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

Jayant Nirmalkar[1], Tsatsral Batmunkh[2], Jinsang Jung[1,*]

[1]Center for Gas Analysis, Korea Research Institute of Standards and Science (KRISS), Daejeon 34113, Republic of Korea

[2]Department of Green Development Policy and Planning, Ministry of Environment and Tourism, UIaanbaatar-15160, Mongolia

Correspondence to: Jinsang Jung (jsjung@kriss.re.kr)

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 $OC_{non\text{-}BB}$ (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 site to quantify OC_{BB} using their own chemical measurements. In addition to OC_{BB}, sources of $OC_{non\text{-}BB}$ were also investigated through multivariate correlation analysis. It was found that $OC_{non\text{-}BB}$ 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 (Sullivan et al., 2019). Coal combustion, thermal power plant, 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 those 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).

A half of residents in Ulaanbaatar lives in 160,000 Gers (traditional Mongolian dwelling) (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 is very few estimate of OC emitted from biomass burning (OC_{BB}) in Ulaanbaatar. Few studies have investigated the chemical characteristics of organic aerosol in Ulaanbaatar (Jung et al., 2010; 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 identify 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 analyte 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 analyzed 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 $\Delta\Theta$, and $m_{\Delta\Theta}$ is the number of times that the source contribution peaked while the wind passed through sector $\Delta\Theta$ (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 $\Delta\Theta$ was set at 10°. To calculate $m_{\Delta\Theta}$, the 75th percentile 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, PC) 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). 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 that 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 plant (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.

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 showed 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 tracers 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 tended 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 belongs 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

303 and cooking purposes. Previous studies have used levoglucosan/mannosan ratios to
304 investigate the BB fuel types (Cheng et al., 2013; Jung et al., 2014).
305 However, the levoglucosan/mannosan ratio can't distinguish crop residuals (29 ± 15)
306 (Sheesley et al., 2003, Sullivan et al., 2008, Engling et al., 2009, Oanh et al., 2011) and
307 hardwood (28 ± 28) (Fine et al. 2001, 2002, 2004a, b; Engling et al., 2006; Schmidl et al.,
308 2008; Bari et al., 2009; Goncalves et al., 2010) due to the overlap of ratios between these
309 fuel types (Cheng et al., 2013; Fine et al. 2001, 2002, 2004a, b; Engling et al., 2006).
310 However, levoglucosan/K^+ ratio can distinguish between the two groups (Jung et al., 2014,
311 Chen et al., 2018). Both levoglucosan/mannosan and levoglucosan/K^+ ratios are therefore
312 useful in distinguishing various types of fuel (Cheng et al., 2013; Puxbaum et al., 2007).
313 A levoglucosan/mannosan–levoglucosan/K^+ scatter plot based on results of the
314 present and previous studies is shown in Fig. 10, using data from Schauer et al. (2001),
315 Fine et al. (2001, 2002, 2004a, b), and Engling et al. (2006) for hardwood grown in the
316 USA; Schauer et al. (2001), Hays et al. (2002), Fine et al. (2001, 2002, 2004a, b), and
317 Engling et al. (2006) for US softwood; Schmidl et al. (2008), Bari et al. (2009) and
318 Goncalves et al. (2010) for hardwood grown in Europe; Iinuma et al. (2007), Schmidl et
319 al. (2008), and Goncalves et al. (2010) for European softwood; Engling et al. (2006) and
320 Sullivan et al. (2008) for needles and duff found in the USA; Sullivan et al. (2008) for US
321 grass; and from Sheesley et al. (2003), Sullivan et al. (2008), Engling et al. (2009) and
322 Oanh et al. (2011) for Asian rice straw.
323 The average levoglucosan/mannosan ratio was 3.6 ± 0.2 (range: 3.4 − 4.1) in winter
324 and 4.1 ± 1.0 (2.12 − 7.05) in spring, whereas the levoglucosan/K^+ ratio was 8.9 ± 1.8
325 (5.5 − 12.4) in winter and 4.2 ± 2.1 (0.58 – 7.49) in spring at the study site (Fig. 10),
326 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. First, candidate OC_{BB} (Fig. 11) in this study was estimated from OC/levoglucosan ratios for softwood burning in a previous chamber experiment (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 levoglucosan concentration at this site. Second, OC_{non-BB} concentration was calculated by subtracting OC_{BB} from corresponding total OC. If calculated OC_{non-BB} doesn't contain OC emitted from biomass burning, both regression slope and R^2 between OC_{non-BB} versus levoglucosan will be close to zero. As shown in Fig. 12, 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 it has 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 and 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 $OC_{non\text{-}BB}$ 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 due mainly 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.

Acknowledgments

This work was funded by a grant (19011057) from the Korea Research Institute of Standards and Science (KRISS) under the Basic R&D Project of Quantification of local and long-range transported pollutants during a severe haze episode over the Korean Peninsula. The authors gratefully acknowledge the NOAA Air Resources Laboratory for the provision of the HYSPLIT transport and dispersion model and access to the READY website (http://www.arl.noaa.gov/ready.html) and the Fire Information for Resource Management System (FIRMS) of the National Aeronautics and Space Administration (NASA), United States (https://firms.modaps.eosdis.nasa.gov/alerts/) used in this study.

Data availability

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

References

Achad, M., Caumo, S., de Castro Vasconcellos, P., Bajano, H., Gómez, D., and Smichowski, P.: Chemical markers of biomass burning: Determination of levoglucosan, and potassium in size-classified atmospheric aerosols collected in Buenos Aires, Argentina by different analytical techniques, Microchem. J., 139, 181−187, https://doi.org/10.1016/j.microc.2018.02.016, 2018.

Acton, W. J. F., Schallhart, S., Langford, B., Valach, A., Rantala, P., Fares, S., and Carriero, G.: Canopy−scale flux measurements and bottom-up emission estimates of volatile organic compounds from a mixed oak and hornbeam forest in northern Italy, Atmos. Chem. Phys., 16, 7149−7170, https://doi.org/10.5194/acp-16-7149-2016, 2016.

Alastuey, A., Querol, X., Chaves, A., Ruiz, C. R., Carratala, A., and Lopez−Soler, A.: Bulk deposition in a rural area located around a large coal-fired power station, northeast Spain, Environ. Pollut., 106(3), 359−367, https://doi.org/10.1016/S0269-7491(99)00103-7, 1999.

Allan, J. D., Morgan, W. T., Darbyshire, E., Flynn, M. J., Williams, P. I., Oram, D. E., Artaxo, P., Brito, J., Lee, J. D., and Coe, H.: Airborne observations of IEPOX-derived isoprene SOA in the Amazon during SAMBBA, Atmos. Chem. Phys., 14, 11393–11407, https://doi.org/10.5194/acp-14-11393-2014, 2014.

Ashbaugh, L.L., Malm, W.C., Sadeh, W.Z.: A residence time probability analysis of sulfur concentrations at Grand Canyon National Park, Atmos. Environ., 19(8), 1263−1270, https://doi.org/10.1016/0004-6981(85)90256-2, 1985.

Bari, M. A., Baumbach, G., Kuch, B., and Scheffknecht, G.: Wood smoke as a source of particle-phase organic compounds in residential areas, Atmos. Environ., 43, 4722−4732, https://doi.org/10.1016/j.atmosenv.2008.09.006, 2009.

Batmunkh, T., Kim, Y. J., Jung, J. S., Park, K., and Tumendemberel, B.: Chemical characteristics of fine particulate matters measured during severe winter haze events in Ulaanbaatar, Mongolia, J. Air & Waste Manag. Assoc., 63, 659−670, https://doi.org/10.1080/10962247.2013.776997, 2013.

Caiazzo, F., Ashok, A., Waitz, I. A., Yim, S. H., and Barrett, S. R.: Air pollution and early deaths in the United States. Part I: Quantifying the impact of major sectors in 2005, Atmos. Environ., 79, 198−208, https://doi.org/10.1016/j.atmosenv.2013.05.081, 2013.

Cao, J. J., Wu, F., Chow, J. C., Lee, S. C., Li, Y., Chen, S. W., An, Z. S., Fung, K. K., Watson, J. G., Zhu, C. S., and Liu, S. X.: Characterization and source apportionment of atmospheric organic and elemental carbon during fall and winter of 2003 in Xi'an, China, Atmos. Chem. Phys., 5, 3127−3137, https://doi.org/10.5194/acp-5-3127-2005, 2005.

Chantara, S., Thepnuan, D., Wiriya, W., Prawan, S., and Tsai, Y. I.: Emissions of pollutant gases, fine particulate matters and their significant tracers from biomass burning in an open-system combustion chamber, Chemosphere, 224, 407−416, 2019.

Chen, H., Yin, S., Li, X., Wang, J., and Zhang, R.: Analyses of biomass burning contribution to aerosol in Zhengzhou during wheat harvest season in 2015, Atmos. Res., 207, 62−73, https://doi.org/10.1016/j.atmosres.2018.02.025, 2018.

Cheng, Y., Engling, G., He, K.-B., Duan, F.-K., Ma, Y.-L., Du, Z.-Y., Liu, J.-M., Zheng, M., and Weber, R. J.: Biomass burning contribution to Beijing aerosol, Atmos. Chem. Phys., 13, 7765−7781, https://doi.org/10.5194/acp-13-7765-2013, 2013.

Chung, S. and Chon, H. T.: Assessment of the level of mercury contamination from some anthropogenic sources in Ulaanbaatar, Mongolia, J. Geochem. Explor., 147, 237−244, https://doi.org/10.1016/j.gexplo.2014.07.016, 2014.

Claeys, M., Kourtchev, I., Pashynska, V., Vas, G., Vermeylen, R., Wang, W., Cafmeyer, J., Chi, X., Artaxo, P., Andreae, M. O., and Maenhaut, W.: Polar organic marker compounds in atmospheric aerosols during the LBA-SMOCC 2002 biomass burning experiment in Rondônia, Brazil: sources and source processes, time series, diel variations and size distributions, Atmos. Chem. Phys., 10, 9319−9331, https://doi.org/10.5194/acp-10-9319-2010, 2010.

Deshmukh, D. K., Deb, M. K., Tsai, Y. I., and Mkoma, S. L.: Water soluble ions in PM2.5 and PM1 aerosols in Durg city, Chhattisgarh, India, Aerosol Air Qual. Res, 11, 696-708, 10.4209/aaqr.2011.03.0023, 2011.

Deshmukh, D. K., Haque, M. M., Kim, Y., and Kawamura, K.: Organic tracers of fine aerosol particles in central Alaska: summertime composition and sources, Atmos. Chem. Phys., 19, 14009–14029, https://doi.org/10.5194/acp-19-14009-2019, 2019.

Duan, F., Liu, X., Yu, T., and Cachier, H.: Identification and estimate of biomass burning contribution to the urban aerosol organic carbon concentrations in Beijing, Atmos. Environ. 38, 1275−1282, https://doi.org/10.1016/j.atmosenv.2003.11.037, 2004

Engling, G., Carrico, C. M., Kreidenweis, S. M., Collett Jr., J. L., Day, D. E., Malm, W. C., Lincoln, L., Hao, W. M., Iinuma, Y., and Herrmann, H.: Determination of levoglucosan in biomass combustion aerosol by high-performance anion-exchange chromatography with pulsed amperometric detection, Atmos. Environ., 40, 299–311, https://doi.org/10.1016/j.atmosenv.2005.12.069, 2006.

Engling, G., Lee, J. J., Tsai, Y. W., Lung, S. C. C., Chou, C. C. K., and Chan, C. Y.: Size resolved anhydrosugar composition in smoke aerosol from controlled field burning of rice straw, Aerosol Sci. Technol., 43, 662–672, https://doi.org/10.1080/02786820902825l1, 2009.

Fang, G. C., Chang, C. N., Chu, C. C., Wu, Y. S., Fu, P. P. C., Yang, I. L., and Chen, M. H.: Characterization of particulate, metallic elements of TSP, $PM_{2.5}$ and $PM_{2.5-10}$ aerosols at a farm sampling site in Taiwan, Taichung, Sci. Tot. Environ., 308, 157–166, https://doi.org/10.1016/S0048-9697(02)00648-4, 2003.

Fehsenfeld, F., Calvert, J., Fall, R., Goldan, P., Guenther, A. B., Hewitt, C. N., Lamb, B., Liu, S., Trainer, M., Westberg, H., and Zimmerman, P.: Emissions of volatile organic compounds from vegetation and the implications for atmospheric chemistry, Global Biogeochem. Cy., 6, 389–430, https://doi.org/10.1029/92GB02125, 1992.

Feng, Y., Chen, Y., Guo, H., Zhi, G., Xiong, S., Li, J., Sheng, G., and Fu, J.: Characteristics of organic and elemental carbon in $PM_{2.5}$ samples in Shanghai, China, Atmos. Res., 92, 434–442, https://doi.org/10.1016/j.atmosres.2009.01.003, 2009.

Fine, P. M., Cass, G. R., and Simoneit, B. R. T.: Chemical characterization of fine particle emissions from fireplace combustion of woods grown in the northeastern United States, Environ. Sci. Technol., 35, 2665–2675, https://doi.org/10.1021/es001466k, 2001.

Fine, P. M., Cass, G. R., and Simoneit, B. R. T.: Chemical characterization of fine particle emissions from the fireplace combustion of woods grown in the southern United States, Environ. Sci. Technol., 36, 1442–1451, https://doi.org/10.1021/es0108988, 2002.

Fine, P. M., Cass, G. R., and Simoneit, B. R. T.: Chemical characterization of fine particle emissions from the fireplace combustion of wood types grown in the midwestern and western United States, Environ. Engin. Sci., 21, 387–409, https://doi.org/10.1089/109287504323067021, 2004a.

559 Fine, P. M., Cass, G. R., and Simoneit, B. R. T.: Chemical characterization of fine particle
560 emissions from the wood stove combustion of prevalent United States tree species,
561 Environ. Engin. Sci., 21, 705−721, https://doi.org/10.1089/ees.2004, 21.705, 2004b.
562 Flues, M., Hama, P., Lemes, M. J. L., Dantas, E. S. K., and Fornaro, A.: Evaluation of the
563 rainwater acidity of a rural region due to a coal-fired power plant in Brazil, Atmos.
564 Environ., 36, 2397−2404, https://doi.org/10.1016/S1352-2310(01)00563-5, 2002.
565 Fu, P. Q., Kawamura, K., Chen, J., Li, J., Sun, Y. L., Liu, Y., Tachibana, E., Aggarwal, S.
566 G., Okuzawa, K., Tanimoto, H., and Kanaya, Y.: Diurnal variations of organic
567 molecular tracers and stable carbon isotopic composition in atmospheric aerosols
568 over Mt. Tai in the North China Plain: an influence of biomass burning, Atmos.
569 Chem. Phys., 12, 8359−8375, doi:10.5194/acp-12-8359-2012, 2012.
570 Gao, S., Hegg D. A., Hobbs P. V., Kirchstetter T. W., Magi B. I., and Sadilek M.: Water-
571 soluble organic components in aerosols associated with savanna fires in southern
572 Africa: Identification, evolution, and distribution, J. Geophys. Res., 108(D13), 8491,
573 doi:10.1029/2002JD002324, 2003.
574 Garcia, J., Beyne-Masclet, S., Mouvier, G., and Masclet, P.: Emissions of volatile organic
575 compounds by coal-fired power stations. Atmos. Environ., Part A, 26, 1589−1597,
576 https://doi.org/10.1016/0960-1686(92)90059-T, 1992.
577 Gonçalves, C., Alves, C., Evtyugina, M., Mirante, F., Pio, C., Caseiro, A., Schmidl, C.,
578 Bauer, H., and Carvalho, F.: Characterisation of PM_{10} emissions from wood stove
579 combustion of common woods grown in Portugal, Atmos. Environ., 44, 4474−4480,
580 https://doi.org/10.1016/j.atmosenv.2010.07.026, 2010.
581 Gu, J., Bai, Z., Li, W., Wu, L., Liu, A., Dong, H., and Xie, Y.: Chemical composition of
582 $PM_{2.5}$ during winter in Tianjin, China, Particuology, 9, 215−221,
583 https://doi.org/10.1016/j.partic.2011.03.001, 2011.
584 Guttikunda, S. K. and Jawahar, P.: Atmospheric emissions and pollution from the
585 coal−fired thermal power plants in India, Atmos. Environ., 92, 449−460,
586 https://doi.org/10.1016/j.atmosenv.2014.04.057, 2014.
587 Guttikunda, S.: Urban air pollution analysis for Ulaanbaatar, Mongolia,
588 http://dx.doi.org/10.2139/ssrn.1288328, 2008.
589 Harrison, R. M., Beddows, D. C. S., Hu, L., and Yin, J.: Comparison of methods for
590 evaluation of wood smoke and estimation of UK ambient concentrations, Atmos.

Chem. Phys., 12, 8271−8283, doi:10.5194/acp−12−8271−2012, 2012.

Hays, M. D., Geron, C. D., Linna, K. J., Smith, N. D., and Schauer, J. J.: Speciation of gasphase and fine particle emissions from burning of foliar fuels, Environ. Sci. Technol., 36, 2281−2295, https://doi.org/10.1021/es0111683, 2002.

Haque, M., Kawamura, K., Deshmukh, D. K., Fang, C., Song, W., Mengying, B., and Zhang, Y. L.: Characterization of organic aerosols from a Chinese megacity during winter: predominance of fossil fuel combustion. Atmos. Chem. Phys., 19, 5147-5164, https://doi.org/10.5194/acp-19-5147-2019, 2019.

Huang, X., Liu, Z., Zhang, J., Wen, T., Ji, D., and Wang, Y.: Seasonal variation and secondary formation of size-segregated aerosol water-soluble inorganic ions during pollution episodes in Beijing, Atmos. Res., 168, 70−79, https://doi.org/10.1016/j.atmosres.2015.08.021, 2016.

Iinuma, Y., Brüggemann, E., Gnauk, T., Müller, K., Andreae, M. O., Helas, G., Parmar, R., and Herrmann, H.: Source characterization of biomass burning particles: the combustion of selected European conifers, African hardwood, savanna grass, and German and Indonesian peat, J. Geophys. Res., 112, D08209. http://dx.doi.org/10.1029/2006JD007120, 2007.

Ji, D., Zhang, J., He, J., Wang, X., Pang, B., Liu, Z., Wang, L., and Wang, Y.: Characteristics of atmospheric organic and elemental carbon aerosols in urban Beijing, China, Atmos. Environ., 125, 293−306, https://doi.org/10.1016/j.atmosenv.2015.11.020, 2016.

Jimenez, J. L., Canagaratna, M. R., Donahue, N. M., Prevot, A. S. H., Zhang, Q., Kroll, J. H., DeCarlo, P. F., Allan, J. D., Coe, H., Ng, N. L., and Aiken, A. C.: Evolution of organic aerosols in the atmosphere, Science, 326, 1525−1529, doi:10.1126/science.1180353, 2009.

Jung, J., Lee, H., Kim ,Y. J., Liu, X., Zhang, Y., Hu, M., and Sugimoto, N.: Optical properties of atmospheric aerosols obtained by in situ and remote measurements during 2006 Campaign of Air Quality Research in Beijing (CAREBeijing-2006), J. Geophys. Res., 114, D00G02, doi:10.1029/2008JD010337, 2009.

Jung, J., Tsatsral, B., Kim, Y. J., and Kawamura, K.: Organic and inorganic aerosol compositions in Ulaanbaatar, Mongolia, during the cold winter of 2007 to 2008: dicarboxylic acids, ketocarboxylic acids, and α-dicarbonyls, J. Geophys. Res.:

Atmos., 115, D22203, https://doi.org/10.1029/2010JD014339, 2010.

Jung, J., Lee, S., Kim, H., Kim, D., Lee, H., and Oh, S.: Quantitative determination of the biomass-burning contribution to atmospheric carbonaceous aerosols in Daejeon, Korea, during the rice-harvest period, Atmos. Environ., 89, 642−650, https://doi.org/10.1016/j.atmosenv.2014.03.010, 2014.

Khan, M. F., Shirasuna, Y., Hirano, K., and Masunaga, S.: Characterization of $PM_{2.5}$, $PM_{2.5–10}$ and $PM_{>10}$ in ambient air, Yokohama, Japan, Atmos. Res., 96, 159-172, https://doi.org/10.1016/j.atmosres.2009.12.009, 2010.

Klejnowski, K., Janoszka, K., and Czaplicka, M.: Characterization and seasonal variations of organic and elemental carbon and levoglucosan in PM10 in Krynica Zdroj, Poland, Atmosphere, 8, 190, doi:10.3390/atmos8100190, 2017.

Lai, C., Liu, Y., Ma, J., Ma, Q., and He, H.: Degradation kinetics of levoglucosan initiated by hydroxyl radical under different environmental conditions, Atmos. Environ., 91, 32−39, https://doi.org/10.1016/j.atmosenv.2014.03.054, 2014.

Lee, T., Sullivan, A. P., Mack, L., Jimenez, J. L., Kreidenweis, S. M., Onasch, T. B., Worsnop, D. R., Malm, W., Wold, C. E., Hao, W. M., and Collett Jr, J. L.: Chemical smoke marker emissions during flaming and smoldering phases of laboratory open burning of wildland fuels, Aerosol Sci. Technol., 44, i-v, https://doi.org/10.1080/02786826.2010.499884, 2010.

Li, Q., Jiang, J., Zhang, Q., Zhou, W., Cai, S., Duan, L., Ge, S., and Hao, J.: Influences of coal size, volatile matter content, and additive on primary particulate matter emissions from household stove combustion. Fuel, 182, 780-787, https://doi.org/10.1016/j.fuel.2016.06.059, 2016.

Liang, L., Engling, G., Du, Z., Cheng, Y., Duan, F., Liu, X., and He, K.: Seasonal variations and source estimation of saccharides in atmospheric particulate matter in Beijing, China, Chemosphere, 150, 365−377, https://doi.org/10.1016/j.chemosphere.2016.02.002, 2016.

Lin, Y.-C., Hsu, S.-C., Lin, C.-Y., Lin, S.-H., Huang, Y.-T., Chang, Y., and Zhang, Y.-L.: Enhancements of airborne particulate arsenic over the subtropical free troposphere: impact of southern Asian biomass burning, Atmos. Chem. Phys., 18, 13865-13879, https://doi.org/10.5194/acp-18-13865−2018, 2018.

Louie, P. K., Watson, J. G., Chow, J. C., Chen, A., Sin, D. W., and Lau, A. K.: Seasonal

characteristics and regional transport of $PM_{2.5}$ in Hong Kong, Atmos. Environ., 39, 1695−1710, https://doi.org/10.1016/j.atmosenv.2004.11.017, 2005.

Maenhaut, W., Nava, S., Lucarelli, F., Wang, W., Chi, X., and Kulmala, M.: Chemical composition, impact from biomass burning, and mass closure for $PM_{2.5}$ and PM_{10} aerosols at Hyytiälä, Finland, in summer 2007, X-Ray Spectrom., 40, 168−171, https://doi.org/10.1002/xrs.1302, 2011.

Nirmalkar, J., Deshmukh, D. K., Deb, M. K., Tsai, Y. I., and Sopajaree, K.: Mass loading and episodic variation of molecular markers in $PM_{2.5}$ aerosols over a rural area in eastern central India, Atmos. Environ., 117, 41–50, https://doi.org/10.1016/j.atmosenv.2015.07.003, 2015.

Nirmalkar, J., Deshmukh, D. K., Deb, M. K., Tsai, Y. I., and Pervez, S.: Characteristics of aerosol during major biomass burning events over eastern central India in winter: A tracer-based approach, Atmos. Pollut. Res., 10, 817−826, https://doi.org/10.1016/j.apr.2018.12.010, 2019.

Oanh, N. T. K., Ly, B. T., Tipayarom, D., Manandhar, B. R., Prapat, P., Simpson, C.D., and Liu, L.J.S.: Characterization of particulate matter emission from open burning of rice straw, Atmos. Environ., 45, 493−502, https://doi.org/10.1016/j.atmosenv.2010.09.023, 2011.

Obrist, D., Moosmüller, H., Schürmann, R., Chen, L. W. A., and Kreidenweis, S. M.: Particulate-phase and gaseous elemental mercury emissions during biomass combustion: controlling factors and correlation with particulate matter emissions. Environ. Sci. Technol., 42, 721-727, https://doi.org/10.1021/es071279n, 2007.

Panda, S., Sharma, S. K., Mahapatra, P. S., Panda, U., Rath, S., Mahapatra, M., Mandal, T. K., and Das, T.: Organic and elemental carbon variation in $PM_{2.5}$ over megacity Delhi and Bhubaneswar, a semi-urban coastal site in India, Nat. Hazards, 80, 1709−1728, https://doi.org/10.1007/s11069-015-2049-3, 2016.

Park, S. S. and Kim, Y. J.: $PM_{2.5}$ particles and size-segregated ionic species measured during fall season in three urban sites in Korea, Atmos. Environ., 38, 1459–1471, https://doi.org/10.1016/j.atmosenv.2003.12.004, 2004.

Park, S. M., Seo, B. K., Lee, G., Kahng, S. H., and Jang, Y.: Chemical composition of water-soluble inorganic species in precipitation at Shihwa Basin, Korea, Atmosphere, 6, 732−750, https://doi.org/10.3390/atmos6060732, 2015.

Pei, B., Wang, X., Zhang, Y., Hu, M., Sun, Y., Deng, J., Dong, L., Fu, Q. and Yan N.: Emissions and source profiles of PM2.5 for coal-fired boilers in the Shanghai megacity, China, Atmos. Pollut. Res. 7, 577-584, https://doi.org/10.1016/j.apr.2016.01.005, 2016.

Pio, C. A., Legrand, M., Alves, C. A., Oliveira, T., Afonso, J., Caseiro, A., Puxbaum, H., Sanchez−Ochoa, A., and Gelencser, A.: Chemical composition of atmospheric aerosols during the 2003 summer intense forest fire period, Atmos. Environ., 42, 7530−7543, https://doi.org/10.1016/j.atmosenv.2008.05.032, 2008.

Puxbaum, H., Caseiro, A., Sánchez-Ochoa, A., Kasper-Giebl, A., Claeys, M., Gelencser, A., Legrand, M., Preunkert, S., and Pio, C.: Levoglucosan levels at background sites in Europe for assessing the impact of biomass combustion on the European aerosol background, J. Geophys. Res.: Atmos., 112, D23S05, https://doi.org/10.1029/2006JD008114, 2007.

Reche, C., Viana, M., Amato, F., Alastuey, A., Moreno, T., Hillamo, R., Teinila, K., Saarnio, K., Seco, R., Penuelas, J., and Mohr, C.: Biomass burning contributions to urban aerosols in a coastal Mediterranean City, Sci. Tot. Environ., 427, 175−190, https://doi.org/10.1016/j.scitotenv.2012.04.012, 2012.

Schauer, J.J., Kleeman, M.J., Cass, G.R., and Simoneit, B. R. T.: Measurement of emissions from air pollution sources. 3. C1-C29 organic compounds from fireplace combustion of wood, Environ. Sci. Technol., 35, 1716−1728, https://doi.org/10.1021/es001331e, 2001.

Schkolnik, G., Falkovich, A. H., Rudich, Y., Maenhaut, W., and Artaxo, P.: New analytical method for the determination of levoglucosan, polyhydroxy compounds, and 2-methylerythritol and its application to smoke and rainwater samples, Environ. Sci. Technol. 39, 2744−2752, https://doi.org/10.1021/es048363c, 2005.

Schmidl, C., Marr, I. L., Caseiro, A., Kotianová, P., Berner, A., Bauer, H., Kasper-Giebl, A., and Puxbaum, H.: Chemical characterisation of fine particle emissions from wood stove combustion of common woods growing in mid-European Alpine regions, Atmos. Environ., 42, 126−141, https://doi.org/10.1016/j.atmosenv.2007.09.028, 2008.

Shao, M., Czapiewski, K. V., Heiden, A. C., Kobel, K., Komenda, M., Koppmann, R., and Wildt, J.: Volatile organic compound emissions from Scots pine: mechanisms

and description by algorithms, J. Geophys. Res.: Atmos., 106(D17), 20483−20491, 10.1029/2000JD000248, 2001.

Sharma, A., Pareek, V., and Zhang, D.: Biomass pyrolysis—A review of modelling, process parameters and catalytic studies. J. Renew. Sustain. Energy, 50, 1081−1096, https://doi.org/10.1016/j.rser.2015.04.193, 2015.

Sheesley, R. J., Schauer, J. J., Chowdhury, Z., Cass, G. R., and Simoneit, B. R. T.: Characterization of organic aerosols emitted from the combustion of biomass indigenous to South Asia, J. Geophys. Res., 108, 4285, http:// dx.doi.org/10.1029/2002JD002981, 2003.

Simoneit, B. R., Schauer, J. J., Nolte, C. G., Oros, D. R., Elias, V. O., Fraser, M. P., Rogge, W. F., and Cass, G. R., 1999. Levoglucosan, a tracer for cellulose in biomass burning and atmospheric particles, Atmos. Environ., 33, 173−182, https://doi.org/10.1016/S1352-2310(98)00145-9, 1999.

Sullivan, A. P., Holden, A. S., Patterson, L. A., McMeeking, G. R., Kreidenweis, S. M., Malm, W. C., Hao, W. M., Wold, C. E., and Collett Jr., J. L.: A method for smoke marker measurements and its potential application for determining the contribution of biomass burning from wildfires and prescribed fires to ambient $PM_{2.5}$ organic carbon, J Geophys. Res., 113, D22302, http:// dx.doi.org/10.1029/2008JD010216, 2008.

Sullivan, A. P., Guo, H., Schroder, J. C., Campuzano-Jost, P., Jimenez, J. L., Campos, T., Shah, V., Jaegle, L., Lee, B. H., Lopez-Hilfiker, F. D., and Thornton, J. A.: Biomass burning markers and residential burning in the winter aircraft campaign. J. Geophys. Res.: Atmos., 124, 1846−1861, https://doi.org/10.1029/2017JD028153, 2019.

Sun, J., Shen, Z., Zhang, Y., Zhang, Q., Wang, F., Wang, T., Chang, X., Lei, Y., Xu, H., Cao, J., and Zhang, N.: Effects of biomass briquetting and carbonization on PM2.5 emission from residential burning in Guanzhong Plain, China, Fuel, 244, 379−387, https://doi.org/10.1016/j.fuel.2019.02.031, 2019.

Tao, J., Zhang, L., Ho, K., Zhang, R., Lin, Z., Zhang, Z., Lin, M., Cao, J., Liu, S., and Wang, G.: Impact of $PM_{2.5}$ chemical compositions on aerosol light scattering in Guangzhou—the largest megacity in South China, Atmos. Res., 135, 48−58, https://doi.org/10.1016/j.atmosres.2013.08.015, 2014.

Thepnuan, D., Chantara, S., Lee, C. T., Lin, N. H., and Tsai, Y. I.: Molecular markers for

biomass burning associated with the characterization of $PM_{2.5}$ and component sources during dry season haze episodes in Upper South East Asia, Sci. Tot. Environ., 658, 708−722, https://doi.org/10.1016/j.scitotenv.2018.12.201, 2019.

Verma, V., Fang, T., Guo, H., King, L., Bates, J. T., Peltier, R. E., Edgerton, E., Russell, A. G., and Weber, R. J.: Reactive oxygen species associated with water-soluble PM2.5 in the southeastern United States: spatiotemporal trends and source apportionment, Atmos. Chem. Phys., 14, 12915-12930, https://doi.org/10.5194/acp-14-12915-2014, 2014.

Wang, T., Tian, M., Ding, N., Yan, X., Chen, S. J., Mo, Y. Z., Yang, W. Q., Bi, X. H., Wang, X. M., and Mai, B. X.: Semivolatile organic compounds (SOCs) in fine particulate matter ($PM_{2.5}$) during clear, fog, and haze episodes in winter in Beijing, China, Environ. Sci. Tech., 52, 5199−5207, https://doi.org/10.1021/acs.est.7b06650, 2018.

Watson, J. G., Chow, J. C., and Fujita, E. M.: Review of volatile organic compound source apportionment by chemical mass balance, Atmos. Environ., 35, 1567−1584, https://doi.org/10.1016/S1352-2310(00)00461-1, 2001a.

Watson, J. G., Chow, J. C., and Houck, J. E.: $PM_{2.5}$ chemical source profiles for vehicle exhaust, vegetative burning, geological material, and coal burning in Northwestern Colorado during 1995, Chemosphere, 43, 1141−1151, https://doi.org/10.1016/S0045-6535(00)00171-5, 2001b.

Zhamsueva, G. S., Zayakhanov, A. S., Starikov, A. V., Balzhanov, T. S., Tsydypov, V. V., Dementyeva, A. L., and Khodzher, T. V.: Investigation of chemical composition of atmospheric aerosol in Ulaanbaatar during 2005–2014. Geography and Natural 39, 270−276, 10.1134/S1875372818030113, 2018

Zhang, F., Wang, Z. W., Cheng, H. R., Lv, X. P., Gong, W., Wang, X. M., and Zhang, G.: Seasonal variations and chemical characteristics of $PM_{2.5}$ in Wuhan, central China, Sci. Tot. Environ., 518, 97−105, https://doi.org/10.1016/j.scitotenv.2015.02.054, 2015.

Zhang, Y. X., Min, S., Zhang, Y. H., Zeng, L. M., He, L. Y., Bin, Z. H. U., Wei, Y. J., and Zhu, X. L.: Source profiles of particulate organic matters emitted from cereal straw burnings. J. Environ. Sci., 19, 167−175, https://doi.org/10.1016/S1001-0742(07)60027-8, 2007.

783 Zhou, Y., Levy, J. I., Hammitt, J. K., and Evans, J. S.: Estimating population exposure to
784 power plant emissions using CALPUFF: a case study in Beijing, China, Atmos.
785 Environ., 37, 815−826, https://doi.org/10.1016/S1352-2310(02)00937-8, 2003.
786 Zhu, C., Kawamura, K., and Kunwar, B.: Effect of biomass burning over the western
787 North Pacific Rim: wintertime maxima of anhydrosugars in ambient aerosols from
788 Okinawa, Atmos. Chem. Phys., 15, 1959−1973, https://doi.org/10.5194/acp-15-
789 1959-2015, 2015.
790
791
792

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

807 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

808

809

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).

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

826 Ulaanbaatar (blue circles and red squares).

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

828 Fig. 12 Graphical determination of optimized OC/levoglucosan ratios used to estimate $PM_{2.5}$ concentrations of OC_{BB} in Ulaanbaatar in

829 winter (a) and spring (b) of 2017.

830 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.

831

832

833

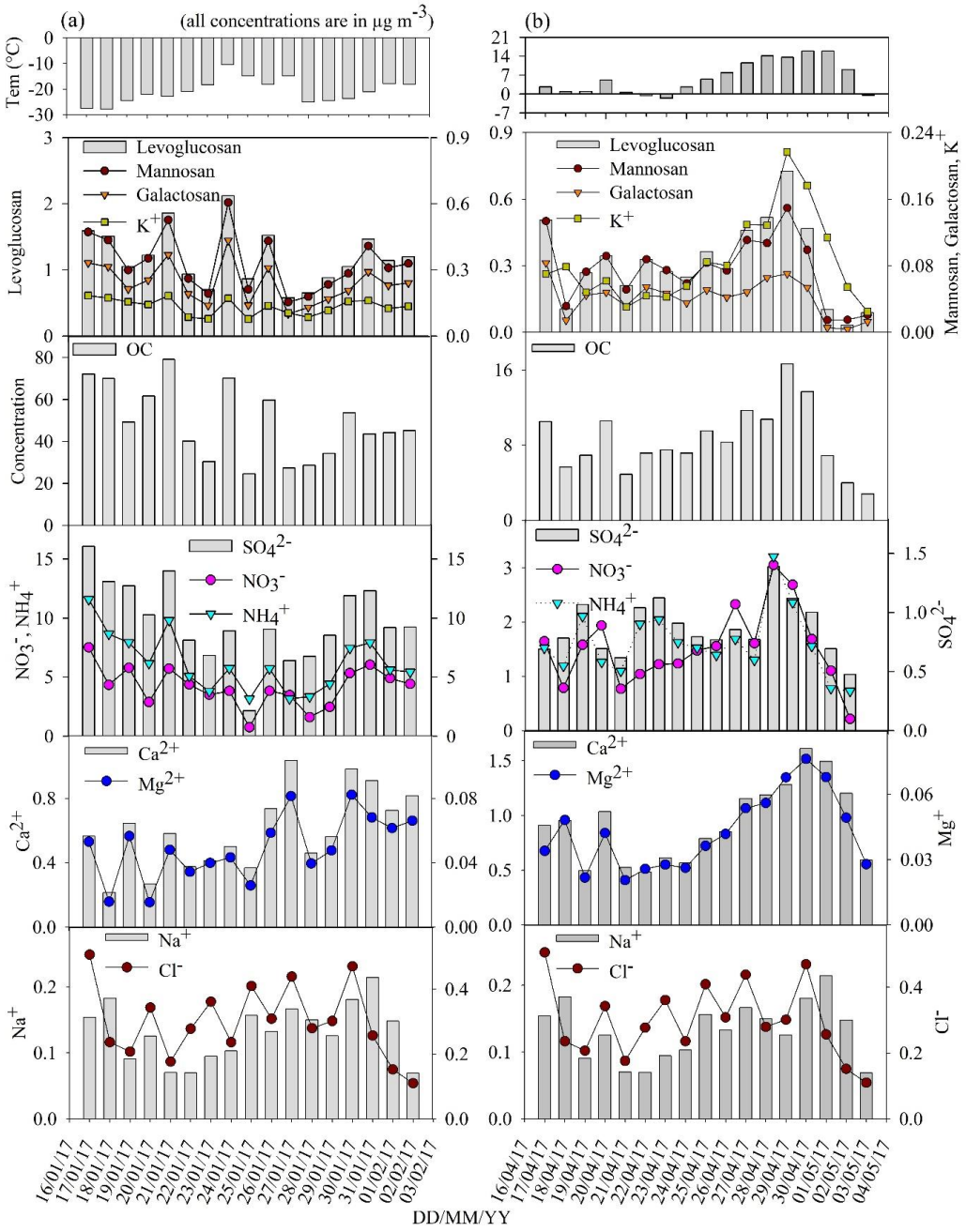
834

Fig. 1


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

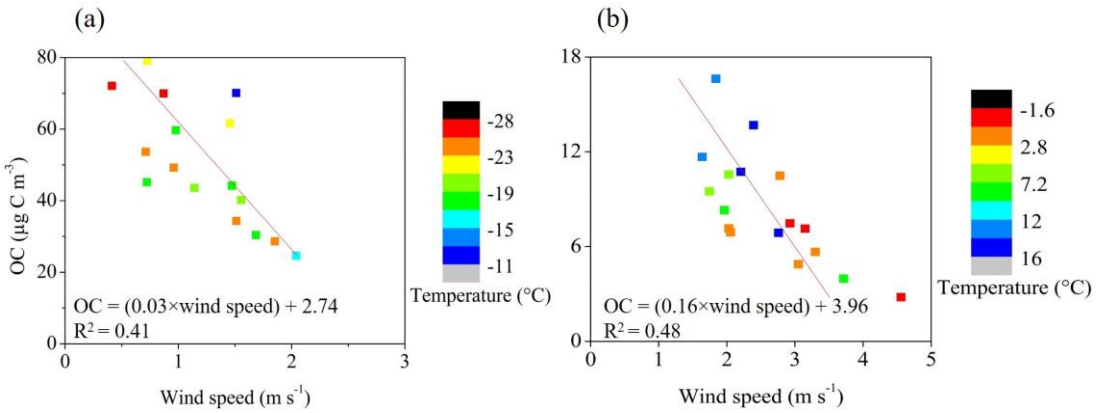
846
847

Fig. 2

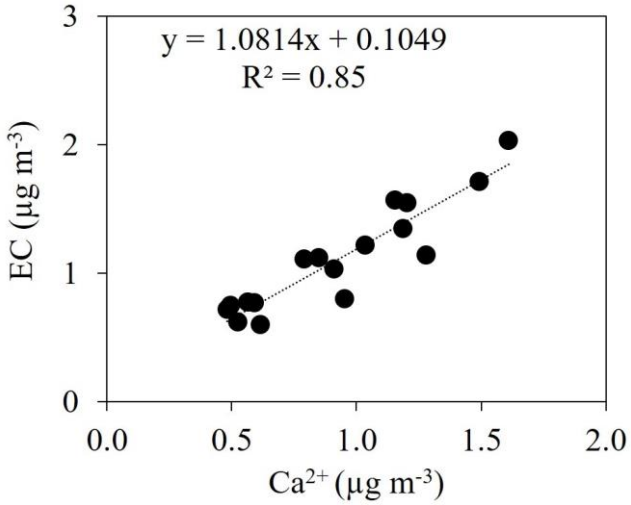


848

Fig. 3

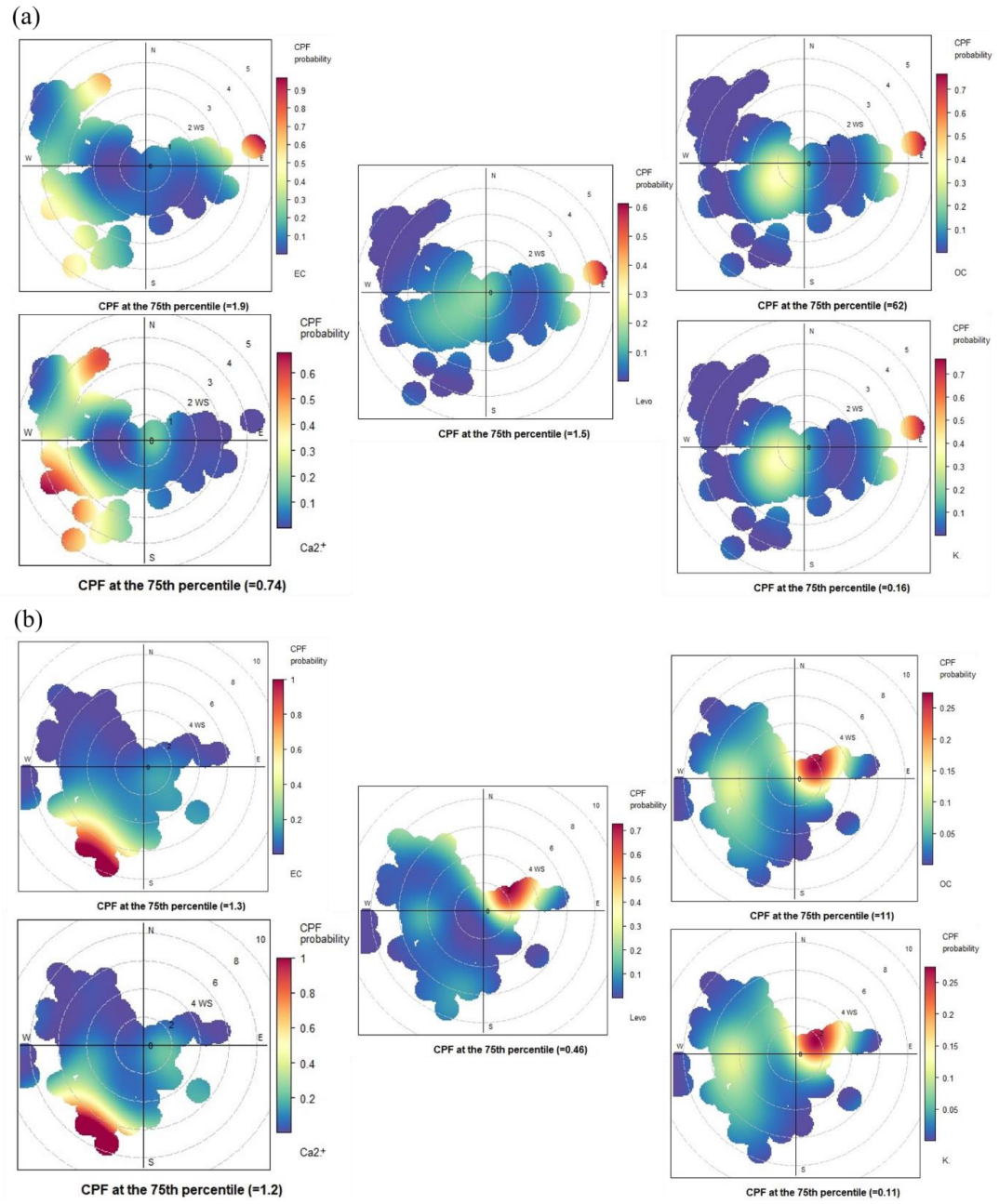

(a)
OC (μg C m-3)
OC = (0.03×wind speed) + 2.74
R2 = 0.41
Wind speed (m s-1)
-28
-23
-19
-15
-11
Temperature (°C)
(b)
OC = (0.16×wind speed) + 3.96
R2 = 0.48
Wind speed (m s-1)
-1.6
2.8
7.2
12
16
Temperature (°C)

857 Fig. 4
858
859

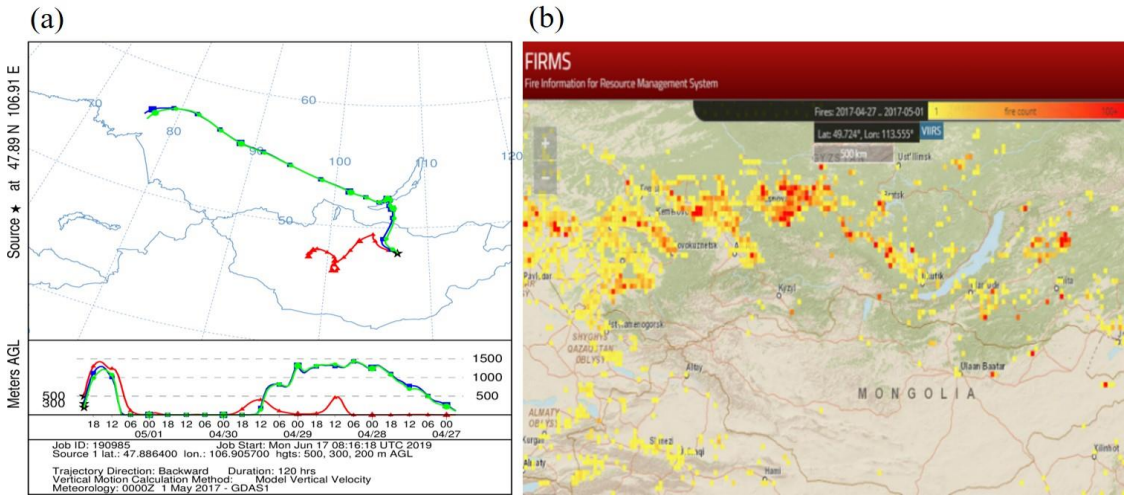


860
861
862

Fig. 5

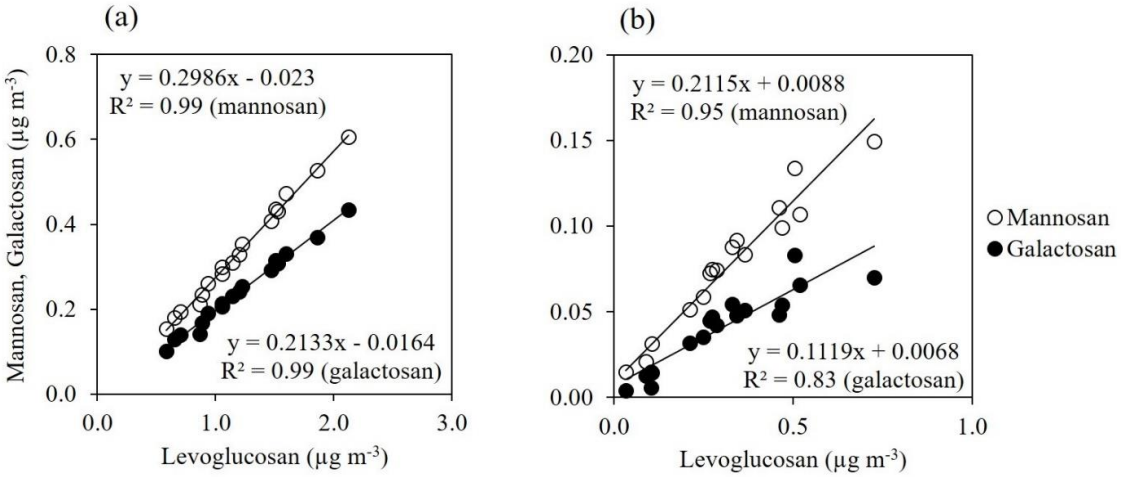

(a)
CPF probability
EC
CPF at the 75th percentile (=1.9)
Levo
CPF at the 75th percentile (=1.5)
OC
CPF at the 75th percentile (=62)
Ca2+
CPF at the 75th percentile (=0.74)
K
CPF at the 75th percentile (=0.16)
(b)
CPF at the 75th percentile (=1.3)
CPF at the 75th percentile (=0.46)
CPF at the 75th percentile (=11)
CPF at the 75th percentile (=1.2)
CPF at the 75th percentile (=0.11)

873 Fig. 6
874



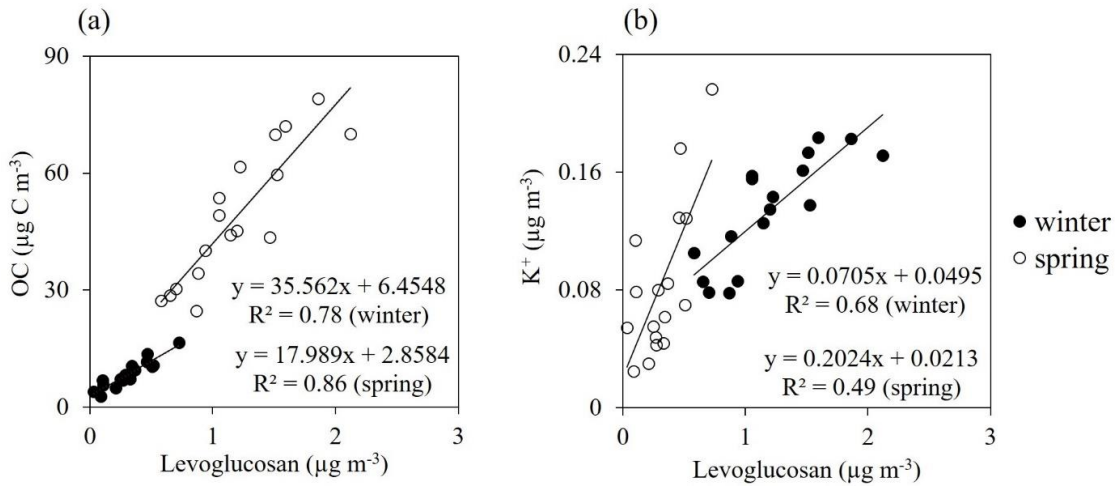
875
876
877

878 Fig. 7
879



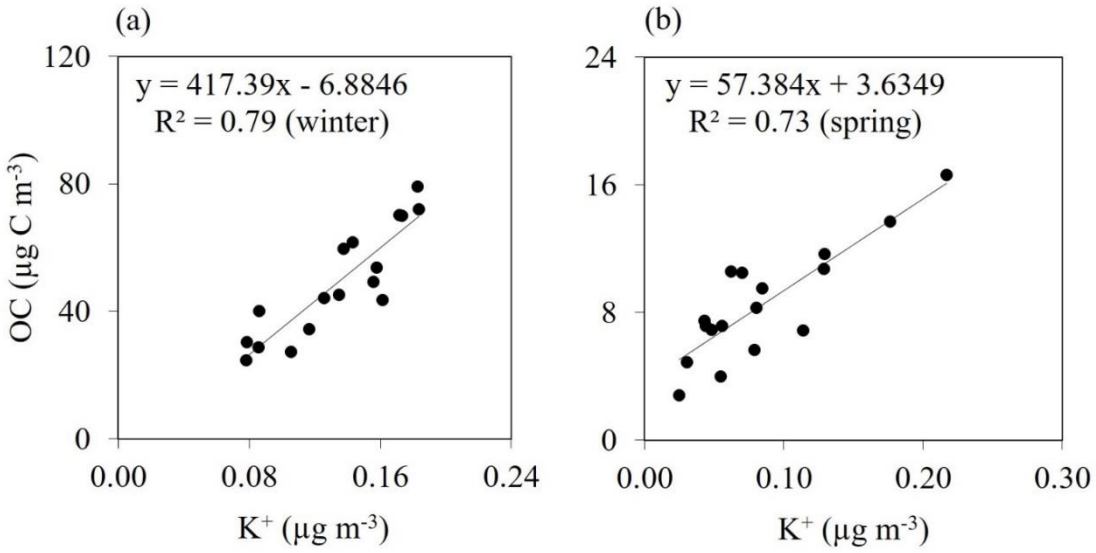
880
881

882 Fig. 8
883



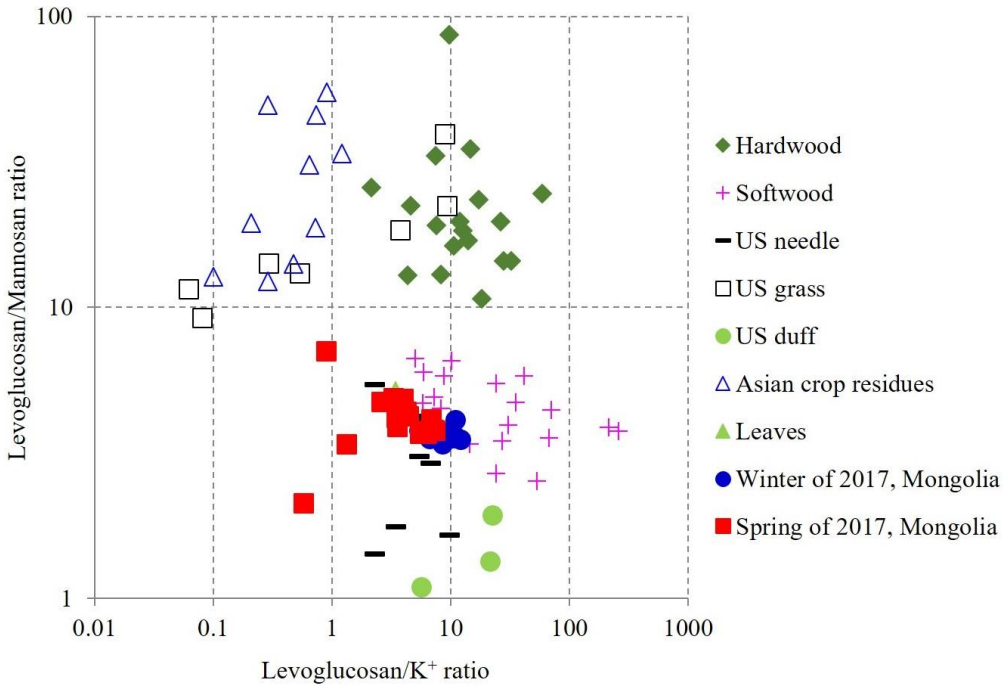
884
885

886 Fig. 9
887



888
889

890 Fig. 10
891



892
893

Fig. 11

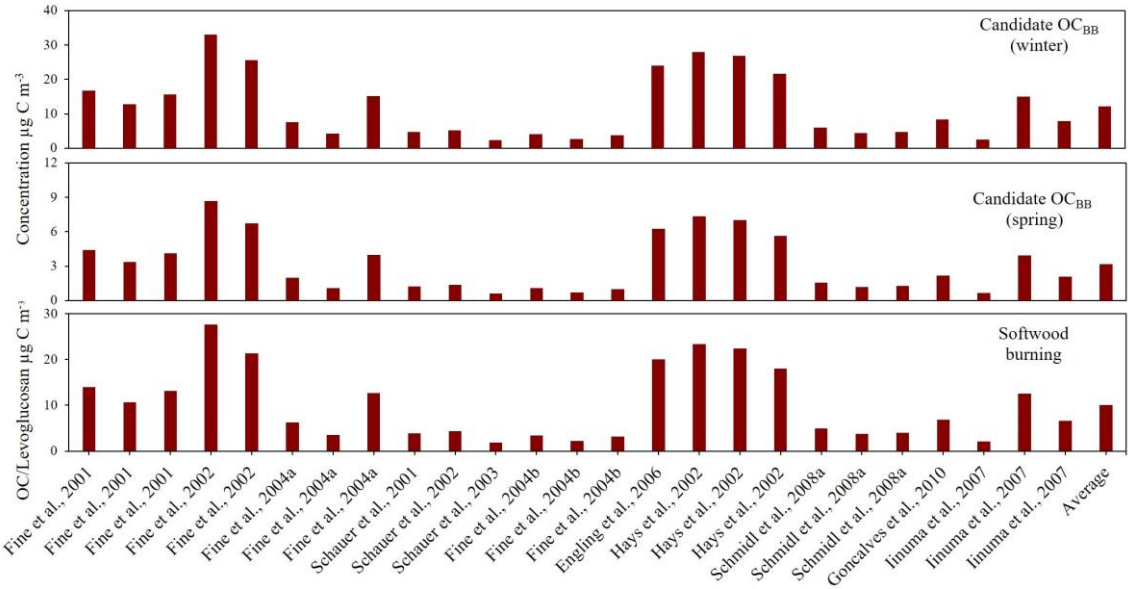
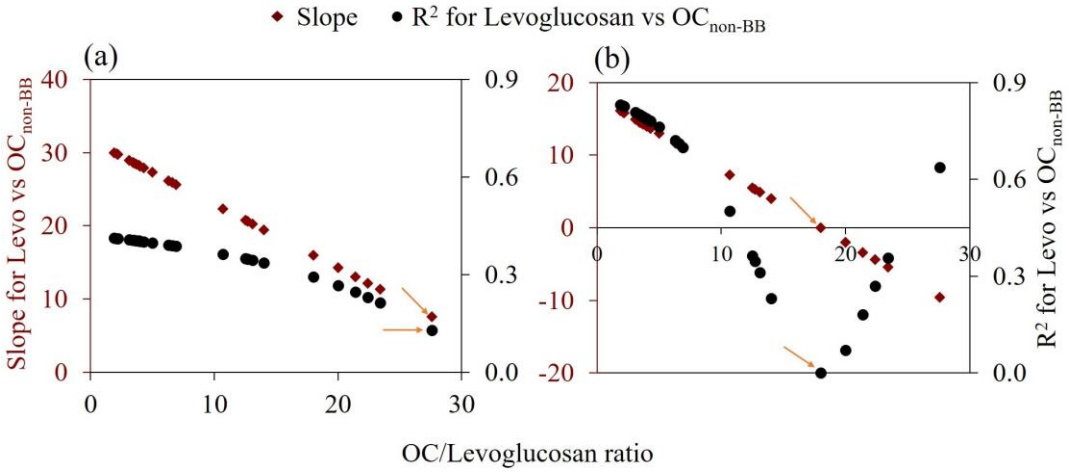
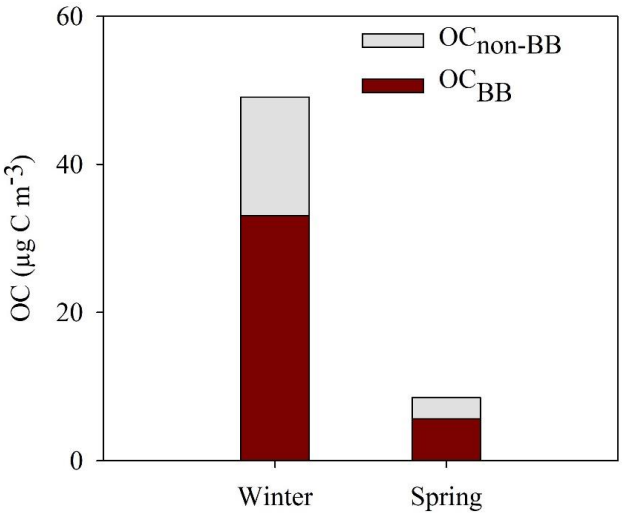

Concentration µg C m-3
OC/Levoglucosan µg C m-3
Candidate OCBB (winter)
Candidate OCBB (spring)
Softwood burning
Fine et al., 2001
Fine et al., 2001
Fine et al., 2001
Fine et al., 2002
Fine et al., 2002
Fine et al., 2004a
Fine et al., 2004a
Fine et al., 2004a
Schauer et al., 2001
Schauer et al., 2002
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

935 Fig. 13
936



937
938
939

Reply to Comments of Anonymous Referee # 1

An optimized tracer-based approach for estimating organic carbon emissions from biomass burning in Ulaanbaatar, Mongolia by Nirmalkar et al This paper by Nirmalkar et al analysed the chemical composition of daily $PM_{2.5}$ filter samples collected in Ulaanbaatar during winter and spring, with the aim of determining the contribution of biomass burning to the $PM_{2.5}$ load. The authors then applied multivariate correlation analysis (PCA) to determine the main sources based on the chemical composition and used diagnostic ratios to apportion the contribution from biomass burning. The authors concluded that biomass burning was a significant source, accounting for 68 and 63% of the organic carbon in winter and spring, respectively and that the very high contributions reflected the practice of wood burning for heating in the city. Ulaanbaatar has a well- known air pollution problem, and this is a nice dataset for investigating the sources during winter. While the dataset appears sound, in my opinion the data interpretation/analysis a bit light. There are much more the authors could do with the dataset to strengthen and support their conclusions.

Furthermore, there have been numerous studies already investigating air pollution in Ulaanbaatar, yet the authors curiously do not mention how their findings relate to this body of work, choosing instead to focus on similar studies in other Asian cities.

Reply: 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.

Following sentences have been added in lines 82-94 in the revised MS.

"A half of residents in Ulaanbaatar lives in 160,000 Gers (traditional Mongolian dwelling) (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 is very few estimate of OC emitted from biomass burning (OC_{BB}) in Ulaanbaatar. Few studies have investigated the chemical characteristics of organic aerosol in Ulaanbaatar (Jung et al., 2010; 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 identify the type of biomass at Ulaanbaatar, Mongolia.

Following sentences have been added in lines 99-103 in the revised MS.

"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}."

Four references have been added in the reference section.

"Sun, J., Shen, Z., Zhang, Y., Zhang, Q., Wang, F., Wang, T., Chang, X., Lei, Y., Xu, H., Cao, J., and Zhang, N.: Effects of biomass briquetting and carbonization on $PM_{2.5}$ emission from residential burning in Guanzhong Plain, China, Fuel, 244, 379−387, https://doi.org/10.1016/j.fuel.2019.02.031, 2019.
Guttikunda, S.: Urban Air Pollution Analysis for Ulaanbaatar, Mongolia, SIM Working Paper No. 2008-005, http://dx.doi.org/10.2139/ssrn.1288328, September 22, 2008.
Guttikunda, S. K. and Jawahar, P.: Atmospheric emissions and pollution from the coal−fired thermal power plants in India, Atmos. Environ., 92, 449−460, https://doi.org/10.1016/j.atmosenv.2014.04.057, 2014."

As the authors mention, the OC/Levoglucosan ratio from biomass burning is highly variable and dependent on many variables such as fuel and burn conditions. I am not entirely convinced by proposed method for optimising the OC/Levoglucosn ratio source apportionment and would have liked to have seen more analysis justifying the proposed 'optimal' ratio. For example, some discussion on how did the optimal OC/Levoglucosan from winter and summer compare to the literature values?

Reply: Thank for the reviewer's comments. We have compared OC/levoglucosan value obtained by literature values using R^2 and slope values of regression analysis between the concentration of levoglucosan and $OC_{non\text{-}BB}$ ($OC\text{-}OC_{BB}$) in this site during winter and spring separately. The literature value of OC/levoglucosan ratio, which gives the lowest coefficient of determinant (R^2) and slope value, is treated as optimised OC/levoglucosan ratio. Based on the regression analysis we found two different optimised OC/levoglucosan ratios for winter (27.6) and spring (18). Further, these ratios are used for estimating OC_{BB} during winter and spring separately for Ulaanbaatar.

Following sentences have been added in lines 368-376 in the revised MS.

"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 it has 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."

A reference has been added in the reference section.

"Obrist, D., Moosmüller, H., Schürmann, R., Chen, L. W. A., and Kreidenweis, S. M. Particulate-phase and gaseous elemental mercury emissions during biomass combustion: controlling factors and correlation with particulate matter emissions. Environ. Sci. Technol., 42, 721-727, https://doi.org/10.1021/es071279n, 2007."

Does the optimal OC/Levoglucosan ratio make sense in terms what would be expected based on the main fuel used in Ulaanbaatar?

Reply: Thank you for the reviewer's comment. The estimation of OC_{BB} in this study is relevant to the main fuel used in Ulaanbaatar. Majority of Ulaanbaatar's population lives in a Ger (traditional dwelling) and each Ger family burns an average of 3 m^3 of wood per year (~6 tons/year) (Guttikunda, 2008). 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). This showed that the softwood burning is one of the major sources in Ulaanbaatar for heating home and cooking food.

Following sentences have been added in lines 82-88 in the revised MS.
"A half of residents in Ulaanbaatar lives in 160,000 Gers (traditional Mongolian dwelling) (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 m3 of wood per year (Guttikunda, 2008; Zhamsueva et al., 2018)."

What about if the source of biomass burning changed over time during the sampling period, and therefore presumably the ambient OC/Levoglucosan?
Reply: Thank you for the comment. Based on scatter plot analysis between levoglucosan/mannosan and levoglucosan/K^+ ratios shown in Fig. 10 in the revised MS, it was found that softwood was the major type of biomass burning during winter and spring. However, OC/levoglucosan ratio of softwood burning can vary depending on burning type such as smoldering or flaming. Thus, we determined optimum OC/levoglucosan ratio during winter and spring.

One potential pitfall in this approach not discussed would be if some of the non-BB sources of OC had similar temporal trends to biomass burning emissions, which would mean that they would also be high when the levoglucosan was high, thus affecting the correlation analysis. For example, coal burning was noted by the authors to be a source of OC, yet I could imagine that during cold periods power station emissions would be also be high at the same time as wood burning due to the heating load.
Reply: To follow the reviewer comments we analysed potential source direction of OC using Conditional Probability Function (CPF). The thermal power plants are situated west side to the study site. Yes, reviewer is rightly pointing out that coal burning is also source of OC. CPF analysis suggested that the potential source direction is west for both OC and levoglucosan but with low wind speed (~≤2 m/s). Therefore, power plant emission (potential source for $OC_{non\text{-}BB}$) may not influencing the concentration of OC. So, OC concentration mainly influenced by the nearby residential biomass emissions. Therefore, OC_{BB} concentration estimated by optimised OC/Levoglucosan ratio was not affected by coal burning in thermal power plant. Further, potential source direction of levoglucosan, K^+ and OC was similar suggested by CPF analysis. The correlation of levoglucosan and K^+ with OC during winter (R^2=0.78 and 0.79, respectively) and spring (R^2=0.86 and 0.73, respectively) was strong which suggested the tight association of OC with biomass burning. This supported the preciseness of this novel approach for estimating the OC_{BB} at Ulaanbaatar.

Following sentences have been added in lines 155-166 in the revised MS.
"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:

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

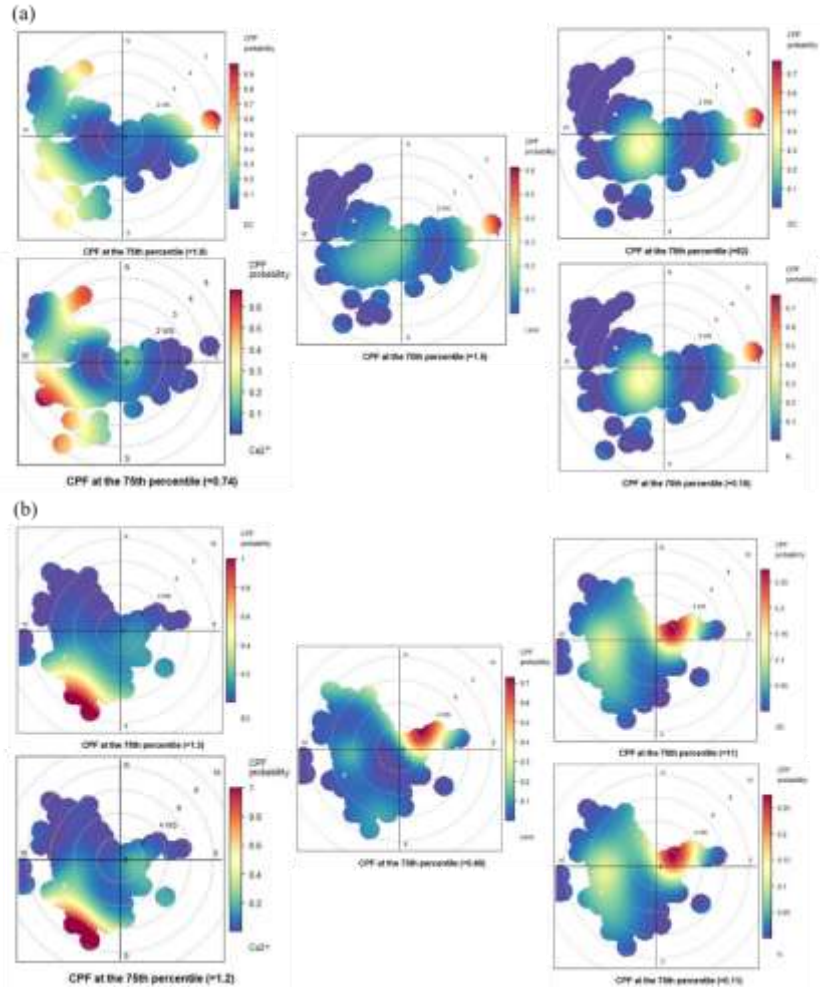
where $n_{\Delta\Theta}$ is the number of times that the wind passed through direction sector ΔΘ, and $m_{\Delta\Theta}$ is the number of times that the source contribution peaked while the wind passed through sector ΔΘ (Ashbaugh et al., 1985). To use CPF with the Ulaanbaatar data, the 24 h

averaged source contribution data have been applied to all 1 h wind direction averages recorded at the site for each date. The angular interval $\Delta\Theta$ was set at 10°. To calculate $m_{\Delta\Theta}$, the 75th percentile 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."

Following sentences have been added in lines 203-209 in the revised MS.

"The potential source direction of EC during winter and spring was west as shown in Fig. 5 that 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 plant (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."

Following figure has been added in Fig. 5.



Two references have been added in the reference section.

"Ashbaugh, L.L., Malm, W.C., Sadeh, W.Z.: A residence time probability analysis of sulfur concentrations at Grand Canyon National Park, Atmos. Environ., 19(8), 1263−1270, https://doi.org/10.1016/0004-6981(85)90256-2, 1985.

Pei, B., Wang, X., Zhang, Y., Hu, M., Sun, Y., Deng, J., Dong, L., Fu, Q. and Yan N.: Emissions and source profiles of $PM_{2.5}$ for coal-fired boilers in the Shanghai megacity, China, Atmos. Pollut. Res. 7, 577-584, https://doi.org/10.1016/j.apr.2016.01.005, 2016."

The uncertainties associated with this approach to determining the optimal ratio should be discussed in detail.
Reply: Thank you for the comment. There is no uncertainty is associated with optimised ratio. In this approach, we screened optimised OC/levoglucosan ratio individually for winter and spring from various ratios reported in BB chamber experiments using regression analysis. The details about the approach is given in section 3.5. However, a large amount of uncertainty associated with OC/levoglucosan ratio for biomass fuel due to different kinds (hard, softwood, crop etc.), burning place (open or inside stove), burning condition (mouldering or flaming) etc. Even sometime same wood type (e.g. softwood) has different OC/levoglucosan ratio might be due to the causes mentioned above (line number 383-388). Therefore, it is important to select a suitable OC/levoglucosan ratio for any of the study site for estimating appropriate concentration of OC_{BB}. This study provides us a novel approach to select the suitable OC/levoglucosan ratio for different study site for understanding the impact of BB in OC fraction.
Please see section 3.4 and 3.5 in the revised MS.

Minor comments
Abstract: The authors could be more explicit that the optimal OC/Levoglucosan ratio determined is specific to Ulaanbaatar, and that it is method for determining it is applicable for other studies.
Reply: The developed approach can be applicable to any study site for screening the appropriate OC/levoglucosan ratio for estimating OC_{BB} contribution to ambient PM. To do so, the regression analysis is required between $OC_{non\text{-}BB}$ [(OC at any study site)-(levoglucosan at any study site*OC/levoglucosan obtained from chamber experiments)] and levoglucosan concentration at any study site. The ratio, which give the lowest value of coefficient of determinant (R^2) and slope, could use as optimised ratio of OC/levoglucosan. This optimised ratio can be applied for estimating OC_{BB} for at any study site.

By the following of reviewer's comments, we have clearly explained the approach and applicability for the other study site.

Following sentence has been added in lines 26-29 in the revised MS.
"The optimum OC/levoglucosan ratio in Ulaanbaatar was obtained by regression analysis between $OC_{non\text{-}BB}$ ($OC_{total}-OC_{BB}$) and levoglucosan concentrations that gives the lowest coefficient of determination (R^2) and slope."
Following sentence has been added in lines 32-33 in the revised MS.
"This novel approach can also be applied to other study site to quantify OC_{BB} using their own chemical measurements."

Page 6, line 98: I presume that you mean it is difficult to determine the most suitable OC/levoglucosan ratio of BB emissions for ambient measurements?
Reply: Our intension is not saying like that. The chosen world "difficult" is creating inappropriate meaning.
The sentence in line 98 in the original MS has been modified as follows.
Please see lines 102-103 in the revised MS.
"Therefore, it is required to determine the most suitable OC/levoglucosan ratio of BB emissions for estimating appropriate concentration of OC_{BB}."

Page 7, line 111: do these thermal power plants burn biomass? If so, emissions from these plants could have affected the results.

Reply: Thank for reviewer's comments. No, they only used coal in thermal power plants. Ulaanbaatar has three coal fired thermal power plants (Chung and Chon, 2014).

Following sentence has been added in lines 114-115 in the revised MS.
"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)."
A reference has been added in the reference section.
"Chung, S. and Chon, H. T.: Assessment of the level of mercury contamination from some anthropogenic sources in Ulaanbaatar, Mongolia, J. Geochem. Explor., 147, 237–244, https://doi.org/10.1016/j.gexplo.2014.07.016, 2014."

Section 3.1: This could perhaps be broken down into a few subsections to help the reader find relevant sections. For example, the PCA analysis could be one sub section.
Reply: As per the reviewer's comments, PCA analysis has been discussed in separate sub section as 3.2 in RMS.

Following sentences have been added in lines 242-258 in the revised MS.
"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 showed that the chemical components of PM2.5 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)."

Page 8, line 153: Are these the average contributions of OC to the total chemical species? It would also be good to give an indication of the variability, perhaps by showing the standard deviation.
Reply: Thank you very much for the reviewer's suggestion. Yes, this is, OC contributed 64±5.1% and 56±6.0% of the quantified aerosol components in $PM_{2.5}$ in winter and spring, respectively. As per the reviewer's suggestion standard deviation is added accordingly.

Please see lines 186-187 in the revised MS.
"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)."

Page 9, line 160-3: The statement that during spring the OC increased with temperature due to SVOC volatilization appears to contradict the earlier statement that high concentrations in the winter due to increased condensation of SVOC at low temperature?
Why would SVOC volatilization account for the relationship of OC with temperature?
Could it maybe be more related to increased biogenic emissions?

Reply: Thank you for the comment. The original statement regarding SVOC during spring in lines 160-163 in the original MS has been deleted.

Page 9, line 179-81: I am not sure I follow the explanation for the relationship between temperature and EC. What is the source/mechanism that would explain the relationship between temperature and re-suspension of soil?
Reply: We thank for the reviewer's comment. The association of temperature and EC was strong during spring. But did not find any explanation for source and mechanism based on temperature. Therefore, to follow the reviewer's comments we removed this line from RMS and modified Fig. 4. Now we interpreted the EC concentration by Conditional Probability Function (CPF) analysis as reviewer's suggested (Fig. 5).

We have rewritten this phrase in lines 203-209 in the revised MS as follows.
"The potential source direction of EC during winter and spring was west as shown in Fig. 5 that 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 plant (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."

Details of CPF analysis has been also added in lines 156-166 in the revise MS.
"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:

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

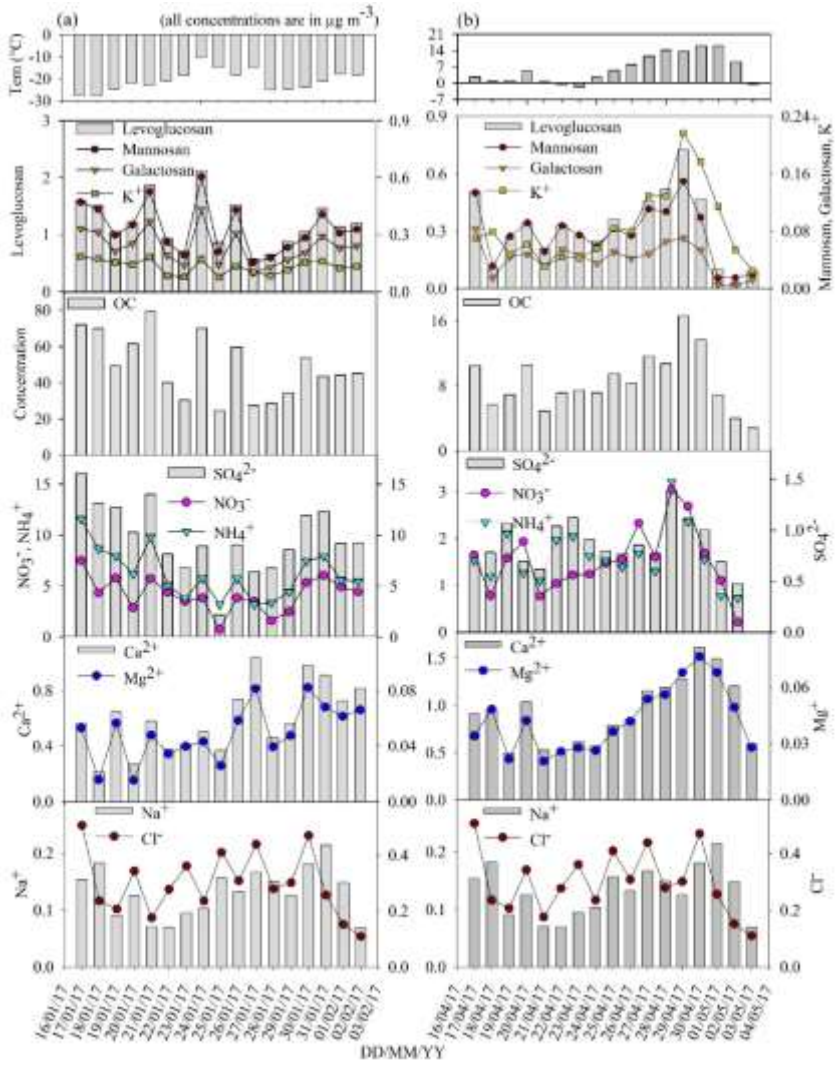
where nΔΘ is the number of times that the wind passed through direction sector ΔΘ, and mΔΘ is the number of times that the source contribution peaked while the wind passed through sector ΔΘ (Ashbaugh et al., 1985). To use CPF with the Ulaanbaatar data, the 24 h averaged source contribution data have 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 mΔΘ, the 75th percentile 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."

Modified Fig. 4 has been modified in Revised MS.

Two references have been added in the reference section in the revised MS.
"Ashbaugh, L., Malm, W. and Sadeh, W. : A Residence Time Probability Analysis of Sulfur Concentrations at Ground Canyon National Park. Atmos. Environ., 19(8): 1263–1270, 1985."
Pei, B., Wang, X., Zhang, Y., Hu, M., Sun, Y., Deng, J., Dong, L., Fu, Q. and Yan N.: Emissions and source profiles of PM2.5 for coal-fired boilers in the Shanghai megacity, China, Atmos. Pollut. Res. 7, 577-584, https://doi.org/10.1016/j.apr.2016.01.005, 2016."

Page 10, line 183: A time series plot of these tracers with temperature would help the arguments in this paragraph

Reply: As per the reviewer suggestion time series of temperature has been added to Fig. 2. Following figure has been added in Fig. 2.



Page10, line 201-203: As power stations are large point sources, the authors could do some wind sector analysis (e.g. polar plots, concentrations as function of wind speed and direction) to test this hypothesis. This could also help to see if any of the OC and EC was also from power stations. In addition, Ca^{2+} has also been associated with coal station emissions (see Pei et al 2016, https://doi.org/10.1016/j.apr.2016.01.005) and may explain the association of EC and Ca from earlier. The authors need to consider the emissions from power stations more closely in order to be confident in the OCbb apportionment later in the paper.

Reply: Thank to reviewer's suggestion for polar plots analysis for investigating potential source directional. As per the reviewer's suggestion we have incorporated the Conditional Probability Function (CPF) analysis for investigating potential source direction of EC and Ca^{2+} (Fig. 5). Based on CPF analysis during winter and spring seasons, levoglucosan, OC and K^+ (Fig. 5), their potential source direction are similar, mostly from west direction with low speed (~2 m/s).

Following sentences have been added in lines 203-209 in the revised MS.

"The potential source direction of EC during winter and spring was west as shown in Fig. 5 that 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 plant (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."

Following figure has been added in Fig. 5.
Please find Fig. 5 in previous response

A reference has been added in the reference section in the revised MS.
"Pei, B., Wang, X., Zhang, Y., Hu, M., Sun, Y., Deng, J., Dong, L., Fu, Q. and Yan N.: Emissions and source profiles of $PM_{2.5}$ for coal-fired boilers in the Shanghai megacity, China, Atmos. Pollut. Res. 7, 577-584, https://doi.org/10.1016/j.apr.2016.01.005, 2016."

Page 11, line 212: Since there was a large regional source of BB on these days, where they removed/accounted for in subsequent optimization of OC/Levoglucosan? Local and regional sources are known to have different ratios, and therefore will affect the analysis.
Reply: We thank to reviewer for the comment. However, wood is one of major fuel used for domestic purposes in Ulaanbaatar as shown by the high concentration of levoglucosan. Therefore, we assume that there is no significant change in the ratio of OC/levoglucosan from local wood burning or wood burning during forest fire. Further, we have also rechecked the OC/Levoglucosan ratio by excluding the data points (27-04-17 to 30-04-17), we did not find any significant change in optimised ratio of OC/levoglucosan.

Page 11, line 214: The details of how the PCA analysis was performed need to be included, perhaps in the method section
Reply: As per the reviewer's suggestion we have added details about PCA analysis method section in section 2.4.

Following sentences have been added in 168-180 in the revised MS.
"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, PC) 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)."

Page 11, line 223: Were there any other reasons for choosing vehicles as the source of PC4 as there were other sources of EC as well (e.g. biomass burning). Furthermore, I am surprised that if biomass burning was such a strong source that EC did not come out in the same PC as the BB tracers. Perhaps the authors could comment on this. I am also curious as to why there was not a vehicle source found in spring, I would have thought that vehicle

source would be consistent across both seasons. Why would there be a combined SIA and vehicles source in spring?

Reply: Thank for the comment. Fine size EC is potentially associated with the traffic emission as suggested by previous study (Lonati et al., 2007; Zhang et al., 2005), thus we chosen PC4 as traffic in original MS. The reason for not chosen EC from biomass is due its weak correlation with levoglucosan during winter and spring. The correlation of EC with levoglucosan and OC was weak during winter (R^2 = 0.07 and R^2 = 0.05, respectively) and spring (R^2 = 0.21 and R^2 = 0.04, respectively) these might be due to their different sources or processing in air.

However, fine mode EC may be associated with various sources including vehicle emission, coal combustion and fine dust (Tao et al., 2013). Further, reviewer also suggested that there might be other sources for EC. By following the another reviewer, we analysed potential source direction of EC using polar plot (conditional probability function, CPF). CFP results indicated that potential source direction of EC was west during winter and spring. This might be suggested due to influenced of stack emission from thermal power plants situated in west direction to study site. In reviewer comment (page 10, line number 201-203 in original manuscript) also supported EC derived from coal burning. We found some study reported EC from both motor vehicle and coal combustion (Lonati et al., 2007; Song et al., 2007; Tao et al., 2013). Therefore, we have now replaced vehicular source from fossil fuel combustion in PC2 in Table 2 and PC3 in Table 3 in revised manuscript.

It should be noted that at this site in spring, the correlation between OC and EC was poor while good correlation between OC and K^+ (R^2 = 0.73 and 0.79) was observed in both seasons. These findings indicated that OC was significantly influenced by biomass burning, while the EC might be mostly from primary coal combustion.

The potential cause of association of EC with secondary inorganic anion (SIA) during spring might be due similar sources. In spring, EC and Ca^{2+} have strong correlation with the total variance of PC2 (Table 3). The potential source direction for both EC and Ca^{2+} was west which might be due to the influence of stalk emission from thermal power plant (Fig. 5). Pie et al., 2011 observed emission of EC and Ca^{2+} from coal combustion.

Following sentences have been added in 203-209 in the revised MS.

The potential source direction of EC during winter and spring was west as shown in Fig. 5 that 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 plant (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.

Following sentences have been added in 249-254 in the revised MS.

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^+).

Lonati, G., Ozgen, S., & Giugliano, M. (2007). Primary and secondary carbonaceous species in $PM_{2.5}$ samples in Milan (Italy). Atmospheric Environment, 41(22), 4599-4610.

Song, Y., Tang, X., Xie, S., Zhang, Y., Wei, Y., Zhang, M., Zeng, L. and Lu, S., 2007. Source apportionment of $PM_{2.5}$ in Beijing in 2004. Journal of hazardous materials, 146(1-2), pp.124-130.

Zhang, F., Wang, Z. W., Cheng, H. R., Lv, X. P., Gong, W., Wang, X. M., & Zhang, G. (2015). Seasonal variations and chemical characteristics of $PM_{2.5}$ in Wuhan, central China. Science of the Total Environment, 518, 97-105.

Tao, J., Zhang, L., Engling, G., Zhang, R., Yang, Y., Cao, J., Zhu, C., Wang, Q. and Luo, L., 2013. Chemical composition of $PM_{2.5}$ in an urban environment in Chengdu, China: Importance of springtime dust storms and biomass burning. Atmospheric Research, 122, pp.270-283.

Pei, Bing, Xiaoliang Wang, Yihua Zhang, Ming Hu, Yanjing Sun, Ji Deng, Li Dong, Qingyan Fu, and Naiqiang Yan. "Emissions and source profiles of $PM_{2.5}$ for coal-fired boilers in the Shanghai megacity, China." Atmospheric Pollution Research 7, no. 4 (2016): 577-584.

Page 11, line 229: Do the authors have any ideas why K^+ was associated with biomass burning in the winter but not in the spring? Was there a source change?

Reply: We appreciated the reviewer for the comment. We apologised for the inappropriate sentences used here regarding the K^+ during spring. We are not saying that K^+ was not associated with biomass burning during spring. We highlighted that K^+ is emitted from biomass in winter and spring. The sentences have been re-phrased in MS for better clarity and some sentences have been deleted which are not relevant to the scope of the manuscript.

Following sentences have been added in 255-258 in the revised MS.

The PCA results showed 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).

Following sentences have been added in 273-281 in the revised MS.

"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 tended 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."

Four new references have been added in MS.

Schkolnik, G., Falkovich, A. H., Rudich, Y., Maenhaut, W., and Artaxo, P.: New analytical method for the determination of levoglucosan, polyhydroxy compounds, and 2-methylerythritol and its application to smoke and rainwater samples, Environ. Sci. Technol. 39, 2744-2752, https://doi.org/10.1021/es048363c, 2005.

Gao, S., Hegg D. A., Hobbs P. V., Kirchstetter T. W., Magi B. I., and Sadilek M.: Water-soluble organic components in aerosols associated with savanna fires in southern Africa: Identification, evolution, and distribution, J. Geophys. Res., 108(D13), 8491, doi:10.1029/2002JD002324, 2003.

Lee, T., Sullivan, A. P., Mack, L., Jimenez, J. L., Kreidenweis, S. M., Onasch, T. B., Worsnop, D. R., Malm, W., Wold, C. E., Hao, W. M., and Collett Jr, J. L.: Chemical smoke marker emissions during flaming and smoldering phases of laboratory open burning of wildland fuels, Aerosol Sci. Technol., 44, i-v, https://doi.org/10.1080/02786826.2010.499884, 2010.

Page 12. Line 243: It would be good to show the intercept as percentage of the total OC.
Reply: As per the reviewer's comments. Because intercept of regression line of OC vs levoglucosan represent the OC, which is not related to biomass burning ($OC_{non\text{-}BB}$). We have already discussed about $OC_{non\text{-}BB}$ in section 3.6. Thus, we have deleted this line 243 from original manuscript.
Please see section 3.6

Page13, line 238: You state here that the correlation between OC and K^+ indicates that biomass burning was a major source but in the previous paragraph you state that K^+ is coming from soil re-suspension in spring? Please clarify.
Reply: Thank you for the reviewer's comment. We apologised for the conflicting sentences. We are not saying like that K^+ was not associated with biomass burning during spring. We highlighted that K^+ is emitted from biomass in winter and spring. Now conflicting sentences have been re-written in both the paragraphs as per the comment for clarity in the explanation.

Following sentences have been added in 255-258 in the revised MS.
The PCA results showed 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).

Following sentences have been added in 273-281 in the revised MS.
"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 tended 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."

Four new references have been added in MS.
Schkolnik, G., Falkovich, A. H., Rudich, Y., Maenhaut, W., and Artaxo, P.: New analytical method for the determination of levoglucosan, polyhydroxy compounds, and 2-methylerythritol and its application to smoke and rainwater samples, Environ. Sci. Technol. 39, 2744-2752, https://doi.org/10.1021/es048363c, 2005.

Gao, S., Hegg D. A., Hobbs P. V., Kirchstetter T. W., Magi B. I., and Sadilek M.: Water-soluble organic components in aerosols associated with savanna fires in southern Africa: Identification, evolution, and distribution, J. Geophys. Res., 108(D13), 8491, doi:10.1029/2002JD002324, 2003.

Lee, T., Sullivan, A. P., Mack, L., Jimenez, J. L., Kreidenweis, S. M., Onasch, T. B., Worsnop, D. R., Malm, W., Wold, C. E., Hao, W. M., and Collett Jr, J. L.: Chemical

smoke marker emissions during flaming and smoldering phases of laboratory open burning of wildland fuels, Aerosol Sci. Technol., 44, i-v, https://doi.org/10.1080/02786826.2010.499884, 2010.

Page 12, line 267: If the excess of K^+ during winter was due to biomass burning for cooking, do you the same in the relationship or similar value for the intercept in the spring? I am assuming that cooking is also happening in spring and not just winter?
Reply: Thank you for the comment. Because R^2 of regression line of K^+ versus OC is moderate (0.79 and 0.73), intercepts of the regression line may have high uncertainty. Thus, we decided to delete the discussion regarding the intercept in the original MS.

Page 14, line 284: it would be good here to give the actual ratios for these different sources from the literature to show how much overlap there is
Reply: As per the reviewer suggestion actual ratios has been provided in revised manuscript.

Please see line number 305-309 in the revised MS.
"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)."

Page 15, line 308: Is the result that the levoglucsaon/mannosan ratio is consistent with softwood expected based on people activity in Ulaanbaatar? That is do people mostly burn softwood at home for heating? Earlier you have stated that coal is mainly burnt for cooking, so it appears that it may not.
Reply: Thank you for the comment. Ulaanbaatar Gers mostly used wood for heating and cooking purposes. 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). As the concentration of levoglucosan was high during winter and significant during spring suggested that wood burning might be one of the major sources in Ulaanbaatar. A total numbers Gers (tradition dwellings) of Ulaanbaatar city consumed ~480, 000 m^3 per year of wood (160, 000 Gers*each Ger consumed 3 m^3 of wood per year) (Guttikunda, 2008; Zhamsueva et al., 2018). In Ulaanbaatar, we identified softwood as major fuel type by regression plot between levoglucosan/mannosan and levoglucosan/K^+ ratio.
Now based on the above explanation we have rewritten the sentences and incorporated in RMS.

Please see line number 82-88 in the revised MS.
"A half of residents in Ulaanbaatar lives in 160,000 Gers (traditional Mongolian dwelling) (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)."

A referene has been added in the reference section in the revised MS.
"Zhamsueva, G. S., Zayakhanov, A. S., Starikov, A. V., Balzhanov, T. S., Tsydypov, V. V., Dementyeva, A. L., and Khodzher, T. V.: Investigation of chemical composition of

atmospheric aerosol in Ulaanbaatar during 2005–2014. Geography and Natural 39, 270−276, 10.1134/S1875372818030113, 2018."

Page 17, line 355: What is uncertainty associated the derived optimal OC/levoglucosan for winter and spring?

Reply: Thank you for the comment. We did not determine uncertainty associated with optimised ratio in this study. In this approach, we screened optimised OC/levoglucosan ratio individually for winter and spring from various ratios reported in BB chamber experiments using regression analysis. The details about the approach is given in section 3.5. However, a large amount of uncertainty associated with OC/levoglucosan ratio for biomass fuel due to different kinds (hard, softwood, crop etc.), burning place (open or inside stove), burning condition (mouldering or flaming) etc. Even sometime same wood type (e.g. softwood) has different OC/levoglucosan ratio might be due to the causes mentioned above (line number 383-388). Therefore, it is important to select a suitable OC/levoglucosan ratio for any of the study site for estimating appropriate concentration of OC_{BB}. This study provides us a novel approach to select the suitable OC/levoglucosan ratio for different study site for understanding the impact of BB in OC fraction.

Please see section 3.4 and 3.5, line number 358-360, 403-415

Page 17, line 357: How do the optimized ratio of 27.6 and 18 compare to the literature for sources. Earlier you stated that levoglucosan/mannosan ratio was consistent with softwood combustion, so are these OC/levoglucosan ratios also consistent for softwood combustion?

Reply: Yes, both levoglucosan/mannosan and OC/levoglucosan ratios are consistent with softwood burning. In this approach firstly, we have determined softwood as a kind of fuel used in Ulaanbaatar by comparing the ratios of levoglucosan/mannosan and levoglucosan/K^+ with these ratios reported in chamber experiments (section 3.5). The average levoglucosan/mannosan ratio was within the ranges reported for softwood burning sources. Thus, we identified softwood as major biomass in this study site. In the previous chamber experiment, OC/levgoglucosan ratio of softwood were highly variable as shown in Fig. 11 mainly due to different burning conditions. Thus, we determined optimised OC/levoglucosan at the Ulaanbaatar during winter and spring for accurate quantification of OC from biomass burning (OC_{BB}).

Reply to Comments of Anonymous Referee # 2

The manuscript is well-written; it is apparently the first study for Mongolia in which use was made of levoglucosan to assess the impact from biomass burning on the $PM_{2.5}$ aerosol and the authors made a noteworthy attempt to obtain the optimum OC/levoglucosan ratio to derive that impact. However, as indicated below, the manuscript has several shortcomings and major revision is definitely needed before it can be published in ACP.

Reply: We thank the reviewer for the encouraging comments and nicely reviewing 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 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.

Major comments:

1. The number of samples in this study is quite limited, i.e., only 34 in total; besides, samples were only taken in two seasons (winter and spring) and it is really unfortunate that no $PM_{2.5}$ mass data were obtained. Because the $PM_{2.5}$ mass is unknown, the authors cannot state, like they do in lines 18-19, "that OC was the predominant species in the total aerosol compositions", in line 271 "that OC is a major contributor to $PM_{2.5}$", and in line 401 that "OC was the major component of $PM_{2.5}$"; "in the total aerosol compositions" should be replaced by "of the quantified aerosol components", and "to $PM_{2.5}$" and "of $PM_{2.5}$" should be replaced by "of the quantified aerosol components in $PM_{2.5}$".

Reply: Thank you for the comment. Although the number of samples are limited, this study provides an important finding about the influence of biomass burning on the quantified aerosol components in $PM_{2.5}$ by new approach. Following the comment of reviewer, we have done all the modification accordingly in phrase in RMS.

Please see lines 21, 169, 292, 420, 422 in the revised MS.

2. With regard to the application of PCA to the separate winter and spring sample sets of 17 samples each: Although the results in Tables 2 and 3 look reasonable, that application is not justified at all. Henry et al. state on page 1512 of their seminal 1984 paper (full reference: Henry, R.C., Lewis, C.W., Hopke, P.K., Williamson, H.J, 1984. Review of receptor model fundamentals. Atmos. Environ. 18, 1507-1515) that, in order for a multivariate receptor model to be acceptable, the number of samples N should be larger than 30 + (V + 3)/2, with V being the number of variables. As V is 13 in Tables 2 and 3, this means that the number of samples in each of the authors' sample sets should be larger than 38. Their number of 17 is very much smaller. Although there are only 34 samples in the combination of the winter and spring sample sets and the criterion of 38 is then also not fulfilled, it might be worthwhile to apply PCA to that combined sample set. Furthermore, instead of using PCA for source apportionment, the preferred method nowadays is positive matrix factorization (PMF). I suggest that PMF be used instead of PCA, although the number of 34 samples may make the use of PMF difficult; many researchers suggest to have at least 50 samples (and some even 100) for PMF. Furthermore, the authors talk in line 217 of "eigenvalues >1". Do they mean here eigenvalues before or after Varimax rotation? More important, the high loadings (or total variance explained by a component) do no tell anything about the importance or

contribution of a component to a species or the aerosol mass. In order to obtain the contribution, one has to apply absolute PCA (APCA) or PMF. Thus, the statements in lines 226, 228 and 272 that "BB is the major source of OC" are not justified.

Reply: Thank you for the comment. The phrase in line 217, the discussed eigenvalues >1 is after Varimax rotation. As per the reviewer suggestions we have added this information in the phrase. Yes, reviewer is rightly point out the PCA results interpretation as it is not quantitative estimates of source contribution. Therefore, to justify the PCA results we have rewritten the related phrase and incorporated in RMS. Also, Yes, the reviewer is right to use PMF for source apportionment studies. But in this study the scope of PCA is only to identify the sources of chemical components. To honor the reviewer's comments, we have also check PCA analysis of total number of samples. PCA results still suggested that the chemical components are highly influence by BB phenomena at Ulaanbaatar.

However, previous published papers they did PCA analysis using similar number of samples. For instance, Pavuluri et al., 2010 where, in winter: V=15, no of samples (N)=15; late winter: V=15, N=10. Therefore, the relevance of the scope of the present study we wish to retain the PCA results for winter and spring.

Following sentence has been added in lines 245-248 in the revised MS.

"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)."

Following sentences have been added in lines 255-258 in the revised MS.

"The PCA results showed 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. The authors' approach to arrive at the optimum OC/levoglucosan ratio needs to be much better explained. In lines 348-350 they write "candidate OC_{BB} in this study was estimated from OC/levoglucosan ratios for softwood burning in the previous chamber experiment (Cheng et al., 2013 and papers cite therein)", but they fail to give actual numbers for the candidate(s) OC_{BB}. Why was only use made of Cheng et al. (2013) and the references cited therein? Why not also of data from other publications or even hypothetical OC/levoglucosan ratios? It seems from Fig. 11a that a higher OC/levoglucosan ratio than the highest one used could lead to better results.

Reply: Thank you for reviewer's suggestion. As per the suggestion we have added and modified the sentences for clear explanation of approach to arrive at the optimum OC/levoglucosan ratio.

In this approach firstly, we have determined the kind of biomass (i.e. softwood wood in Ulaanbaatar) by comparing the ratios of levoglucosan/mannosan and levoglucosan/K^+ in this site with these ratios reported in chamber experiments (section 3.5). The average levoglucosan/mannosan and levoglucosan/K^+ ratios were within the ranges reported for softwood burning. Thus, we identified softwood as major biomass in this study site. Then we determine OC emitted from biomass burning (OC_{BB}) using levoglucosan in this site and OC/levoglucosan ratios reported in chamber experiments. Then we calculated $OC_{non\text{-}BB}$ (OC_{total}-OC_{BB}) and further regress this with levoglucosan concentration. The OC/levoglucosan ratio that gives lowest R^2 and slope value is treated as optimized OC/levoglucosan. Then appropriate OC_{BB} concentration is estimated by multiplying optimized OC/levoglucosan ratio obtained from chamber experiment and levoglucosan

concentration in this site. As per suggestion relevant references for OC/levoglucosan ratios of softwood have cited (please see line number 367).

The OC/levoglucosan ratios for softwood burning are consolidated from the previous chamber experiments and all relevant publication have cited in text relevant (Fig. 11). Candidate OC_{BB} is now added in Figure 11 as per the reviewer suggestion.

In this site higher OC/levoglucosan gave better result for estimating OC_{BB} during winter. Therefore, as per reviewer' suggestion we also check some higher hypothetical values of OC/levoglucosan ratio for estimating OC_{BB} during winter. We found that although the R^2 and slope value goes closer to zero by taking high hypothetical value of OC/levoglucosan ratio but the estimated OC_{BB} is start exceeding from the total OC concentration after 28. Thus, for this site higher hypothetical value of OC/levoglucosan is 28 or below, where estimated OC_{BB} within the range of total OC. So, always it might not be appropriate to take higher values as optimum OC/levoglucosan. In case of hypothetical value, we highlighted to take care of optimized OC/levoglucosan for estimating OC_{BB} so that it should be within the range of total OC in any study site. This also support that 27.6 is might be the optimum ratio for OC/levoglucosan for estimating appropriate concentration of OC_{BB} at Ulaanbaatar during winter. However, it was not same for spring, here we found 18 as the optimized OC/levoglucosan ratio which is not a highest value among OC/levoglucosan ratios consolidated from chamber experiments. Therefore, the optimized ratio might be vary based on the concentration of OC and levoglucosan at the any study site.

4. The authors should refer 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). In that study coarse ($PM_{10-2.5}$) and fine ($PM_{2.5}$) aerosol samples were collected twice a week from 2004 to 2008 and analysed by ion beam analysis techniques. PMF was used for source apportionment. For $PM_{2.5}$ (see Fig. 10 in that publication) and winter, coal combustion (2 factors) was by far the major contributor and the contribution from biomass burning was almost two orders of magnitude smaller. This is in very large contrast with what is concluded in the authors' manuscript. Possible explanations for that discrepancy are definitely needed.

Reply: We thank to reviewer to highlight the results regarding sources apportionment study at Ulaanbaatar during 2004-2008 using PMF by Davy et al., 2011. This study used K (potential tracer for biomass burning) to apportion the BB contribution to $PM_{2.5}$ aerosol. The concentration of K in Davy et al., 2011 is 0.324 $\mu g/m^3$ at Ulaanbaatar.

However, in the present study, we investigated the influence of BB in $PM_{2.5}$ aerosol using levoglucosan (specific tracers for biomass burning). The concentration of K^+ in present study was 0.13 $\mu g\ m^{-3}$, whereas the levoglucosan concentration (1.2 $\mu g\ m^{-3}$) was ~9 times higher that of K^+. Therefore, from the best of our experience, might be in Davy et al., 2011, the contribution of BB source is underestimated without levoglucosan as an input variable in PMF analysis. Moreover, the association of levoglucosan and K^+ (BB tracers) is strong with OC (major constituent in this study) showed significant influence of BB phenomena in atmospheric PM.

Minor comments:

1. Lines 26-27: R^2 is not a "correlation coefficient" but a "coefficient of determination".

Reply: As per reviver comment correlation coefficient has been corrected as coefficient of determination.

Please see line number 29 in the revised MS.

2. Section 2.1 and Fig. 1: There is no reference made to Fig. 1 within the text; it should be made within this Section.
Reply: Thank for the reviewers' comments, now Fig. 1 has cited.
Please see section 2.1, line number 114 in the revised MS.

3. Section 2.1 and line 128: It is unclear whether blank filter samples were taken. If so how? And does the "blanks" in line 128 refer to "blank filters" or simply to procedural blanks without the use of any blank filter?
Reply: Yes, field blank filters are collected and used for correcting background interferences in concentration of chemical constituents.

Following sentences have been added in lines 118-120 in the revised MS.
"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 analyte has been corrected using blank filters concentration."

4. Lines 160-162 and Fig. 3: It is unclear to me what the R^2, Intercept and Slope within the boxes in the Figure denote. Also, in contrast to what the authors state, the characteristics (R^2 = 0.36, slope = 1.04) of the relationship between OC and temperature are not shown in Fig. 3b.
Reply: Thank you for the comment. Now, for more clarity in explanation, we have re-phrased this paragraph and incorporated in RMS.
We are sorry for the inconvenience to the reviewer to understand the Figure, here R^2 denotes of coefficient of determination of linear fit equation y (OC)=m*x(wind speed)+c, intercept the denotes the wind speed not associated with OC, slope represents the variation of OC with the wind speed, different color represents the temperature values. This equation was added in Fig. 3.

Furthermore, I do not understand how "volatilization of SVOCs during periods of elevated temperature" can lead to increased OC in the particle phase.
Reply: Thank you for the comment. We agreed that discussion about SVOCs depending on temperature is not clear. Thus, we decided to delete the discussion regarding SVOCs with temperature during spring.

5. Lines 171-174: Do the numeric data given in parentheses pertain to spring and winter, respectively? If so, "In winter and spring" should be replaced by "In spring and winter". If, in contrast, the numeric data pertain to winter and spring, respectively, then, what is written here is in contrast with what the authors wrote in lines 169-171.
Reply: Thank you for reviewer comments. Here we wish to compare the both winter and spring concentration at this study site with suburban and urban site at China. We have modified the phrase as per the reviewer's comments and incorporated.

Following sentence has been added in lines 198-202 in the revised MS.
"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)."

6. Lines 257-259: I cannot follow why the "correlation between OC and K^+ suggests that BB is one of the major sources of ambient aerosol in Ulaanbaatar".

Reply: Thank you for the comment. Water soluble K^+ is used as tracer for biomass burning in various studies (Park et al., 2004; Lee et al., 2010; Deshmukh et al., 2011). In this site OC was major constituent of the quantified aerosol components in $PM_{2.5}$. OC and K^+ concentrations are correlated well during winter (R^2 = 0.79; Fig. 8a) and spring (R^2 = 0.73; Fig. 8b). Thus, good correlation between K^+ and OC suggested the BB phenomena primarily influences the ambient aerosol at this site. Further, influence of BB phenomena on OC is also supported by its tight association with levoglucosan.

Following sentences have been added in lines 282-289 in the revised MS.
"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 belongs 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)."

Three new references have been added in the reference section.
"Lee, T., Sullivan, A. P., Mack, L., Jimenez, J. L., Kreidenweis, S. M., Onasch, T. B., Worsnop, D. R., Malm, W., Wold, C. E., Hao, W. M., and Collett Jr, J. L.: Chemical smoke marker emissions during flaming and smoldering phases of laboratory open burning of wildland fuels, Aerosol Sci. Technol., 44, i-v, https://doi.org/10.1080/02786826.2010.499884, 2010.

Deshmukh, D. K., Deb, M. K., Tsai, Y. I., and Mkoma, S. L.: Water soluble ions in $PM_{2.5}$ and PM_1 aerosols in Durg city, Chhattisgarh, India, Aerosol Air Qual. Res, 11, 696-708, 10.4209/aaqr.2011.03.0023, 2011.

Park, S. S., Kim, Y. J.: $PM_{2.5}$ particles and size-segregated ionic species measured during fall season in three urban sites in Korea, Atmos. Environ., 38, 1459–1471, https://doi.org/10.1016/j.atmosenv.2003.12.004, 2004."

7. Lines 317-318: Why are the K^+ concentrations similar in both seasons? Possible explanations should be given.
Reply: Thank for the reviwer's comments. We aggred with the comments but we did not find any proper explation for similar concentration of K^+ during winter and spring therfore we have deleted line number 317-318 from original manucript.

8. Line 369: replace "where aerosols" by "where BB aerosols".
Reply: Thank you for reviewer's suggestion, modification has been incorporated.
Please see line number 388 in the revised MS.

9. Lines 379-380: It is unclear to me why it should be that "the similarity between seasons indicates that $OC_{non\text{-}BB}$ originated mainly from local background sources".
Reply: As per the reviewer comments of clear explanation for seasonal variability of $OC_{non\text{-}BB}$. In previous sentence the meaning of statement is not appropriate about the seasonal trend of $OC_{non\text{-}BB}$. Therefore, we have re-phrased this sentence as

"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."
Please see line number 397-400 in the revised MS.

10. Line 399: replace "Conclusion" by "Conclusions".
Reply: As per reviewer's suggestion modification has been done.
Please see line number 419 in the revised MS.

11. Pages 21-29, Reference list: There are several problems:
- Titles of journal articles should all be in lower case instead of in Title Case.
Reply: Thank you for the comment. However, we could not find any serious problem about format of journal articles in the reference section compared to ACP reference format. Thus, no modification was done.

- For references with three of more authors, there should be ", and" preceding the last author.
Reply: We apologies by typo in reference section. Modification has been incorporated as per the suggestion in MS.

For references with only two authors, there should be " and" (without a comma) preceding the second author.
Reply: As per the reviewer's suggestion correction has been made in reference section in MS.

- Journal names should be properly abbreviated and the abbreviated words should end on a period (".").
Reply: Thank you for reviver's comments. Abbreviation of journals have been carefully checked and modification has been incorporated in references in MS.

- The reference "Jung et al., 2009" is incomplete; the article number is missing.
Reply: Article number is added to references of Jung et al., 2009.
Please line number 620 in the revised MS.

- "Jung et al., 2010" should come before "Jung et al., 2014". Besides, the reference
Reply: As per the reviewer suggestions modification has been done in the references and incorporated in the RMS.
Please see line number 617-624 in the revised MS.

"Jung et al., 2010" is incomplete; the article number is missing.
Reply: As per the reviewer's suggestion article number is added in the Jung et al., 2010.
Please see line number 624 in the revised MS.

- "Nirmalkar et al., 2015" should come before "Nirmalkar et al., 2019".
Reply: As per the reviewer's suggestion modification has been incorporated.
Please line number 662-669 in the revised MS.

- There is not referred to "Pio et al., 2008" within the text.
Reply: Pio et al., 2008 has been cited in test.
Please see line number 62-63 in the revised MS.

- *The reference "Puxbaum et al., 2007" is incomplete; the article number is missing.*
Reply: Modification has been incorporated in Puxbaum et al., 2007, and incorporated in reference section.
Please see line number 699 in the revised MS.

- *"Sullivan et al., 2008" should come before "Sullivan et al., 2019".*
Reply: As per the reviewer's suggestion modification has been done for both mentioned references.
Please see line number 733-742 in the revised MS.

Technical corrections:
- *Lines 15 and 112: replace "quartz filters" by "quartz fibre filters".*
Reply: Thank for reviewers' suggestion. Quartz filters is replaced by Quartz fiber filters.

- *Line 30: replace "of OC" by "of the OC".*
Reply: As per the reviewer's suggestion correction has been made.
Please line number 31 in the revised MS.

- *Line 32: replace "and indicate" by "and it was found".*
Reply: As per reviewer's suggestion modification done.
Please see line number 35 in the revised MS.

- *Line 46: replace "in future" by "in the future".*
Reply: Replacement is done, thank you for suggestion.
Please see line number 46 in the revised MS.

- *Line 47: replace "power-plant" by "power plants".*
Reply: Now power-plant is written as power plants in entire revised MS.
Please line number 48 in the revised MS.

- *Line 118: replace "of quartz filter" by "of each quartz fibre filter".*
Reply: The phrase "of quartz filter" is replaced by "of each quartz fibre filter", thank you for reviewer suggestions.
Please line number 125 in the revised MS.

- *Line 130: replace "were analyzed" by "were measured".*
Reply: Thank for reviewer's comments. "were analyzed" is more appropriate thus we retain it in revised MS.

- *Lines 141-142: replace "of quartz filter" by "of the quartz fibre filter".*
Reply: The phrase "of quartz filter" is replaced by "of the quartz fibre filter".
Please see number 147-148 in the revised MS.

- *Line 146: replace "analytical errors" by "analytical uncertainties".*
Reply: The phrase "analytical errors" is replaced by "analytical uncertainties".
Please line number 152 in the revised MS.

- *Line 182: replace "Table 4" by "Fig. 4".*
Reply: Thank for reviewer's comments, Table 4 is replaced by Fig. 4
Please line number line number 205 in the revised MS.

- *Line 350: replace "the previous" by "a previous" and replace "cite therein" by "cited therein".*
Reply: Replacement has been incorporated in RMS. In place of cited therein we incorporated relevant references.
Please see line number 358, 359-361 in the revised MS

- *Line 351: replace "in this" by "at this".*
Reply: Replacement has been done.
Please see line number 361 in the revised MS.

- *Line 354: replace "closed to" by "close to".*
Reply: Replacement done.
Please see line number 364 in the revised MS.

- *Line 374: replace "likely to be due" by "likely due".*
Reply: Replacement has been incorporated.
Please see line number 394 in the revised MS.

- *Line 409: replace "of OC" by "of the OC".*
Reply: Re-placement is done accordingly.
Please line number 430 in the revised MS.

- *Line 412: replace "spring due" by "in spring due".*
Reply: Replacement has been done.
Please line number 433 in the revised MS.

- *Line 419: replace "Batmunkh Tsatsral" by "Tsatsral Batmunkh".*
Reply: Thank for reviewers' comments. Name has been corrected.
Please line number 440 in the revised MS.

- *Line 642: replace "2008a" by "2008".*
Reply: Replacement has been done.
Please line number 707 in the revised MS.

- *Line 679: replace "Asia. Sci." by "Asia, Sci.".*
Reply: Replacement has been made.
Please see line number 757 in the revised MS.

- *Page 34, caption of Fig. 7: replace "in during" by "during".*
Reply: Thank you for the reviewer's suggestion. Caption of Fig. 8 (previously Fig. 7 in original manuscript) has been modified by incorporating during in place of in during.

Modified caption has been added in page 37, lines 827-828 in the revised MS.
"Fig. 8 (previously Fig. 7) 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."