Saccharide composition in atmospheric fine particulate matter during spring at the remote sites of Southwest China and estimates of source contributions

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Abstract. Based on source-specific saccharide tracers, the characteristics of biomass burning (BB) and biogenic emissions to saccharides were investigated in three rural sites at Lincang, where covered with 65% of forest in the southwest border of China. The total saccharides accounted for 8.4 ± 2.7% of organic compound (OC), and 1.6 ± 0.6% of $PM_{2.5.}$ The measured anhydrosugars accounted for 48.5% of total saccharides, among which levoglucosan was the most dominant species. The high level of levoglucosan was both attributed to the local BB activities and biomass combustion smoke transported from the neighboring regions of Southeast Asia (Myanmar) and the northern Indian Peninsula. The measured mono (di) saccharides and sugar alcohols accounted for 24.9 ± 8.3% and 26.6 ± 9.9% of the total saccharides, respectively, were both proved to be mostly emitted by direct biogenic volatilization from plant materials/surface soils, rather than byproducts of polysaccharides breakdown during BB processes. Five sources of saccharides were resolved by non-negative matrix factorization (NMF) analysis, including BB, soil microbiota, plant senescence, airborne pollen, and plant detritus with contributions of 34.0%, 16.0%, 21.0%, 23.7%, and 5.3%, respectively. The results provide information on the magnitude of levoglucosan and contributions of BB, as well as the characteristic of biogenic saccharides, at the remote sites of Southwest China, which can be further applied to regional source apportionment models and global climate models.

1 Introduction

Biomass burning (BB) and biogenic aerosols are thought to play important roles on air quality, human health, and climate through direct or indirect effects (Jacobson et al., 2000; Christner et al., 2008; Pöschl et al., 2010; Després et al., 2012; Chen et al., 2017, Tang et al., 2019). Atmospheric saccharide components have been extensively reported to originate from natural or anthropogenic biomass burning (BB), suspended soil or dust and primary biological aerosol particles (PBAPs), e.g., fungal and fern spores, pollens, algae, fungi, bacteria, and plant debris, and biogenic secondary organic aerosol (SOA) (e.g., Rogge et al., 1993; Graham et al., 2003; Jaenicke, 2005; Medeiros et al., 2006; Elbert et al., 2007; Fu et al., 2013). As one of the major class of water-soluble organic compounds, saccharides in atmospheric aerosols have been detected over urban areas, forests, mountains, and remote marine regions (Pashynska et al., 2002; Yttri et al., 2007; Fu et al., 2009; Burshtein et al., 2011; Jia and Fraser, 2011; Chen et al., 2013; Pietrogrande et al., 2014； Li et al., 2016a, b). It has been reported that saccharides account for 13–26% of the total organic compound mass identified in continental aerosols and up to 63% in oceanic aerosols (Simoneit et al., 2004).

Levoglucosan and related anhydrosugar isomers (mannosan and galactosan), produced from pyrolysis of cellulose and hemicellulose, are considered to be relatively stable in the atmosphere (Schkolnik et al., 2005; Puxbaum et al., 2007), and thus have been recognized as specific molecular markers for BB source emissions (Simoneit et al., 1999, 2000; Fraser and Lakshmanan, 2000; Sullivan et al., 2014; Du et al., 2015). However, some studies have challenged this knowledge and proved that levoglucosan alone was unsuitable to be a distinct marker for BB in various regions and periods. Because there were evidences that levoglucosan was also emitted from non-BB sources (Wu et al., 2021), such as coal burning (Rybicki et al., 2020; Yan et al., 2018), open waste burning (Kalogridis et al., 2018), incense burning (Tsai et al., 2010), and food cooking (Reyes-Villegas et al., 2018). It was reported that the levoglucosan emission contribution of BB sources ranged from 21.3% to 95.9% (Wu et al., 2021). The current studies in China have reported the value of 2.6–289.1 ng m^{-3} and 11.6–1803.1 ng m^{-3} respectively over Beijing and Wangdu in summer (Yan et al., 2019), 2.4–1064.1 ng m^{-3} over Shanghai all year round (Xiao et al., 2018), 15.6–472.9 ng m^{-3} over Guangzhou (Zhang et al., 2010), 21.1–91.5 ng m^{-3} over Hongkong (Sang et al., 2011), 60.2–481.9 ng m^{-3} over Xi 'an (Yang et al., 2012), 36.0–1820.9 ng m^{-3} over Chengdu (Yang et al., 2012) and 10.1–383.4 ng g^{-1} dry weight in cryoconites over the Tibetan Plateau (Li et al., 2019). In north China, the high concentration of levoglucosan was a serious problem due to the drastic enhancement of coal and BB for house heating in winter and autumn (Zhang et al., 2008; Zhu et al., 2016). The BB pollution might be exacerbated under unfavorable meteorological conditions, such as in the Chengdu plain (Chen and Xie, 2014). In general, BB with a notable contribution to OC was an important source of fine particulate matter in China (Zhang et al., 2008; Cheng et al., 2013; Chen et al., 2017). Controls on BB could be an effective method to reduce pollutant emissions. Recently study reported that total levoglucosan emission of China exhibited a clear decreasing trend from 2014 (145.7 Gg) to 2018 (80.9 Gg) (Wu et al., 2021), suggesting BB activities might reduce in China.

Saccharide compounds including a variety of primary saccharides (monosaccharides and disaccharides) and sugar alcohols (reduced sugars) have been measured to estimate the contribution of biogenic aerosols, including fungi, viruses, bacteria, pollen, as well as plant and animal debris (Simoneit et al., 2004; Jaenicke et al., 2007). For instance, arabitol and mannitol have been proposed as biomarkers for airborne fungal spores (Bauer et al., 2008; Zhang et al., 2010; Holden et al., 2011; Liang et al., 2013a, b), because both of them can function as storage or transport carbohydrates to regulate intracellular osmotic pressure (Bauer et al., 2008). Glucose and sucrose are thought to originate from natural biogenic detritus, including numerous microorganisms, plants, and animals (Simoneit et al., 2004; Tominaga et al., 2011). As the oxidation products of isoprene, methyltetrols (including 2-methylthreitol and 2-methylerythritol) have been suggested as tracers of isoprene-derived SOA (Claeys et al., 2004; Kleindienst et al., 2007; Ding et al., 2016). In the previous study, the contributions of fungal spores to OC were estimated to be 14.1 ± 10.5% and 7.3 ± 3.3% respectively at the rural and urban sites of Beijing (Liang et al., 2013b). Airborne pollen and fungal spores contributed 12%–22% to the total OC in ambient aerosols collected in Toronto (Womiloju et al., 2003). Jaenicke (2005) found that PBAPs could account for 20%–30% of the total atmospheric PM (>0.2 mm) from Lake Baikal (Russia) and Mainz (Germany). However, studies on quantifying the abovementioned biogenic aerosol contributions to ambient aerosol are inadequate.

Lincang, located on the southwest border of China, is a traditional agricultural area of Yunnan province, where planting a large area of tea, sugar cane, rubber, macadamia nut, etc. It is the largest production base of black tea and macadamia nut in China. Referring to the official website of Lincang Municipal People's Government, the forest coverage rate of Lincang reaches to 65%. It has a wide variety of plant species, and has 6 nature reserves covering an area of ~222,000 hectares, accounting for 8.56% of the total area. As a residential area for ethnic minorities, Lincang has unique culture, humanity and living habits. The proportion of houses that employ wood burning for cooking is very high in villages in proximity and a large area of Southeast Asia, and forest fires frequently occur in this area, especially in the dry seasons (March–April). These imply that there are abundant biogenic aerosols in Lincang, and BB pollution may be an essential potential source of air pollution. However, little information on the magnitude of biogenic and BB tracers in this area is available. The contributions of biogenic aerosol and BB, as well as BB types, are poorly understood.

In this study, the sampling was conducted from 8 March to 9 April, 2019 at three mountaintop sites of Lincang, where is an ideal site for investigating the BB emission characteristics. BB tracers, including anhydrosugars and K^+, as well as biogenic aerosol tracers (primary saccharides and sugar alcohols) were measured to gain the information on source and contributions of BB and biogenic emissions in $PM_{2.5}$ over the rural Lincang. This study would be useful and valuable for providing reliable information on sources and magnitudes of saccharides involving rural BB and biological emissions in China.

2 Experimental section

2.1 Aerosol sampling

$PM_{2.5}$ samples were simultaneously collected on three mountaintop sites in Lincang, respectively of Datian (24.11◦ N, 100.13◦ E, 1960 m asl), Dashu (24.12◦ N, 100.11◦ E, 1840 m asl) and Yakoutian (24.12◦ N, 100.09◦ E, 1220 m asl), which are located ~300 km west of Kunming (the capital of Yunnan province in China) and ~120 km east from the Burma border (shown in Figure S1). These sites are surrounded by massive mountains and scattering villages without obvious nearby traffic or major industry emissions. Each sampling was performed over a 23.5 h period every day, and was collected on quartz by high-volume air samplers (Thermo) equipped with a size selective inlet to sample $PM_{2.5}$ at a flow rate of 1.13 m^3 min^{-1}. Altogether, 91 samples were collected.

Quartz filters (Whatman, 8 × 10 in.) were prebaked at 550 °C for 4 h in a muffle furnace to remove organic material, and were then stored in pre-baked aluminum foils. The samples were stored at about −20 °C in a refrigerator until analysis. Field blanks were collected by mounting filters in the sampler without air flow to replicate the environmental exposure. The data reported were corrected by the blanks at the sampling sites.

2.2 Measurements

The concentrations of OC and elemental carbon (EC) were measured using a Multiwavelength Carbon Analyzer (DRI Model 2015; Aerosol Inc., USA). Typically, a 0.58 cm^2 punch of the filter was placed on a boat inside the thermal desorption chamber of the analyzer, and then stepwise heating was applied. Carbon fractions were obtained following the Interagency Monitoring of Protected Visual Environments (IMPROVE-A) thermal/optical reflectance (TOR) protocol (Chow et al., 2007). Replicate analyses were conducted once every ten samples. Blank sample was also analyzed and used to correct the sample results.

A punch (4.7 cm^2) of each quartz filter was ultrasonically extracted with 10.0 mL deionized water (resistivity = 18.2 MU) for 40 min. The aqueous extracts were filtrated through syringe filters (PTFE, 0.22 μm) to remove insoluble materials. Ion chromatography (Metrohm, Switzerland) coupling with Metrosep C6-150 and A6-150 columns was used to detect water-soluble ions (Cl^-, NO_3^-, PO_4^-, SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}) with a detection limit (DL) range of 0.001-0.002 μg m^{-3}.

Five saccharide alcohols (glycerol, erythritol, inositol, arabitol and mannitol) and five primary saccharides (fructose, glucose, mannose, sucrose and trehalose), together with three anhydrosugars (levoglucosan, mannosan and galactosan) were quantified by an improved high-performance anion-exchange chromatography coupled with a pulsed amperometric detector (Engling et al., 2006; Caseiro et al., 2007; Zhang et al., 2013). This method developed by Engling et al. (2006) was validated to be a powerful method for the detection of carbohydrates without derivatization techniques, and has been successfully applied for the atmospheric tracers (e.g., Zhang et al., 2010; Holden et al., 2011; Liang et al., 2013a, b; Li et al., 2016a, b; Kalogridis et al., 2018; Yan et al., 2018). The separation of the saccharides was performed on an ion chromatograph (Metrohm, Switzerland) equipped with a Metrosep Carb 4–250 analytical column and a guard column. The aqueous eluent of sodium hydroxide and sodium acetate was pumped by a dual pump module at a flow rate of 0.4 mL min^{-1}. The low concentration of 50 mM sodium hydroxide and 10 mM sodium acetate (eluent A) was applied to pump 1, while the high concentration of 250 mM sodium hydroxide and 50 mM sodium acetate (eluent B) was applied to pump 2. The gradient generator was set as: 0–10 min, 100% of eluent A; 10–20 min, 50% of eluent A and 50% of eluent B; 20–50 min, 100% of eluent B; 50–60 min, 100% of eluent A for equilibration. The extraction efficiency of this analytical method was determined to be better than 90% based on analysis of quartz filters spiked with known amounts of mannitol. The method DL of the referred carbohydrate compounds were 0.005–0.01 mg L^{-1}. All carbohydrate species were below detection limits in the field blanks.

2.3 Other data

The meteorological parameters, including temperature (T), relative humidity (RH), solar irradiation (W m^{-2}), and rainfall (mm) were obtained from the Physical Sciences Laboratory of NOAA (https://psl.noaa.gov). The temporal changes in meteorological variables over the observation sites during the sampling periods are shown in Figure S2.

In order to characterize the origin and transport pathway of the air masses to the sampling sites, 72 h back-trajectories of the aerosol were calculated using Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model developed by NOAA/ARL (Draxler and Hess, 1998) via NOAA ARL READY Website (http://ready.arl.noaa.gov/HYSPLIT.php) with an endpoint height of 1500 m. To investigate the influence of BB emissions, fire pixel counts were obtained from Moderate Resolution Imaging Spectroradiometer (MODIS) observations on NASA satellites (https://earthdata.nasa.gov/).

2.4 Statistical analysis

A Pearson's correlation test was performed using the statistical product and service solutions software for the dataset containing ambient concentrations of the measured saccharides, inorganic ions and solar irradiation. Non-negative matrix factorization (NMF) analysis was utilized to resolve potential emission source and estimate their contribution to atmospheric saccharides. NMF introduced by Lee and Seung (1999) was similar to positive matrix factorization (PMF). Both methods find two matrices (termed the contribution matrix of W and the source profile matrix of H) to reproduce the input data matrix (V) using the factorization approach ($V = WH$) as a positive constraint ($W \geq 0$ and $H \geq 0$). However, PMF forces the negative factors to be positive, but NMF method only retains nonnegative factors. NMF minimizes the conventional least-squares error and the generalized Kullback-Leibler divergence (Shang et al., 2018). Therefore, the results obtained from NMF are more responsive to the original characteristics of input data set and fewer number of factors will be extracted (Zhang et al., 2019). Half of the DL was used for the value below the detection limit. In this study, galactosan, mannose and inositol were excluded because their concentration in most samples was below the DL. Concentrations of the other ten saccharide species for a total of 91 samples were subjected to NMF analysis. The uncertainties in NMF analysis were estimated as 0.3 plus the analytical detection limit according to the method of Xie et al. (1999). The constant 0.3 corresponding to the log (geometric standard deviation) was calculated from the normalized concentrations for all measured species, and was used to represent the variation of measurements.

3 Results and discussion

3.1 Saccharides concentration and composition

The temporal variations of $PM_{2.5}$ mass, OC, EC, and various saccharides measured in all samples are shown in Figure 1. A statistical summary of all the data is listed in

Table S1. During the sampling periods, the $PM_{2.5}$ mass concentrations ranged between 13.7 and 87.8 µg m^{-3} with an average value of 41.8 µg m^{-3}. The concentrations of OC and EC, respectively, were in the range of 2.5–22.4 and 0.3–4.3 µg m^{-3} with average values of 8.4 and 1.7µg m^{-3}. OC accounted for 19.9 ±3.7% of total $PM_{2.5}$ mass. The ambient concentrations of the total saccharides varied between 244.5 and 1291.6 ng m^{-3} with an average value of 638.4 ng m^{-3}. The total saccharides quantified in $PM_{2.5}$ accounted for 8.4 ± 2.7% (range: 3.8%–20.6%) of the OC, and accounted for 1.6 ± 0.6% (range: 0.6%–3.0%) of the $PM_{2.5.}$ Figure 2 presents the average concentration levels of 12 measured saccharide compounds, categorized into anhydrosugar, mono (di) saccharide and sugar alcohol, as well as the relative contribution of these saccharides for all samples. The values for each site are shown in Figure S3.

3.1.1 Anhydrosugars

The average concentrations of levoglucosan and mannosan were 287.7 and 31.6 ng m^{-3}, respectively, with respective ranges of 95.6–714.7 and 0–134.7 ng m^{-3} for all 91 samples. Galactosan was detected only in six samples, with a range of 2.5–5.5 ng m^{-3}. The anhydrosugars accounted for 50.1% of the total measured saccharides. Levoglucosan was the most dominant specie among all the saccharides. The average levoglucosan concentration in this study was comparable to the value at urban Beijing collected in spring of 2012 (above 200 ng m^{-3}) (Liang et al., 2016) and at urban Xi’an collected in winter of 2015 (268.5 ng m^{-3}) (Wang et al., 2018). It was higher than the value at rural Tengchong mountain site (193.8 ng m^{-3}) (Sang et al., 2013), at urban Shanghai collected in spring of 2012 (66.0 ng m^{-3}) (Li et al., 2016) and at urban Hong Kong collected in spring of 2004 (36.0 ng m^{-3}) (Sang et al., 2011), as well as at urban Beijing collected in summer of 2013 (49.4 ng m^{-3}) (Yan et al., 2019), but was lower than that at a rural site of Xi’an (0.93 mg m^{-3}) (Zhu et al., 2017) and at a rural site in eastern central India (2258 ng m^{-3}) (Nirmalkar et al., 2015). During the observation period, several instances of elevated levoglucosan occurred, peaked on 8, 16, 23 March and 1 April. It was thought that the ambient levoglucosan was primarily attributed to domestic biomass fuel burning; the high levoglucosan emission on these peak days might be from open BB events.

Regression analyses of levoglucosan and the other two anhydrosugars (mannosan, galactosan) are shown in Figure 3a. Levoglucosan was highly correlated with mannosan and galactosan, with coefficients of determination (R) of 0.81 ($P < 0.01$) and 0.89 (P <

0.01), respectively, indicating similar combustion sources of them. The ratios of levoglucosan to mannosan (L/M) and mannosan to galactosan (M/G) had been employed to identify the specific types of BB, although these ratios were quite variable (Fabbri et al., 2009; Sang et al., 2018). Previous studies suggested that L/M ratios for the burning of softwood were 3–10, hardwood were 15–25, and those from crop residues were often above 40 (Cheng et al., 2013; Zhu et al., 2015; Kang et al., 2018). The average L/M and M/G ratios were statistically reported as 32.6 and 1.2 for crop residues combustion, 4.0 and 3.9 for softwood combustion, 21.5 and 1.5 for hardwood combustion, respectively (Sang et al., 2013; Shen et al., 2018). In this study, the ratios of L/M and M/G ranged from 4.7 to 16.1 (average: 9.7, n = 91) and from 3.9 to 6.1 (average: 4.8, n = 6), respectively, crudely indicating major contribution from softwood burning. The samples collected from 31 March to 1 April and from 8 to 10 March, respectively, had considerably lower and higher concentrations of mannosan than predicted by the levoglucosan-mannosan regression model (Figure 3a). The results suggested that BB aerosols collected from 31 March to 1 April (L/M = 11.52 ± 1.34) and from 8 to 10 March (L/M = 6.57 ± 0.53) originated from different types of BB compared with the remaining sampling periods (L/M = 9.34 ± 1.20). Therefore, the high levoglucosan emission from 31 March to 1 April and from 8 to 10 March might be from different open BB events, possibly an open agricultural waste burning event or a forest fire, while the BB of most sampling days originated from biomass fuel for domestic cooking and heating. It was worth noting that the peak days from 31 March to 1 April (L/M = 11.52 ± 1.34) neared the Qingming Festival. A possibility of BB events was that people burned ghost money as sacrifice to their ancestors according to Chinese tradition.

Anhydrosugars and water-soluble potassium (K^+) have been both widely utilized as source tracers of BB emissions (e.g., Puxbaum et al., 2007; Wang et al., 2007; Zhang et al., 2008; Engling et al., 2011). The daily variation on concentrations of levoglucosan and K^+ are shown in Figure S4, the regression analysis of K^+ and three anhydrosugars is shown in Figure 3b. K^+ was weakly correlated with levoglucosan, mannosan, and galactosan, with R values of 0.33, 0.28, and 0.74, respectively. It could be explained by the additional emissions of K^+ from soil and sea water. Since Lincang is far from the coast, sea salt could be negligible. Because the inhomogeneity of crustal K^+ associated with soil types, it was difficult to fully account for crustal K^+ contributions from soil (Harrison et al., 2012; Cheng et al., 2013). The ratio of levoglucosan to K^+ (L/K^+) was also used to track possible sources of BB in the previous studies. The ratios of L/K^+ strongly depended on BB processes, namely smoldering and flaming. Studies suggested that relatively high L/K^+ ratios were obtained from smoldering combustion at low temperatures compared with flaming combustion (Schkolnik et al., 2005; Lee et al., 2010). Previous results showed the emissions from the combustion of crop residuals such as rice straw, wheat straw and corn straw exhibited comparable L/K^+ ratios, typically below 1.0. The average of L/K^+ ratios in this study was 0.48 ± 0.20, which was higher than the ratio for wheat straw (0.10 ± 0.00) and corn straw (0.21 ± 0.08), but was lower than the ratio for Asian rice straw (0.62 ± 0.32) (Cheng et al., 2013). In this study, higher L/K^+ ratios were observed during 8-10 March (1.20 ± 0.19) than those from 31 March to 1 April (0.40 ± 0.13), which suggested that the open fire event from 8 to 10 March was more possibly due to smoldering combustion of residues at low temperatures.

Figure 3c and 3d show the scatter plots and regression analyses of K^+ versus $PM_{2.5}$, OC, and EC, and levoglucosan versus $PM_{2.5}$, OC, and EC, respectively. Linear regression of K^+ on $PM_{2.5}$, OC, and EC resulted in R values of 0.64, 0.63, and 0.62, respectively, which were generally higher than those of levoglucosan on $PM_{2.5}$, OC, and EC, with R values of 0.40, 0.54, and 0.48, respectively. It showed that K^+ was more highly correlated with $PM_{2.5}$, OC, and EC, which could be explained by either the photo-oxidative decay of levoglucosan (Hennigan et al., 2010) and/or different types of BB processes (Schkolnik et al., 2005; Lee et al., 2010). Even so, the results supported that the BB posed great impact on fine aerosols. The ratio of levoglucosan to $PM_{2.5}$ (L/$PM_{2.5}$) was also helpful in distinguishing the contributions of different levoglucosan sources (Wu et al., 2021). The ratios of L/$PM_{2.5}$ in this study was 0.0041–0.0162 (average: 0.0072), indicating that the levoglucosan emission in the areas might mainly come from woods (0.01–0.09) and crop straws (0.001–0.008), not excluding incense burning (0.001–0.007), ritual item burning (0.004–0.086), and meat cooking (0.005–0.06). However, certainly, it was not from coal burning (0.0001–0.001) and waste incineration (0.0022).

An empirical ratio of levoglucosan to OC (8.2%), calculated from main types of Chinese cereal straw (rice, wheat and corn) based on combustion chamber experiments (Zhang et al., 2007), was used to estimate the BB-derived OC. The average mass concentration of BB-derived OC was 3534.4 ng m^{-3}, whilst the contributions of BB to OC was 41.3%, with a large range of 19.1%–73.9%. The contributions were higher than those previous reported, such as 6.5%–11% in Hong Kong (Sang et al., 2011), 18%–38% in Beijing (Zhang et al., 2008), 18.9%–45.4% over southeastern Tibetan Plateau (Sang et al., 2013), 26.4%–30.2% in Xi'an (Zhang et al., 2014). The large range of 19.1% –73.9% revealed that the daily contribution of BB varied greatly, suggesting open BB event or forest fire happened occasionally. The contribution apportionment of primary BB might be underestimated due to the degradation of levoglucosan during atmospheric aging of BB influenced air mass after long-range transport (Hennigan et al., 2010; Mochida et al., 2010; Lai et al., 2014). Moreover, Wu et al (2021) have reported that the total levoglucosan emission of China exhibited a clear decreasing trend in China. However, it was noteworthy that the average concentration of levoglucosan (287.7 ng m^{-3}) and the BB contributions to OC (41.3%) at Lincang mountain site were both higher than the values of 191.8 ng m^{-3} and 28.4% at Tengchong mountain site in 2004 spring (Sang et al., 2013). The result suggested that no significant reduction in BB emissions in Southwest Yunnan Province.

3.1.2 Mono (di) saccharides

The total concentrations of five mono (di) saccharides, including glucose, fructose, mannose, sucrose and trehalose, were in the range of 25.2–373.7 ng m^{-3} (average: 158.9 ng m^{-3}), which contributed 24.9 ± 8.3% of the total measured saccharides. The average values of glucose, fructose, mannose, sucrose and trehalose were 31.2, 24.6, 2.7, 86.4, and 13.8 ng m^{-3}, respectively. Sucrose was the dominant mono (di) saccharides. The results agreed with those of previous studies (Yttri et al., 2007; Jia et al., 2010; Fu et al., 2012), which had found that sucrose was one of the dominate specie in spring fine aerosols. The ruptured pollen may be an important source of sucrose, especially in the spring blossom season (Yttri et al., 2007; Fu et al., 2012; Miyazaki et al., 2012). In spring and early summer, farmland tilling after wheat harvest causes an enhanced exposure of soil containing wheat roots to the air, which is beneficial to the release of sucrose stored in the root (Medeiros et al., 2006), thus resulting in a sharply increased sucrose.

It was reported that sugars, such as glucose, sucrose and fructose, could be emitted from developing leaves (Graham et al., 2003). Glucose could be released from both soils and plant materials (e.g., pollen, fruits and their fragments) (Graham et al., 2003; Simoneit et al., 2004; Fu et al., 2012). Glucose and sucrose were rich in biologically active surface soils (Rogge et al., 2007). In this study, the positive correlations were found between sucrose and glucose (R = 0.52) (Table S2), suggesting a similar origin of glucose and sucrose in this study. Glucose and fructose have also been identified as a minor product of cellulose pyrolysis, because they were found to be enrich in BB emission (Nolte et al., 2001), and correlated well with K^+ (Graham et al., 2002) and levoglucosan (Kang et al., 2018). Herein, no significant correlation was found between K^+, levoglucosan, and these mono (di) saccharides. Therefore, the detected glucose, fructose and sucrose might mostly be emitted by direct volatilization from plant materials/surface soils, rather than as products of polysaccharides breakdown during BB processes. The high abundance of sucrose, as well as glucose and fructose were responsible for biogenic aerosols associated with developing leaves and flowers, and surface soil suspension.

Trehalose as a stress protectant of various microorganisms and plants (Medeiros et al., 2006; Jia and Fraser, 2011) was found to be abundant in the fine mode soil, and has been proposed as a marker compound for fugitive dust from biologically active surface soils (Simoneit et al., 2004; Medeiros et al., 2006; Rogge et al., 2007; Fu et al., 2012). Previous study found a positive correlation between trehalose and calcium (Nishikawa et al., 2000). In this study, there was no significant correlation between trehalose and calcium. Besides, mannose has been reported to be one of the major monosaccharide components in phytoplankton, which is originate from marine biological fragments (Tanoue and Handa, 1987). Mannose was detected in only a few samples and presented in low concentrations in this study.

3.1.3 Sugar alcohols

Five sugar alcohol compounds, including glycerol, threitol, mannitol, arabitol and inositol were detected in $PM_{2.5}$. These reduced sugars are often reported to be related to plant senescence and decay by microorganisms (Simoneit et al., 2004; Tsai et al., 2013); and they are produced by fungi, lichens, soil biota and algae (Elbert et al., 2007; Bauer et al., 2008). The average concentration of total sugar alcohols was 159.9 ng m^{-3} with a range of 53.1−254.0 ng m^{-3}, which accounted for 25.1 ± 9.9% of the total measured saccharides. Glycerol has been widely found in soil biota (Simoneit et al., 2004). Previous studies suggested that the source of glycerol was not be specific to biological emissions, biomass combustion might increase atmospheric glycerol concentrations (Jia et al., 2010; Graham et al., 2002; Wang et al., 2011). Herein, glycerol was the second most abundant saccharide, with an average concentration of 123.7 ng

m^{-3} accounting for 5.1%–44.6% (average: 22.6%) of the total measured saccharides.

Mannitol and arabitol have been proposed as tracers for airborne fungal spores (Elbert et al., 2007; Bauer et al., 2008; Zhang et al., 2010; Burshtein et al., 2011). Mannitol and arabitol were detected with a concentration range of 0.0–38.6 ng m^{-3} (14.7 ng m^{-3}) and 0.0–21.1 ng m^{-3} (5.8 ng m^{-3}), respectively. The average concentrations of mannitol and arabitol were comparable to those (average: 11.3 and 9.1 ng m^{-3}) reported in the Beijing spring aerosols (Liang et al., 2013b), but were lower than those (average: 21.9 and 8.43 ng m^{-3}) in the Mediterranean summer aerosols (Burshtein et al., 2011) and (30 and 24 ng m^{-3}) at Hyytiälä, Finland in summer (Yttri et al., 2011). Poor correlations ($r = 0.38$) were found among mannitol and arabitol in this study. Nevertheless, a positive correlation was found between trehalose and mannitol ($r = 0.79$, $P < 0.05$) (Table S2).

In the previous studies, the total measured mannitol has been measured and used for estimating the contribution of fungal spores to organic carbon (Elbert et al., 2007; Bauer et al., 2008; Zhang et al., 2010). A factor of mannitol per spore (0.49 ± 0.20 pg) was used to calculate the number concentrations of fungal spores (Liang et al., 2013a), then the carbon content of fungal spores could be calculated using a conversion factor of 13 pg C per spore obtained earlier as the average carbon content of spores from nine airborne fungal species, with an uncertainty of 20% (Bauer et al., 2008). The diagnostic tracer ratio of mannitol to OC was calculated to be 0.0377 according to these researches (Bauer et al., 2008; Liang et al., 2013a), and then used to estimate the contribution of fungal spores to the OC. The contribution of fungal spores might be underestimated because previous results had indicated that mannitol and arabitol were mainly associated with the coarse PM fraction (Samaké et al., 2019). The average mannitol concentrations were 14.7 ± 11.2 ng m^{-3} during the observation period. The average spore-derived OC was calculated to be 390.3 ng C m^{-3}, which contributed of 4.9% of the total OC.

Claeys et al. (2004) firstly identified two diastereoisomeric 2-methyltetrols as oxidation products of isoprene in the Amazonian rain forest aerosols. Henceforward, 2-methyltetrols has been used as tracers for isoprene-derived SOA (Liang et al., 2012; Fu et al., 2016; Yan et al., 2019). In the previous studies, erythritol was often quantified as surrogate of 2-methyltetrols (2-methylthreitol and 2-methylerythritol) due to lack of standards (Claeys et al., 2004; Ding et al., 2013; Ding et al., 2016). In this study, the concentration range of erythritol was 0.4–19.8 ng m^{-3} (average: 11.1 ng m^{-3}). The values of inositol ranged from 0.0 to 22.8 ng m^{-3} with average values of 5.8 ng m^{-3}. Moreover, the sugar alcohols not only originate from biological emissions but also from BB (Wan and Yu, 2007; Jia et al., 2010). Different levels of glycerol, arabitol, mannitol, erythritol, and inositol in fine particles have been found during the burning of crop residues and fallen leaves as well as indoor biofuel usage for heating and cooking (Graham et al., 2002; Burshtein et al., 2011; Wang et al., 2011; Yang et al., 2012; Kang et al., 2018). In this study, only inositol correlated with levoglucosan (R = 0.42), suggesting that inositol might be linked to biomass combustion sources. Hence, the primary source of sugar alcohols associated with fine particles was biogenic aerosols at observation sites.

3.2 Sources and transport

Since the distinct concentration of the studied compounds was due to different emission sources arising from different wind directions, the 72 h backward trajectories for the samples at the Dashu site (24.12◦ N, 100.11◦ E) and the spatial distribution of the fire spots (8 March to 9 April, 2019) were calculated to understand the source of saccharides in aerosol (Figure 4). The analysis of air mass backward trajectories suggested that the air mass over Lincang were almost from the westerlies during the sampling periods, and could be separated into two episodes of remote western source above 2000 meters and local western source below 2000 meters, as shown in wine red and green lines. 51.6% of air mass backward trajectories were generally above 2000 meters, whereas 48.4% of them were below 2000 meters.

The average concentrations of saccharide compounds, as well as the contribution of them, for the episodes above and below 2000 meters are shown in Figure 5. The average concentrations of levoglucosan and mannosan for the above 2000 meters samples (327.4 and 35.6 ng m^{-3}) were higher than those for the below 2000 meters samples (250.3 and 27.3 ng m^{-3}). The anhydrosugars accounted for 49.2% and 36.9% of total saccharides, respectively for the above and below 2000 meters samples. It implied that the levoglucosan at the observation site was both attributed to the local BB activities and BB smoke transported from the neighboring regions of Southeast Asia (Myanmar) and the northern Indian Peninsula. The southwest wind from the Indian Ocean prevailed at Lincang all year-round. In spring, the southwest wind was often affected by the low temperature downhill wind blowing from the snow-covered Hengduan Mountains. The weather frequently alternated between hot and cold, with unstable air pressure and strong wind. Therefore, the lower air could be diluted by the relatively clean cold air over the plateau. The upper air mainly came from the westerlies. These results were in agreement with the fact that residents across Southeast Asia use wood as an energy source to cook and generate heat.

While for glucose, fructose and sucrose, it was a little higher in the below 2000 meters samples (average: 33.5, 26.4, and 106.2 ng m^{-3}) than that in the above 2000 meters samples (average: 29.2, 22.9, and 67.8 ng m^{-3}). It implied that biogenic aerosols (such as ruptured pollen) carrying sugars could pass long distance, which was supported by previous study, which have observed long-range atmospheric transport of fine pollen from the Asian continent to the remote island Chichi-Jima under the influence of westerlies (Rousseau et al., 2008). Although the pollens are usually coarse with various shapes and hard shells, which results in the relatively short retention time in the atmosphere. Therefore, it could be concluded that, in addition to the local pollen, the concentration of sucrose in Lincang was also influenced by the transport of airborne pollen derived from South Asia areas.

3.3 Source apportionment of saccharides

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

The pie chart in Figure 6b shows the contribution of each source to total saccharides. BB of factor 1 (34.0%) was found as the dominant contributor to total saccharides. Factors 2-5 could all be labeled to a biogenic source accounting for a total contribution of 66.0%. The sources of soil microbiota (factor 2), plant senescence (factor 3), airborne pollen (factor 4), and plant detritus (factor 5), respectively, contributed 16.0%, 21.0%, 23.7%, and 5.3% to total saccharides. During the sampling periods, daily variations in the proportion of the five factors are shown in Figure S5. Factor 2 soil microbiota emissions could be associated to soil reclamation and cultivation of farming periods, whereas factors 3 plant senescence and factor 5 plant detritus could be associated to the harvesting of vegetation or crops. During the observation period of a month, along with the weather warming as sunshine enhanced, humans left two obvious traces of cultivated soil from 9 to 17 March and from 27 March to 8 April and a trace of vegetation or crop harvest from 17 to 30 March. The stronger pollen discharge occurred in March, probably due to the flowering of certain plants. The BB emissions peaked on 9, 16 March, and 1 April were more prone to be open burnings.

Since there is still some uncertainty of the factor apportionment, the proportion of sources are only relative and uncertain. It is still difficult to distinguish the contributions of microorganisms and plants to biogenic aerosols. Furthermore, all the above speculations about farming and harvesting periods are based on only one month's observation, and long-term observations are needed to obtain more accurate and effective information.

4 Conclusion

With the help of various atmospheric saccharides, this study presents the characteristics of BB and various biogenic emissions to ambient aerosol in the rural sites of Southwest China. Levoglucosan was the most dominant specie among all the saccharides, with a concentration of 287.7 ng m^{-3}. The ratios of levoglucosan/OC were 1.9%–8.9% (average: 3.7%). BB contributed to 19.1%–73.9% of OC (average: 41.3%).

The results indicated that domestic biomass fuel burning, open BB events, possibly open agricultural waste burning, forest fire, or sacrificial activity are significant during the spring in this area. The total concentrations of five mono (di) saccharides and five sugar alcohols respectively contributed 24.9 ± 8.3% and 26.6 ± 9.9% of the total measured saccharides. Based on the regression analysis, these mono (di) saccharides and sugar alcohols were mostly emitted by direct biogenic volatilization from plant materials/surface soils, rather than BB processes. The sampling sites suffered from both local emissions and BB via long-range transport from Southeast Asia (Myanmar, Bangladesh) and the northern Indian Peninsula. Five sources of saccharides were resolved by NMF analysis, including BB (34%), soil microbiota (16.0%), plant senescence (21.0%), airborne pollen (23.7%) and plant detritus (5.3%) at rural Lincang in spring.

The data indicated that biofuel and open BB activities in rural Southwest China and neighboring regions could have a significant impact on ambient aerosol levels. In addition to the residential biofuel usage, field burning of agricultural residues, fallen leaves, and forest fire, were non-negligible. Some new technical measures of biomass resource utilization are urgently needed to improve the open burning emission scenario in rural areas, along with strict prohibition policy of BB. Meanwhile, the characteristics analysis of saccharides in the region can serve as a valuable reference for future studies to evaluate temporal variations of biomass combustion and biogenic emission during modeling predictions and policy making.

ASSOCIATED CONTENT

Supporting Information

The location of the sampling sites is shown in Figure S1. Temporal variations of RH, temperature, solar irradiation and rainfall are shown in Figure S2. Average concentrations of saccharide compounds and the contribution of them for the Datian, Dashu, and Yakoutian samples are shown in Figure S3. Daily variation on average concentrations of levoglucosan and K^+ (a), arabitol and mannitol (b), $PM_{2.5}$, Ca^{2+} and trehalose (c) at the three sites throughout the sampling period are shown in Figure S4. Figure S5 showed daily variations on proportion of the five factors to the total saccharides in $PM_{2.5}$ sampled at three sites during the sampling periods. Table S1 lists the concentrations of the carbonaceous components and soluble inorganic ions in $PM_{2.5}$

during the sampling periods of spring 2019. Correlation matrix for the dataset of the determined saccharides compounds in $PM_{2.5}$ samples is shown in Table S2.

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Captions of Figure and Table

Figure 1. Temporal variations of OC, EC, $PM_{2.5}$ and total sugars at the three sites during the sampling periods.

Figure 2. The absolute concentration (bar chart) and the relative contribution (pie chart) of various saccharide compounds during the sampling periods.

Figure 3. (a) Regression analyses of levoglucosan versus the other two anhydrosugars, (b) K^+ versus three anhydrosugars, (c) levoglucosan versus $PM_{2.5}$, OC and EC, and (d) K^+ versus $PM_{2.5}$, OC and EC.

Figure 4. Spatial distribution of the fire spots observed by MODIS, as well as the corresponding 72 h backward air-mass trajectory clusters arriving at 1500 m above ground level （AGL） during the sampling periods for the collected samples. The backward trajectories were separated into two episodes of remote western source above 2000 meters and local western source below 2000 meters, as shown in wine red and green lines.

Figure 5. Average concentrations and contributions of saccharide compounds for the aerosol samples separated as above and below 2000 meters.

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

Figure 1

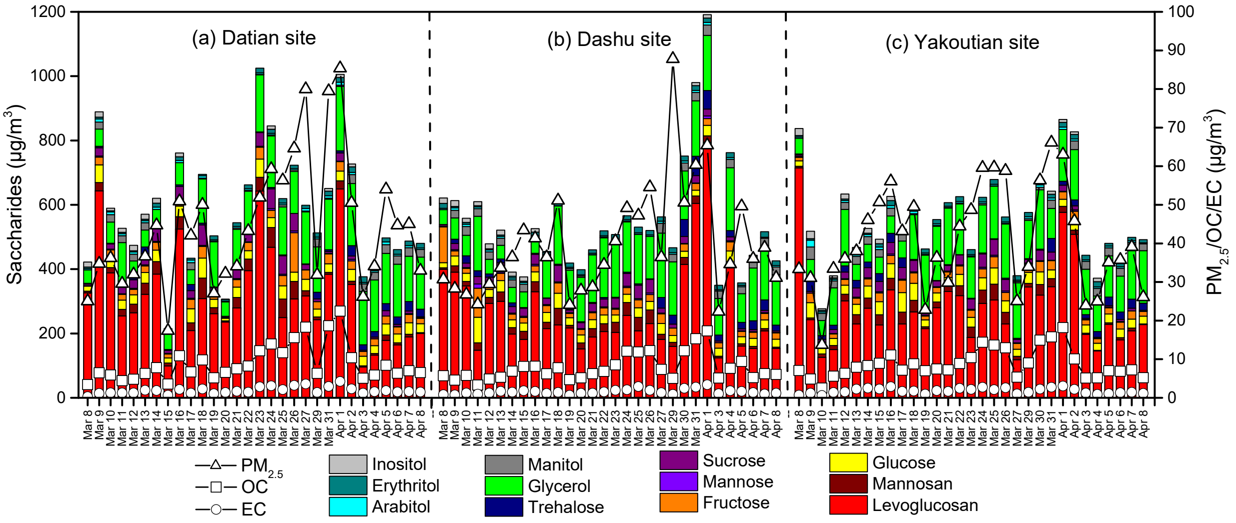

(a) Datian site
(b) Dashu site
(c) Yakoutian site
Saccharides (μg/m^3)
PM$_{2.5}$/OC/EC (μg/m^3)
1200
1000
800
600
400
200
0
100
90
80
70
60
50
40
30
20
10
0
Mar 8
Mar 9
Mar 10
Mar 11
Mar 12
Mar 13
Mar 14
Mar 15
Mar 16
Mar 17
Mar 18
Mar 19
Mar 20
Mar 21
Mar 22
Mar 23
Mar 24
Mar 25
Mar 26
Mar 27
Mar 29
Mar 30
Mar 31
Apr 1
Apr 2
Apr 3
Apr 4
Apr 5
Apr 6
Apr 7
Apr 8
PM$_{2.5}$
OC
EC
Inositol
Erythritol
Arabitol
Manitol
Glycerol
Trehalose
Sucrose
Mannose
Fructose
Glucose
Mannosan
Levoglucosan

Figure 2

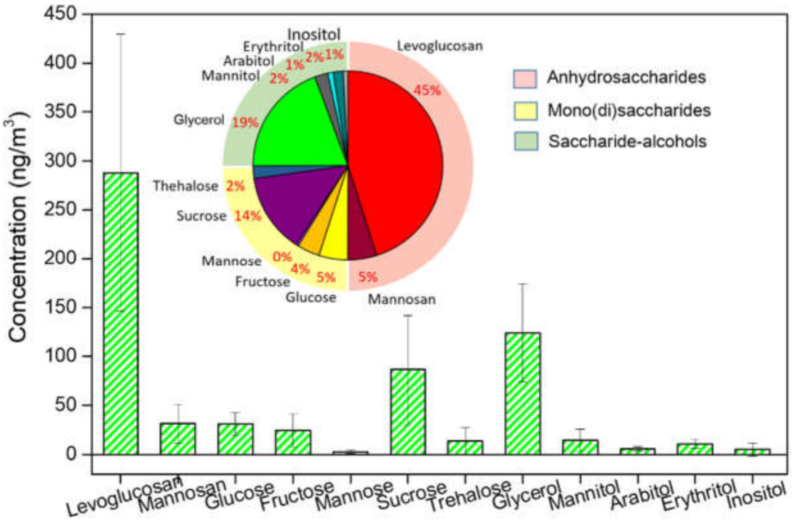

Concentration (ng/m^3)
450
400
350
300
250
200
150
100
50
0
Levoglucosan
Mannosan
Glucose
Fructose
Mannose
Sucrose
Trehalose
Glycerol
Mannitol
Arabitol
Erythritol
Inositol
Inositol
Erythritol
Arabitol
Mannitol
Glycerol
Thehalose
Sucrose
Mannose
Fructose
Glucose
Mannosan
Levoglucosan
45%
5%
5%
4%
0%
14%
2%
19%
2%
1%
2%
1%
Anhydrosaccharides
Mono(di)saccharides
Saccharide-alcohols

Figure 3

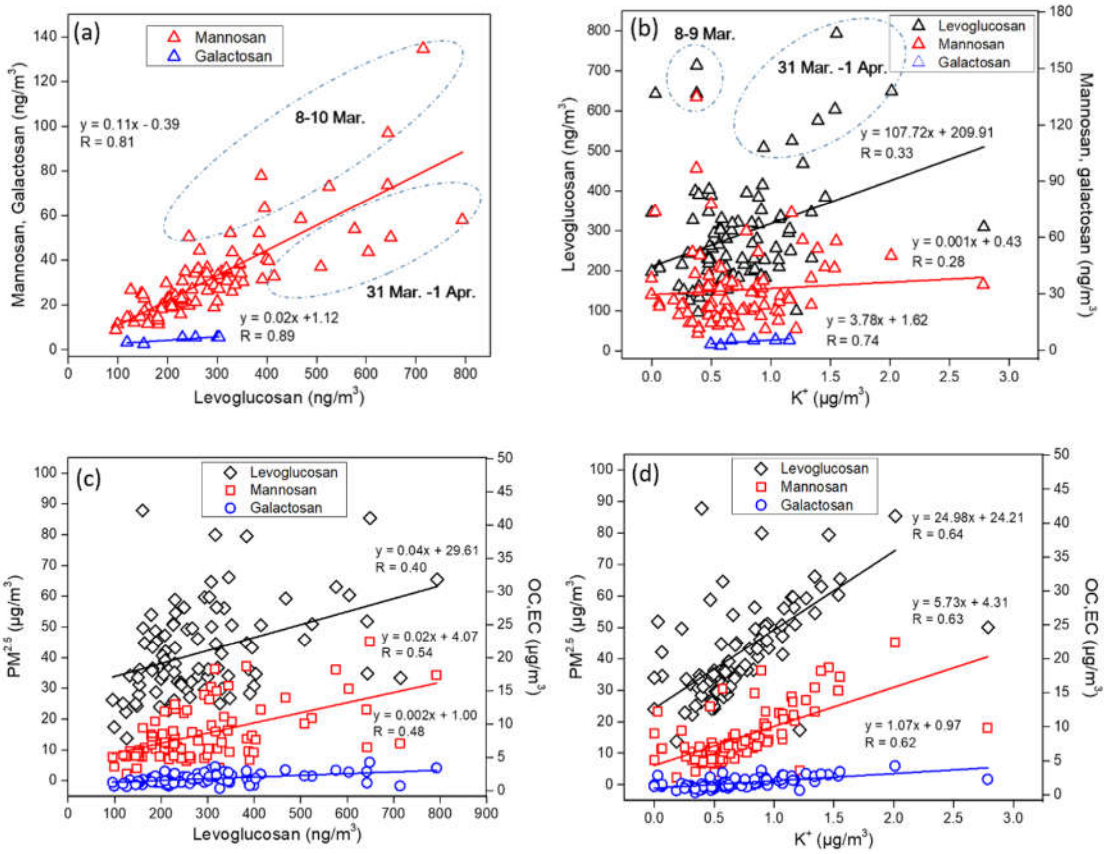

(a)
Mannosan
Galactosan
y = 0.11x - 0.39
R = 0.81
8-10 Mar.
31 Mar. -1 Apr.
y = 0.02x +1.12
R = 0.89
Mannosan, Galactosan (ng/m^3)
Levoglucosan (ng/m^3)
0
20
40
60
80
100
120
140
0
100
200
300
400
500
600
700
800
(b)
8-9 Mar.
31 Mar. -1 Apr.
Levoglucosan
Mannosan
Galactosan
y = 107.72x + 209.91
R = 0.33
y = 0.001x + 0.43
R = 0.28
y = 3.78x + 1.62
R = 0.74
Levoglucosan (ng/m^3)
Mannosan, galactosan (ng/m^3)
K$^+$ (μg/m^3)
0
100
200
300
400
500
600
700
800
0
30
60
90
120
150
180
0.0
0.5
1.0
1.5
2.0
2.5
3.0
(c)
Levoglucosan
Mannosan
Galactosan
y = 0.04x + 29.61
R = 0.40
y = 0.02x + 4.07
R = 0.54
y = 0.002x + 1.00
R = 0.48
PM$^{2.5}$ (μg/m^3)
OC,EC (μg/m^3)
Levoglucosan (ng/m^3)
0
10
20
30
40
50
60
70
80
90
100
0
5
10
15
20
25
30
35
40
45
50
0
100
200
300
400
500
600
700
800
900
(d)
Levoglucosan
Mannosan
Galactosan
y = 24.98x + 24.21
R = 0.64
y = 5.73x + 4.31
R = 0.63
y = 1.07x + 0.97
R = 0.62
PM$^{2.5}$ (μg/m^3)
OC,EC (μg/m^3)
K$^+$ (μg/m^3)
0
10
20
30
40
50
60
70
80
90
100
0
5
10
15
20
25
30
35
40
45
50
0.0
0.5
1.0
1.5
2.0
2.5
3.0

Figure 4

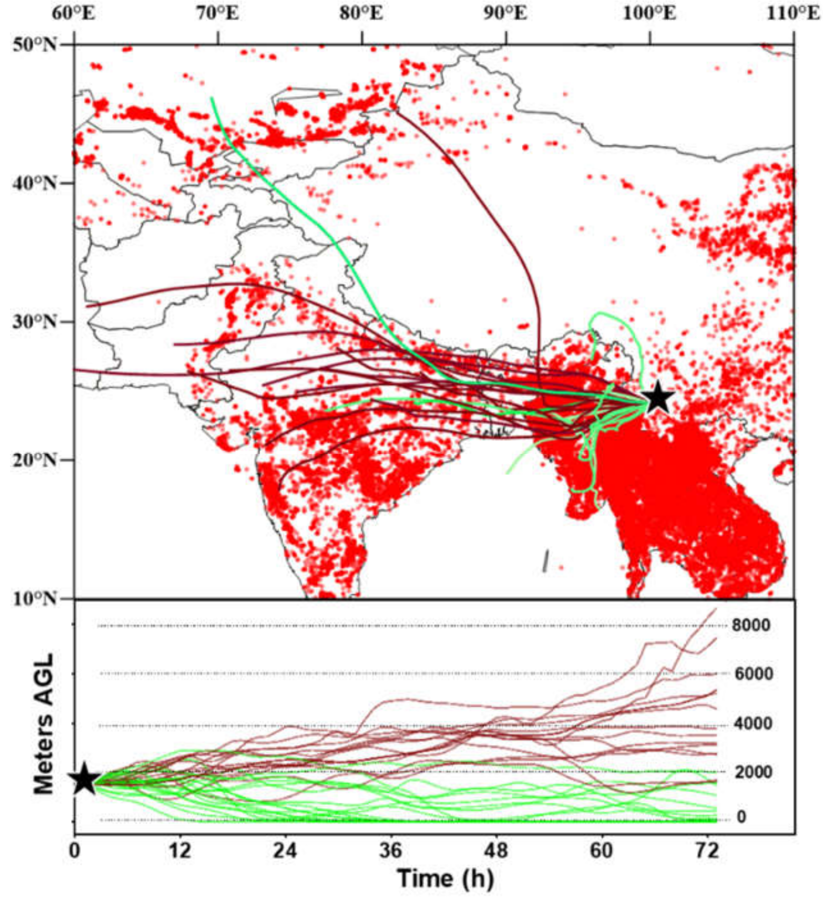

60°E
70°E
80°E
90°E
100°E
110°E
50°N
40°N
30°N
20°N
10°N
Meters AGL
8000
6000
4000
2000
0
0
12
24
36
48
60
72
Time (h)

Figure 5

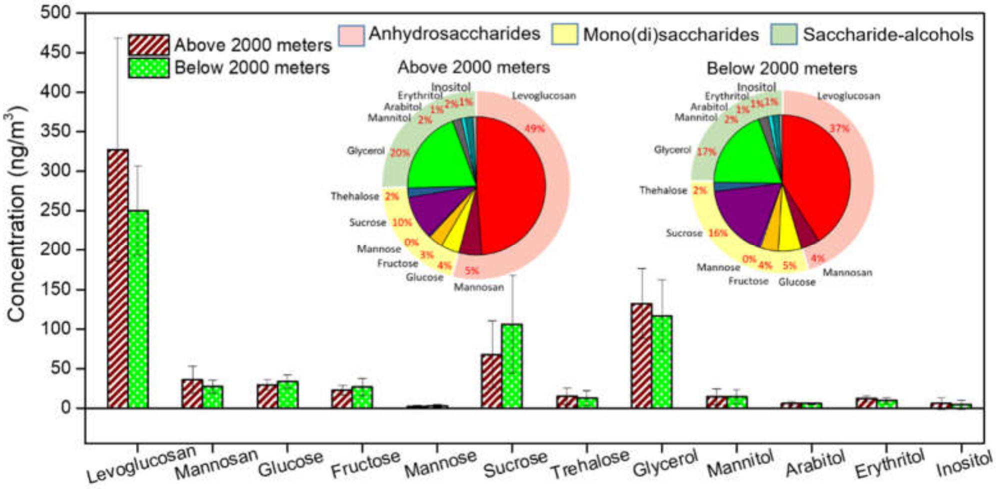
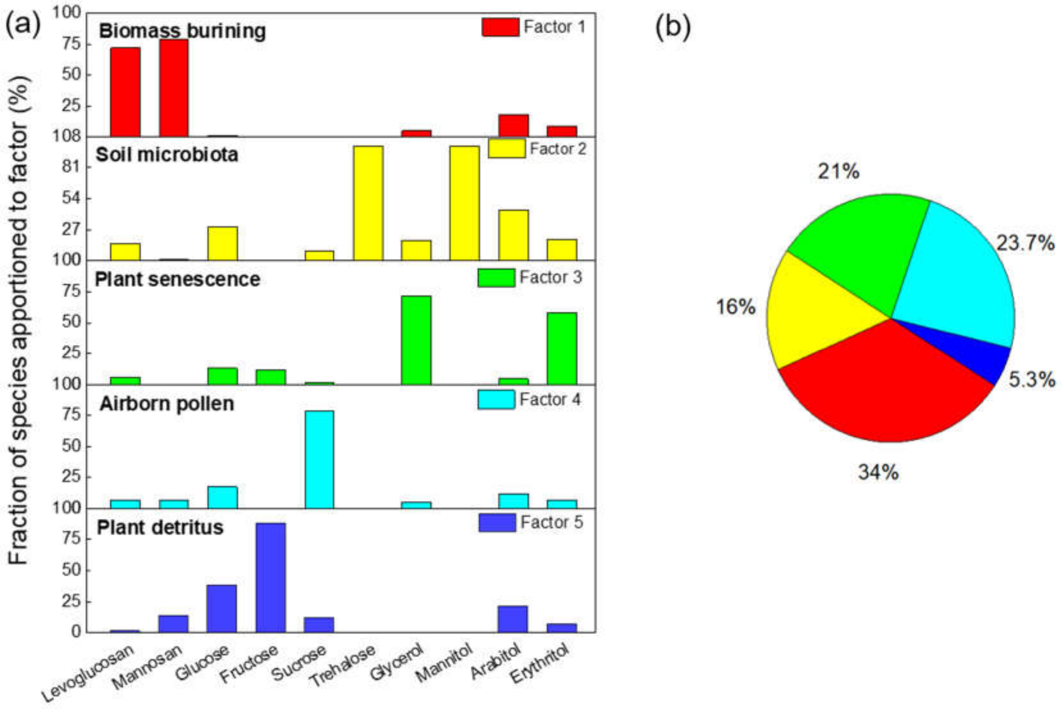

500
450
400
350
300
250
200
150
100
50
0
Concentration (ng/m³)
Above 2000 meters
Below 2000 meters
Anhydrosaccharides
Mono(di)saccharides
Saccharide-alcohols
Above 2000 meters
Inositol
Erythritol
Arabitol
Mannitol
1%
2%
1%
2%
Levoglucosan
49%
Glycerol
20%
Thehalose
2%
Sucrose
10%
Mannose
0%
Fructose
3%
Glucose
4%
5%
Mannosan
Below 2000 meters
Inositol
Erythritol
Arabitol
Mannitol
1%
1%
1%
2%
Levoglucosan
37%
Glycerol
17%
Thehalose
2%
Sucrose
16%
Mannose
0%
Fructose
4%
Glucose
5%
4%
Mannosan
Levoglucosan
Mannosan
Glucose
Fructose
Mannose
Sucrose
Trehalose
Glycerol
Mannitol
Arabitol
Erythritol
Inositol

Figure 6


(a)
Fraction of species apportioned to factor (%)
Biomass burining
Factor 1
Soil microbiota
Factor 2
Plant senescence
Factor 3
Airborn pollen
Factor 4
Plant detritus
Factor 5
100
75
50
25
108
81
54
27
0
Levoglucosan
Mannosan
Glucose
Fructose
Sucrose
Trehalose
Glycerol
Mannitol
Arabitol
Erythritol
(b)
21%
23.7%
16%
5.3%
34%