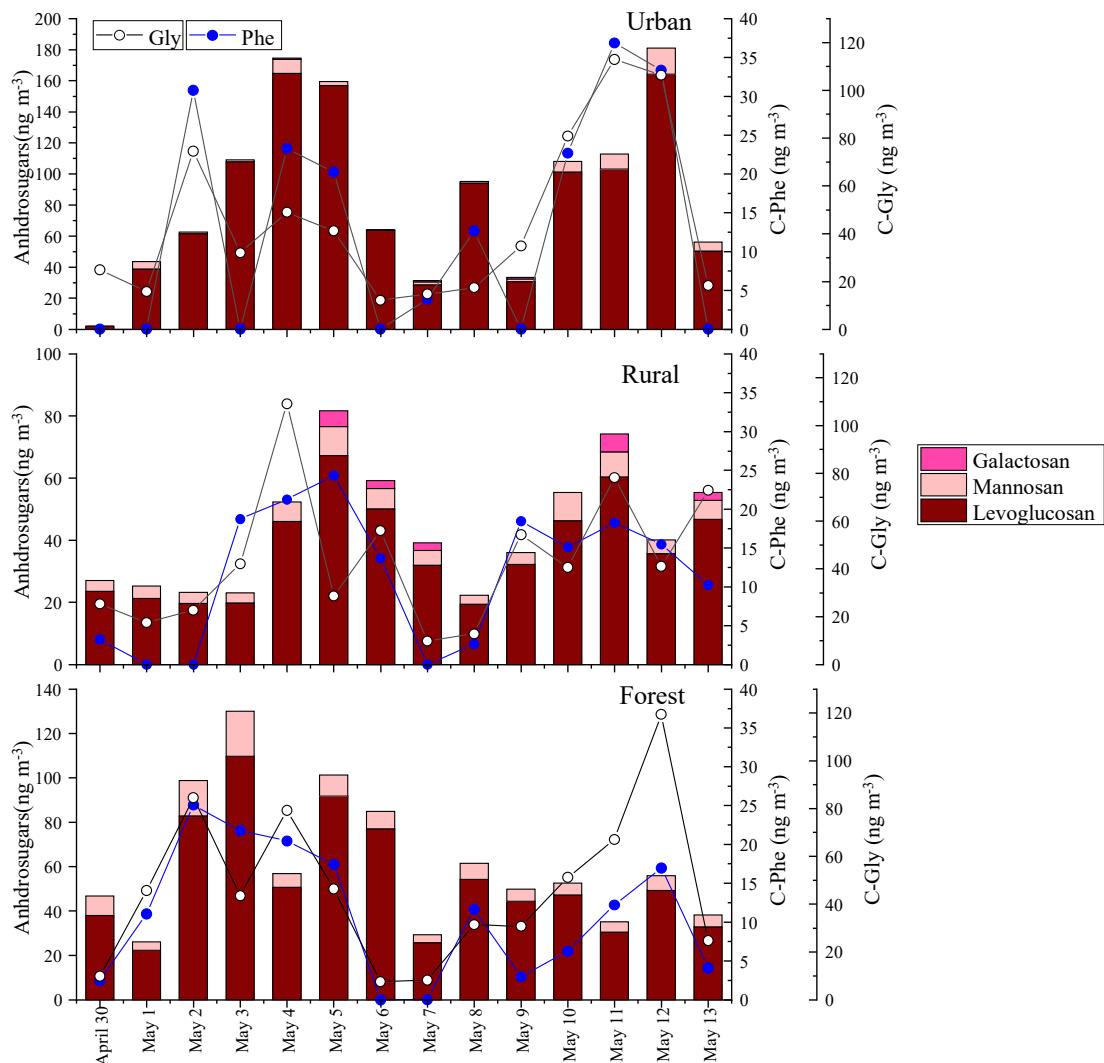
Figure 2. Concentrations of anhydrosugars, combined Phe and Gly in urban, rural and forest. The concentrations of anhydrosugars and CAAs for each sample were normalized for the total volume of air sampled.

Nine primary sugars (fructose, glucose, xylose, sucrose, and trehalose) were measured in this study. The average concentration of total primary sugars measured in PM$_{2.5}$ was comparable in the urban, rural and forest location, with the averaged value of 120 ± 69, 71 ± 47 and 120 ± 82 ng m^{-3}, respectively (Figure 3). It is interesting to note that the average concentration of trehalose was higher in the urban location (28±66 ng m^{-3}) than those in the rural (2±4 ng m^{-3}) and forest locations (7±14 ng m^{-3}) ($p < 0.05$) (Figure S3). Trehalose has been reported to be the most abundant sugar in soils and be used as a tracer for the resuspension of surface soil and unpaved road dust (Fu et al., 2012; Medeiros et al., 2006). Thus, high

levels of trehalose observed in the urban location indicates that soil resuspension and unpaved road dust

225   are important sources in the urban location.
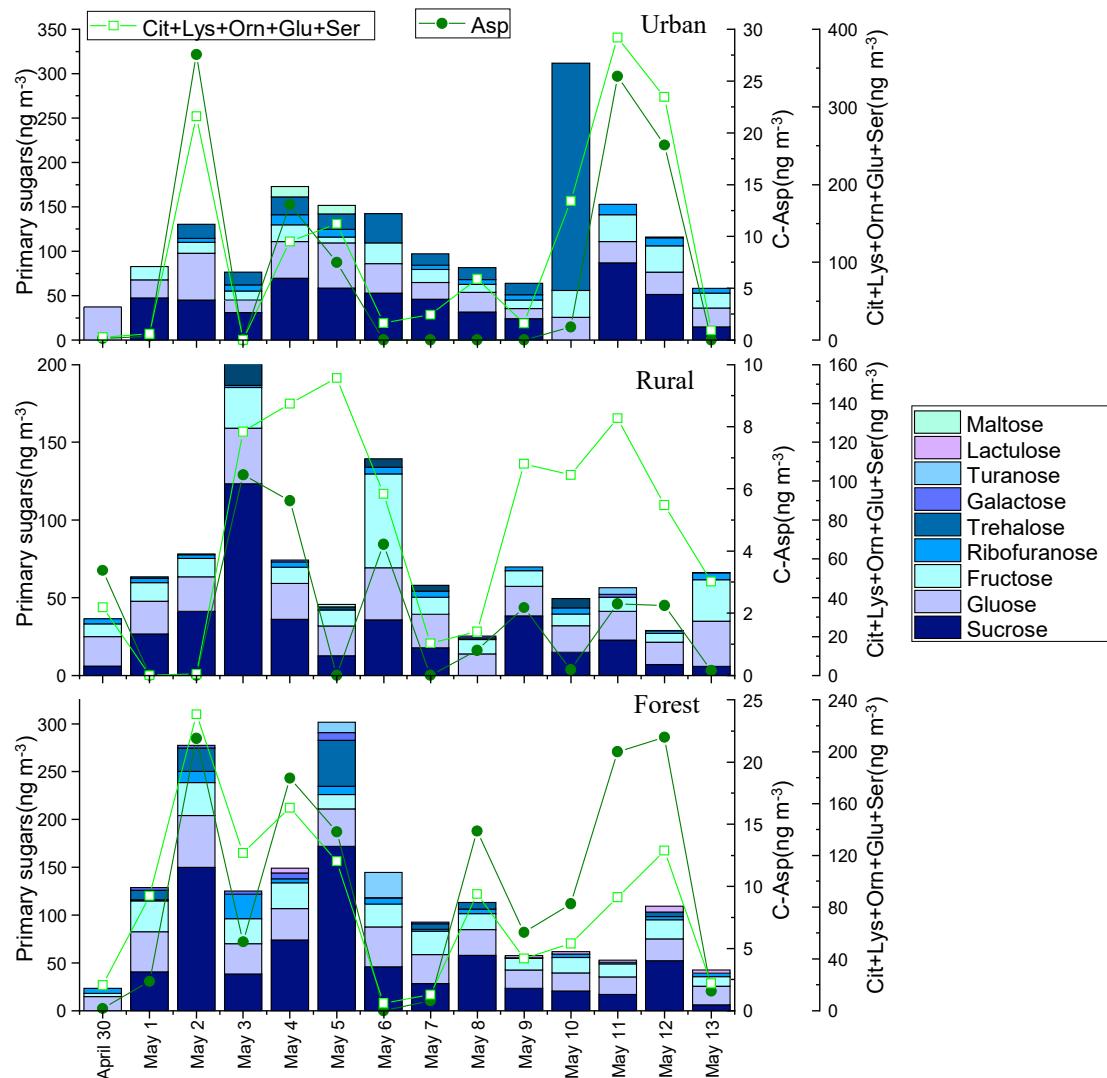
Figure 3. Concentrations of primary saccharides, combined Asp and the sum of Cit, Lys, Orn, Glu and Ser in urban, rural and forest. The concentrations of primary saccharides and CAAs for each sample were normalized for the total volume of air sampled.

230   We also determined four sugar alcohols, consisting of mannitol, arabitol, pinitol and inositol. The

concentrations of total sugar alcohols averaged 110 ± 50, 66 ± 28 and 93 ± 54 ng m^{-3} in the urban, rural

and forest locations, respectively (Figure 4). Average of the sum of mannitol and arabitol concentrations

measured in the three sampling locations (75 ng m^{-3}) were higher than those reported in megacity Beijing

(21 ng m^{-3}) (Kang et al., 2018) and rural site at Lincang, China (21.5 ng m^{-3}) (Wang et al., 2021). Since

235   these sugar polyols are reported to be abundant in fungi, trees, branches, and leaves (Wang et al., 2021;

Fu et al., 2008), the high abundance of mannitol and arabitol found in this study can be attributed to large

amount of biological origin.
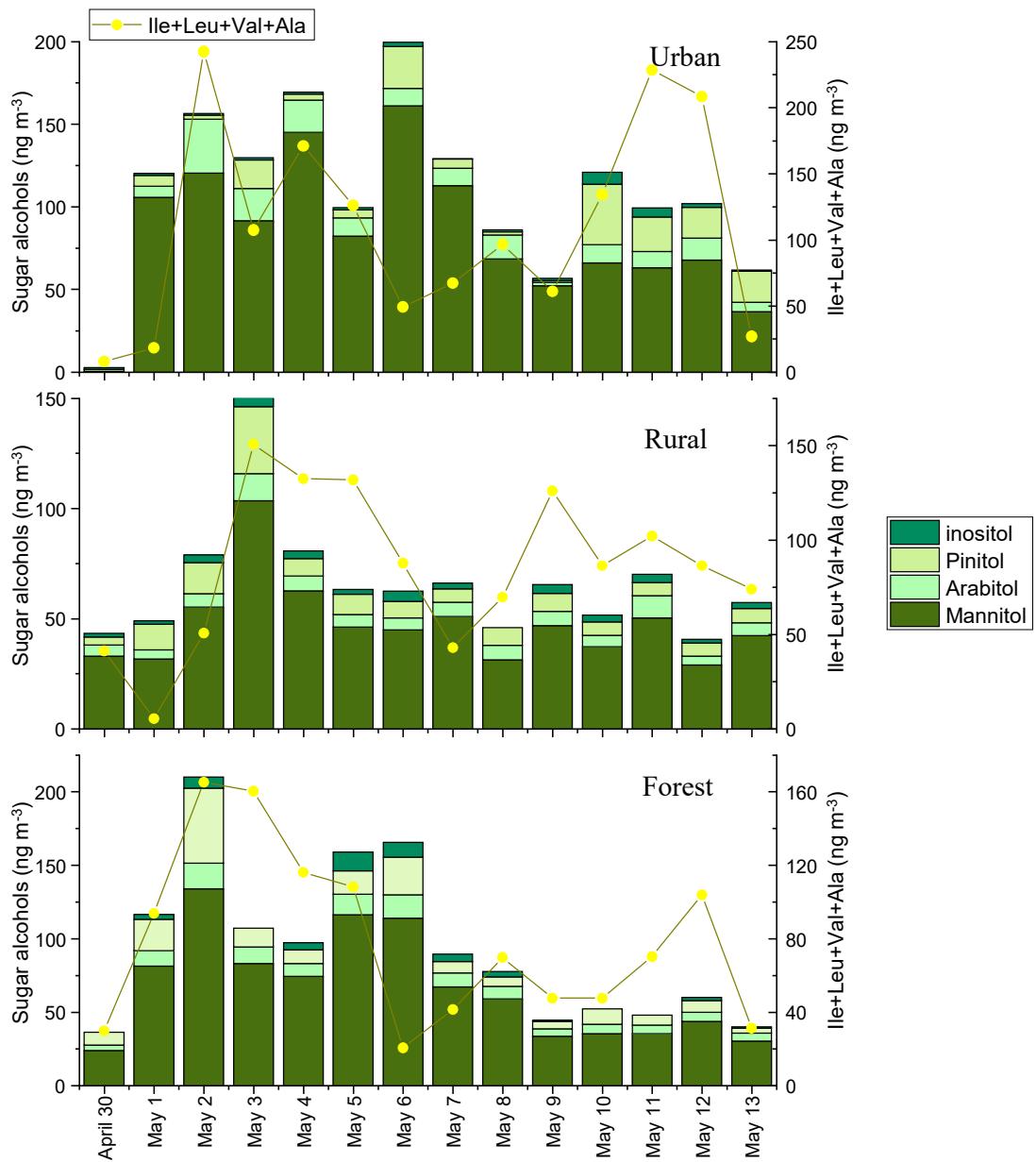
Figure 4. Concentrations of sugar alcohols the sum of Ile, Leu, Val and Ala in urban, rural and forest. The concentrations of sugar alcohols for each sample were normalized for the total volume of air sampled.

The average Σsugars concentration measured in the three sampling locations were comparable to several previous studies, including those measured at urban Beijing, China (average: 200 ± 100 ng m^{-3}) (Yan et al., 2019), the urban HongKong, China (292 ng m^{-3}) (Wan and Yu, 2007) and Mangshan National Forest Park, China (325 ng m^{-3}) (Verma et al., 2021), lower than those observed at rural Linchang, China (638 ng m^{-3}) (Wang et al., 2021), rural Wangdu, China (487 ± 567 ng m^{-3}) (Yan et al., 2019), but much higher compared to those measured in a natural forest area in India (156 ng m^{-3}) (Fu et al., 2010b) and Arctic aerosol (0.6 ± 0.5 ng m^{-3}) (Feltracco et al., 2020). In summary, the concentrations of aerosol sugars span a large range in variety regions by different research groups, which may be caused by many factors, including PM$_{2.5}$ concentration, the fraction of organic carbon in PM$_{2.5}$, vegetation and BB activities, and anthropogenic activities (Jia et al., 2010; Verma et al., 2021). To further elucidate the differences in sugar levels in aerosols collected from different regions, source apportionment is necessary.

3.1.2 The composition of sugars in PM2.5

The distribution pattern of sugar compounds in PM2.5 was generally similar among the urban, rural and forest locations (Figure 5). The average contribution of anhydrosugars to total measured sugars in the urban, rural and forest locations was 25.8%, 25.9% and 25.0%, respectively (Figure 5). Levoglucosan was the most dominant species among anhydrosugars in all sampling locations (Figure S4), accounting for 24.4%, 22.0% and 21.7% of the total sugars pool in PM2.5 in the urban, rural and forest locations, respectively, indicating biomass burning is common in all locations. However, the average contribution of levoglucosan to the total measured sugars in all locations were lower than those of the aerosol samples sampled from other regions in China, such as Mountain Tai (46.1 ± 20.4% of total sugars in daytime versus 39.0 ± 18.0% in nighttime) and fourteen cities in China (around 90%) (Fu et al., 2008; Wang et al., 2006). This suggests that besides BB sources, other sources are also important sources for sugar compounds in PM2.5 in this study.
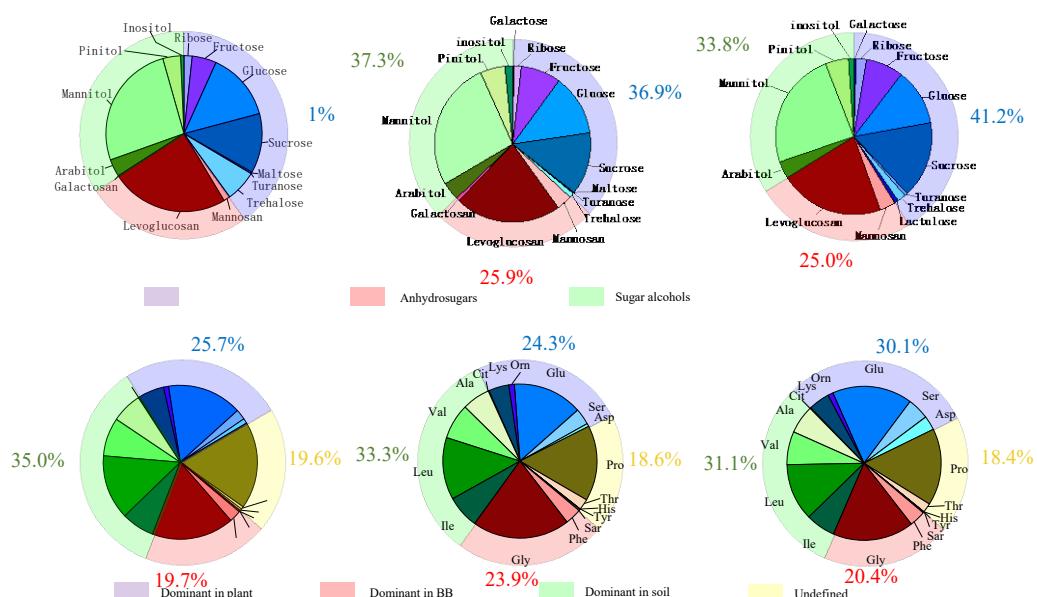
Figure 5. Illustrations of the average contributions of individual species to total sugars and those of each CAA to total combined amino acids in urban, rural and forest sites.

The contribution of primary sugars to total measured sugars measured at urban (40.1%) and forest locations (41.2%), were slightly higher than that in the rural location (36.9%) (Figure 5). Fructose, glucose and sucrose were the dominant primary sugar species in the three sampling locations (Figure S5). It is noted that PM2.5 sampled from the urban location had a larger contribution from trehalose (an average 6.5% of the total sugars) compared to those sampled from the rural (0.2%) and forest (0.8%) locations (Figure 5). Since trehalose is proved to be the most abundant saccharide in soils, the higher fraction of trehalose in total sugars pool in the urban location than those in the rural and forest locations may indicate PM2.5 collected in the urban location may be more impacted by resuspension of surface soil or road dust outflow than the rural and forest locations (Simoneit et al., 2004; Medeiros et al., 2006; Rogge et al., 2007; Fu et al., 2012). This is further supported by the significantly higher average concentration of nss-Ca^{2+} observed in urban site (1834.3 ng m^{-3}) than that of rural (1027.8 ng m^{-3}) and forest (1130.7 ng m^{-3}).

nss-Ca^{2+} is reported to be mainly derived from resuspended dust/soil in the ambient air (Barbaro et al., 2019; Rathnayake et al., 2016).

The average contribution of sugar alcohols to the total sugars pool in $PM_{2.5}$ was also close in the urban (34.1%), rural (37.2%) and forest (33.8%) locations (Figure 5). Mannitol was the major sugar alcohol specie in $PM_{2.5}$ (Figure S6), with an average contribution of 26.7%, 26.1% and 24.4% to the total sugars pool in the urban, rural and forest locations, respectively. Primary sugars and sugar alcohols are reported to be generally abundant in several animals, vegetative debris, microorganisms, fungal spores and pollen (Jaenicke, 2005; Jia et al., 2010). The large proportion of primary sugars and sugar alcohols observed in $PM_{2.5}$ collected from the three locations, further implying ambient sugars in this study were greatly influenced by primary biological sources.

3.2 AAs in the urban, rural and forest locations

3.2.1 Concentration of specific CAAs

As exhibited in Figure 1, the temporal variation pattern of ambient total free amino acids (TFAA) was different from that of Σsugar concentrations in all locations. Therefore, the concentration and composition of FAAs will not be compared with sugar compounds in the following sections. In contrast, the temporal variation pattern of Σsugar concentrations was consistent with that of TCAA measured in the three locations during the sampling period (Figure 1). Furthermore, the temporal variations in the concentrations of source-specific sugars (including anhydrosugars, primary sugars and sugar alcohols) were compared with those of some specific CAA species in $PM_{2.5}$. CAA species were initially grouped by their correlation with source-specific sugars at three sampling sites (Figure 6) and their abundance in local primary biological sources (Figure S7).
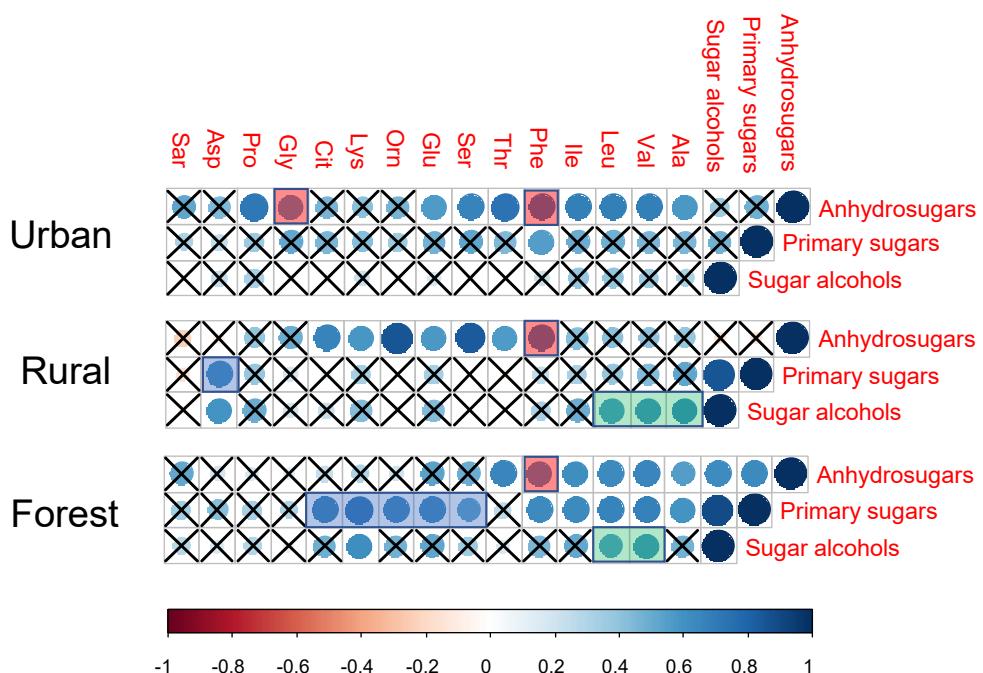
Figure 6. Pearson correlations between three target sugars (anhydrosugars, primary sugars and sugar alcohos) and individual combined amino acids. The cross indicates a *p*-value higher than 0.05. The ball indicates a p-value less than 0.05. The larger a ball is, the more significant the correlation is.

In our previous study, the composition profiles of CAAs in the biomass burning (honeycomb briquette, pine, and straw burning), local soil, and plant sources (pine and straw) were investigated. Gly was found to be the predominant CAA in the aerosol from biomass burning sources, hydrophobic species (Ala, Val, Leu, and Ile) were the major CAA species in soil sources, and hydrophilic species (Glu, Lys, and Asp) were the abundant CAA species in plant sources (Zhu et al., 2020b). Based on these, temporal variation pattern between source specific sugar marker and CAA species abundant in this specific primary biological source were compared. It is interesting to note that there were similar time variation patterns in the concentrations of anhydrosugars (BB tracer), combined Gly and Phe (Figure 2). The temporal variation pattern of primary sugars (primary bioaerosol tracer) concentrations was similar to those of the Asp and the sum of combined Cit, Lys, Orn, Glu and Ser (Figure 3). Additionally, the time variation pattern of sugar alcohols (soil and associated biota tracer) concentrations was consistent with that of the sum of Ala, Val, Leu and Ile concentrations (Figure 4)

Finally, according to the positive correlation between source-specific sugar markers and specific CAA species, and CAAs species predominant in each local primary biological source, as well as the similar temporal variation patterns between source-specific sugars and specific CAA species (Figure S8), ambient CAAs were finally divided into 4 groups: the first group includes Gly and Phe, which may be closely related to the BB source; the second group includes Ala, Val, Leu, and Ile, which are greatly impacted by soil source; the third group including Asp, Cit, Lys, Orn, Glu, Ser, which is close with plant sources, and the other CAAs are classified into the fourth group (Figure 5).

The sum of Phe and Gly concentrations (group 1) in $PM_{2.5}$ averaged 58 ± 46 ng m^{-3}, 55 ± 33 ng m^{-3} and 57 ± 38 ng m^{-3} in the urban, rural and forest locations, respectively (Figure 2). Average concentration of the sum of Ala, Val, Leu, and Ile (group 2) in urban, rural and forested mosses were 110 ± 75 ng m^{-3}, 85 ± 40 ng m^{-3} and 79 ± 45 ng m^{-3}, respectively, and average concentration of the sum of Asp, Cit, Lys, Orn, Glu, Ser (group 3) were 123 ± 135 ng m^{-3}, 78 ± 53 ng m^{-3} and 93 ± 68 ng m^{-3}, respectively (Figure 3 and Figure 4). By comparing the concentration of CAAs in different groups, it can be found that the average of the sum of CAAs concentrations in group 2 and group 3 is higher than that in group 1. This suggested that similar to sugar compounds, CAAs in $PM_{2.5}$ collected from the three sampling locations were greatly impacted by plant and soil sources in addition to biomass burning source.

3.2.2 Composition profiles of specific CAAs at the three sampling sites

To better understand the influence of primary sources on the composition profiles of sugar compounds and CAAs, the percentage of CAA species abundant in specific primary sources was compared with that of the corresponding sugar molecular markers. As discussed above, combined Gly and Phe are the predominant CAA species in the aerosols from BB sources and they exhibited similar time variation patterns with molecular markers for BB sources (anhydrosugars). Therefore, their percentage were compared with the contribution of BB molecular marker to the total sugar compounds in three sampling sites. These two CAAs together accounted for an average of 19.7%, 23.9% and 20.4%, respectively, of the TCAAs pool in $PM_{2.5}$ collected from the urban, rural and forest areas. It is interesting to note that the percentage of Gly and Phe in total CAAs pool was generally similar to the percentage of the anhydrosugars in total sugar compounds in $PM_{2.5}$. Ala, Val, Leu, and Ile (abundant in local soil source)

together contributed an average of 35.0%, 33.3% and 31.1%, respectively, to the TCAAs pool in the urban, rural and forest locations, which was also similar to the contribution of sugar alcohols to the total sugar compounds in the three locations. Furthermore, the average contribution of the combined CAA species in groups 3 and 4 to the TCAA pool was similar to the percentage of primary sugars to total sugar compounds in the three sampling locations (Figure 5). It is likely that composition profiles of sugar compounds and CAAs in $PM_{2.5}$ are mainly controlled by their primary sources.

3.3 Temporal variation of ambient sugars tracers and AAs

The temporal variations of sugar and AA compounds in the PM2.5 samples collected during the sampling periods are quite different at the three sites (Figure 1). In the urban area, the total sugar concentrations showed two major peaks, one during 4-6 May and the other during 10-12 May. Anhydrosugars, the marker of BB source, showed similar trend with the highest concentration occurred during 4-6 May and 10-12 May, suggesting urban sites may be more influenced by BB sources. In the rural area, the temporal variations of the total sugar concentrations showed different patterns, with two major peaks on 3 May and 6 May. On these two days, sugar alcohols and primary sugars exhibited the highest concentration, indicating a significant contribution from primary biogenic sources in the rural location. In the forest area, total sugars showed its concentration maximum on 2 May and 5 May. The concentrations of sugar alcohols and primary sugars in the forest area showed similar trends to total sugars, with the highest concentration occurring on 2 May and 5 May, implying that this area may be greatly influenced by primary biological sources.

Figure S8 presents the overall temporal variations of three classes of sugar compounds (anhydrosugars, primary sugars and sugar alcohols) in $PM_{2.5}$ collected at the three sampling sites. The temporal patterns of biomass burning tracers (anhydrosugars), primary bioaerosols tracers (primary saccharides) and surface soil and associated biotas tracer (sugar alcohols) are similar in the forest area. In the rural area, the temporal variation of primary saccharides is similar to that of sugar alcohols, but it is not consistent with that of anhydrosugars. In contracts, anhydrosugars, primary sugars and sugar alcohols showed different trends in the urban area. This further supported that the difference in the temporal variations of sugar compounds in $PM_{2.5}$ may point to the contribution of sources along the sampling period. The sugar compounds in $PM_{2.5}$ collected in the urban area might be more influenced by combustion sources whereas ambient sugars in the forest area are more influenced by primary biogenic sources.

The covariation of TCAA concentration with total sugar concentration was found in the three sampling areas. On the contrary, the temporal variations of TFAA concentrations showed different patterns from total sugar concentrations. These suggest that CAA may have similar sources and atmospheric processes to sugar compounds in $PM_{2.5}$, whereas FAA may have different sources and atmospheric processes from them, which will be discussed further in the following section.

3.4 Comparative behaviour of sugars and AAs in aerosols

3.4.1 Implications of formation pathways and atmospheric processes of ambient sugars and AAs

Pearson's correlation coefficients were performed based on sugar and AAs concentrations in $PM_{2.5}$ samples aggregated by sampling sites, and the results are shown in the Figure 7. It is interesting to note

that no correlation was observed between total free amino acids (TFAA) and the other individual measured sugar and total sugars in the three sampling locations (Figure 7a, yellow shadow). This could be explained by the formation pathways and atmospheric processes of aerosol FAAs and sugars. The fact that secondary processes is an important formation pathway of FAAs in the atmosphere has been corroborated by many studies (Wang et al., 2019; Xu et al., 2020b). Both laboratory simulations and field measurements demonstrated that the oxidation of proteins and peptides with hydroxyl radicals ($\cdot$OH) and O_3 can release of FAAs (Liu et al., 2017; Mcgregor and Anastasio, 2001; Song et al., 2017). On the other hand, sugar compounds are suggested as atmospheric primary organic aerosol, which directly emitted from sources such as plant material, soil dust, microorganisms and BB, and then aerosolized (Fröhlich-Nowoisky et al., 2016; Joung et al., 2017). Moreover, compared to FAAs, sugars are not derived from secondary photochemical processes in the atmosphere.
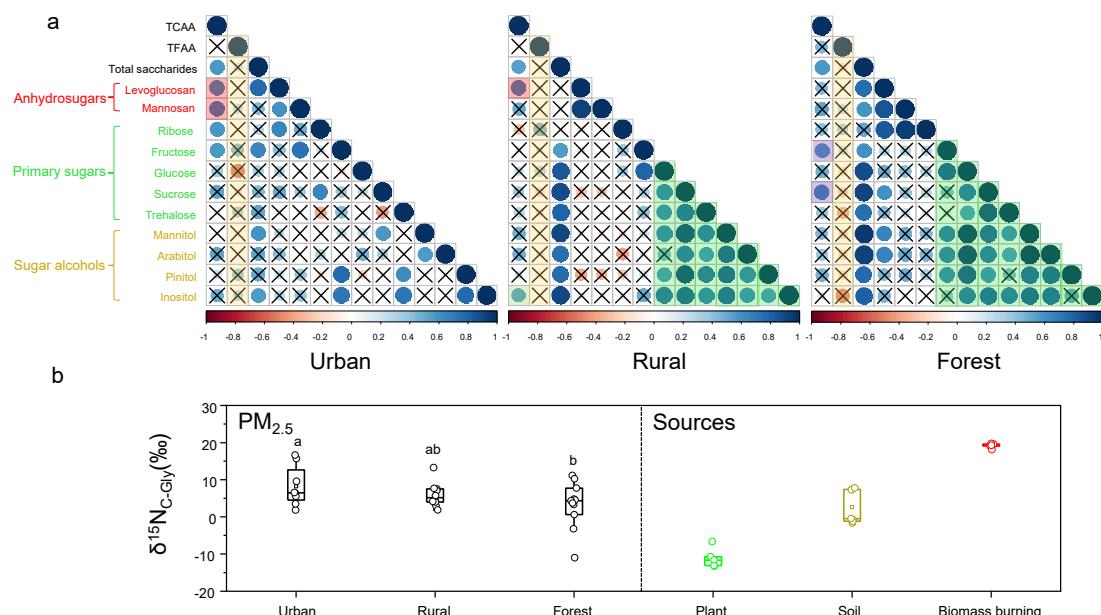
Figure 7. (a) Pearson correlations between TFAA and sugar species, and between TCAA and sugar species in $PM_{2.5}$ collected in urban, rural and forest sites. The cross indicates a *p*-value higher than 0.05. The ball indicates a *p*-value less than 0.05. The larger a ball is, the more significant the correlation is; (b) The range of $\delta^{15}N$ for combined Gly in $PM_{2.5}$ and major emission sources. The data of combined Gly was cited from Zhu et al. (2020). Open circles represent the isotopic composition data of combined Gly.

To further investigate the correlation between ambient Σsugar concentrations and FAAs, Pearson's correlation coefficients were calculated based on total sugar concentrations and individual FAA concentrations in $PM_{2.5}$ aggregated by sampling locations, which are exhibited in Figure S9. Generally, no correlation was recorded at the three sampling sits (Figure S9). Only Gln, Cit, Phe and Thr were weakly correlated with the concentration of total sugar concentrations in the forest location, and Glu exhibited a weak correlation with total sugar compounds in the rural location. These individual FAAs are known to be relatively high reactive with short half-lives in the atmosphere (Mcgregor and Anastasio, 2001). It is likely that these individual reactive FAAs are directly emitted from biological sources rather than secondary processes. Accordingly, we infer that there are differences in the formation pathways and atmospheric processes of sugars and FAAs in fine particles.

On the contrary, significant correlations at 95% confidence level were found between total combined amino acids (TCAA) and total sugars (r = 0.56~0.57, p < 0.05, n=14) at each sampling site in this study

(Figure 7a). Although the relationship between CAAs and sugars in the atmosphere has not been directly analysed, their sources have been explored in previous studies. For instance, Feltracco et al. (2019) investigated the possible sources of AAs in Artic aerosol and proved FAAs and CAAs in aerosols might have different sources. They suggest that similar to aerosol sugars, ambient CAA derive from PBAPs, including viruses, algae, fungi, bacteria, protozoa, spores and pollen, fragments of plants and insects, and epithelial cells of animals and humans. Previous studies also demonstrated that unlike FAAs greatly influenced by atmospheric processes, composition profiles of aerosol CAAs are highly depended on the origin of CAAs and it has been used to trace emission sources including primary biological sources and BB sources (Abe et al., 2016; Zhu et al., 2020b). Thus, significant correlations between TCAA and total sugars in aerosols found in this study further corroborate that they may have common sources and are less impacted by atmospheric photochemical processes.

3.4.2 Evaluate the contribution of primary emission sources in different locations.

Sugars in the atmosphere have been demonstrated to be derived from numerous sources. Three target sugar categories (anhydrosugars, primary sugars and sugar alcohols) can be used as tracers for specific source information (Simoneit, 2002; Simoneit et al., 2004). Anhydrosugars (including levoglucosan, mannosan and galactosan), generated by the thermal decomposition of cellulose and hemicellulose, are regarded as the specific markers for BB in the atmosphere (Simoneit et al., 2004; Zangrando et al., 2016). In this study, TCAA was found to be positively correlated with anhydrosugars at the urban and rural locations whereas no correlation between TCAA and anhydrosugars was observed in the forest location (Figure 7a, red shadow), indicating the urban and rural areas were more influenced by BB sources compared to the forest area. These conclusions were also corroborated by the $\delta^{15}N$ signatures of combined Gly ($\delta^{15}N_{C-Gly}$) in $PM_{2.5}$ measured at these three sites. Our previous study found that more positive $\delta^{15}N_{C-Gly}$ values were observed in $PM_{2.5}$ at the urban (average = +8.2 ± 5.4‰) and the rural (average = +5.9 ± 3.2‰) site than the forest site (average = +3.3 ± 6.2‰) (Zhu et al., 2020b) (Figure 7b). According to the $\delta^{15}N$ inventories of emission sources of atmospheric combined Gly, the urban location was highly influenced by combustion sources, followed by the rural location, while the forest location was more affected by natural sources.

Primary sugars are reported to be derived from microorganisms, plants, animals, lichens, and bacteria and have been considered to be good tracers for PBAPs (Fu et al., 2013; Zangrando et al., 2016). Particularly, sucrose and fructose are predominantly produced from plant fragments (e.g., bark, pollen grain, flowers buds, fruit and leaves) and soil dust (Fu et al., 2012; Xu et al., 2020a). In this study, TCAA only exhibited positive correlation with primary sugars (fructose and sucrose) in the forest location (Figure 7a, purple shadow), pointing to aerosols in the forest area were mainly influenced by intensive biological activities and vegetation growth during sampling period. Previous studies mentioned that increased biological activities and active vegetation such as developing flower buds in the spring can emit abundant primary sugars into the air (Jia et al., 2010). Hence, the sampling period (spring) of this study and intensive biological sources in the forest location may explain the positive correlation between TCAA and primary sugars in the forest area.

Sugar alcohols primarily come from fungal spores, bacteria, plants, algae and detritus of mature leaves (Bauer et al., 2008; Feltracco et al., 2020). Xu et al. (2020a) suggest that in spring mannitol and arabitol may be closely related to the activities of the terrestrial biosphere. When comparing the correlation results of sugars classes between sites, it is important to note that the correlation of primary sugars with sugar alcohols varied degrees at different sampling sites (Figure 7a, green shadow). More species of primary sugars and sugar alcohols in $PM_{2.5}$ positively correlated at the rural and forest sites ($p < 0.05$) compared to the urban site. Moreover, the stronger correlation between primary sugars with sugar alcohols were observed for $PM_{2.5}$ samples collected from the rural and forest sites than that at the urban site. This may imply that the impact from local biogenic sources at the rural and forest sites was greater than that of the urban site, which is also supported by the temporal patterns of anhydrosugars, primary sugars and alcohol-sugar at the three sites (Figure S8). We can thus conclude that the influence of primary biogenic and BB sources on aerosols in different locations can be evaluated by the correlations between TCAA and source specific sugar tracers (anhydrosugars, primary sugars and sugar alcohols).

3.4.3 Identify burning tissues, local vegetation classes and soil-related biological sources

A Pearson's correlation test was also calculated for the dataset containing ambient concentrations of sugar compounds and individual CAAs in $PM_{2.5}$ collected at three sites, which are exhibited in Figure 6. In the urban location, the concentrations of anhydrosugars showed a positive correlation with that of combined Gly, but no correlation was found between these two compounds in the other two locations. As we discussed above, combined Gly is major CAA specie in the honeycomb briquette, pine, and straw burning aerosols. Therefore, anhydrosugars and combined Gly both are closely related to biomass burning sources. A possible explanation for the positive correlation found between these two BB tracers is that the urban region is mainly affected by local combustion sources, while rural and forest regions may be more influenced by BB source from long-transport.

Significant positive correlations of anhydrosugars with Phe were observed simultaneously in the urban (r=0.68, $p<0.05$), rural (r=0.65, $p<0.05$) and forest (r=0.57, $p<0.05$) locations, serving as statistical support for a common source for the combined Phe and anhydrosugars in aerosols (Figure 6). Previous studies indicate that the amino acid Phe is not only used as vital constituent of proteins but also a critical metabolic node that plays an essential role in lignin biosynthesis in vascular plants (Bonawitz and Chapple, 2010; Wallace and Fry, 1994). It is estimated that nearly 30–45% of photosynthetically fixed carbon is channelled through Phe for the biosynthesis of lignin during wood formation (Pascual et al., 2016). In addition, the vascular plant cell wall is a heterogeneous natural nanocomposite of cellulose, hemicelluloses and lignin (Agarwal, 2006; Jin et al., 2017). The lignin, the second most abundant polymer in plants, deposited in the secondary wall in which cross-linked to the structural polysugars, such as hemicelluloses and cellulose found there (Bonawitz and Chapple, 2010; Wallace and Fry, 1994). During the combustion processes of plant cell wall, the thermal breakdown of lignin takes place along with cellulose/hemicellulose. Therefore, the combustion of plant cell wall may explain the observed concentration coupling between combined Phe and anhydrosugars.

In this study, primary sugars were found to be highly correlated with combined Asp in $PM_{2.5}$ samples collected in the rural location, but primary sugars were correlated with combine Glu, Lys, Orn, Cit and

Ser in the forest location (Figure 6, blue shadow), which is consistent with the temporal variation pattern of primary sugars and these CAA species (Figure 3). This may be attributed to the difference vegetation types in these two regions. The rural site is a typical agricultural area where grasses and crops are the main vegetation classes. Asp is reported to be the most abundant free and combined amino acids in Gramineae (Barnett and Naylor, 1966) (Burkholder et al., 1959; Wilson and Tilley, 1965). On the other hands, conifers dominate ecosystems at the forest site. In particular, hydrophilic species (Glu, Lys, Orn, and Ser) have been shown to be the predominant CAA species in pine needles (Zhu et al., 2020b). Hence, the correlation between primary sugars (primary bioaerosols tracers) and specific CAAs may be used to track the predominant local vegetation types.

Consistent correlations were observed between sugar alcohols and combined Ala, Val and Leu at the rural and forest sites (Figure 6, green shadow), pointing to the significant contribution of biological sources associated with surface soil to aerosols at these two sites (Simoneit et al., 2004; Fu et al., 2013; Moura et al., 2013; Zhu et al., 2020b). As above mentioned, sugar alcohols indicate soil and associated biota contribute to ambient aerosols through resuspension by wind erosion or agricultural activities (Fu et al., 2013; Fu et al., 2010a). Similarly, Friedel and Scheller (2002) showed that Ala, Val and Leu were the main hydrolysable amino acid species in soil organic matter and soil microbial biomass. Grass land environments are characterized by higher content of Ala, Asp and Leu (Moura et al., 2013). Our previous studies also demonstrated that hydrophobic Ala, Val, Leu, and Ile were the major CAA species in local crop soil (Zhu et al., 2020b). On the contrary, primary sugars and sugar alcohols exerted no correlation with individual CAA at the urban site except for Phe, further supporting that aerosol at urban location are less influenced by biogenic source than combustion sources. Thus, it can be concluded that the correlations between individual CAAs and target category sugars can provide insight into various source characteristics such as plant tissues used for combustion, local vegetation classes (based on the correlation of primary sugars with specific CAAs dominant in local plants) and soil-related biological sources vs combustion sources (based on the correlation of sugar alcohols with specific CAAs dominant in soil source).

3.5 Identify classes of combustion sources

Levoglucosan has been tested and confirmed as an excellent tracer for biomass burning, which produces from the pyrolytic decomposition of cellulose and hemicellulose, or thermal stripping of sugar polyols during the combustion processes (Jia et al., 2010; Kang et al., 2018; Simoneit, 2002), but it could not distinguish different classes biomass combustion sources. Detailed fingerprint tracers should be measured to get comprehensive information to identify different biomass burnings. To accurately qualify the BB source in different locations, Nss-K^+ have been also used as a typical tracer of BB in previous studies (Kunwar & Kawamura, 2014). However, previous studies also indicated that soil dust could be another source of water-soluble Nss-K^+ in aerosols (Urban et al., 2014; Urban et al., 2012; Wang et al., 2021). Nss-K^+ concentrations averaged 633.1 ng m^{-3} in the urban area which was higher than that observed in the rural (549.6 ng m^{-3}) and forest (590.2 ng m^{-3}) areas. Using levoglucosan as a representative BB tracer, the correlations between anhydrosugars and Nss-K^+ were further analyzed at three sampling sites to investigate the contribution of BB sources at each site (Table S3). Nss-K^+ showed

a statistically significant linear correlation with levoglucosan in the urban location (r=0.6, p < 0.05, n=14), whereas no correlation between levoglucosan and $Nss-K^+$ was observed in the rural and forest locations (Table S3). These may suggest the rural and forest areas were more impacted by the other sources of $Nss-K^+$, such as soil resuspension and fertilizers, while the urban area was more influenced by BB sources. This conclusion can also be confirmed by the fact that the stronger positive correlations between TCAA and anhydrosugars and between combined Gly and anhydrosugars were observed in the urban area compared to rural and forest areas. Most positive $\delta^{15}N_{C-Gly}$ values observed in the urban location. In addition, different temporal variation pattern of anhydrosugars from that of primary sugars and sugar alcohols was only found in the urban area.

The average ratio of levoglucosan to potassium (lev/K^+) was 0.07 given by the slope of the linear regression equation, which is close to the value of aerosols collected from a rural region of São Paulo State, Brazil (average = 0.08), where is strongly influenced by burning sugar cane (Urban et al., 2012). Furthermore, these ratios observed in this study were also comparable with the ratio found in Beijing, China during the typical summer period (0.05), which is greatly impacted by the combustion of crop residuals (Cheng et al., 2013). However, this value was lower than those for aerosol samples collected in the Amazon (average = 1.01) and those in Austria (ranged 0.22 to 1.7), where are dominated by wood burning. 2013). The ratio of lev/K^+ was used to characterize the type of biomass burned (Wang et al., 2021) (Table S3). Cheng et al. (2013) found that that levoglucosan to K^+ ratios in emissions from the combustion of crop residuals were below 1.0, which is much lower than the ratio for hardwood burning (average = 25.69±9.11) and softwood burning (average = 105.58±102.60). Therefore, the contribution of wood burning was negligible for the biomass burning aerosol in the urban area. However, using lev/K^+ ratio to identify the type of combustion source is severely limited in the presence of potassium derived from non-biomass burning sources (Roberta Cerasi Urban et al., 2012). It is therefore that biomass combustion sources in the rural and forest locations cannot be distinguished by the lev/K^+ ratio.

L/M ratios have been employed to identify specific burning substrates in numerous studies (Fabbri et al., 2009; Fan et al., 2020; Zhu et al., 2015). L/M ratios for smoke particles from the burning of crop residues (~40 to 55.7) were high, while L/M ratios for the burning of hardwood (15-33) and softwood (3 to 10) were low (Engling et al., 2014; Engling et al., 2009; Sheesley et al., 2003; Wang et al., 2021). Besides that, recently studies demonstrate that levoglucosan is also present in lignite smoke particle matter (Rybicki et al., 2020a). Moreover, pyrolysis experiments confirm that lignite produce levoglucosan concentrations fully comparable to that of hardwoods and softwoods (Fabbri et al., 2009; Sheesley et al., 2003). Burning of lignite has been proved to be an additional input of levoglucosan to the atmosphere in regions where brown coal is utilized. Rybicki et al. (2020b) reported that the L/M ratios in lignites smoke (ranged from 31 to 189) were generally higher than those found for other fuel types because cellulose is more resistant to diagenetic (bio)degradation than hemicelluloses.

To better understand the different classes of combustion sources, the L/M ratio in the three areas were investigated. In this study, the ratios of L/M in the rural and forest areas ranged from 5.1 to 8.4 (average: 6.9) and from 4.3 to 9.8 (average: 7.2), respectively. According to Figure 8, the L/M ratios for rural and forest areas fall within the range for softwood burning. Thus, biomass burning aerosol in the rural and forest areas were mainly derived from softwood burning. However, the extremely high L/M ratios found

in the urban area (ranged: 7.9 to 359.1; average: 59.9). These ratios were even higher than the values in the smoke particles released by the burning of crop residuals (>57) reported in the previous literature but fell within the range of lignite combustion, implying the urban area may be influenced by lignite combustion besides burning of crop residuals. Further support for this result was provided by the fact that a great quantity of lignite was used at the urban site (averaged 3803646 tons per year, the data are from the statistical yearbook of Nanchang city, http://tjj.nc.gov.cn//zbft/front/tjjnjnew/2020/mobile/index.html). Besides that, the average concentration of Cl^- in the urban location (107.3±68.4 ng m^{-3}) was higher than those in the rural (99.9±71.3 ng m^{-3}) and forest (62.5±29.9 ng m^{-3}). Cl^- has been reported to be a major chemical composition of coal combustion (Guo et al., 2020).
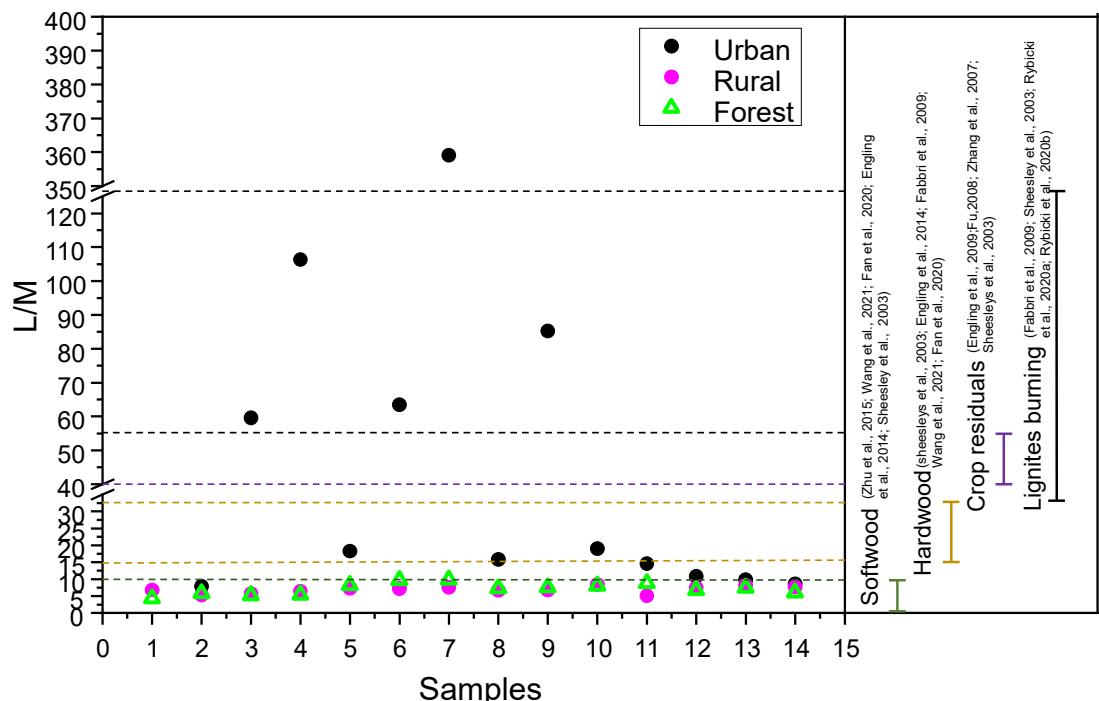
Figure 8. Ratios of levoglucosan to mannosan in PM$_{2.5}$ and source test emissions from softwood, hardwood, crop residuals and lignites burning.

4 Conclusions

Determining the presence and signatures of source specific sugars and AAs in aerosol samples can be of great value to estimate the contribution of primary biogenic and BB sources to aerosols at different sampling locations. Anhydrosugar, a confirming BB tracer, highly correlated with combined Phe suggests the burning of plant cell wall. The correlations between primary sugars (primary bioaerosol tracer) and specific CAAs dominant in local plant classes can help to identify potential local plant classes. Combined sugar alcohols (soil and associated biota tracer) with specific CAAs species dominant in soil source can help to distinguish soil-related biological sources contribution. Previous work on aerosols focused on using only sugars or AAs biomarker alone to trace the aerosol sources. To our knowledge, this is the first report combining source specific sugar tracers with amino acids species dominant in primary emission sources to trace primary sources signatures at different locations. Moreover, this paper

clearly demonstrates combining these two molecular biomarkers in atmospheric particles may provide
590   more source characteristics than using a single biomarker alone. Future work should be aimed at
comprehensively characterizing physical, chemical, and biological properties of sugars and amino acids
in aerosols, assessing possible interaction mechanisms between them, and extensively addressing the
distribution of these two compound classes in potential aerosol sources.

595   *Data availability.* The data for lignite consumption at the urban site were obtained from the statistical
yearbook of Nanchang city (http://tjj.nc.gov.cn/zbft/front/tjjnjnew/2020/mobile/index.html) (Nanchang
Statistics Bureau, 2020). Raw data sets (Zhu, 2021) used in this manuscript were available at
https://figshare.com/articles/dataset/Concentration_of_saccharides_in_PM2_5_xlsx/17158661.

600   *Supplement.* The supplement related to this article is available online.

Author contributions. This research was designed RGZ and HYX. Laboratory measurements were
performed by HXZ, LQC and YYG. The paper was prepared by RGZ, HYX and HWX.

605   *Competing interests.* The authors declare that they have no conflict of interest.

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