An optimized tracer-based approach for estimating organic carbon emissions from biomass burning in Ulaanbaatar, Mongolia

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Abstract

The impact of biomass burning (BB) on atmospheric particulate matter of <2.5 μm diameter ($PM_{2.5}$) at Ulaanbaatar, Mongolia, was investigated using an optimized tracer-based approach during winter and spring, 2017. Integrated 24 h $PM_{2.5}$ samples were collected on quartz fiber filters using a 30 L min^{-1} air sampler at an urban site in Ulaanbaatar. The aerosol samples were analyzed for organic carbon (OC) and elemental carbon (EC), anhydrosugars (levoglucosan, mannosan, and galactosan), and water-soluble ions. OC was found as the predominant species, contributing 64% and 56% to the quantified aerosol components in $PM_{2.5}$ in winter and spring, respectively. BB was identified as a major source of $PM_{2.5}$, followed by dust and secondary aerosols. Levoglucosan/mannosan and levoglucosan/K^+ ratios indicate that BB in Ulaanbaatar was mainly originated from burning of softwood. Because of the large uncertainty associated with quantitative estimation of OC emitted from BB (OC_{BB}), a novel approach was developed to optimize the OC/levoglucosan ratio for estimating OC_{BB}. The optimum OC/levoglucosan ratio in Ulaanbaatar was obtained by regression analysis between OC_{non-BB} (OC_{total}–OC_{BB}) and levoglucosan concentrations that gives the lowest coefficient of determination (R^2) and slope. The optimum OC/levoglucosan ratio was found to be 27.6 and 18.0 for winter and spring, respectively, and these values were applied in quantifying OC_{BB}. It was found that 68% and 63% of the OC were emitted from BB during winter and spring, respectively. This novel approach can also be applied to other study sites to quantify OC_{BB} using their own chemical measurements. In addition to OC_{BB}, sources of OC_{non-BB} were also investigated through multivariate correlation analysis. It was found that OC_{non-BB} was originated mainly from coal burning, vehicles, and vegetative emissions.

***Keywords*:** Source identification, Biomass burning, Optimized organic-carbon/levoglucosan ratio

1. Introduction

Organic aerosol (OA) contributes a significant fraction (10%–90%) of atmospheric particulate matter (PM), which can affect human health and air quality (Jimenez et al., 2009; Maenhaut et al., 2011; Fu et al., 2012; Allan et al., 2014; Chen et al., 2018). An understanding of the sources of PM is highly relevant for air-quality remediation. Biomass burning (BB) is a major source of organic carbon (OC) in $PM_{2.5}$ (PM with aerodynamic diameter ≤2.5 μm) and it may become more significant in the future as air-quality regulations restrict other anthropogenic emissions (Davy et al., 2011; Allan et al., 2014; Sullivan et al., 2019). Coal combustion, thermal power plants, and traffic emissions also make potential contributions to the OC content of PM (Watson et al., 2001a, b; Pei et al., 2016; Deshmukh et al., 2019; Haque et al., 2019), modifying PM characteristics such as hygroscopicity, light-attenuating properties, and health impacts (Jung et al., 2009; Sullivan et al., 2019). Previous studies have observed that the toxicity of $PM_{2.5}$ increases with the oxidation potential of BB species because of the water-soluble fraction of OC (Verma et al., 2014).

Previous studies have identified and quantified OC emitted from BB (OC_{BB}) using the BB tracers (levoglucosan, mannosan, galactosan, and K^+). Levoglucosan is produced from the pyrolysis of cellulose at temperatures of >300°C (Simoneit et al., 1999; Claeys et al., 2010; Maenhaut et al., 2011; Nirmalkar et al., 2015; Achad et al., 2018); and two isomers of levoglucosan, mannosan and galactosan are produced by the burning of hemicellulose (Reche et al., 2012). The atmospheric concentration of levoglucosan is higher than that of the two isomers because of the lower content of hemicellulose (20%–30%, dry weight) than cellulose (40%–50%) in softwood and hardwood (Reche et al., 2012; Sharma et al., 2015). Water-soluble K^+ can also be used as a BB tracer (Pio et al., 2008; Cheng et al., 2013; Nirmalkar et al., 2015; Chen et al., 2018; Chantara et al., 2019). The proportion of these BB tracers in PM depends on various factors such as the type of biomass (softwood, hardwood, crop, grass, etc.), where it is burnt (traditional stoves, fireplaces, field burning, burning in closed chambers, etc.), the type of burning (smoldering, flaming, etc.), and the burning season (Fu et al., 2012; Cheng et al., 2013; Jung et al., 2014). Levoglucosan/mannosan, levoglucosan/K^+, and OC/levoglucosan ratios were used to identify major biomass types and quantify OC_{BB} (Reche et al., 2012; Cheng et al., 2013; Jung et al., 2014; Chen et al., 2018). However, OC/levoglucosan ratios are quite variable even with the same type of BB because of variations in burning type, place, and season (Cheng et al., 2013; Thepnuan et al., 2019 and references therein). It is therefore essential to optimize the OC/levoglucosan ratio to better estimate OC_{BB}.

Ulaanbaatar, with a population of about 1 million, is an atmospheric pollution 'hotspot' because of its topography, being situated in the Tuul river valley and surrounded by the Khentei mountains, with a high elevation (1300 m−1949 m above sea level) and large variations in temperature (–28°C to +16°C) and relative humidity (17.7%–72.7%; Table 1; Batmunkh et al., 2013; Jung et al., 2014). As the world's coldest capital city during winter, it requires additional fuel for space heating. The topography and low-temperature conditions cause an increase in PM concentrations, which are exacerbated by low wind speeds and atmospheric temperature inversions (Jung et al., 2010).

Half of the residents in Ulaanbaatar lives in 160,000 Gers (traditional Mongolian dwellings) (Guttikunda and Jawahar, 2014). Biomass is used as fuel for cooking and heating in many of low-income Gers at Ulaanbaatar. The common tree species in

Mongolia are larch, pine, cedar, spruce, birch; these are mostly softwood (http://www.fao.org/3/w8302e/w8302e05.htm; http://www.fao.org/3/a-am616e.pdf, excess date 17-12-2019). Each Ger burns an average of 3 m^3 of wood per year (Guttikunda, 2008; Zhamsueva et al., 2018). Organic carbon (OC) has severe effects on human health and global climate change (Sun et al., 2019). But there are very few estimates of OC emitted from biomass burning (OC_{BB}) in Ulaanbaatar. Few studies have investigated the chemical characteristics of aerosol in Ulaanbaatar (Jung et al., 2010; Davy et al., 2011; Batmunkh et al., 2013), with none examining the contribution of OC_{BB} and type of biomass. Therefore, this study estimated appropriate concentration of OC_{BB} and identified the type of biomass at Ulaanbaatar, Mongolia.

In this study, we quantified the BB tracers levoglucosan, mannosan, galactosan, K^+, and other chemical species. Potential sources of $PM_{2.5}$ were identified by principal component analysis (PCA), with levoglucosan/K^+ and levoglucosan/mannosan ratios being used to identify major biomass types. OC_{BB} can be quantified from OC/levoglucosan ratios and levoglucosan concentrations in PM. However, uncertainties of OC_{BB} are high because OC/levoglucosan ratios can vary depending on fuel type, burning conditions, and burning place (Duan et al., 2004; Cheng et al., 2013; Jung et al., 2014). Therefore, it is required to determine the most suitable OC/levoglucosan ratio of BB emissions for estimating appropriate concentration of OC_{BB}. Here, for the first time, optimized OC/levoglucosan ratios were investigated for estimating concentrations of OC_{BB} during winter and spring. $OC_{non\text{-}BB}$ sources were also investigated using multivariate correlation analysis with ions and elemental carbon (EC).

2. Methods

2.1 Sampling site and aerosol sampling

Aerosol sampling was carried out in Ulaanbaatar during the winter (17 January to 03 February) and spring (17 April to 4 May) of 2017, with 24 h periods commencing daily at 11:00 local time. An aerosol sampler was installed on the rooftop of the National Agency for Meteorology and Environmental Monitoring station in Ulaanbaatar (47°92' N, 106°90' E, Fig. 1), 10 m above ground level. The sampling site was located at 8 km–10 km far from two coal based thermal power plants to the west (Chung and Chon, 2014). $PM_{2.5}$ samples were collected on 47 mm diameter quartz fiber filters (Pall-Life Sciences, USA) using an aerosol sampler (Murata Keisokuki Service, Japan) at a flow rate of 30 L min^{-1}. Field blank filter was collected during winter (n=1) and spring (n=1). The quartz fiber filter was loaded in the sampler for 5 minutes without operating a pump. The concentration of all chemical analytes has been corrected using blank filters concentration. Sampled filters were wrapped in aluminum foil and heated at 550°C for 12 h to remove adsorbed impurities before use and stored at –20°C before and after sampling.

2.2 Filter analysis

A one-fourth part of each quartz fiber filter sample was extracted in 10 mL ultrapure water (resistivity 18.2 MΩ, total OC content < 1 ppb,) under ultrasonication for 30 min. The water extract was then filtered using a syringe filter (Millipore, Millex−GV, 0.45μm) and stored at 4°C pending analysis. Water-soluble cations (K^+, Na^+, Ca^{2+}, Mg^{2+}, and NH_4^+) were quantified by an ion chromatograph (Dionex ICS 5000, Thermo Fisher Scientific, USA). Water-soluble cations were separated using an IonPac CS−12A column (Thermo Fisher Scientific, USA) with 20 mM methanesulfonic acid as eluent at a flow rate of 1.0 mL min^{-1}. Water-soluble anions (Cl^{-}, NO$_3^{-}$, and SO$_4^{2-}$) were separated using an IonPac AS−15 column (Thermo Fisher Scientific, USA) with 40 mM KOH as eluent at a flow rate of 1.2 mL min^{-1}. The detection limits for major inorganic ions based on 3σ of blanks were 0.01 μg m^{-3}, 0.01 μg m^{-3}, and 0.03 μg m^{-3} for NO$_3^{-}$, SO$_4^{2-}$, and NH$_4^{+}$, respectively.

Levoglucosan, mannosan, and galactosan were measured by a high-performance anion-exchange chromatograph (Dionex, ICS−5000, Thermo Fisher Scientific, USA) with pulsed amperometric detection involving an electrochemical detector with a gold working electrode. Details of the method are given elsewhere (Jung et al., 2014). In brief, separation involved a CarboPak MA1 (4 × 250 mm, Thermo Fisher Scientific, USA) analytical column and NaOH eluent (360 mM, 0.4 mL min^{-1}). Limits of detection were 3.0 ng m^{-3}, 0.7 ng m^{-3}, and 1.0 ng m^{-3} for levoglucosan, mannosan, and galactosan, respectively.

Aerosol samples were analyzed for OC and EC using a thermal optical OC/EC analyzer (Sunset Laboratory Inc. Forest Grove, OR, USA) with laser transmittance-based correction of pyrolysis. Details of the analyzer and quality-control parameters are reported elsewhere (Jung et al., 2014). In brief, 1.5 cm^2 punch samples of the quartz fiber filter were placed in a quartz dish inside the thermal desorption oven of the analyzer. OC and EC were quantified using a temperature program developed by the US National Institute for Occupational Safety and Health (NIOSH) in an inert atmosphere (100% He) and in an oxidizing atmosphere (98% He + 2% O$_2$), respectively. Detection limits of OC and EC were 0.04 and 0.01 μg C m^{-3}, and analytical uncertainties of them were 1.3% and 3.7%, respectively.

2.3. Conditional Probability Function

The Conditional Probability Function (CPF) calculates the probability that a source is located within a particular wind direction sector, ΔΘ:

$$CPF = \frac{m_{\Delta\Theta}}{n_{\Delta\Theta}}$$

where $n_{\Delta\Theta}$ is the number of times that the wind passed through direction sector ΔΘ, and $m_{\Delta\Theta}$ is the number of times that the source contribution peaked while the wind passed through sector ΔΘ (Ashbaugh et al., 1985). To use CPF with the Ulaanbaatar data, the 24 h averaged source contribution data have been applied to all 1 h wind direction averages recorded at the site for each date. The angular interval ΔΘ was set at 10°. To calculate $m_{\Delta\Theta}$, the 75th percentiles of source contribution concentrations were counted. CPF is useful in determining the direction of a source from a receptor site; however, it cannot determine the actual location of the source.

2.4 Principal component analysis

In order to identify the source groupings of chemical species in $PM_{2.5}$, principal component analysis (PCA) was applied. PCA is done using a commercially available software package (SPSS, version 10.0). PCA applies projection dimension reduction methods, converting several concentrations sets into significant sets of columns (principal components, PCs) without damaging the original data. PCA is a widely used statistical technique to quantitatively identify a small number of independent factors among the species concentrations, which can explain the variance of the data, by using the eigenvector decomposition of a matrix of pair-wise correlations. PCA with varimax rotation and retention of principal components having eigenvalues >1.0 was used to identify major species associated with different sources. It was widely used for identification of pollution sources in the atmosphere (Fang et al., 2003, Nirmalkar et al., 2015).

3. Results and Discussion

3.1 Chemical characteristics of $PM_{2.5}$ and source identification

Mass concentrations of carbonaceous aerosol, BB tracers, and water-soluble ions in $PM_{2.5}$ samples collected at Ulaanbaatar during winter and spring of 2017 are summarized in Table 1. OC contributed 64 ± 5.1% and 56 ± 6.0% of the quantified aerosol components in $PM_{2.5}$ in winter and spring, respectively (Table 1). Average concentrations of OC during winter were five times those obtained in spring (Fig. 2). Previously, OC has been observed as major component in $PM_{2.5}$ in Ulaanbaatar during winter period (Jung et al., 2010; Batmunkh et al., 2013). This may be attributed to additional BB emission for home heating, and temperature inversions with low wind speeds (average wind speed of 1.43 ± 0.73 m s^{-1}; Table 1 and Fig. 3a). OC concentrations decreased with increasing wind speed during winter (Fig. 3a) and spring (Fig. 3b), over all air temperature ranges. The inverse relationship between OC and wind speed during winter (Fig. 3a) and spring (Fig. 3b) suggests a predominance of local sources, with higher wind speeds flushing air pollutants out of the area whereas low wind speeds allow them to accumulate (Khan et al., 2010; Wang et al., 2018).

Average concentration of EC during winter (1.71 ± 0.58 μg m^{-3}) was higher than that in spring (1.11 ± 0.42 μg m^{-3}) (Table 1), consistent with general urban observations in cities of China (Ji et al., 2016) and India (Panda et al., 2016). During both winter and spring, EC concentrations at the study site were lower and having different trends compared to those observed in a suburban site (2.3 ± 1.0 μg m^{-3} and 3.1 ± 1.5 μg m^{-3}, respectively) and an urban site (2.3 ± 1.0 μg m^{-3} and 3.3 ± 1.2 μg m^{-3}, respectively) in Shanghai, China (Feng et al., 2009).

The potential source direction of EC during winter and spring was west as shown in Fig. 5; this can be explained by the influence of emission from thermal power plants. Correlation of EC was strong with Ca^{2+} during spring as shown in Fig. 4. CPF analysis suggested that potential source direction of EC and Ca^{2+} was similar (Fig. 5). High abundances of Ca^{2+} and EC is observed from stack emission of coal fired thermal power plants (Pei et al., 2016; Zhang et al., 2015). Thus, EC and Ca^{2+} in Ulaanbaatar might be strongly related to emission from thermal power plants.

Daily concentrations of levoglucosan, mannosan and galactosan have similar trends during winter and spring (Fig. 2), possibly because of combustion of similar biomass fuels in both seasons. Changes in concentrations of these BB tracers might be attributed to changes in relative proportions of cellulose and hemicellulose in different biomass fuels (Zhu et al., 2015; Nirmalkar et al., 2015). Concentrations of anhydrosugars were four times higher in winter than in spring (Table 1) due to increased heating requirements in winter. The higher relative humidity (58.5%–72.7%) and lower temperature (–10.5°C to –27.8°C; Table 1) in winter can also contribute to longer atmospheric residence times due to increased levoglucosan stability (Lai et al., 2014). Higher concentrations of BB tracers in winter than spring have previously been observed in Beijing, China, (Liang et al., 2016) and were attributed to meteorological conditions similar to those of Ulaanbaatar. Average K^+ concentration (0.08±0.05 μg m^{-3}) in this study is significantly lower than the K (0.32 μg m^{-3}) observed in Ulaanbaatar during 2004-2008 (Davy et al., 2011).

Among water-soluble ions, SO_4^{2-} (9.7 ± 3.4 µg m^{-3}) was the most dominant $PM_{2.5}$ species during winter, followed by NH_4^+ (6.2 ± 2.4 µg m^{-3}) and NO_3^- (4.2 ± 1.7 µg m^{-3}), whereas SO_4^{2-} (1.9 ± 0.5 µg m^{-3}) was the dominant species during spring, followed by Ca^{2+} (0.9 ± 0.4 µg m^{-3}) and NH_4^+ (0.7 ± 0.3 µg m^{-3}). The total SO_4^{2-} + NH_4^+ + NO_3^- content accounted for 27% and 23% of the total measured chemical species during winter and spring, respectively (Fig. 2 and Table 1). SO_4^{2-} is the most prevalent water-soluble ion in $PM_{2.5}$ in Wuhan, Guangzhou, and Tianjin (China) due to industrial emissions and coal burning (Gu et al., 2011; Tao et al., 2014; Huang et al., 2016; Pei et al., 2016). This suggests that the higher SO_4^{2-} concentration in Ulaanbaatar may be attributable to emissions from the three major coal-fired thermal power plants near the study site.

The atmospheric concentrations of OC (11–17 µg m^{-3}) and levoglucosan (0.46–0.73 µg m^{-3}) were higher for samples collected during 27–30 April 2017 than on almost all remaining days in spring (Fig. 2b). Backward atmospheric trajectories based on the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model provided by the US National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory (ARL) indicate that during those days' air masses originated from a region where a significant number of fires were detected [US Fire Information for Resource Management System (FIRMS); National Aeronautics and Space Administration (NASA); Fig. 6a, b)]. Thus, the elevated OC and levoglucosan concentrations during 27–30 April might be influenced by long-range transport of BB from north of Mongolia.

3.2 Principal Component Analysis

Principal component analysis (PCA) is a useful tool for reducing the dimensionality of large aerosol datasets to principal components using varimax rotation for source identification (Cao et al., 2005; Lin et al., 2018; Nirmalkar et al., 2019). Four principal components (PCs) in winter and three in spring were identified with eigenvalues >1 after varimax rotation explaining 96% and 92%, respectively, of the total variance (Tables 2 and 3). The PCs were categorized on the basis of loadings of chemical components as follows. In winter, PC1 includes BB characterized by high loadings of levoglucosan, mannosan, and galactosan; PC2 includes dust characterized by Ca^{2+} and Mg^{2+} content; PC3 includes secondary formation characterized by SO_4^{2-}, NO_3^-, and NH_4^+ content; and PC4 includes fossil fuel combustion characterized by EC. In spring, PC1 includes BB (levoglucosan, mannosan, and galactosan); PC2 includes dust (Ca^{2+} and Mg^{2+}) and fossil fuel combustion (EC); and PC3 includes secondary formation (SO_4^{2-}, NO_3^-, and NH_4^+). The PCA results show that the chemical components of $PM_{2.5}$ in Ulaanbaatar were mainly affected by BB during winter and spring. Further, OC was primarily influenced by BB because it correlated well with the total variance of PC1 during winter (0.82; Table 2) and spring (0.77; Table 3).

3.3 Relationship among BB tracers

The correlations among the three BB tracers levoglucosan, mannosan, and galactosan are shown in Fig. 7a (winter) and 7b (spring). The correlations between levoglucosan and mannosan and between levoglucosan and galactosan are strong during winter ($R^2 = 0.99$ for both pairs) and spring ($R^2 = 0.95$ and 0.83, respectively; Fig. 7a, b). Concentrations of levoglucosan and OC are strongly correlated during both winter ($R^2 = 0.78$) and spring ($R^2 = 0.86$; Fig. 8a), suggesting that a major fraction of OC might be originated from BB in Ulaanbaatar. The similar strong correlation and steep slope observed in OC–levoglucosan plots for PM collected in Chiang Mai Province (Thailand) and Daejeon (Korea) were attributed mainly to BB (Jung et al., 2014; Thepnuan et al., 2019).

Fine mode K^+ is considered as biomass burning tracer in previous studies (Louie et al., 2005; Deshmukh et al., 2011; Cheng et al., 2013). The moderate correlation between levoglucosan and K^+ concentrations (R^2 = 0.68) in winter indicates that they are produced from similar sources (Fig. 8b), with BB contributing most of the K^+. However, the correlation between levoglucosan and K^+ was weak in spring (R^2 = 0.49; Fig. 8b). Because K^+ is typically emitted at a higher mass fraction in flaming phase combustion compared to smoldering (Lee et al., 2010), smoldering combustion tends to have higher levoglucosan/K^+ emission ratio compared to flaming combustion (Schkolnik et al., 2005; Gao et al., 2003). High levoglucosan/K^+ ratio was observed during winter (8.92) compared to spring (4.21) in this site. Thus, week correlation between levoglucosan and K^+ concentrations at Ulaanbaatar in spring can be explained by mixed burning condition such as smoldering and flaming.

OC and K^+ concentrations correlated well during winter (R^2 = 0.79; Fig. 9a) and spring (R^2 = 0.73; Fig. 9b), suggesting that they might be originated from similar sources. Because most of the aerosol particles emitted from BB belong to $PM_{2.5}$, the correlation between OC and K^+ as well as levoglucosan suggests that BB is one of the potential sources of OC in winter and spring. Because biomass fuel is burned in traditional stoves with no pollution control devices in Ulaanbaatar (Batmunkh et al., 2013), soil and ash particles are entrained in convective processes and uplifted in the atmosphere together with smoke particles (Deshmukh et al., 2011; Nirmalkar et al., 2019).

3.4 Tracing the source of BB aerosol

OC is a major contributor of the quantified aerosol components in $PM_{2.5}$ in Ulaanbaatar during spring and winter (Table 1). To quantify the OC_{BB}, it is necessary to identify the BB fuel type. Several investigators used levoglucosan/mannosan and levoglucosan/K^+ ratios to identify BB fuel types (Puxbaum et al., 2007; Cheng et al., 2013; Jung et al., 2014; Chen et al., 2018; Thepnuan et al., 2019).

The levoglucosan/mannosan ratio is source-specific and can be used to identify BB fuel types due to the unique cellulose and hemicellulose compositions of different biomass fuels (Zhang et al., 2007; Cheng et al., 2013). A previous study suggested that the levoglucosan/mannosan ratio is strongly dependent on wood type, rather than on the site where the wood is grown (Cheng et al., 2013). Therefore, the levoglucosan/mannosan ratio was used to trace the type of wood burnt during winter and spring for indoor heating and cooking purposes. Previous studies have used levoglucosan/mannosan ratios to investigate the BB fuel types (Cheng et al., 2013; Jung et al., 2014).

However, the levoglucosan/mannosan ratio can't distinguish crop residuals (29 ± 15) (Sheesley et al., 2003, Sullivan et al., 2008, Engling et al., 2009, Oanh et al., 2011) and hardwood (28 ± 28) (Fine et al. 2001, 2002, 2004a, b; Engling et al., 2006; Schmidl et al., 2008; Bari et al., 2009; Goncalves et al., 2010) due to the overlap of ratios between these fuel types (Cheng et al., 2013; Fine et al. 2001, 2002, 2004a, b; Engling et al., 2006). However, levoglucosan/K^+ ratio can distinguish between the two groups (Jung et al., 2014, Chen et al., 2018). Both levoglucosan/mannosan and levoglucosan/K^+ ratios are therefore useful in distinguishing various types of fuel (Cheng et al., 2013; Puxbaum et al., 2007).

A levoglucosan/mannosan–levoglucosan/K^+ scatter plot based on results of the present and previous studies is shown in Fig. 10, using data from Schauer et al. (2001), Fine et al. (2001, 2002, 2004a, b), and Engling et al. (2006) for hardwood grown in the USA; Schauer et al. (2001), Hays et al. (2002), Fine et al. (2001, 2002, 2004a, b), and Engling et al. (2006) for US softwood; Schmidl et al. (2008), Bari et al. (2009) and Goncalves et al. (2010) for hardwood grown in Europe; Iinuma et al. (2007), Schmidl et al. (2008), and Goncalves et al. (2010) for European softwood; Engling et al. (2006) and Sullivan et al. (2008) for needles and duff found in the USA; Sullivan et al. (2008) for US grass; and from Sheesley et al. (2003), Sullivan et al. (2008), Engling et al. (2009) and Oanh et al. (2011) for Asian rice straw.

The average levoglucosan/mannosan ratio was 3.6 ± 0.2 (range: 3.4 − 4.1) in winter and 4.1 ± 1.0 (2.12 − 7.05) in spring, whereas the levoglucosan/K^+ ratio was 8.9 ± 1.8 (5.5 − 12.4) in winter and 4.2 ± 2.1 (0.58 – 7.49) in spring at the study site (Fig. 10), within the ranges reported for softwood burning sources (2.5 − 6.7 and 4.6 − 261, respectively) (Fine et al., 2001; Schauer et al., 2001; Fine et al., 2002, 2004a, b; Hays et al., 2002; Engling et al., 2006; Iinuma et al., 2007; Schmidl et al., 2008; Goncalves et al., 2010; Cheng et al., 2013). During winter and spring, the levoglucosan/K^+ and levoglucosan/mannosan ratios in Ulaanbaatar appeared in the softwood region (Fig. 10).

Therefore, softwood burning seems to be the major source of BB aerosol in Ulaanbaatar during both winter and spring, consistent with previously reported softwood-burning emissions from fireplaces of northern and southern regions of the USA (Fine et al., 2001, 2002), from household combustion in Zhengzhou, China (Chen et al., 2018), and from stove wood combustion in the mid-European region (Austria;

Schmidl et al., 2008).

3.5 Optimization of OC/levoglucosan ratio for estimating OC_{BB} emission

OC_{BB} was estimated by multiplying OC/levoglucosan ratio and levoglucosan concentration. Previous studies have used the OC/levoglucosan ratio obtained from sources of BB aerosol to estimate OC_{BB}. A ratio of 7.35 reported for burning of four types of US hardwood (Fine et al., 2002) was used for estimating OC_{BB} at four background sites in Europe (Puxbaum et al., 2007). Later, mean value of 11.2 of OC/levoglucosan ratio derived from ratios ranged between 4.5 – 24.6 was used for estimating OC_{BB} in the UK (Harrison et al., 2012). However, such estimates may not be accurate as the OC/levoglucosan ratio is highly variable in BB emissions. For example, the average OC/levoglucosan ratio from softwood burning (23.8) is much higher than that of hardwood burning (7.35) (Fine et al., 2002; Schmidl et al., 2008), differences are more than ten-fold among studies of softwood-burning OC/levoglucosan ratios (Fine et al., 2002; Hays et al., 2002; Engling et al., 2006; Iinuma et al., 2007; Goncalves et al., 2010). Combustion conditions may also significantly influence OC/levoglucosan ratios. For example, the OC/levoglucosan ratio varied by a factor of about seven between burning the same wood (Loblolly pine) in a fireplace (27.6; Fine et al., 2002) and in a stove (3.4; Fine et al., 2004b). Therefore, it is necessary to optimize the OC/levoglucosan ratio for use in estimating OC_{BB}.

This study has used an optimized OC/levoglucosan ratio to estimate precise concentration of OC_{BB} for the Ulaanbaatar study site. We have used a range of different OC/levoglucosan ratios obtained from previous literatures (Fig. 11) for regression analysis with measured levoglucosan concentrations to estimate optimum

OC/levoglucosan ratio (Fig. 12a, b). First, candidate OC_{BB} (Fig. 11) in this study was estimated from OC/levoglucosan ratios for softwood burning in a previous chamber experiments (Cheng et al., 2013; Schauer et al., 2001; Hays et al., 2002; Fine et al., 2001, 2002, 2004a, b; Engling et al., 2006; Iinuma et al., 2007; Schmidl et al., 2008; Goncalves et al., 2010, Fig 11) and measured levoglucosan concentration at this site. Second, $OC_{non\text{-}BB}$ concentration was calculated by subtracting OC_{BB} from corresponding total OC. If calculated $OC_{non\text{-}BB}$ doesn't contain OC_{BB}, both regression slope and R^2 between $OC_{non\text{-}BB}$ versus levoglucosan will be close to zero. As shown in Fig. 12a and 12b, the lowest R^2 and regression slope were observed when OC/levoglucosan ratios of 27.6 and 18.0 in winter and spring, respectively. Thus, the optimized OC/levoglucosan ratios for our site were determined to be 27.6 and 18.0 in winter and spring, respectively.

During winter higher optimum ratio of OC/levoglucosan might be due to incomplete combustion during smoldering phenomena. As smoldering fires are characterized by lower temperatures and thus have lower combustion efficiency, they release more un-combusted condensable products, resulting in the production of more unbroken organic compounds (Engling et al., 2006). Smoldering combustion generally leads to increased emissions of volatile organic compounds (VOCs) and particulate organic matter (OM) (Obrist et al., 2007). In contrast, the relatively lower optimum ratio of OC/levoglucosan during spring might be due to the higher combustion efficiency during flaming phenomena.

The OC_{BB} concentrations at the Ulaanbaatar study site were calculated from the optimized OC/levoglucosan ratios (winter: 27.6 and spring: 18.0) and measured levoglucosan concentrations. The OC_{BB} concentration was estimated to be 33.1 ± 11.9 μg C m^{-3} (range 16.0−58.5 μg C m^{-3}) and 5.64 ± 3.29 μg C m^{-3} (range 0.57−13.1 μg C m^{-3}), accounting for 68% and 63% of the total OC in winter and spring, respectively (Fig. 13). The average of previously published OC/levoglucosan ratios, 10.1 ± 7.9 (range 1.90 − 27.6), gives an estimated OC_{BB} concentration of 12.1 ± 4.4 μg C m^{-3} (range 5.9–21.4 μg C m^{-3}) and 3.2 ± 1.8 μg C m^{-3} (0.32–7.34 μg C m^{-3}) in winter and spring, respectively. Their values are 2.7 (winter) and 1.8 (spring) times lower than values estimated using our optimized OC/levoglucosan ratio.

Our estimated contribution of OC_{BB} was higher than that in Daejeon, South Korea (24%–68% of total OC, mean 45% ± 12%; Jung et al., 2014) and Beijing, China (50% of total OC; Cheng et al., 2013), where BB aerosols are produced mainly by the burning of crop residues. The contribution of OC_{BB} to total OC is 57% and 31% during heating (average temperature 0.6°C) and non-heating (average temperature 14°C) seasons in Krynica Zdroj, Poland (Klejnowski et al., 2017), significantly lower than that of Ulaanbaatar during both winter (average temperature –21°C) and spring (average temperature 6°C). Such high concentrations of OC_{BB} in Ulaanbaatar and Krynica Zdroj are likely due to intense wood burning for heating during winter.

3.6 Tracing sources of $OC_{non\text{-}BB}$

High concentration of $OC_{non\text{-}BB}$ was found during winter compared to spring (Fig. 13). Elevated $OC_{non\text{-}BB}$ could be attributed to enhanced emission from combustions and favorable meteorological conditions (cold temperatures and inversion conditions, etc.) during the winter. There is strong correlation between $OC_{non\text{-}BB}$ and SO_4^{2-}, NH_4^+, and K^+ in winter and $OC_{non\text{-}BB}$ and NO_3^-, Na^+, K^+, Mg^{2+}, Ca^{2+}, and EC in spring (Table 4). Residential combustion of coal emits significant amounts of OC, EC, and inorganic species (SO_4^{2-} and metals) due to incomplete combustion and lack of pollution control devices (Garcia et al., 1992; Li et al., 2016; Watson et al., 2001a, b). Garcia et al. (1992) studied emissions of volatile organic compounds from coal burning and vehicle engines.

In Ulaanbaatar, the use of wood and coal for cooking and heating, and emissions from old vehicles are reported as potential sources of OC (Batmunkh et al., 2013; Zhamsueva et al., 2018). The three thermal power plants in Ulaanbaatar are point sources for emissions of carbonaceous aerosol (Batmunkh et al., 2013), burning ~5 million tons of coal per year (Batmunkh et al., 2013). High concentrations of anions (SO_4^{2-} and NO_3^-) and cations (NH_4^+ and Na^+) are reported in China (Zhou et al., 2003), the USA (Caiazzo et al., 2013), Brazil (Flues et al., 2002), India (Guttikunda and Jawahar, 2014), Korea (Park and Kim, 2004; Park et al., 2015), and Spain (Alastuey et al., 1999) near coal-fired thermal power plants. Emissions of volatile organic compounds from vegetation have also been observed in previous studies (Fehsenfeld et al., 1992; Shao et al., 2001; Acton et al., 2016). The correlations of $OC_{non\text{-}BB}$ with ions and EC are thus likely due to volatile organic compounds emitted from coal-burning and vehicles, and vegetative emissions.

4. Conclusions

BB was identified as a major source of the quantified aerosol components in $PM_{2.5}$ in Ulaanbaatar, Mongolia, during the winter and spring of 2017, based on PCA. OC was the major component of the quantified aerosol components during the entire sampling period, winter and spring. For determination of OC_{BB}, the fuel type must be identified and levoglucosan/mannosan and levoglucosan/K^+ ratios obtained from previous studies and our on-site measurements were used for this purpose.

Softwood burning was identified as a major source of OC_{BB}. However, OC/levoglucosan ratios from softwood burning are highly variable, and an optimum ratio was derived by regression analysis between daily concentrations of OC_{non-BB} and levoglucosan, yielding values of 27.6 and 18.0 for winter and spring, respectively. The application of these ratios indicates that 68% and 63% of the OC originated from BB during winter and spring, respectively, which is about double that estimated using average values of previous studies. The atmospheric concentration of OC_{BB} was higher in winter than in spring mainly due to additional BB for heating and cooking. BB aerosols in Ulaanbaatar originate mainly from local softwood burning. The approach developed here may be applied elsewhere for screening region-specific OC/levoglucosan ratios for estimating atmospheric appropriate concentrations of OC_{BB}, aiding the establishment of BB control measures.

Author contribution

Jinsang Jung and Tsatsral Batmunkh designed the study and carried out the field work. Jinsang Jung performed chemical analyses and quality-control measures. Jayant Nirmalkar wrote the manuscript under the guidance of Jinsang Jung. All authors commented on and discussed the manuscript.

Competing interests

The authors declare that they have no conflict of interests.

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Data availability

The data used in this study are available from the corresponding author upon request (jsjung@kriss.re.kr).

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Table 1. Concentrations (μg m^{-3}) of organic carbon, elemental carbon, levoglucosan, mannosan, galactosan, and water-soluble ions in $PM_{2.5}$ samples collected from Ulaanbaatar, Mongolia, during the winter (n = 17) and spring (n = 17) of 2017.

	OC	EC	Levoglucosan	Mannosan	Galactosan	Cl^-	SO_4^{2-}	NO_3^-	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Temperature (°C)	Wind Speed (m sec^{-1})	RH (%)
Winter																
Mean	49.06	1.71	1.20	0.33	0.24	1.69	9.74	4.17	0.64	6.18	0.13	0.05	0.60	-20.8	1.36	66.1
SD	17.32	0.58	0.43	0.13	0.09	0.76	3.37	1.69	0.44	2.42	0.04	0.02	0.24	4.74	0.73	4.56
Min	24.62	0.79	0.58	0.15	0.10	0.26	2.17	0.76	0.10	3.16	0.08	0.02	0.22	-27.8	0.41	58.5
Max	79.07	3.34	2.12	0.61	0.43	2.89	16.06	7.51	1.34	11.59	0.18	0.08	1.04	-10.5	3.55	72.7
Spring																
Mean	8.50	1.11	0.31	0.08	0.04	0.30	1.90	0.70	0.13	0.74	0.08	0.04	0.93	6.11	2.60	35.1
SD	3.55	0.42	0.18	0.04	0.02	0.11	0.50	0.32	0.04	0.28	0.05	0.02	0.36	6.16	0.79	13.9
Min	2.80	0.60	0.03	0.01	0.00	0.11	1.04	0.10	0.07	0.33	0.02	0.02	0.48	-1.52	1.64	17.8
Max	16.63	2.03	0.73	0.15	0.08	0.51	3.02	1.40	0.21	1.47	0.22	0.08	1.61	15.9	4.56	65.2

Table 2. Source identification of chemical species using principal component (PC) analysis and varimax rotation at Ulaanbaatar, Mongolia, during winter of 2017.

Winter	Component			
Chemical species	PC1 (Biomass Burning)	PC2 (Dust)	PC3 (Secondary formation)	PC4 (Fossil fuel combustion)
Levoglucosan	**0.96**	−0.06	0.24	0.06
Mannosan	**0.95**	−0.08	0.27	0.06
Galactosan	**0.95**	−0.07	0.28	0.04
Cl^-	0.19	**0.94**	−0.05	−0.07
SO_4^{2-}	0.43	0.01	**0.88**	0.09
NO_3^-	0.28	0.20	**0.87**	0.20
Na^+	−0.27	**0.87**	−0.33	−0.17
NH_4^+	0.48	−0.12	**0.86**	0.07
K^+	**0.70**	0.11	0.61	0.25
Mg_2^+	−0.15	**0.90**	0.25	0.26
Ca_2^+	−0.12	**0.92**	0.19	0.24
OC	**0.82**	−0.17	0.47	0.07
EC	0.14	0.14	0.19	**0.95**
Eigenvalues	4.54	3.44	3.30	1.20
% of Variance	34.95	26.49	25.37	9.21
Cumulative %	34.95	61.44	86.81	96.02

Table 3. Source identification of chemical species using PCA and varimax rotation at Ulaanbaatar, Mongolia, during spring of 2017.

Spring	Component		
Chemical species	PC1 (Biomass Burning)	PC2 (Dust and Fossil fuel combustion)	PC3 (Secondary formation)
Levoglucosan	**0.88**	0.13	0.39
Mannosan	**0.94**	0.00	0.30
Galactosan	**0.95**	−0.11	0.20
Cl^-	**0.81**	0.32	−0.03
SO_4^{2-}	0.18	0.12	**0.93**
NO_3^-	0.59	0.54	0.52
Na^+	0.08	**0.91**	−0.09
NH_4^+	0.44	0.05	**0.88**
K^+	0.41	0.67	0.55
Mg^{2+}	0.05	**0.90**	0.35
Ca^{2+}	0.10	**0.97**	0.15
OC	**0.77**	0.41	0.46
EC	0.10	**0.94**	0.01
Eigenvalues	4.59	4.53	2.87
% of Variance	35.30	34.84	22.04
Cumulative %	35.30	70.14	92.18

Table 4. Correlation coefficients (r) from Spearman correlation analysis for $OC_{non\text{-}BB}$ and water-soluble ions during winter and spring of 2017 at Ulaanbaatar, Mongolia.

		Cl^-	SO_4^{2-}	NO_3^-	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	EC
$OC_{non\text{-}BB}$	Winter	−0.26	0.71**	0.44	−0.58*	0.72**	0.64**	−0.16	−0.16	0.15
	Spring	0.29	0.37	0.59*	0.74**	0.23	0.65**	0.78**	0.77**	0.74**

*Correlation is significant at the .05 level (2-tailed); **Correlation is significant at the .01 level (2-tailed).

Figure captions

Fig. 1 Sampling site in Ulaanbataar, Mongolia (https://www.google.com/earth/versions/#earth-pro, © Google Earth).

Fig. 2 Daily variations in atmospheric concentrations (μg m^{-3}) of chemical species in Ulaanbaatar during winter (a) and spring (b) of 2017.

Fig. 3 Daily atmospheric concentrations of OC (μg C m^{-3}) as a function of wind speed (m s^{-1}) and temperature (°C) during winter (a) and spring (b) of 2017.

Fig. 4 Relationship between $PM_{2.5}$ concentrations of $Ca^{2+}$and EC (μg m^{-3}) during spring of 2017.

Fig. 5 Conditional Probability Function (CPF) of levoglucosan (levo), OC, K^+, EC, Ca^{2+} during winter (a) and spring (b) of 2017.

Fig. 6 (a) Five-day backward air-mass trajectories (https://ready.arl.noaa.gov/HYSPLIT.php) and (b) FIRMS fire counts (https://firms.modaps.eosdis.nasa.gov/alerts/) around Ulaanbaatar during spring of 2017.

Fig. 7 Correlations of $PM_{2.5}$ concentrations (μg m^{-3}) of mannosan and galactosan with levoglucosan during winter (a) and spring (b) of 2017.

Fig. 8 Correlation between $PM_{2.5}$ concentrations of (a) OC (μg C m^{-3}) and levoglucosan (μg m^{-3}) and (b) K^+ and levoglucosan (μg m^{-3}) during winter and spring of 2017.

Fig. 9 Correlation between $PM_{2.5}$ concentrations of OC (μg C m^{-3}) and K^+ (μg m^{-3}) during winter (a) and spring (b) of 2017.

Fig. 10 Scatter plot of levoglucosan/K$^+$ versus levoglucosan/mannosan from different types of BB emissions, including those measured in Ulaanbaatar (blue circles and red squares).

Fig. 11 Comparison of previously reported OC/levoglucosan ratios for softwood burning.

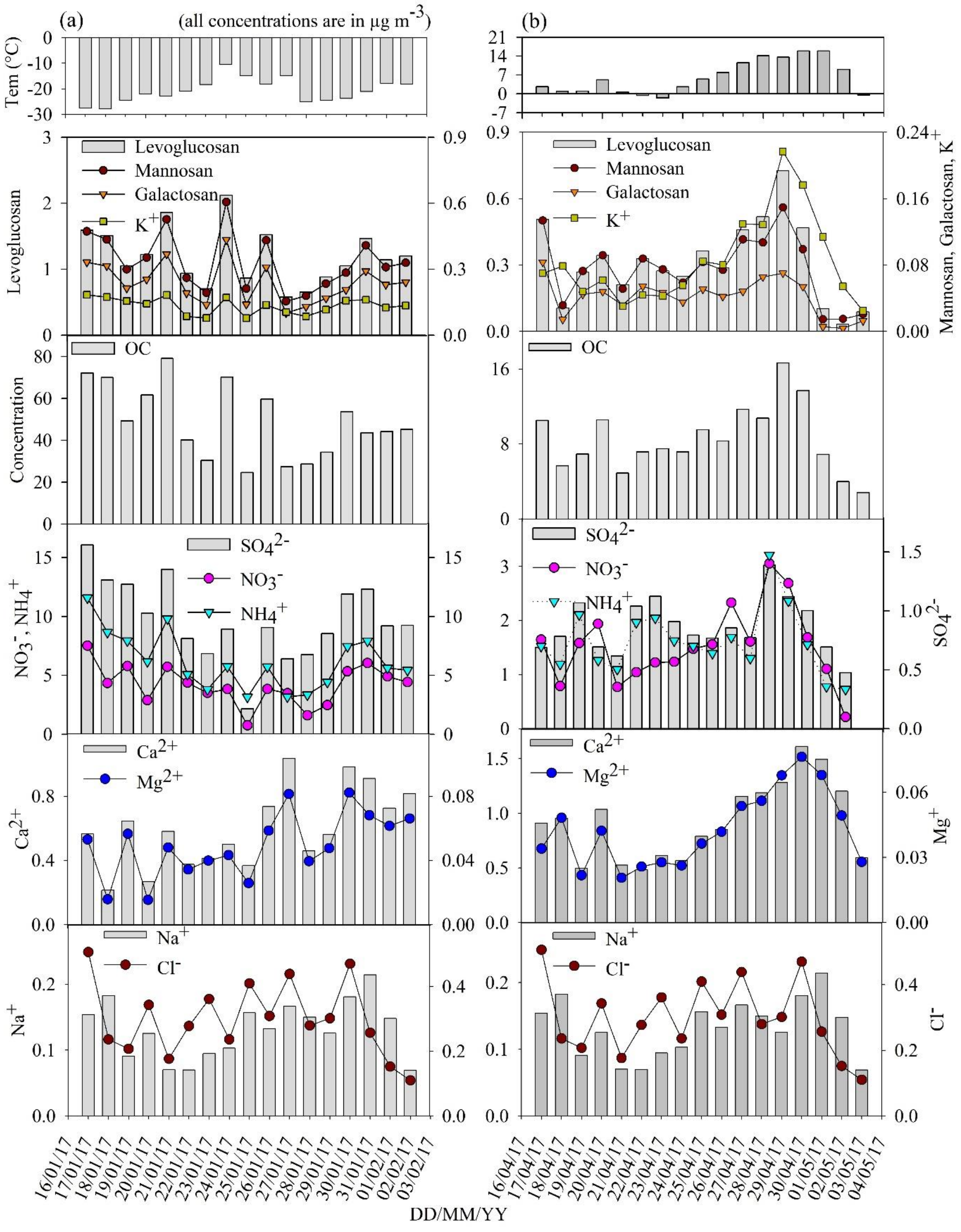
Fig. 12 Graphical determination of optimized OC/levoglucosan ratios used to estimate $PM_{2.5}$ concentrations of OC_{BB} in Ulaanbaatar in winter (a) and spring (b) of 2017.

Fig. 13 Relative contributions (μg C m^{-3}) of OC_{BB} and $OC_{non\text{-}BB}$ to $PM_{2.5}$ in Ulaanbaatar during winter and spring of 2017.

Fig. 1


Ulaanbaatar, Mongolia
Aerosol sampling site, NAMEM
(47.92° N, 106.90° E)
Khentii mountains areas
N
10 km
Thermal power plant 1
National Agency for Meteorology and Environment Monitoring of Mongolia
Thermal power plant 3
Thermal Power Plant 4
Tuul River
Bogdkhan Uul
Google Earth

Fig. 2



(a)
(all concentrations are in μg m-3)
(b)
Tem (°C)
Levoglucosan
Mannosan
Galactosan
K+
Mannosan, Galactosan, K+
OC
Concentration
SO4 2-
NO3-
NH4+
NO3-, NH4+
Ca2+
Mg2+
Mg+
Na+
Cl-
16/01/17
17/01/17
18/01/17
19/01/17
20/01/17
21/01/17
22/01/17
23/01/17
24/01/17
25/01/17
26/01/17
27/01/17
28/01/17
29/01/17
30/01/17
31/01/17
01/02/17
02/02/17
03/02/17
16/04/17
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23/04/17
24/04/17
25/04/17
26/04/17
27/04/17
28/04/17
29/04/17
30/04/17
01/05/17
02/05/17
03/05/17
04/05/17
DD/MM/YY

Fig. 3

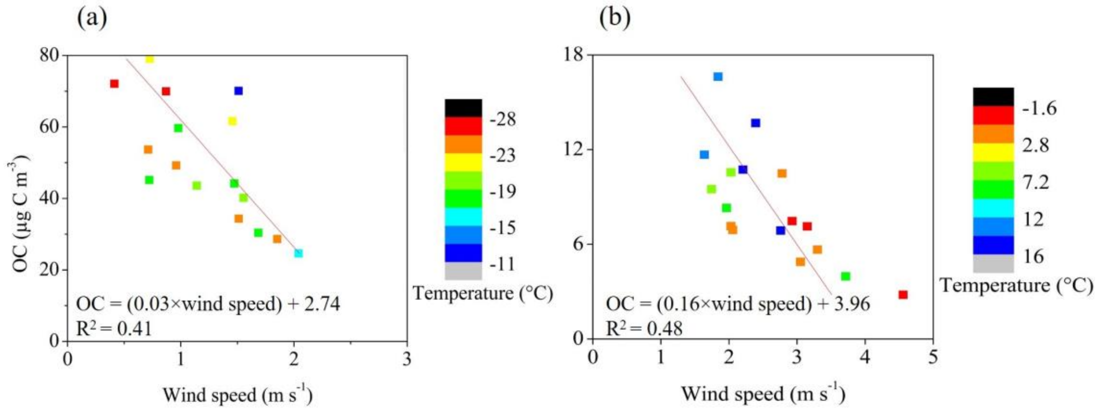

(a)
OC (μg C m^{-3})
80
60
40
20
0
0
1
2
3
Wind speed (m s^{-1})
OC = (0.03×wind speed) + 2.74
R^2 = 0.41
-28
-23
-19
-15
-11
Temperature (°C)
(b)
18
12
6
0
0
1
2
3
4
5
Wind speed (m s^{-1})
OC = (0.16×wind speed) + 3.96
R^2 = 0.48
-1.6
2.8
7.2
12
16
Temperature (°C)

Fig. 4

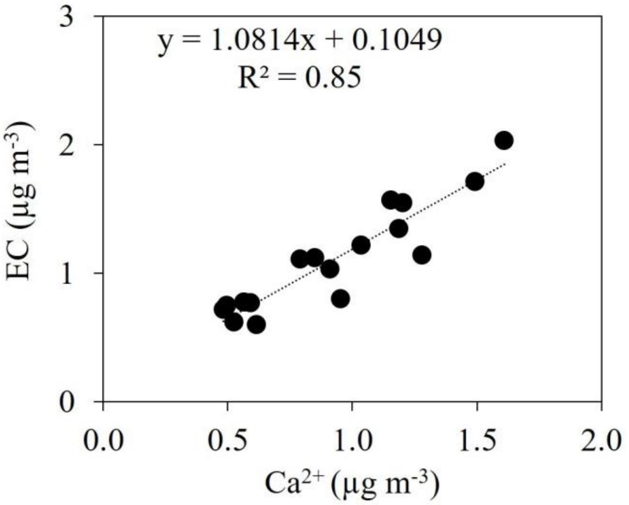


Fig. 5

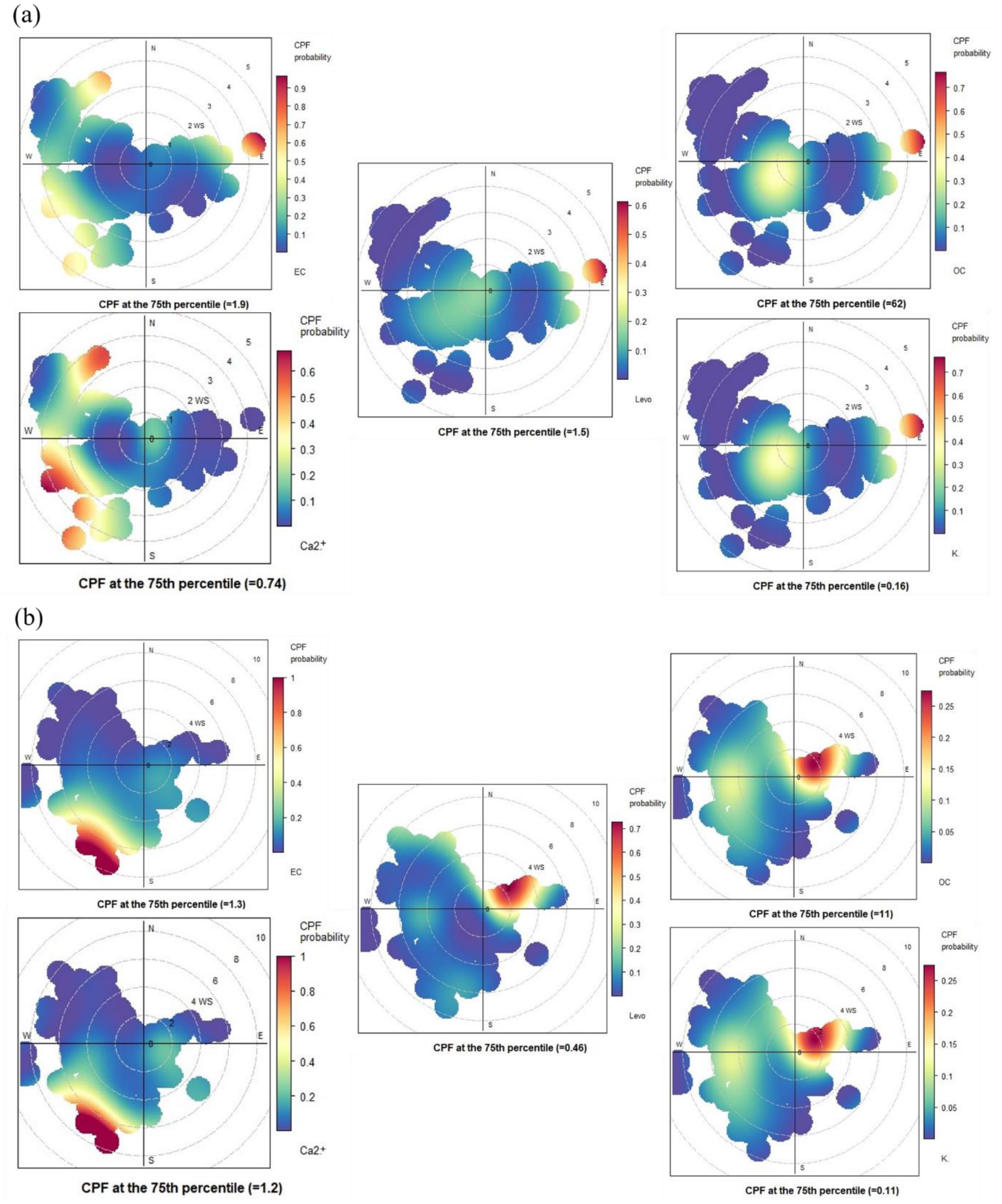

(a)
CPF probability
EC
CPF at the 75th percentile (=1.9)
Levo
CPF at the 75th percentile (=1.5)
OC
CPF at the 75th percentile (=62)
Ca2+
CPF at the 75th percentile (=0.74)
K.
CPF at the 75th percentile (=0.16)
N
S
E
W
2 WS
(b)
EC
CPF at the 75th percentile (=1.3)
Levo
CPF at the 75th percentile (=0.46)
OC
CPF at the 75th percentile (=11)
Ca2+
CPF at the 75th percentile (=1.2)
K.
CPF at the 75th percentile (=0.11)
4 WS

Fig. 6

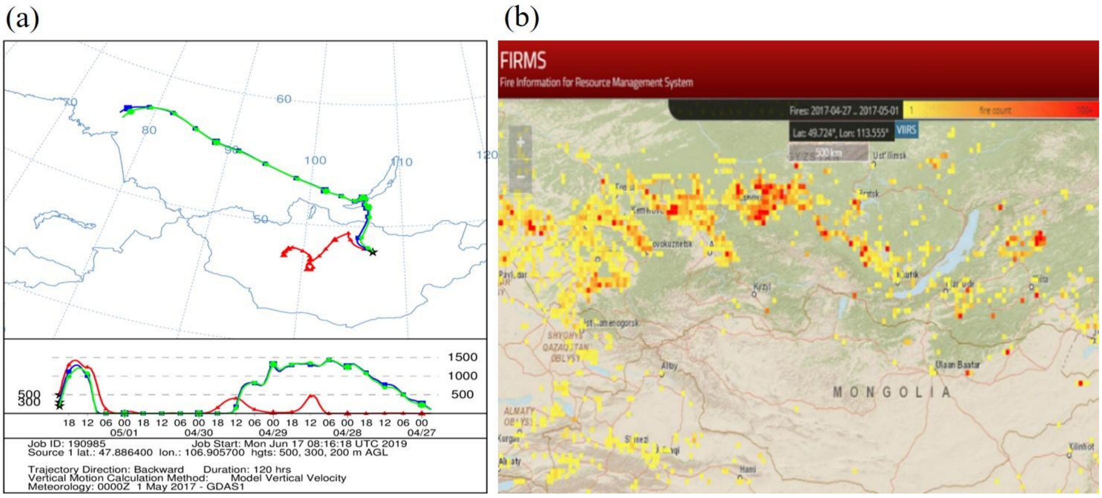

(a)
(b)
Source ★ at 47.89 N 106.91 E
70
80
90
100
110
120
60
50
Meters AGL
1500
1000
500
500
300
18 12 06 00 18 12 06 00 18 12 06 00 18 12 06 00 18 12 06 00
05/01
04/30
04/29
04/28
04/27
Job ID: 190985 Job Start: Mon Jun 17 08:16:18 UTC 2019
Source 1 lat.: 47.886400 lon.: 106.905700 hgts: 500, 300, 200 m AGL
Trajectory Direction: Backward Duration: 120 hrs
Vertical Motion Calculation Method: Model Vertical Velocity
Meteorology: 0000Z 1 May 2017 - GDAS1
FIRMS
Fire Information for Resource Management System
Fires: 2017-04-27 .. 2017-05-01
1
fire count
100+
Lat: 49.724°, Lon: 113.555°
VIIRS
500 km
Ust'Ilimsk
Bratsk
Kemerovo
Novokuznetsk
Pavlodar
Kyzyl
Irkutsk
Ulan Ude
Chita
Ust-Kamenogorsk
Altay
Ulaan Baatar
MONGOLIA
ALMATY OBLYSY
Kurgan
Almaty
Shihezi
Hami
Kilinbot

Fig. 7

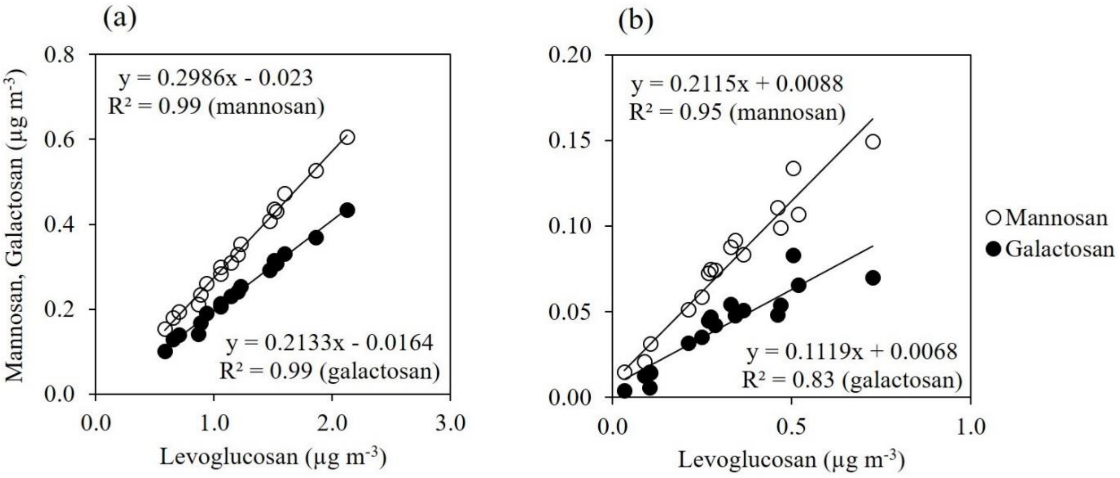

(a)
y = 0.2986x - 0.023
R² = 0.99 (mannosan)
y = 0.2133x - 0.0164
R² = 0.99 (galactosan)
Mannosan, Galactosan (μg m-3)
0.0
0.2
0.4
0.6
0.8
0.0
1.0
2.0
3.0
Levoglucosan (μg m-3)
(b)
y = 0.2115x + 0.0088
R² = 0.95 (mannosan)
y = 0.1119x + 0.0068
R² = 0.83 (galactosan)
0.00
0.05
0.10
0.15
0.20
0.0
0.5
1.0
Levoglucosan (μg m-3)
○ Mannosan
● Galactosan

Fig. 8

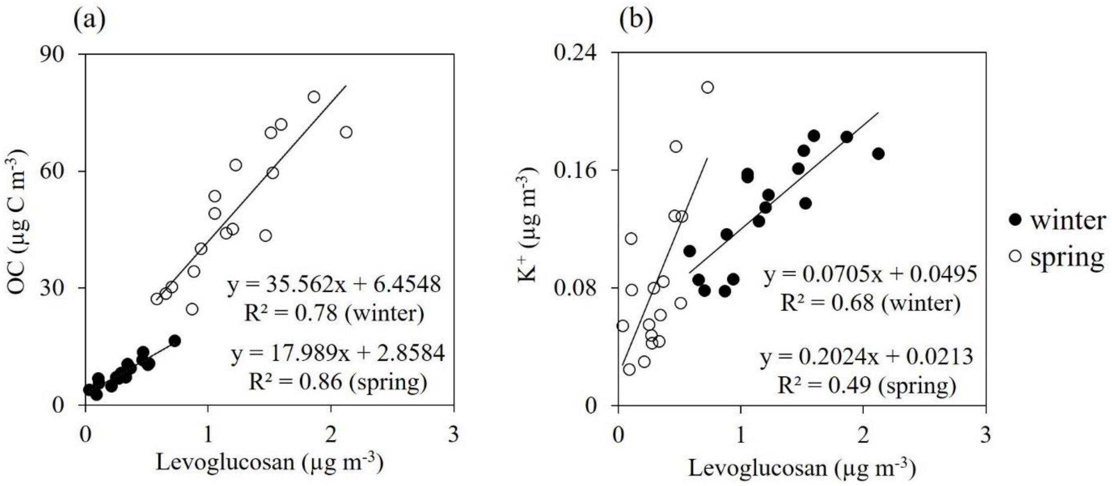
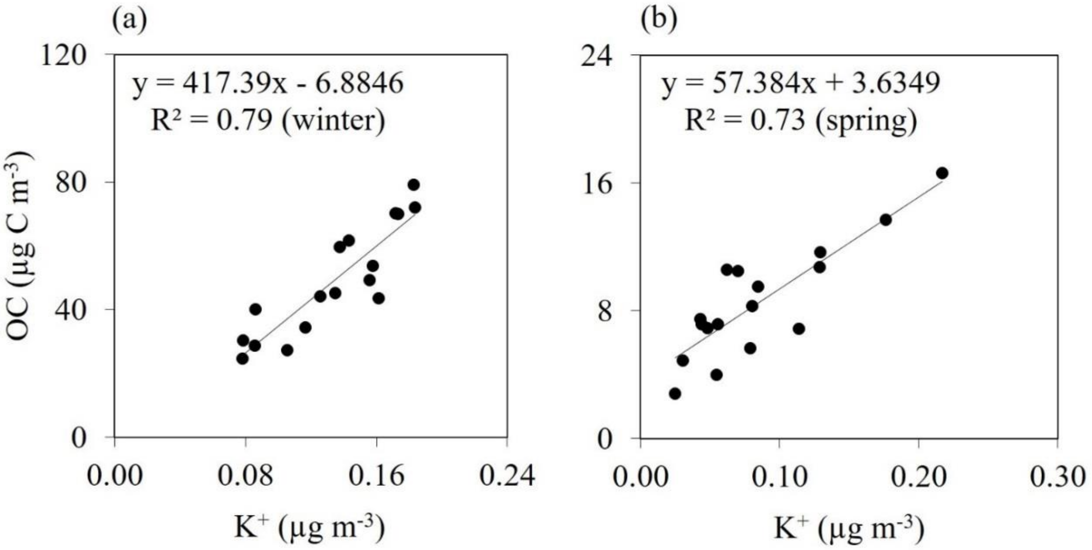
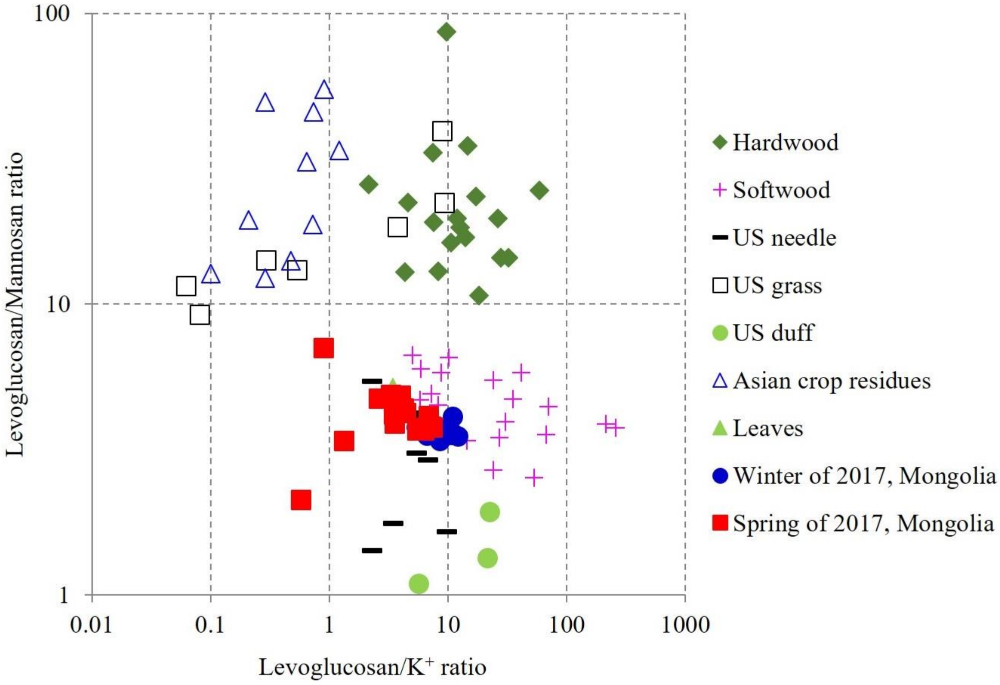

(a)
90
60
30
0
OC (μg C m-3)
0
1
2
3
Levoglucosan (μg m-3)
y = 35.562x + 6.4548
R² = 0.78 (winter)
y = 17.989x + 2.8584
R² = 0.86 (spring)
(b)
0.24
0.16
0.08
0
K+ (μg m-3)
0
1
2
3
Levoglucosan (μg m-3)
y = 0.0705x + 0.0495
R² = 0.68 (winter)
y = 0.2024x + 0.0213
R² = 0.49 (spring)
● winter
○ spring

Fig. 9


(a)
y = 417.39x - 6.8846
R² = 0.79 (winter)
OC (µg C m-3)
120
80
40
0
0.00
0.08
0.16
0.24
K+ (µg m-3)
(b)
y = 57.384x + 3.6349
R² = 0.73 (spring)
24
16
8
0
0.00
0.10
0.20
0.30
K+ (µg m-3)

923 Fig. 10

924



925

926

Fig. 11

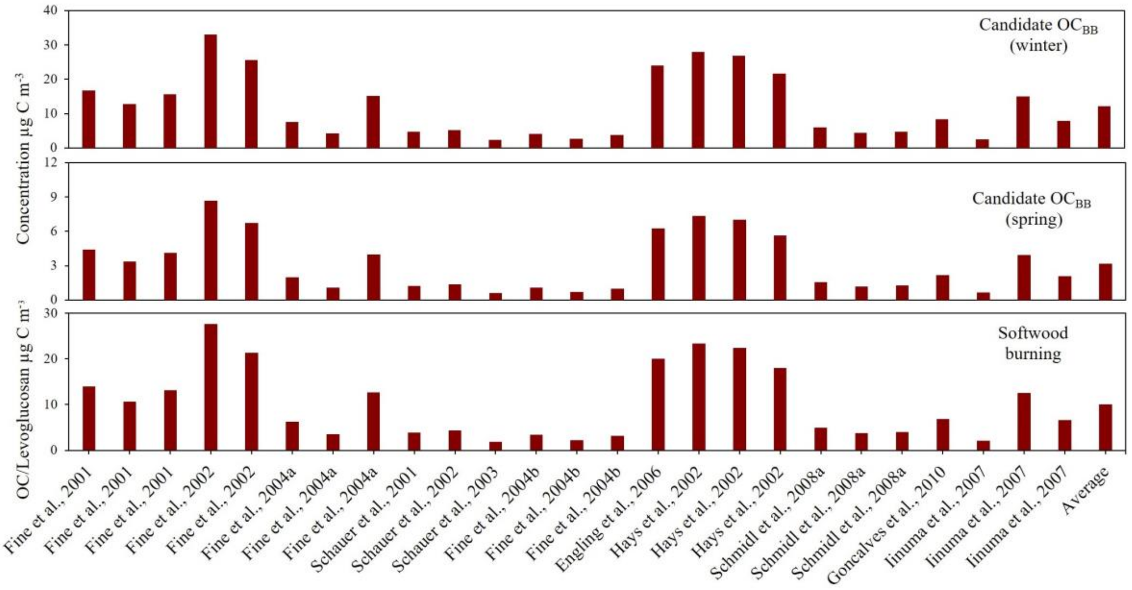

Candidate OC_{BB} (winter)
Candidate OC_{BB} (spring)
Softwood burning
Concentration µg C m^{-3}
OC/Levoglucosan µg C m^{-3}
0
10
20
30
40
0
3
6
9
12
0
10
20
30
Fine et al., 2001
Fine et al., 2001
Fine et al., 2001
Fine et al., 2002
Fine et al., 2002
Fine et al., 2004a
Fine et al., 2004a
Fine et al., 2004a
Schauer et al., 2001
Schauer et al., 2002
Schauer et al., 2003
Fine et al., 2004b
Fine et al., 2004b
Fine et al., 2004b
Engling et al., 2006
Hays et al., 2002
Hays et al., 2002
Hays et al., 2002
Schmidl et al., 2008a
Schmidl et al., 2008a
Schmidl et al., 2008a
Goncalves et al., 2010
Iinuma et al., 2007
Iinuma et al., 2007
Iinuma et al., 2007
Average

Fig. 12

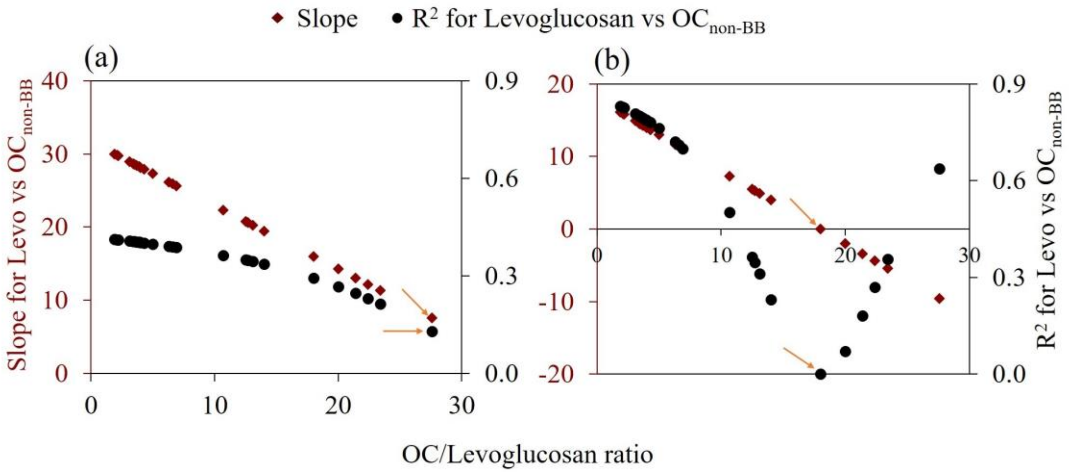
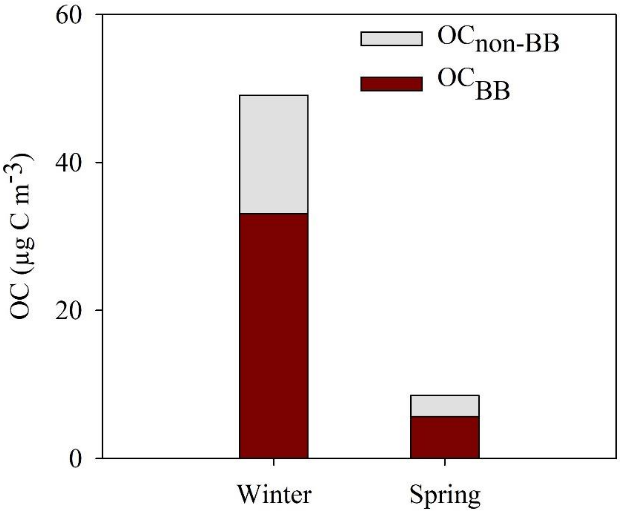

◆ Slope
● R² for Levoglucosan vs OC_non-BB
(a)
(b)
Slope for Levo vs OC_non-BB
R² for Levo vs OC_non-BB
OC/Levoglucosan ratio
40
30
20
10
0
-10
-20
0.9
0.6
0.3
0.0
0
10
20
30

Fig. 13



Report #1

Comments:

Thank very much for the reviewer for the appreciating comments and important inputs to improve the quality of the manuscript. We follow all the reviewer's comments very carefully and answer accordingly. We have incorporated all the modifications in the revised manuscript (RMS). Please refer to the revised manuscript where we highlighted the changes by turquoise color. We provide here below a reply to the specific comments and modifications made in the revised manuscript based on the line number provided in RMS. Please follow the line numbers to reviewing the changes in RMS.

1. The revised Fig 2: It is noteworthy to me that the coldest periods in the winter do not coincide with the highest concentrations of levoglucosan, mannosan etc (e.g 16-17th Jan and 29th Jan). If my reading of the paper is correct and biomass burning for heating is important in Ulaanbaatar, then one would expect the highest concentration of these tracer to be high when it is cold. Perhaps the authors could comment.

Reply: We thank the reviewer for highlighting this point. During winter the ambient temperature was consistently low (less than -10 °C) in Ulaanbaatar therefore residential biomass burning occurred continuously for space heating. Thus, there would be no dependence of emission rates of levoglucosan and mannosan with ambient temperature during the sampling period. The concentration of levoglucosan and mannosan may be influenced by local wind speed as shown in Fig. 3 rather than ambient temperature.

2. Revised Fig 5: I am not sure that the authors have enough samples to perform CPF analysis. I think they need to justify applying a 24 hr average concentration measurement to each 1 hour wind measurement in order to do this analysis. I would have instead done a simpler analysis, like a wind rose plot with concentration data.

Reply: Thank you very much for the suggestion. Already we have calculated CPF using 24h concentrations of measured species and, hourly wind speed and wind direction data in revised manuscript. Also we already mentioned this phrase in section 2.3 of first revised manuscript.

Following phrase added in first revised manuscript.

"To use CPF with the Ulaanbaatar data, the 24 h averaged source contribution data have been applied to all 1 h wind direction averages recorded at the site for each date."

Please see line number 166-168

3. I am still not sure I quite follow the method for determining the optimized OC/levoglucosan (Section 3.5). Is it that you selected a range of different OC/levoglucosan ratios from the literature (as shown in Fig 11?) to calculate the OC from biomass burning using the measured levoglucosan. Then the slope and r2 of the relationship between the resultant OCnon-bb and levoglucosan was calculated to find the one that gave the minimum? This may need further clarification in the text, including indicating which ratio were used in Fig 12.

If that is correct why did you settle on the ratio for winter (27.8) when it did not give a slope of zero unlike observed in the spring (Fig 12)? Why not try a higher ratio to get to zero?

Reply: Thank for the reviewer's suggestions. We have selected a different OC/levoglucosan ratios of softwood burning from the literature (Fig. 11) to calculate the OC from biomass burning (OC_{BB}) from multiplying with measured levoglucosan concentration. The highest value of OC/levoglucosan ratio of softwood burning was reported to be 27.6 from the previous chamber experiment as shown in Fig. 11. Thus, we did not include OC/levoglucasan ratio of higher than 27.6 for optimum OC/levoglucosan ratios estimation.

OC_{non-BB} is calculated by subtracting estimated OC_{BB} with measured total OC of each corresponding day. Then the slope and R^2 of the relationship between resultant OC_{non-BB} and measured levoglucosan concentration is calculated to find the one OC/levoglucosan ratio that gave the minimum. That OC/levoglucosan ratio is treated as optimized ratio. Finally, this optimized OC/levoglucosan ratio is used for estimating OC_{BB} for this site. OC/levoglucosan ratios of 27.6 and 18.0 have optimized for winter and spring, respectively, for this site to estimate OC_{BB}. We have modified phrase in section 3.5 for clarity about the approach for obtaining optimized OC/levoglucosan ratio.

Following sentences have been added and modified:

"We have used a range of different OC/levoglucosan ratios obtained from previous literatures (Fig. 11) for regression analysis with measured levoglucosan concentrations to estimate optimum OC/levoglucosan ratio (Fig. 12a, b)."

"The OC_{BB} concentrations at the Ulaanbaatar study site were calculated from the optimized OC/levoglucosan ratios (winter: 27.6 and spring: 18.0) and measured levoglucosan concentrations."

Please see line number 372-375, 379, 381, 395-397

Report #2

Comments:

I was Referee #2 for the ACPD manuscript. The authors have reasonably addressed the first 3 of my 4 major comments. As to my major comment 4, reference to the study of Davy et al. (full reference: Davy, P.K., Gunchin, G., Markwitz, A., Trompetter, W.J., Barry, B.J., Shagjjamba, D., Lodoysamba, S., 2011. Air particulate matter pollution in Ulaanbaatar, Mongolia: determination of composition, source contributions and source locations, Atmos. Poll. Res., 2, 126-137) should definitely be made. The authors' explanation for the large discrepancy between the contribution from biomass burning in PM2.5 in that study and in the current study is not very convincing. But if the authors do not want to elaborate on this discrepancy in their manuscript, they should at least refer to Davy et al. (2011) in the Introduction as an earlier study in Ulaanbatar, together with the studies of Jung et al. (2010) and Batmunkh et al. (2013), thus in lines 91-92. Since OC was not measured in Davy et al. (2011), "organic aerosol" in line 91 could then be replaced by "aerosol". Also, further within the manuscript's section 3.1, the authors could compare the PM2.5 K data of Davy et al. (2011) and the PM2.5 OC data of Jung et al. (2010) and of Batmunkh et al. (2013) with their own data.

Reply: Thank for the reviewers' comments and suggestion for improving the quality of manuscript. Please refer to the revised manuscript where we highlighted the changes by green color. We provide here below a reply to the specific comments and modifications made in the revised manuscript based on the line number provided in RMS. Please follow the line numbers to reviewing the changes in RMS.

We have replaced "organic aerosol" in revised manuscript by "aerosol". We have modified and added following sentences in revised manuscript as per reviewer suggestions:

"Few studies have investigated the chemical characteristics of aerosol in Ulaanbaatar (Jung et al., 2010; Davy et al., 2011; Batmunkh et al., 2013), with none examining the contribution of OCBB and type of biomass."

Please see line number 93-96.

"Previously, OC has been observed as major component in $PM_{2.5}$ in Ulaanbaatar during winter period (Jung et al., 2010; Batmunkh et al., 2013)."

Please see line number 194-195.

"Average K^+ concentration (0.08±0.05 $\mu g\ m^{-3}$) in this study is significantly lower than the K (0.32 $\mu g\ m^{-3}$) observed in Ulaanbaatar during 2004-2008 (Davy et al., 2011)."

Please see line number 228-230.

Besides, I have the following (minor) comments:

Reply: We thank to reviewer for his appreciable comments. We have attempted all the suggestions and modified the revised manuscript accordingly.

1. *Line 33: replace "study site" by "study sites".*

   Reply: Replacement done, please see line number 33.

2. *Line 48: replace "power plant" by "power plants".*

   Reply: Replacement done, please see line number 48.

3. *Line 60: replace "than those" by "than that".*

    Reply: Replacement done, please see line number 61.

4. *Line 82: replace "A half of" by "Half of the".*

    Reply: Replacement done, please see line number 85.

5. *Line 83: replace "dwelling)" by "dwellings)".*

    Reply: Replacement done, please see line number 86.

6. *Line 85: replace "birch these" by "birch; these".*

    Reply: Replacement done, please see line number 88.

7. *Line 89: replace "is very few estimate" by "are very few estimates".*

    Reply: Replacement done, please see line number 92-93.

8. *Line 93: replace "and identify" by "and identified".*

    Reply: Replacement done, please see line number 97.

9. *Line 120: replace "analyte has" by "analytes has".*

    Reply: Replacement done, please see line number 123.

10. *Line 136: I disagree with the authors' reply that '"were analyzed" is more appropriate thus we retain it in revised MS'; samples are analyzed, but species, as is the case here, are measured; therefore, "were analyzed" should be replaced by "were measured".*

    Reply: Replacement done, please see line number 141.

11. *Line 164: replace "percentile of" by "percentiles of".*

    Reply: Replacement done, please see line number 169.

12. *Line 173: replace "PC)" by "PCs)".*

    Reply: Replacement done, please see line number 178.

13. *Line 204: replace "5 that can" by "5; this can".*

    Reply: Replacement done, please see line number 211.

14. *Line 208: replace "plant (Pei" by "plants (Pei".*

    Reply: Replacement done, please see line number 215.

15. *Line 255: replace "results showed" by "results show".*

    Reply: Replacement done, please see line number 266.

16. *Line 270: replace "as biomass burning tracers in" by "as a biomass burning tracer in".*

    Reply: Replacement done, please see line number 282.

17. *Line 276: replace "tended to" by "tends to".*
    Reply: Replacement done, please see line number 288.

18. *Line 284: replace "belongs to" by "belong to".*
    Reply: Replacement done, please see line number 296.

19. *Line 359: replace "experiment (Cheng" by "experiments (Cheng".*
    Reply: Replacement done, please see line number 377.

20. *Line 370: replace "thus it has lower" by "thus have lower".*
    Reply: Replacement done, please see line number 388.

21. *Line 433: replace "mainly to" by "mainly due to".*
    Reply: Replacement done, please see line number 453.

22. *Line 433: replace "Ashbaugh, L.L., Malm, W.C., Sadeh, W.Z." by "Ashbaugh, L. L., Malm, W. C., and Sadeh, W. Z."*
    Reply: Replacement done, please see line number 502-504.

23. *Lines 584-588: "Guttikanda, S., 2008" should come before "Guttikanda and Jawahar, 2014"; furthermore, for "Guttikanda, S., 2008", the authors should insert "SIM Working Paper No. 2008-005," after "Ulaanbaatar, Mongolia, "*
    Reply: Modification done, please see line number 615-619.

24. *Line 687: replace "Q. and Yan" by "Q., and Yan".*
    Reply: Modification done, please see line number 718.