Biomass burning is an important source of organic aerosols in Interior Alaska

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Abstract

Biomass burning (BB) affects air quality, the global cycling of carbon, climate, and human health. Intensive BB activities occur throughout the year due to wildfires and domestic wood burning in Fairbanks. We collected total suspended particle samples from the air in Fairbanks from June 2008 to June 2009. Here, we report seasonal variations in the molecular composition of organic aerosols and its BB contributions in Fairbanks. Levoglucosan is the dominant BB tracer (annual mean 67 ± 77 ng m^{-3}), showing a winter maximum (145 ± 47 ng m^{-3}) and spring minimum (12 ± 12 ng m^{-3}). Levoglucosan showed significant correlations ($p<0.001$) with organic carbon (OC), water-soluble organic carbon (WSOC) and $PM_{2.5}$, indicating a significant contribution of BB to hazes in Fairbanks. However, no correlation exists between levoglucosan and non-sea salt potassium (nss-K^+), which has been proposed as a BB tracer. We hypothesize that nss-K^+ is removed by deposition on the inner surfaces of woodstove chimneys. Levoglucosan contributes 3.6% to OC and 6.3% to WSOC in winter, indicating that BB significantly affects carbonaceous aerosols in central Alaska. Moreover, positive matrix factorization analysis demonstrates that BB is an important source (47.5%) of Fairbanks year-round aerosols. We conclude that domestic wood burning is an important source of atmospheric particles that impact the air quality of Fairbanks, especially in winter. We presumed that Fairbanks BB products might be transported to the remote Arctic, potentially affecting the chemical composition of Arctic aerosols. These findings will be useful to better understand the seasonal influence of BB on the Arctic and subarctic aerosols.

Keywords: Domestic wood burning, anhydrosugars, lignin and resin acids, seasonal variation, potassium, woodstove

1. Introduction

Particulate matter (PM) is a complex mixture of small solid particles and liquid droplets suspended in the air, many of which are known as hazardous pollutants. Biomass burning (BB) emits various gases such as CO_2, CO, CH_4, C_{2-4} hydrocarbons, NH_3, NO_x, and aerosol components such as organic carbon (OC), black carbon (BC), sugars, alcohols, organic acids, polycyclic aromatic hydrocarbons (PAHs) and persistent organic pollutants (POPs), and thus plays an important role in affecting air quality, climate and health (McMeeking et al., 2009; Hu et al., 2013; Burnett et al., 2018). On a global scale, BB accounts for up to 85% of primary organic aerosol emissions (Andreae, 2019). Carbonaceous components in fire smoke cause human respiratory diseases (Laumbach & Kipen, 2012). A laboratory study suggests that secondary organic aerosol (SOA) can be formed substantially in the BB plumes via aging processes (Grieshop et al., 2009). The SOA formation and modification have been confirmed by field observations (Adler et al., 2011; Garofalo et al., 2019; Lee et al., 2008). Organic aerosols associated with BB contain large amounts of oxygenated and polar compounds and thus can act as cloud condensation nuclei (CCN), influencing the microphysical properties of clouds (Kondo et al., 2011).

There are several types of BB, including open-fires in forests, savannas, peatlands, field burning of agricultural wastes, residential heating and cooking, and industrial biofuel burning (Akagi et al., 2011; Engling et al., 2014). Forest fires frequently occur globally. Burning areas increased from 1 million ha yr^{-1} in 1950 to almost 3 million ha yr^{-1} in 2000 in the North American boreal forests (Kasischke & Stocks, 2000), where black spruce (*Picea mariana*) is an important wildfire fuel type (e.g., Amiro et al., 2001). Northern boreal forests, including permafrost areas, represent almost 35% of the world's forest, containing 66% of the world's forest soil carbon pools (Kim & Tanaka, 2003). Recent studies have reported a significant contribution of boreal forest fires in summer to the aerosol burden over the Arctic, which is sensitive to climate change (Iziomon et al., 2006; Stohl et al., 2006; Kaplan & New, 2006). Moreover, fire incidents and areas burned have been increasing in Alaska in recent decades (French et al., 2002). BB for residential heating also plays an important role in the generation of PM (Busby et al., 2016). In the countries bordering the Arctic, i.e., Scandinavia, Canada, United States of America (USA), and Russia, substantial amounts of wood are frequently used for domestic heating (Yttri et al., 2014). Domestic wood burning significantly contributes to the ambient aerosols (Gelencsér et al., 2007; Puxbaum et al., 2007; Lanz et al., 2010; Maenhaut et al., 2012; Genberg et al., 2013). Biopolymers such as cellulose, lignin, hemicellulose, suberin, sporopollenin, chitin are major constituents of biomass, whereas several tracers such as

anhydrosugars (Engling et al., 2006, 2009; Fu et al., 2012a; Mochida et al., 2010), and resin acids (Kawamura et al., 2012; Zhang et al., 2013) are emitted to the atmosphere by BB.

Fairbanks is located south of the Arctic Circle and is in the subarctic climate region. This city has suffered from severe air pollution, particularly in winter (Ward et al., 2012; Ward et al., 2015; Busby et al., 2016). Ward et al. (2012) pointed out that 60-80% of the ambient $PM_{2.5}$ in Fairbanks originated from residential wood burning. Wang and Hopke (2014) proposed that winter heating is the major factor controlling the air quality of Fairbanks. Schmale et al. (2018) reported that OC species are a large part of $PM_{2.5}$ in Fairbanks. Benson (1969) first reported that air pollution in Fairbanks is also associated with meteorological inversions because the downtown is in a valley. In 2017, this city was classified as a severe non-attainment area due to serious air pollution in winter (Schmale et al., 2018). Several modeling studies on the air pollution have been conducted (Tran & Mölders, 2011, 2012; Leelasakultum et al., 2012; Mölders et al., 2012; Kotchenruther, 2016). Although Busby et al. (2016) directly measured bulk parameters combined with levoglucosan and ^{14}C in winter aerosols, organic tracer-based studies are still limited in Fairbanks particularly for year-round.

Wang and Hopke (2014) reported a source apportionment study based on PM and OC measurements and encouraged further studies to confirm their results by performing organic tracer-based source apportionment. The results of chemical tracers and their ratios can explain the photochemistry and characteristic burning substances, respectively. To better understand the specific emission sources in Fairbanks for different seasons, we conducted a source apportionment study based on specific organic tracers and major ions. Here, we present, for the first time, one-year time series of organic BB tracer observations in the North American subarctic region, with detailed chemical composition of BB tracers in the ambient aerosols from Fairbanks. The contribution of BB to organic aerosols is estimated by positive matrix factorization (PMF) analysis to demonstrate the importance of forest fire in summer and woodstove burning in winter.

2. Experimental

2.1. Aerosol sampling

Total suspended particle samples (n = 32) were collected on the rooftop of the International Arctic Research Center (IARC) building at the University of Alaska Fairbanks (64.51°N; 147.51°W), Alaska, USA, from June 2008 to June 2009. The IARC is a five-story building (ca. 20 m above the ground). The planetary boundary layer height (PBLH) ranged from 100-744 m during the campaign, and was lowest in winter, with some daily averages as low as 32 m (Table S1). Such low PBLHs could impact concentrations of BB tracers in Fairbanks

during the winter to an extent not fully captured at the sampling location, since the university campus is located on the outskirts of Fairbanks near a natural forest (Figure 1). The altitude of IARC is 192 m above sea level. Sampling was conducted at one to three week intervals on pre-combusted (450°C for 6 h) quartz fiber filters (47 mm, Pall Corporation, USA) using a low volume air sampler (NIER) at a flow rate of 16.7 L min^{-1}. Field blanks were collected by placing blank filters into the sampler for 5 min without sucking air. After sampling, each sample filter was placed in a pre-combusted (450°C for 6 h) glass vial with a Teflon-lined screw cap. The samples were transported to the laboratory in Sapporo, Japan, and stored in a dark freezer room at -20°C until the analysis.

2.2. Extraction and derivatization

Filter aliquots (disk diameter of 20 mm) were cut into pieces and extracted three times with 7 mL of dichloromethane/methanol (2:1; v/v) under ultrasonication for 10 min. The solvent extracts were filtered through a Pasteur pipette packed with quartz wool to remove filter debris and particles, concentrated using a rotary evaporator under vacuum and then blown dry with nitrogen (purity >99.99%). The total extracts were then reacted with 50 μL of N,O-bis(trimethylsilyl)trifluoroacetamide (BSTFA) with 1% trimethylsilyl chloride and 10 μL of pyridine in a glass vial (1.5 mL) with a Teflon-lined screw cap at 70°C for 3 h to derive trimethylsilyl (TMS) derivatives: COOH and OH groups are converted to the corresponding TMS esters and ethers, respectively (Simoneit et al., 2004a; Simoneit et al., 2004b; Wang and Kawamura, 2005). The derivatives were diluted with 40 μL of *n*-hexane containing an internal standard (C_{13} *n*-alkane, 1.43 ng μL^{-1}) prior to injection into a gas chromatography-mass spectrometry (GC-MS) for identification and quantification (Fu et al., 2008).

2.3. Gas chromatography-mass spectrometry

The derivatized total extracts were analyzed using a Hewlett-Packard model 6890 GC coupled to Hewlett-Packard model 5973 Mass-Selective Detector (MSD). The GC separation was performed on a DB-5MS fused silica capillary column (30 m × 0.25 mm i.d., 0.5 μm film thickness) with helium as a carrier gas at a flow rate of 1.0 mL min^{-1}. The GC oven temperature was programmed from 50°C (2 min) to 120°C at 30°C min^{-1} and then to 300°C at 6°C min^{-1} with a final isotherm hold at 300°C for 16 min. The sample was injected at 280°C on a splitless mode. The mass spectrometer was operated on an electron impact (EI) mode at 70 eV and scanned from 50 to 650 Daltons. Mass spectral data were acquired and processed using the Hewlett-Packard Chemstation software.

Organic compounds were identified individually by comparing retention times and mass spectra with authentic standards, literature and Chemstation library data of mass fragmentation

patterns (Medeiros & Simoneit, 2007). Mass fragment ions were used for quantification: m/z 217, 204 and 333 for levoglucosan, mannosan and galactosan; m/z 239 for dehydroabietic acid; m/z 297, 312 and 342 for vanillic acid; m/z 327, 312 and 342 for syringic acid; m/z 193, 223, 267 and 282 for 4-hydroxybenzoic acid. GC-MS relative response factors (RRF) of individual compounds were determined using authentic standards and surrogate compounds. Authentic standards were used for the measurements of levoglucosan, dehydroabietic, vanillic, syringic and 4-hydroxybenzoic acids, whereas levoglucosan was used as a surrogate compound for the determination of mannosan and galactosan by comparing their RRF values.

Each standard (100-200 ng) was spiked to the 20 mm punch of pre-combusted quartz fiber filters, which were then analyzed as a real sample. This recovery experiment was repeated three times, and the results showed average recoveries of more than 80% for target compounds. The field (n = 3) and laboratory blank (n = 3) filters were also analyzed by the procedures described above. Target compounds were not detected in the field blanks. The data reported here were not corrected for recoveries. The analytical errors based on replicate analyses (n = 3) of real samples were generally <10%, in which uncertainties in the recovery test (up to 20%) were not taken into consideration. The limits of detection (LOD) for target compounds in the injected samples ranged from 0.007 to 0.079 ng μL^{-1}, which are equivalent to atmospheric concentrations of 0.12 ng m^{-3} (galactosan), 0.22 ng m^{-3} (mannosan), 1.07 ng m^{-3} (levoglucosan), 0.35 ng m^{-3} (4-hydroxybenzoic acid), 0.04 ng m^{-3} (vanillic acid), 0.01 ng m^{-3} (syringic acid), and 0.12 ng m^{-3} (dehydroabietic acid).

2.4. Carbonaceous components

Organic carbon (OC) and elemental carbon (EC) were measured using a thermal-optical carbon analyzer (Sunset Lab., USA) (Birch and Cary, 1996) following the Interagency Monitoring Protected Visual Environments (IMPROVE) thermal evolution protocol. Aliquots of quartz filter samples (14 mm diameter) were punched and put in a quartz tube inside the thermal desorption chamber of the analyzer and then combusted with stepwise heating as described in Wang et al. (2005). Helium was applied in the first ramp and then switched to a He/O_2 mixture in the second ramp. During the oxidation process at each temperature step, the evolved CO_2 was measured with a non-dispersive infrared (NDIR) detector. The transmittance of light at 660 nm through the filter punch was used for setting up OC/EC split point and thereby OC correction. The analytical errors in replicate analyses (n = 5) were less than 7% for OC and 5% for EC. The reported concentrations of OC and EC in this study were corrected for the field blanks.

To determine water-soluble organic carbon (WSOC), an aliquot of filter (18 mm diameter) was extracted with 15 mL of organic free pure water by ultrasonication for 30 mins

and then filtered with a syringe filter (Millex-GV, 0.22 μm pore size, Millipore, USA). WSOC in water extracts was measured using a total organic carbon (TOC) analyzer (Model TOC-Vcsh, Shimadzu, Kyoto, Japan) as described in Miyazaki et al. (2011). The concentrations of WSOC reported here were corrected for the field blanks. Duplicate analysis of filter samples showed uncertainties of 5% for WSOC.

2.5. Major inorganic ions

For the determination of major inorganic ions, another filter cut (14 mm diameter) was extracted with 10 mL ultrapure water (resistivity >18.2 MΩ cm, Sartorius arium 611 UV) in a plastic container using ultrasonication for 15 mins. The total extracts were filtered through a membrane disc filter (Millex-GV, 0.45 μm, Millipore, USA) and injected into ion chromatography (Model 761 compact IC, Metrohm, Switzerland) for measuring major anions and cations. Anions were isolated on a SI-90 4E Shodex column (Showa Denko, Tokyo, Japan) with a suppressor using a mixture of 1.8 mM Na_2CO_3 and 1.7 mM $NaHCO_3$ as an eluent at a flow rate of 1.2 mL min^{-1}. For cations, a Metrosep C2-150 (Metrohm) column was used with 4 mM tartaric acid + 1 mM 2,6-pyridinedicarboxylic acid as an eluent at a flow rate of 1.0 mL min^{-1}. The injection loop volume was 200 μL for both columns. All ions were quantified against a standard calibration curve.

The concentrations of ions reported here were corrected for the field blanks. The analytical errors for inorganic ions were estimated to be less than 5% based on triplicate analysis of ambient samples. We calculated the fractions of Na^+ and Ca^{2+} derived from sea salt (ss) and non-sea salt (nss) sources in Fairbanks samples using the following equations.

$$ss\text{-}Na^+ = Na^+ - nss\text{-}Na^+$$

$$= Na^+ - nss\text{-}Ca^{2+}(Na^+/Ca^{2+})_{crust}$$

$$nss\text{-}Ca^{2+} = Ca^{2+} - ss\text{-}Ca^{2+}$$

$$= Ca^{2+} - ss\text{-}Na^+(Ca^{2+}/Na^+)_{seawater}$$

$$(Na^+/Ca^{2+})_{crust} = 0.56 \text{ (w/w)}$$

$$(Ca^{2+}/Na^+)_{seawater} = 0.038 \text{ (w/w)}$$

, where $(Na^+/Ca^{2+})_{crust}$ means ratios in the Earth's crust and $(Ca^{2+}/Na^+)_{seawater}$ means ratio in bulk sea water (Bowen, 1979), whereas w/w means weight by weight ratios. nss-K^+ was calculated by the method of George et al. (2008) as follows.

$$nss\text{-}K^+ = K^+ - ss\text{-}Na^+(K^+/Na^+)_{seawater}$$

$$(K^+/Na^+)_{seawater} = 0.037 \text{ (w/w)}$$

More details of the calculation are described elsewhere (Becagli et al., 2005; Benassai et al., 2005; George et al., 2008). The contributions of ss-Na^+ and ss-Ca^{2+} were minor in Fairbanks

aerosols. Therefore, Ca^{2+} in the samples are mainly derived from crust. The measured Na^{+} and K^{+} were also considered as non-sea salt from the continent, being consistent with the results of McMeeking et al. (2009).

2.6. Air mass trajectories, fire counts and PBLHs

In order to investigate the source regions of BB tracers and the influence of long-range atmospheric transport, 7-day air mass backward trajectories were calculated on a daily basis at 00.00 UTC time from June 2008 to June 2009 using the NOAA Hybrid Single-Particle Lagrangian Integrated Trajectory model (http://ready.arl.noaa.gov/HYSPLIT.php) (Figure S1) and meteorological data (6 h archived 1°×1° Global Data Assimilation System) from the National Centers for Environmental Prediction (http://ready.arl.noaa.gov/gdas1.php). The starting heights of the trajectories are 50 m (winter) and 100 m (summer) above the ground, which is within the boundary layer in most of cases. The sensitivity of back trajectories was investigated for different starting elevations at 50, 100, 500 and 1000 m.

PBLHs were calculated using MeteoInfo software tools (Wang, 2014). Cluster analyses were further applied to better interpret the air mass origins, in which three seed clusters (centroids) were derived in each season (Figure 2). Based on the source regions of air parcels, we considered three dominant source regions for the air mass back trajectories, namely, the Arctic Ocean, the Pacific Ocean and the Bering Sea in summer and spring, and the Arctic Ocean, the Bering Sea and Alaska in winter and autumn. However, all groups of air masses passed over continental Alaska before arriving at the sampling site in Fairbanks. The Pacific Ocean was a dominant (43%) source region of air clusters in summer, while air parcels from the Bering Sea showed the highest contribution in spring. Furthermore, Alaskan air masses contributed abundantly (50%) in winter, whereas the air cluster from the Arctic Ocean was dominant in autumn (44%).

For understanding the influence of open BB in the Arctic and subarctic regions, combustion activities were illustrated by fire spots, whose data were obtained from Fire Information for Resource Management System (FIRMS) operated by the National Aeronautics and Space Administration (NASA) of the United States (https://earthdata.nasa.gov/data/near-real-time-data/firms) (Figures 2 and S1). Monthly composite images of the Moderate Resolution Imaging Spectroradiometer (MODIS) active fire spots were also obtained using the Earth Observing System Data and Information System (EOSDIS) from the Terra and Aqua satellites (https://earthdata.nasa.gov/data/near-real-time-data/firms/) (Figure S2).

2.7. Chimney deposit samples from a woodstove

Chimney deposit samples (n = 18) were collected from an inner surface of the chimney (height of 6 m) connected to a woodstove (Morso model 1126CB). The woods used as fuels were dried broadleaf trees (mostly oak). At the beginning of burning, the stovetop temperature was low (less than 100°C). Within 10 minutes, the temperature increased to 200°C. The estimated temperature inside of the stove was 500-800°C with an ideal condition of 200-250°C at the stovetop. We collected deposits from different heights of the chimney after the end of the winter season. The chimney deposit samples were analyzed for major ions and organic compounds using the methods described above.

2.8. PMF analysis

To better understand the importance of BB products to organic aerosols in central Alaska, we performed a PMF analysis using the EPA-PMF model (version 5.0) (http://www.epa.gov/heasd/products/pmf/pmf.html). PMF is a powerful statistical tool to verify the sources of atmospheric particles (Paatero & Tapper, 1994; Kim et al., 2003; Jaeckels et al., 2007; Miyazaki et al., 2012). We used the uncertainties of 15% for the error estimates of the measured organic compounds in this study. Half of the LOD was used for the values below the detection limit, and 5/6 of the LOD was used for the corresponding error estimate (Fu et al., 2012b). PMF runs were performed with 3-7 factors. Based on Q values (the objective function to be minimized) (Paatero & Tapper, 1994) and the interpretable factors identified by PMF, four factors appeared to be the optimal solution to the probable origin of organic aerosols in the Fairbanks atmosphere.

3. Results and Discussion

3.1. Characterization of BB tracers in central Alaskan aerosols

3.1.1. Molecular compositions of BB tracers

Two types of BB tracers: (a) anhydrosugars (levoglucosan, mannosan, and galactosan) and (b) lignin and resin acids (dehydroabietic, vanillic, syringic, and 4-hydroxybenzoic acids) were identified in the Alaskan aerosol samples (Table 1). The concentrations of levoglucosan, mannosan, and galactosan were found to be 1.1-273 ng m^{-3} (avg. 66.7 ng m^{-3}), 0.2-63.1 ng m^{-3} (15.0 ng m^{-3}) and 0.1-39.4 ng m^{-3} (9.9 ng m^{-3}), respectively. These values are much higher than those reported in other Arctic sites such as Alert in the Canadian High Arctic (0.003-1.08 ng m^{-3}, Fu et al., 2009), the Arctic Ocean (1.1-41 ng m^{-3}, Hu et al., 2013), Svalbard in Norway (0.07-3.20 ng m^{-3}, Yttri et al., 2014), and the southern Beaufort Sea (0.01-0.93 ng m^{-3}, Fu et al., 2013), suggesting a large impact of BB in the central Alaskan aerosols. Levoglucosan was detected as the dominant BB tracer, followed by its two isomers: mannosan and galactosan. Time series concentrations for anhydrosugars are shown in Figure 3e-g, where all the compounds present

very similar temporal trends. The concentrations of anhydrosugars seem to increase in September with the highest peak in November and then dramatically decrease toward March, and again increase in June. Levoglucosan showed strong correlations with mannosan ($r = 0.98$, $p<0.001$) and galactosan ($r = 0.94$, $p<0.001$), indicating that they were derived from similar emission sources (Figure 4a-b). The concentration level of levoglucosan in Fairbanks is comparable to the levels reported from other sites in the world (Table 2).

Dehydroabietic acid is another tracer of BB that has been recognized as a more specific tracer of the burning of conifer trees (Kawamura et al., 2012). This acid is produced by the incomplete combustion of conifer resin (Simoneit, 2002). The concentrations of dehydroabietic acid ranged from below detection limit (BDL) to 42.6 ng m^{-3} with an average of 7.4 ng m^{-3}. Vanillic, syringic, and 4-hydroxybenzoic acids are produced by the incomplete combustion of lignin (Simoneit, 2002). The concentration ranges of vanillic, syringic, and 4-hydroxybenzoic acids were BDL-15.7 ng m^{-3} (avg. 2.3 ng m^{-3}), BDL-6.6 ng m^{-3} (0.7 ng m^{-3}), and 0.4-5.7 ng m^{-3} (1.7 ng m^{-3}), respectively. Dehydroabietic acid was more abundant than other lignin and resin products. The lignin and resin products exhibited temporal variations similar to levoglucosan (Figure 3a-e). Significant correlation coefficients were also obtained between levoglucosan and lignin and resin products, indicating that they originated from similar sources (Figure 4c-f). Major BB tracers exhibited a seasonal variation, as discussed below.

3.1.2. Seasonal variations of BB tracers

Seasonal variations were observed in the concentrations of anhydrosugars detected in the ambient aerosol samples from Fairbanks (Figure 5a). Seasons are defined as summer (June-August), autumn (September-November), winter (December-February), and spring (March-May). The average concentrations of levoglucosan were significantly higher in winter (avg. 145 ± 47 ng m^{-3}, n = 5) and autumn (129 ± 80 ng m^{-3}, n = 7) compared to summer (34.2 ± 45 ng m^{-3}, n = 13) and spring (12.3 ± 12 ng m^{-3}, n = 7). The seasonal variations of mannosan and galactosan were similar to levoglucosan, although their concentrations are 4-5 times lower than levoglucosan (Table 1). Dehydroabietic, vanillic, syringic, and 4-hydroxybenzoic acids showed seasonal trends similar to levoglucosan with maxima in winter followed by autumn, summer, and spring (Table 1, Figure 5b). Smaller numbers of fire spots were observed in autumn (total 188) and winter (not detected) than summer (1896) and spring (404) in Alaska during the campaign.

Indoor burning for residential heating may have largely contributed to BB tracers during cold seasons in Fairbanks. Moreover, a decreased dispersion due to the lower PBLH may explain the high levels of BB tracers in winter. Lower PBLHs in the range from 32.1 to 744 m were observed in winter (daily average), although in summer, they could reach 1264 m (Table S1).

Previous studies also pointed out that local emissions and poor dispersion of the pollutants caused severe air pollution in Fairbanks in winter (Tran & Mölders, 2011; Schmale et al., 2018). A reduced photo-oxidation during the dark winter might decrease the degradation of BB tracers. The high latitudinal Northern Hemisphere acts as a cold sink to receive aerosols, whereas aerosol removal rates are decreased under a stagnant condition due to a lack of solar radiation (Shaw et al., 1995). Photochemical oxidations at high latitudes are decreased due to extreme cold and dry conditions and long periods of darkness, followed by a prolonged time of sunlight at high solar zenith angles (Olson et al., 2012). Moreover, scavenging rates of organic compounds may decrease in winter due to the lower relative humidity of cold air (Li et al., 2018; Pant et al., 2018).

High loadings of levoglucosan were observed during the sampling period of 13-21 November 2008 (241 ng m^{-3}) and 21 November to 1 December 2008 (273 ng m^{-3}) when no fire counts or relatively small numbers of fire counts were recorded in Alaska, East Asia and North America (Figure S3a). During this period (13 November to 1 December), most of the air masses that originated within Alaska did not pass through fire-detected regions (Figure S3a), indicating that domestic burning in central Alaska is the main source for BB tracers detected during the cold season. Strong inversions occurred during these two pollution episodes (Table S1), which may enhance the concentration level of BB tracers. Dehydroabietic acid is more abundant in winter, although its average concentration is 9 times lower than that of levoglucosan in Alaskan samples. The residence time of dehydroabietic acid (2.3-4.4 days) is somewhat longer than levoglucosan (1.2-3.9 days) under the same environmental conditions (Lai et al., 2014, 2015).

BB tracers are derived from open burning such as forest fire, as well as burning of agricultural wastes, lignite, etc., during warmer seasons. Backward trajectories in summer and spring reveal that air masses mainly originate from the oceanic region while fire counts were larger within the Alaskan region (Figures S1 and S2). High levels of levoglucosan were detected from 5 June 2009 to 4 July 2009 (Figure 3e) when most of the air masses came from the Pacific Ocean and more fire counts were observed near the sampling site in Alaska (Figure S3b). Air masses from the ocean are expected to bring clean air without any contribution of BB products, suggesting that open burning in the boreal forest in Alaska is the major source for BB tracers in the Fairbanks aerosols in summer and spring. However, lower concentrations of BB tracers were observed in these seasons even though more fire counts were recorded compared to winter. Two possible processes may explain this result: (1) more deposition and/or higher atmospheric mixing, and (2) photodegradation of BB tracers in the warmer season.

Wet deposition is one parameter that reduces the atmospheric levels of BB tracers (Hu et al., 2013). However, wet deposition alone cannot explain such an obvious seasonal variation of

355 BB tracers in central Alaska because we found a high level of levoglucosan (146 ng m^{-3}) with
356 more rainfall (1.4 mm) during the sampling period of 5-6 June 2009 while a low level of
357 levoglucosan (98.3 ng m^{-3}) was observed with less rainfall (1.1 mm) on 25 June-4 July 2009.
358 Moreover, we found greater atmospheric mixing in summer due to the higher PBLHs (daily
359 average: 338-1264 m) compared to winter (32-744 m), which may be the reason for lower
360 loadings of BB tracers in summer. Levoglucosan can be degraded by OH radicals (Hennigan et
361 al., 2010) to result in a large underestimation of summertime BB emission, which could explain
362 lower levels of BB tracers in summer and spring. The OH concentrations are expected to be
363 highest in summer due to higher UV radiation flux, ambient temperature, and relative humidity
364 (Stone et al., 2012), which would enhance the photochemical degradation of levoglucosan
365 (Hennigan et al., 2010; Hoffmann et al., 2010; Mochida et al., 2010). This may be another reason
366 for the lower levels of BB tracers in summer and spring. Furthermore, residential wood burning
367 that occurs substantially in the cold season compared to summer may be another reason for the
368 higher levels of BB tracers in winter. The sources of BB tracers in the central Alaskan aerosols
369 are further interpreted in section 3.2.

370 **3.1.3. Characteristics of burning substrates**

371 The combustion of lignite that contains fossilized cellulose also emits a substantial
372 proportion of levoglucosan, whereas the emission of its isomer (mannosan) is limited (Fabbri et
373 al., 2008, 2009; Kuo et al., 2011). Thus, levoglucosan/mannosan (L/M) ratios may provide
374 crucial information to differentiate the contributions of biomass and lignite burning to the
375 atmosphere. We calculated L/M ratios to distinguish a possible source difference of BB tracers in
376 the Fairbanks aerosol samples. The L/M ratios in this study ranged from 2.3 to 7.8, which are
377 much lower than those reported for lignite combustion (30-40) (Kuo et al., 2011), suggesting that
378 lignite burning is not an important contributor to aerosol in Interior Alaska.

379 Because hardwood contains a higher amount of cellulose (55-65%) compared to
380 hemicellulose (20-30%) (Klemm et al., 2005; Sjostrom, 1993), high levels of levoglucosan are
381 mainly derived by thermal decomposition of hardwood. Hence, higher L/M ratios indicate
382 hardwood burning, whereas lower ratios indicate more contribution from softwood. The L/M
383 ratios can be used to distinguish softwood and hardwood combustions (Kuo et al., 2011),
384 although L/M ratios in this study did not exhibit any seasonal difference (Figure 6a). In a
385 chamber experiment, Schmidl et al. (2008) observed L/M ratios of 3-5 for softwood and 14-15
386 for hardwood burning. The L/M ratios of 3-5 for softwood, >10 for hardwood, and up to 50 for
387 some herbaceous plants were reported by Engling et al. (2009). Previous BB studies also
388 reported different L/M ratios for various burning substrates, e.g., >40 for crop residues (Zhu et

al., 2015), >100 for Africa savannah grass (Engling et al., 2006, 2009), 40-46 for wheat straw (Fu et al., 2008), 42 for rice straw (Sheesley et al., 2003), and 56 for cereal straw (Zhang et al., 2007). Fine et al. (2004) and Engling et al. (2006) reported L/M ratios of 3-7 from conifer trees (softwood). The lower L/M ratios in Fairbanks aerosols are similar to those of softwood burning, implying a predominance of softwood burning in central Alaska.

The levoglucosan/dehydroabietic acid (L/DHAA) ratios can also be used to distinguish softwood from hardwood burning (Kuo et al., 2011; Leithead et al., 2006). During the cold season, paper birch (*Betula papyrifera*) (deciduous tree) and white spruce (*Picea glauca*) (conifer tree) are frequently used for woodstove burning in central Alaska. These softwood species occur widely in the Alaskan boreal forest, suggesting that softwoods may be dominant in both forest fire and domestic wood burning in central Alaska. Significant levels of DHAA (avg. 7.4 ng m^{-3}) were observed in the present study. Fine et al. (2001) reported L/DHAA ratios of 6.7-60.6 for softwood burning, which is consistent with the ratios of 4.1-47.5 obtained in this study, again indicating dominant softwood burning in the central Alaskan region. Based on the above discussions, we hypothesize that BB tracers in the central Alaskan aerosols are mainly derived from softwood burning, being consistent with lower L/M ratios as discussed above.

3.2. Source regions of BB for Fairbanks aerosols

Saunders et al. (2013) pointed out a strong sensitivity of back trajectories against starting elevations. Thus, we evaluated the sensitivity of back trajectories at the different starting heights of 50, 100, 500 and 1000 m above the ground in Fairbanks (Figure S4). We found that horizontal separations of the trajectories did not show a significant difference with different starting heights. These results suggest that estimations of BB source regions were not affected by the sensitivity of back trajectories due to the starting height in Fairbanks. Forest fires frequently occur in Alaska with averaged annual burned areas of 0.4 M ha (Soja et al., 2007), although the total burned area in 2008 and 2009 was 0.16 M ha (https://fire.ak.blm.gov). Backward trajectory analyses showed a mixed air mass origin for all seasons at the sampling site (see Figure S1). Most air masses come with lower heights and shorter transport distances, suggesting a small probability of long-range atmospheric transport to Fairbanks. Hence, current backward trajectories demonstrate that there are no significant air mass contributions to the sampling site from East Asia and North America, where dense fire spots were observed throughout the campaign (Figure S1).

Monthly fire counts detected in Alaska (140-180°W; 50-75°N) were highest in June 2009 (total counts 1026), followed by July 2008 (697), which is not consistent with two peaks of levoglucosan in November 2008 (Figure 3e). In contrast, no fire counts were recorded around Alaska from October 2008 to March 2009, although higher levels of BB tracers were observed.

This result suggests that indoor burning is a likely source of BB tracers in Fairbanks, which cannot be detected by satellite (Yan et al., 2006). The weather of Alaska is very cold from October to March (monthly average: -22°C to -4°C). Backward trajectory clusters indicate that 50% of the air masses in winter originated regionally with very short distance transport within Alaska, whereas the remaining air masses had long-range transport from oceanic regions (Figure 2). Interestingly, levoglucosan showed no correlation with fire counts in East Asia and western North America in autumn/winter, suggesting that high levels of BB tracers in those seasons were associated with residential wood burning in central Alaska. Fairbanks is located at the bottom of the Tanana Valley, where stagnant air is present in the dark winter, further supporting that domestic burning is an important source of BB in Fairbanks during the cold season.

In contrast, backward trajectory clusters in summer and spring indicate that almost all air masses originated from oceanic regions when dense fire spots were observed in East Asia and North America together with the Alaskan region (Figure 2). A significant correlation ($r = 0.68$, $p<0.001$) was obtained between levoglucosan and fire counts in Alaska (140-180°W; 50-80°N) during the late spring and summer seasons (Figure S5), although the levels of levoglucosan could be decreased due to photochemical oxidation. Thus, forest fires significantly contribute to central Alaskan aerosols in summer and spring.

3.3. Relations among levoglucosan, inorganic ions, and carbonaceous components

BB is an important source of reactive nitrogen, which can produce NH_3 and NO_x in the atmosphere (Goode et al., 2000). Nance et al. (1993) reported nitrogen-containing trace gases, e.g., N_2O, NH_3, and NO_x from the burning of Alaskan black spruce. NH_4^+ could be formed by the protonation of NH_3 in the atmosphere, which is abundantly emitted from BB, livestock excreta, agricultural soil and plants, and fertilizer application, etc. (Huang et al., 2012; Paulot et al., 2014; Sutton et al., 2008). A positive correlation ($r = 0.65$, $p<0.001$) was observed between levoglucosan and NH_4^+ (Figure S6a), indicating that BB could also be an important source of NH_4^+ (Andreae & Merlet, 2001; Akagi et al., 2011). Levoglucosan showed a significant correlation ($r = 0.82$, $p<0.001$) with NO_3^- (Figure S6b), suggesting large emission of NO_x during BB activities over central Alaska. Nevertheless, NO_x can also be derived from fossil fuel combustion and automobiles (Agarwal et al., 2010; Reddy et al., 2012).

Levoglucosan showed a significant correlation with OC ($r = 0.51$, $p<0.001$), WSOC ($r = 0.61$, $p<0.001$) and EC ($r = 0.67$, $p<0.001$), indicating that BB is an important source for carbonaceous components (Figure 7). A good correlation of WSOC with levoglucosan implies that WSOC in the central Alaskan aerosols is substantially derived from BB. Mkoma et al. (2013) reported that the main contributors to WSOC are probably BB and secondary organic

aerosols due to their preferential association with the fine fraction. BB is frequently accompanied by abundant emissions of EC (Akagi et al., 2011). Other sources of EC are coal combustion and motor vehicle exhaust (Zhu et al., 2015). We found a significant correlation between levoglucosan and EC, indicating that BB may be an important source of EC in Fairbanks. The large carbon preference index (CPI) for *n*-alkanes (avg. 7.04) in Fairbanks aerosols (Boreddy et al., 2018) indicates that *n*-alkanes are mainly emitted from higher plant waxes rather than petroleum residues (Fu et al., 2012a, 2013). Furthermore, PAHs and hopanes, which are specific tracers for fossil fuel combustion (Fu et al., 2008), were not detected in Fairbanks aerosols. We thus assume that the emission of EC was more influenced by BB than fossil fuel combustion in central Alaska. The concentration ratios of levoglucosan to EC and OC (L/EC and L/OC) exhibited similar temporal variations with levoglucosan. We found higher concentration ratios in winter (Figures 3e and 6b-c), suggesting that BB activity largely contributes to carbonaceous aerosols in Fairbanks.

3.4. Underestimation of nss-K^+ from BB emission

nss-K^+ in the Alaskan aerosols peaked in spring (Table 1). nss-K^+ is considered as a typical tracer of BB (Sullivan et al., 2011). However, we discovered that nss-K^+ showed no correlation with levoglucosan (r = -0.17) and fire counts (r = -0.04) in Fairbanks. Zhang et al. (2010) pointed out that K^+ has additional sources other than BB, including soil dust, vegetation, and meat cooking. Previous studies also reported that soil dust could be another source of nss-K^+ (Arimoto et al., 2004; Duvall et al., 2008; Shen et al., 2009). In summer and spring, these different sources of nss-K^+ may be possible in central Alaska. However, additional emission of nss-K^+ from non-BB sources is not likely in winter and autumn due to the snow coverage over Fairbanks. The emission of K^+ from BB depends on burning conditions (Schkolnik et al., 2005) and types of burning materials (McMeeking et al., 2009). Smoldering combustion produces more organic compounds compared to the flaming process, yielding more levoglucosan in comparison to nss-K^+ (Cheng et al., 2013). Sullivan et al. (2008) noted that there is no overall correlation between levoglucosan and nss-K^+ in ambient aerosols from the upper Midwest.

Here, we propose a hypothesis to better explain the underestimated emission of nss-K^+ from BB in the winter season; that is, K^+ produced by woodstove burning can be deposited on the inner surface of a chimney due to a rapid decrease of the chimney temperature with height. Potassium and silicon are abundantly present in woody biomass (Paneru et al., 2016). During the burning of woody materials in the stove, K can react with Si to produce potassium silicate (K_2O_3Si) in the liquid phase that can deposit on the chimney wall (Paneru et al., 2016), which could lead to underestimating the contribution of nss-K^+ to wintertime ambient aerosols. In

contrast, we presume that unburned organics such as anhydrosugars, lignin, and resin products from wood burning can be emitted to the atmosphere through the woodstove chimney because no correlation was found between levoglucosan and nss-K^+ in the Fairbanks aerosols as discussed earlier.

To test the above hypothesis, we analyzed chimney deposits for major ions and organic tracers. We found that K^+ in the deposits showed the highest value (51%) relative to the mass abundance of total cations. Concentrations of K^+ and concentration ratios of K^+/levoglucosan both decrease with an increase of chimney heights (Figure S7). Substantial amounts of K^+ were found in the lower deposit samples (10-80 cm), but concentrations drastically decreased with height. Interestingly, K^+/levoglucosan ratios showed a substantial increase near the bottom of the chimney (10-80 cm). This result suggests that K^+ was preferentially deposited on the wall of the chimney near the stovetop. Similar deposition of K^+ is known to occur in a biomass boiler (Paneru et al., 2016). In contrast, concentrations of levoglucosan in the chimney deposits increased with height (Figure S7c), suggesting that levoglucosan is efficiently evaporated in the chimney near the stovetop but largely deposited with height when the temperature declined.

It is of interest to note that K^+/levoglucosan ratios were more than one order of magnitude lower in the dark winter (0.19) than in the summer (5.61) in the ambient aerosols from Fairbanks. These results demonstrate that K^+ in Fairbanks aerosols mainly originated from woodstove burning in which K^+ was potentially scavenged on the inner surface of the chimney, resulting in a wall loss of K^+ and its lower concentrations in the ambient aerosols in winter. Hence, we conclude that domestic (woodstove) burning is a source of significant removal of nss-K^+ in BB emissions from central Alaska in the dark winter, which has not been considered before.

3.5. Contributions of BB to WSOC, OC and PM

Levoglucosan-carbon to OC (L/OC) ratios have been used to evaluate the contribution of different BB sources (Sullivan et al., 2008; Mkoma et al., 2013) and possible aging of levoglucosan (Mochida et al., 2010). The contribution of BB to OC could be estimated based on BB tracers such as levoglucosan because levoglucosan is the most abundant BB-derived compound. Previous studies often used L/OC ratios to evaluate the contribution of BB to OC (Zdrahal et al., 2002; Puxbaum et al., 2007; Zhang et al., 2008; Zhang et al., 2012; Reche et al., 2012; Ho et al., 2014). We examined the contributions of BB to WSOC and OC year-round and found a clear winter maximum of L/OC in Fairbanks (Figure 6c). L/OC ratios are different depending on biomass types; branches > straw > needles > leaves, etc. (Sullivan et al., 2008). In the Fairbanks aerosols, we observed a higher L/OC ratio in winter than the growing season

525 (Figure 6c), which is reasonable because forest fire occurs in spring/summer where branches
526 burn together with leaves. In contrast, branches and/or woods are used for domestic heating in
527 the cold season.

528 We further calculated the contributions of levoglucosan-carbon to WSOC and OC in
529 Fairbanks aerosols. In this study, levoglucosan annually accounted for 0.1-9.2% (avg. 2.9%) of
530 WSOC and 0.03-4.9% (avg. 1.6%) of OC, which are higher than those reported from the Pearl
531 River Delta sites that are directly influenced by BB in southern China (levoglucosan-C/WSOC,
532 0.59-3.12% (avg. 1.50%) (Ho et al., 2014). The largest contributions of levoglucosan to WSOC
533 (8.4%) and OC (4.4%) in Fairbanks were observed during the period of 21 November to 1
534 December 2008, when high levels of levoglucosan were measured.

535 Moreover, higher loadings of levoglucosan (146 ng m^{-3}) and higher contributions to
536 WSOC (4.5%) and OC (2.0%) were observed on 5-12 June 2009. The highest contribution of
537 levoglucosan to OC was found in winter (3.6%), followed by autumn (3.2%), summer (0.6%),
538 and spring (0.5%). The high contributions of BB to OC are comparable to previous studies from
539 various sites in the world (Andreae & Merlet, 2001; Zhang et al., 2008, 2012; Reche et al., 2012).
540 The contributions of levoglucosan to WSOC also exhibited similar seasonal variations; highest in
541 winter (6.3%) followed by autumn (5.6%), summer (1.3%), and spring (0.8%). Furthermore, we
542 calculated the contributions of other BB tracers (galactosan, mannosan, and dehydroabietic,
543 vanillic, syringic, and 4-hydroxybenzoic acids) to OC and WSOC, following the method of
544 L/OC ratios. The contributions of BB tracers to OC and WSOC also exhibited similar seasonal
545 patterns with the predominance in winter (OC: 6.3%, WSOC: 10.7%) followed by autumn (5.1%,
546 9.0%), summer (0.9%, 2.0%) and spring (0.8%, 1.2%, respectively). These results suggest that
547 BB emission has a strong influence on the chemical compositions of Fairbanks aerosols during
548 the cold seasons. The lower contributions of BB to OC in summer and spring suggest that other
549 organic sources probably contribute more to organic aerosol during the growing season. This
550 point is consistent with the higher concentrations of biogenic SOA tracers in summer and spring
551 (Haque et al., 2016).

552 We have collected $PM_{2.5}$ data from the US Environmental Protection Agency (EPA)
553 archive. The $PM_{2.5}$ level in Fairbanks (annual mean 12.8 μg m^{-3}) is higher than National Ambient
554 Air Quality Standards set by EPA (12.0 μg m^{-3}), particularly in winter (27.7 μg m^{-3}), implying
555 that the air quality of Fairbanks is a health risk. The levels in Fairbanks are higher than those
556 from other Alaskan cities such as Anchorage (5.2 μg m^{-3}) and Juneau (7.1 μg m^{-3}), suggesting
557 more air pollution in Fairbanks. The $PM_{2.5}$ level of Fairbanks is also comparable with those from
558 different important cities in the USA (Table 3). Levoglucosan showed a significant correlation

559 with $PM_{2.5}$ ($r = 0.84$, $p<0.001$) in central Alaska (Figure 7d). Hence, we propose that BB is a
560 significant source of carbonaceous aerosols and pollutants in Fairbanks, especially in winter.

561 **3.6. Implications from PMF analysis**

562 The detected organic tracers (total 24 species) along with BB tracers were subjected to
563 PMF analysis to constrain their source classifications (Figure S8). OC, EC and some ionic
564 species (total 7 species) were also used to investigate possible sources. Figure S8 shows
565 composition profiles for the four factors resolved by PMF. Factor 1 is abundantly loaded with
566 arabitol (45%), mannitol (33%), fructose (39%), sucrose (34%), and methanesulfonic acid (43%),
567 indicating that chemical species derived from microbial activities (Yttri et al., 2007; Jia et al.,
568 2010; Fu et al., 2012b). It is of interest to note that methanesulfonic acid could be derived from
569 the forest (Yi et al., 2010; Miyazaki et al., 2012; Pavuluri et al., 2015). It should be noted that
570 mannitol and sucrose also showed loadings in Factor 3, implying that vegetation can partly
571 contribute to these sugar compounds. Factor 2 is characterized by levoglucosan (80%), xylose
572 (84%), erythritol (84%), vanillic acid (89%), dehydroabietic acid (86%), *β*-caryophyllinic acid
573 (68%), and NO_3^- (89%), suggesting that the components are associated with BB. Factor 3 is
574 characterized by 2-methyltetrols (81%), C_5-alkenetriols (80%), 3-methyl-1,2,3-
575 butanetricarboxylic acid (MBTCA) (56%), trehalose (63%), mannitol (56%), sucrose (55%), and
576 glucose (53%), which are considered as tracers of biogenic emission. This factor is associated
577 with primary sugars, sugar alcohols, and biogenic SOA tracers. Factor 4 is dominated by
578 inorganic ions, probably originating from soil dust and some anthropogenic activities.

579 As resolved by PMF analysis, the average contributions of each factor to the measured
580 ambient OC are shown in Figure 8. The emission of BB (Factor 2) was found to be the dominant
581 contributor (47.5%) to OC, followed by vegetation (Factor 3: 23.9%), soil dust (Factor 4: 15.6%),
582 and microbial sources (Factor 1: 13.1%) during the study period. These results are comparable
583 with high loadings of levoglucosan and significant relation between carbonaceous components
584 and levoglucosan as discussed above. It can be noted that the contribution of BB is evaluated
585 based on tracers in the PMF analysis, although fossil fuel combustion (another important primary
586 source) was found to be minor as discussed in section 3.3. Based on the PMF results, we
587 conclude that BB was a major source for atmospheric aerosols in central Alaska during the study
588 period.

589 **4. Summary and Conclusions**

590 We report three anhydrosugars and lignin and resin products as BB tracers in ambient
591 aerosols from central Alaska. Levoglucosan was observed as the dominant species (avg. 66.7 ng
592 m^{-3}) among the measured BB tracers. The concentrations of BB tracers increased dramatically in

593 the cold seasons, being 4 times higher than those of boreal fire seasons and reflecting a large
594 contribution of BB to organic aerosols in central Alaska. Relatively low L/M ratios (2.3-7.8) and
595 high L/DHAA ratios (4.1-47.5), indicating that BB tracers in the Fairbanks aerosols are mainly
596 from the burning of softwoods.

597 During the cold seasons, levoglucosan shows no correlation with fire counts in Siberian
598 and North American regions when no fire counts were recorded in Alaska, suggesting that BB
599 pollutants in winter and autumn were mainly from domestic wood burning. However, a
600 significant correlation of levoglucosan with fire spots was detected in the Alaskan region during
601 summer and late spring, attributing regional forest fires as important sources of organic aerosols
602 during the growing season. A strong relationship between levoglucosan and NH_3/NO_x
603 demonstrates that nitrogenous aerosols are substantially influenced by BB emissions.

604 Our results reveal that local BB emissions from domestic burning with a stagnant air
605 condition largely contribute to carbonaceous aerosols and PM pollutants over central Alaska,
606 particularly in the dark winter. Considering the measured components in this study, PMF
607 analysis demonstrated that BB contributed almost 50% to OC. We proposed a hypothetical
608 removal of K^+ from the BB products via the deposition on the wall of woodstove chimneys,
609 resulting in an underestimation of nss-K^+ in the Fairbanks aerosols.

610 Our findings draw an actual picture of BB that would help to understand the effect of BB
611 over central Alaska. Previous investigations considered Fairbanks an urban source of Arctic air
612 pollution, possibly because Fairbanks aerosols can transport to the remote Arctic. This study
613 identified and quantified the molecular composition and source contribution of organic aerosols
614 over Fairbanks, which would be valuable to evaluate the air quality and climate changes in the
615 Arctic and subarctic atmosphere.

616 **Data Availability Statement**

617 The data used in this manuscript are listed as tables, figures, and supporting information.
618 The data archive is available at https://data.mendeley.com/datasets/publish-
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Table 1. Concentrations of anhydrosugars, lignin and resin acids, nss-K^+ (ng m^{-3}) and carbonaceous components (μg m^{-3}) with some ratios in Fairbanks aerosols.

Species	Annual	Summer*	Autumn	Winter	Spring
	Avg.[a] ± S.D.[b] Min.[c], Max.[d]	Avg. ± S.D. Min., Max.	Avg. ± S.D. Min., Max.	Avg. ± S.D. Min., Max.	Avg. ± S.D. Min., Max.
		Anhydrosugars			
Levoglucosan (L)	67 ± 77 1.1, 273	34 ± 45 1.1, 146	129 ± 80 25, 273	145 ± 47 95, 214	12 ± 12 1.4, 36
Mannosan (M)	15 ± 19 0.2, 63	7.1 ± 9.6 0.4, 31	28 ± 18 4.0, 63	36 ± 15 17, 54	2.1 ± 2.2 0.2, 6.3

Galactosan	9.9 ± 12 0.1, 39	4.7 ± 7.2 0.1, 25	18 ± 12 2.0, 39	24 ± 11 11, 36	1.3 ± 1.5 0.1, 3.9
			Lignin & resin acids		
Dehydroabietic acid	7.4 ± 12 BDL[e], 43	1.3 ± 2.0 0.3, 7.5	16 ± 16 1.2, 43	21 ± 14 2.7, 40	0.6 ± 0.5 BDL, 1.3
Vanillic acid	2.3 ± 3.8 BDL, 16	0.4 ± 0.7 0.1, 2.5	4.1 ± 3.8 0.2, 9.7	7.7 ± 5.5 1.3, 16	0.3 ± 0.2 BDL, 0.5
Syringic acid	0.7 ± 1.4 BDL, 6.5	0.1 ± 0.1 BDL, 0.3	1.1 ± 1.3 0.03, 2.9	2.4 ± 2.5 0.2, 6.5	0.04 ± 0.1 BDL, 0.1
4-Hydroxybenzoic acid	1.7 ± 1.2 0.4, 5.6	1.2 ± 0.8 0.38, 2.7	2.1 ± 1.2 0.6, 3.4	3.3 ± 1.5 1.9, 5.6	1.1 ± 0.6 0.4, 2.2
			Carbonaceous components and ion		
nss-K^+	67 ± 80 22, 491	55 ± 18 29, 91	37 ± 17 22, 66	53 ± 13 44, 76	131 ± 161 40, 491
OC[f]	1.7 ± 0.8 0.6, 3.3	1.9 ± 0.7 1.1, 3.3	1.6 ± 0.8 0.8, 2.8	1.9 ± 0.7 1.0, 2.9	1.5 ± 1.0 0.6, 3.3
EC[g]	0.4 ± 0.2 0.2, 1.0	0.3 ± 0.1 0.2, 0.4	0.5 ± 0.3 0.3, 1.0	0.5 ± 0.2 0.3, 0.9	0.4 ± 0.2 0.2, 0.7
WSOC[h]	0.9 ± 0.5 0.3, 1.9	0.8 ± 0.4 0.4, 1.7	1.0 ± 0.6 0.4, 2.0	1.1 ± 0.5 0.6, 1.7	0.9 ± 0.5 0.3, 1.8
			Ratios		
L/M	5.2 ± 1.4 2.3, 7.8	5.0 ± 1.6 2.3, 7.8	5.1 ± 0.9 3.9, 6.1	4.3 ± 1.1 2.6, 5.6	6.3 ± 0.8 5.0, 7.3
L/OC x 10^{-2}	1.6 ± 1.6 0.03, 4.9	0.6 ± 0.6 0.03, 1.9	3.2 ± 1.2 1.0, 4.4	3.6 ± 1.0 2.7, 4.9	0.5 ± 0.6 0.1, 1.8
L/EC x 10^{-2}	6.3 ± 5.9 0.2, 19	4.3 ± 5.6 0.2, 19	9.8 ± 4.5 2.8, 16	13 ± 4.4 6.5, 19	1.6 ± 1.7 0.2, 4.7

[a]Average, [b]Standard deviation, [c]Minimum, [d]Maximum, [e]Below detection limit, [f]Organic carbon, [g]Elemental carbon, [h]Water-soluble organic carbon.

[*] Seasons are divided as June-August (summer), September-November (autumn), December-February (winter), March-May (spring).

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1049 **Table 2.** Comparisons of the concentration (ng m^{-3}) of levoglucosan in Fairbanks aerosols with those
1050 from different sites in the Arctic and other regions.

Location	Sample type	Sampling time	Levoglucosan	References
Alaska, USA	**TSP[a]**	Summer	1.0-146 (Mean: 34.2)	**This study**
		Autumn	24.8-273 (129)	
		Winter	95.1-214 (145)	
		Spring	1.41-36.1 (12.3)	
Alert, Canada	TSP	Spring	0.003-1.08 (0.09)	Fu et al., 2009

Svalbard, Norway	PM_{10}	Summer Winter	0.13 (Mean) 1.02	Yttri et al., 2014
Beaufort Sea, Canadian Arctic	TSP	Summer	0.01-0.93 (0.37)	Fu et al., 2013
Northern Hemisphere	TSP	Summer	1.1-41 (6.5)	Hu et al., 2013
Texas, USA	PM_{10}	May	200-1200 (600)	Fraser & Lakshmanan, 2000
Southeast Asia	TSP	Winter	1.2-4.3 (2.8)	Hu et al., 2013
Rondonia, Brazil (Forest site)	$PM_{2.5}$	Autumn	39.9-2660 (1180)	Graham et al., 2002
Okinawa, Western North Pacific	TSP	Summer Winter	BDL[b]-3.89 (0.57) 0.78-27.2 (5.30)	Zhu et al., 2015
Howland Forest Maine	TSP	Summer	1.0-55.1 (18.1)	Medeiros et al., 2006
Mt. Hua, China	PM_{10}	Spring	13-106 (48)	Wang et al., 2012
Sapporo, Japan	TSP	Summer Winter	6.4 (Mean) 56	Simoneit et al., 2004a
Gosan, Jeju Island, Korea	TSP	Spring	2.8-102 (36.3)	Wang et al., 2009

[a]Total suspended particle, [b]Below detection limit

Table 3. Comparisons of mean values (μg m^{-3}) of $PM_{2.5}$ in the Fairbanks atmosphere with those from other important cities in the USA (https://www.epa.gov/outdoor-air-qualitydata/download-daily-data).

City	Annual mean	Summer	Autumn	Winter	Spring
Fairbanks, Alaska (this study)	**12.8**	**4.4**	**15.6**	**27.7**	**7.9**
Anchorage, Alaska	5.2	3.5	5.2	9.1	5.2
Juneau, Alaska	7.1	3.8	6.0	14.1	5.6
Albany, New York	9.3	11.0	7.9	10.9	7.3

Cook, Illinois	12.0	12.6	12.4	13.8	9.3
Galveston, Texas	11.0	11.1	13.4	9.1	10.4
Maricopa, Arizona	10.3	8.3	11.0	14.1	7.9
New Castle, Delaware	13.1	16.8	10.5	14.1	10.8
San Diego, California	14.9	13.6	14.9	14.9	16.2
Dallas, Texas	10.4	11.7	11.1	8.8	10.1
Los Angeles-Long Beach-Anaheim, California	14.7	17.2	15.3	11.5	13.0
Santa Clara, California	11.7	13.2	13.5	13.5	6.4

Figure Captions

Figure 1. Map showing the sampling location of Fairbanks, AK, USA.

Figure 2. 7-day backward air mass trajectories arriving at 100 m (summer and spring) and 50 m (autumn and winter) above the ground level to Fairbanks, AK, as shown by cluster during June 2008–June 2009. The trajectories of the panel a, b, c, and d indicate the percentages of daily trajectories for summer, autumn, winter, and spring, respectively. Red color indicates the fire spots. (AO refers to Arctic Ocean, BS refers to Bering Sea, PO refers to Pacific Ocean, AR refers to Alaska Region).

1084 **Figure 3.** Time series of the concentrations of (a-d) lignin & resin acids and (e-g) anhydrosugars during
1085 the period of study in Fairbanks, AK.
1086 **Figure 4.** The relationships of levoglucosan with (a) mannosan, (b) galactosan, (c) dehydroabietic acid,
1087 (d) syringic acid, (e) vanillic acid, and (f) 4-hydroxy benzoic acid in Fairbanks aerosols.
1088 **Figure 5.** Seasonal variations of (a) anhydrosugars and (b) lignin and resin acids in Fairbanks aerosols.
1089 **Figure 6.** Temporal trends of (a) levoglucosan/mannosan (L/M), (b) levoglucosan/EC (L/EC) and (c)
1090 levoglucosan/OC (L/OC) ratios during sampling time in central Alaska.
1091 **Figure 7.** The relationship of levoglucosan with (a) OC, (b) EC, (c) WSOC and (d) $PM_{2.5}$ during the
1092 sampling period.
1093 **Figure 8.** PMF results showing annually averaged source contributions of four factors to the organic
1094 carbon (OC) in central Alaska aerosols.
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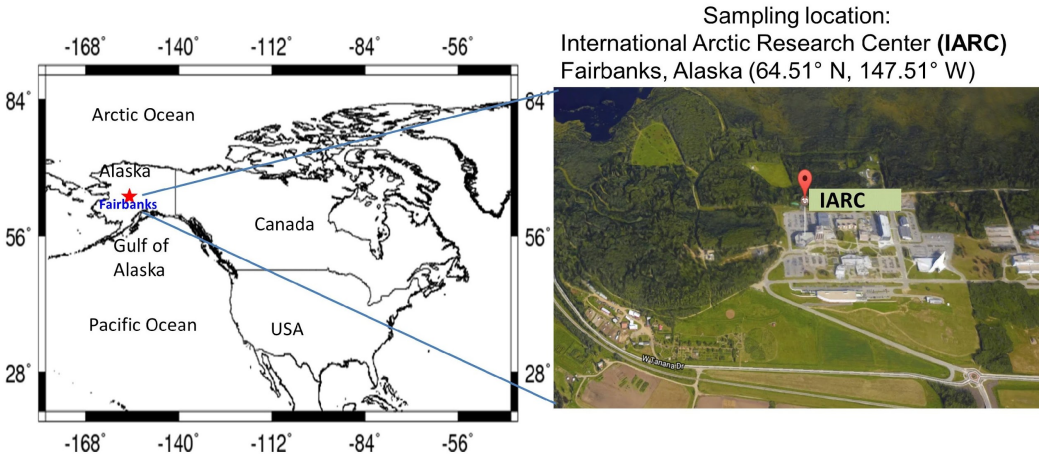

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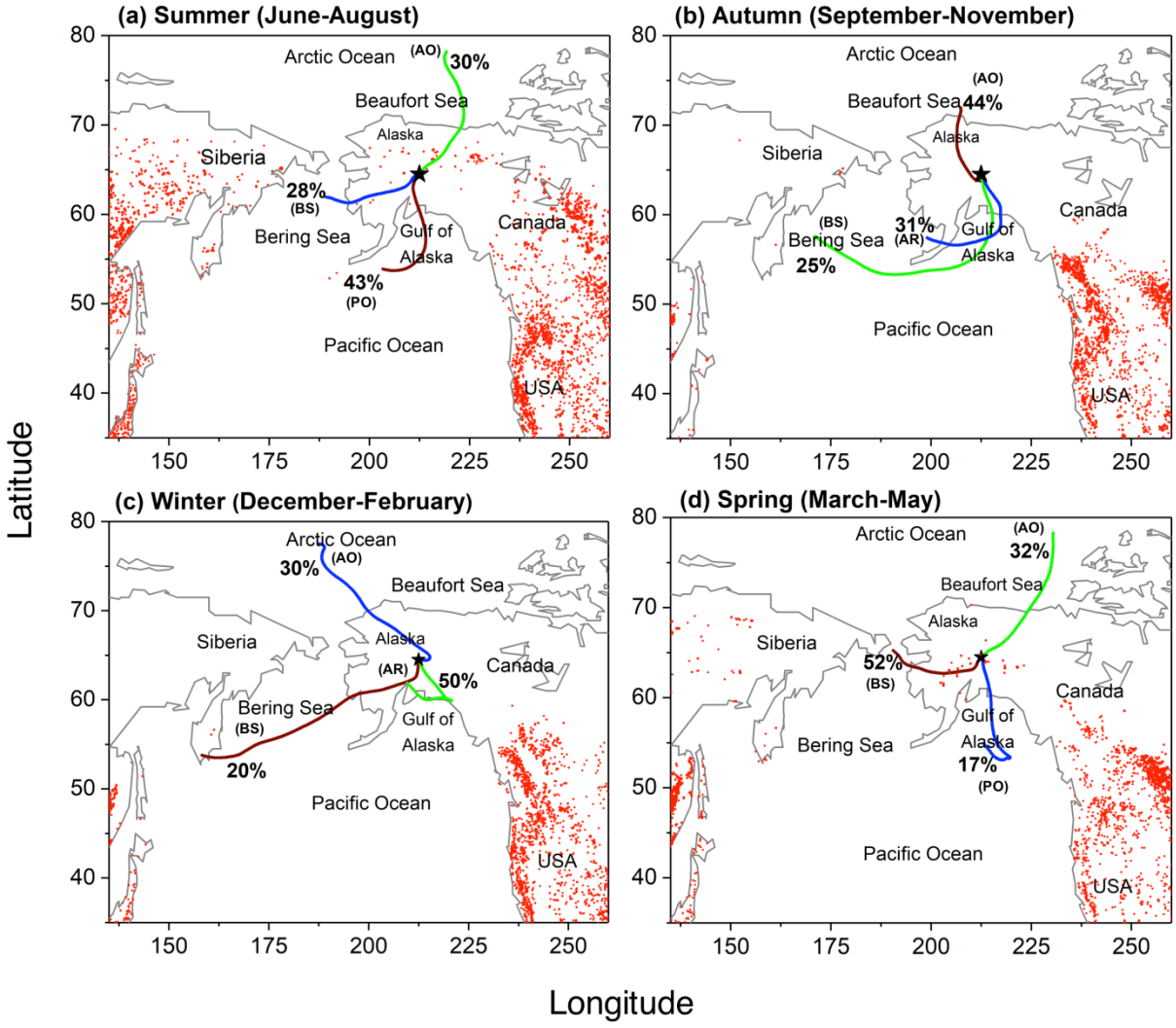

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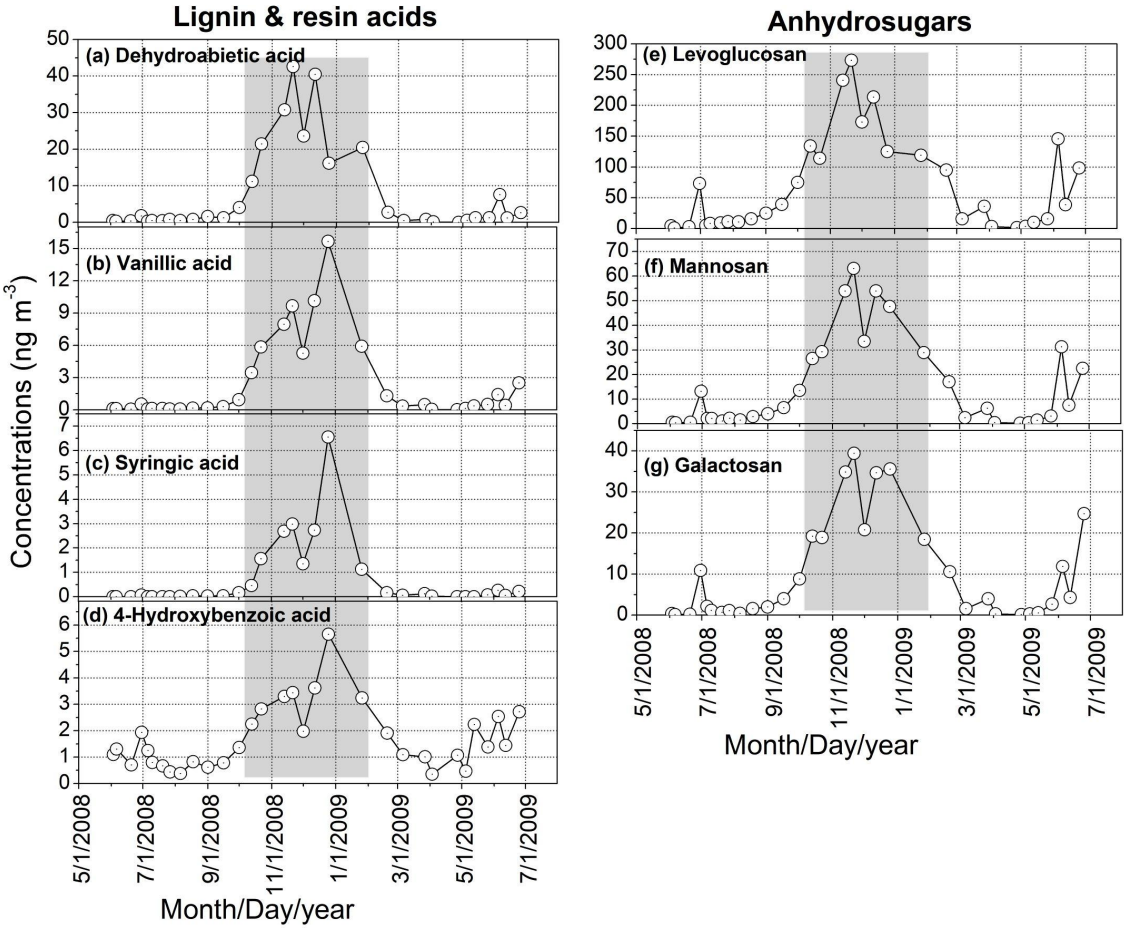

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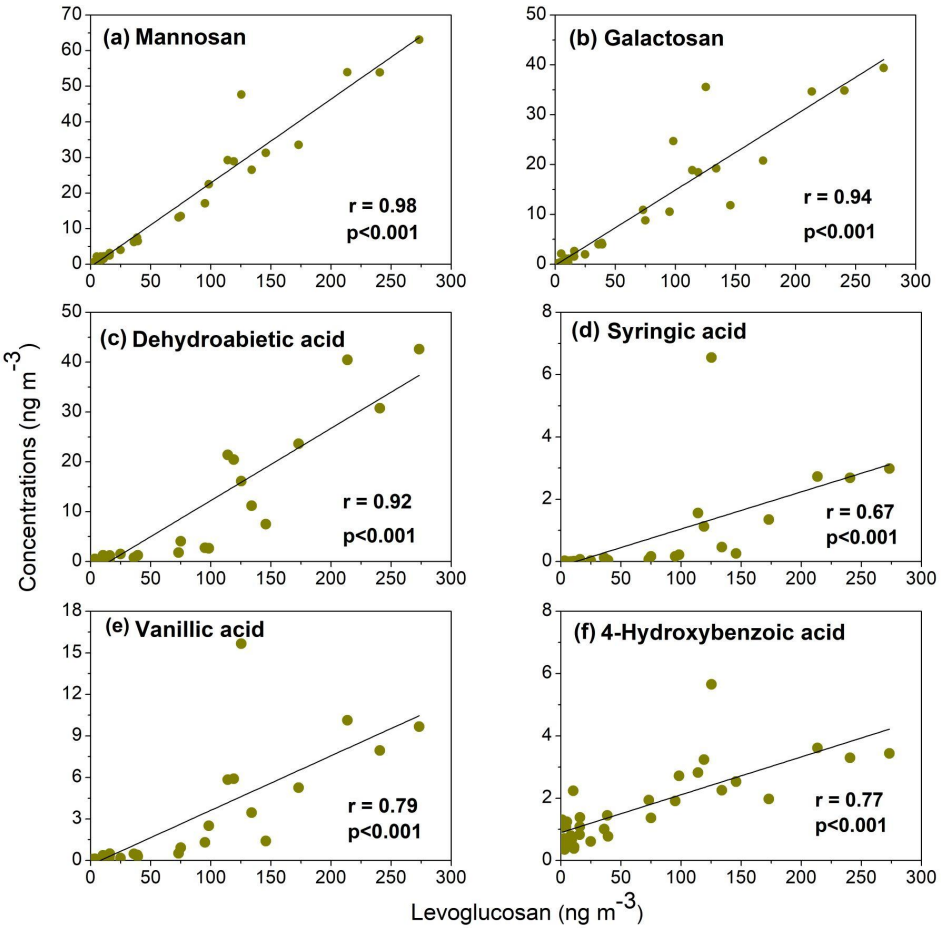

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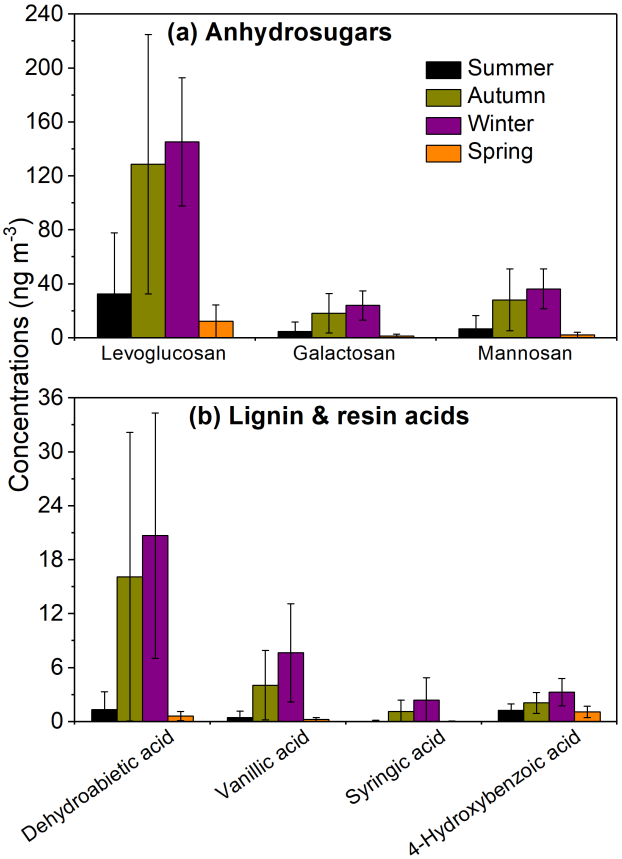

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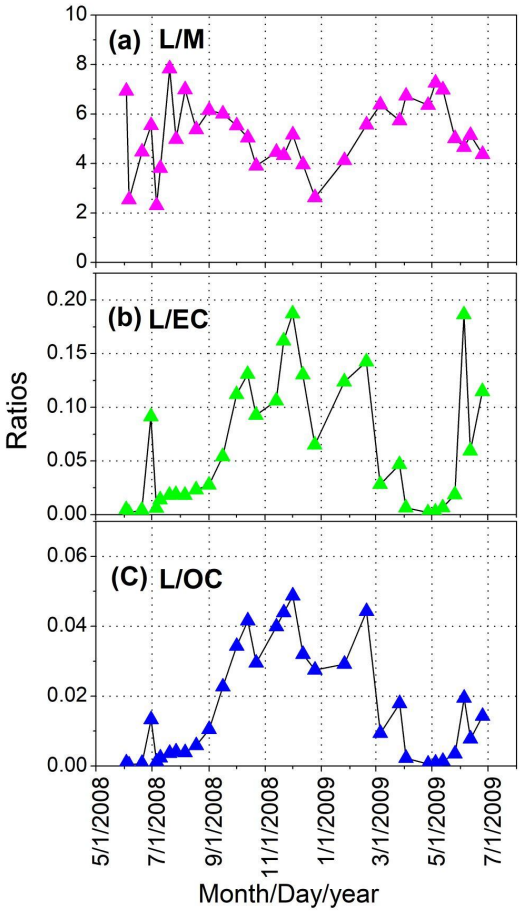

Figure 6. Temporal trends of (a) levoglucosan/mannosan (L/M), (b) levoglucosan/EC (L/EC) and (c) levoglucosan/OC (L/OC) ratios during sampling time in central Alaska.

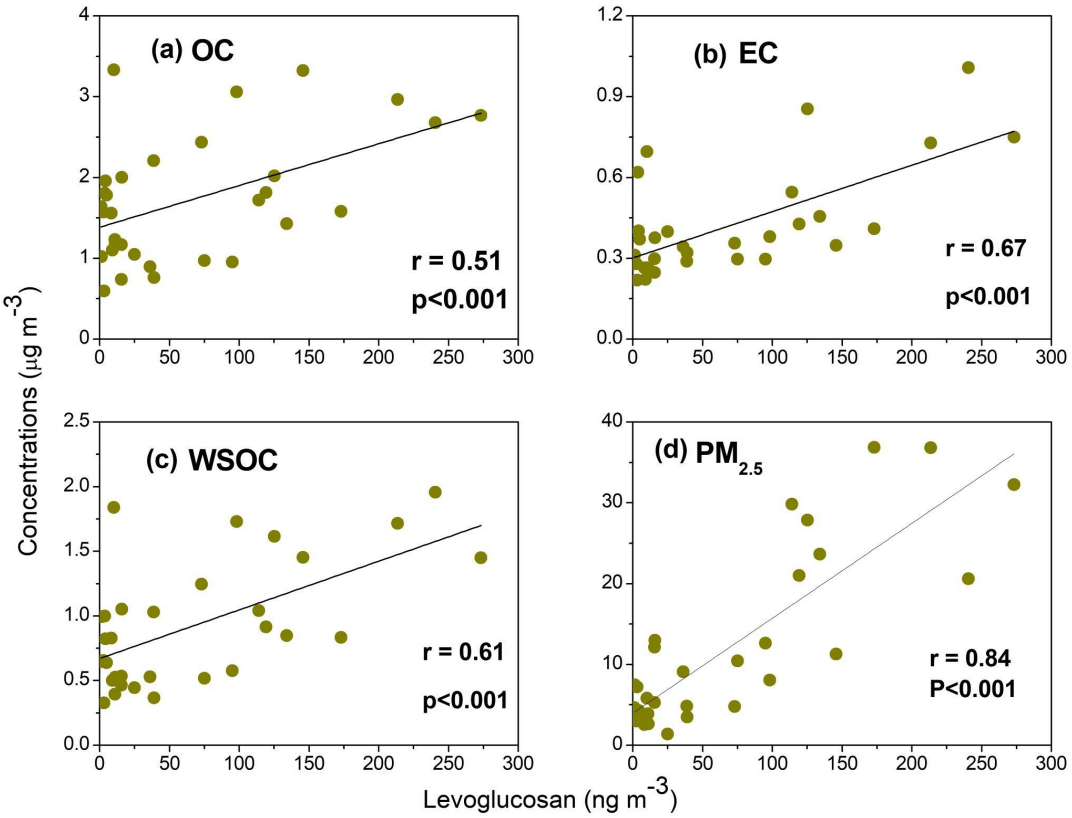

Figure 7. The relationship of levoglucosan with (a) OC, (b) EC, (c) WSOC and (d) $PM_{2.5}$ during the sampling period.

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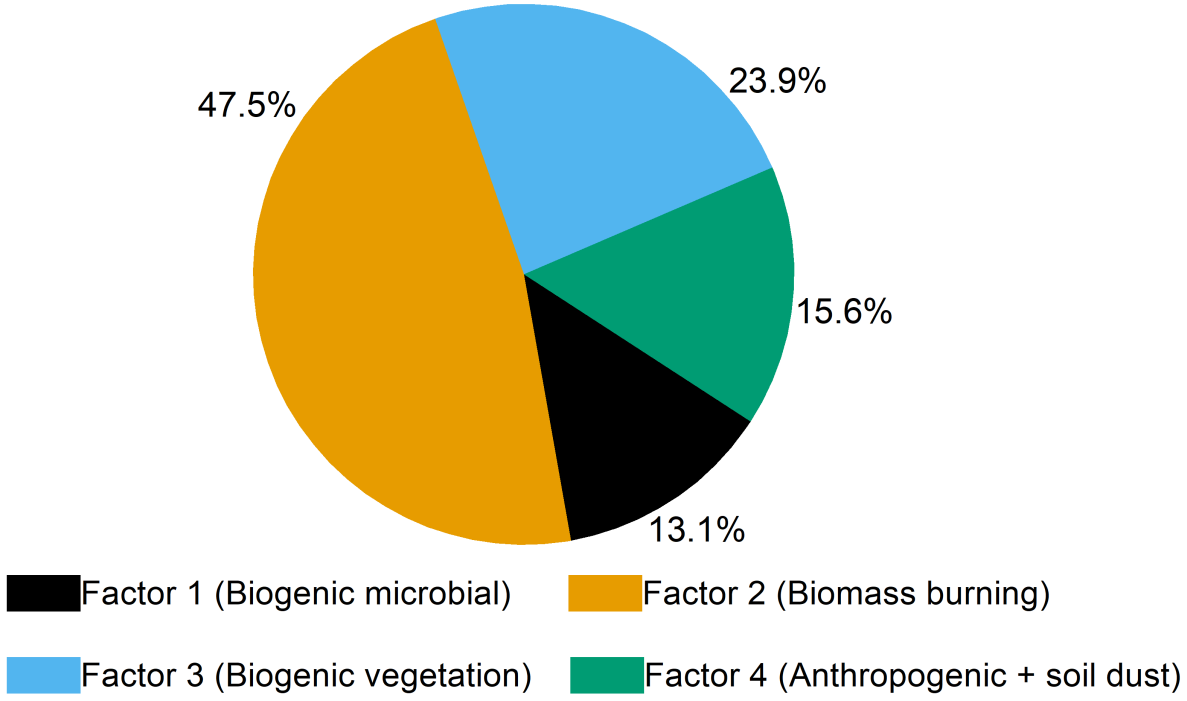

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1242 **Figure 8.** PMF results showing annually averaged source contributions of four factors to the organic
1243 carbon (OC) in central Alaska aerosols.