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-BB} ($OC-OC_{BB}$) 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-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:

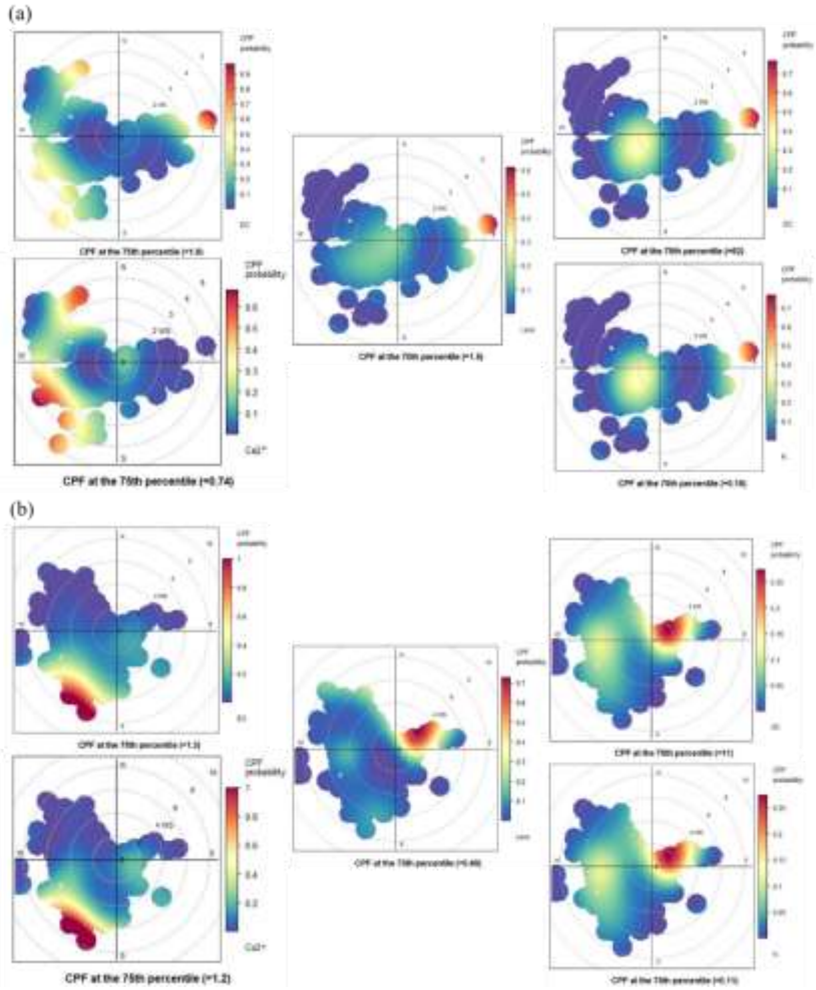
$$CPF = \frac{m_{\Delta\Theta}}{n_{\Delta\Theta}}$$

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 $OC_{non\text{-}BB}$ (OCtotal–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, $\Delta\Theta$:

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

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."

Modified Fig. 4 has been modified in Revised MS.

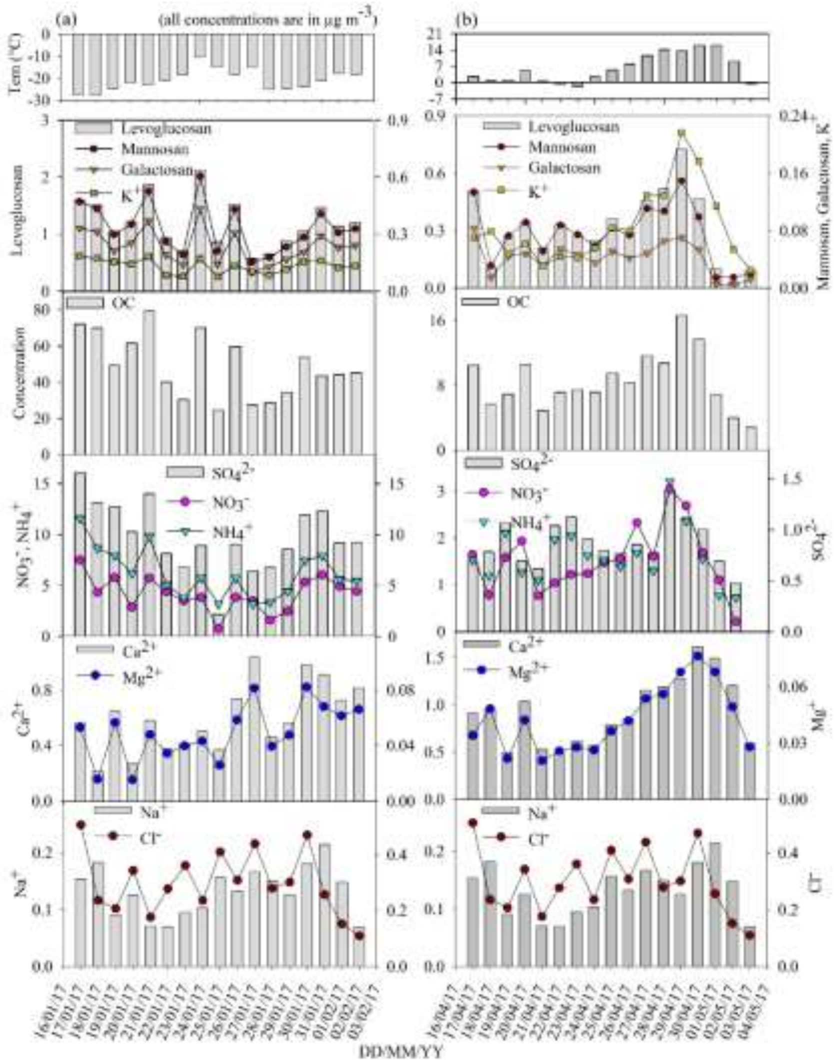
Two references have been added in the reference section in the revised MS.

"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-BB}). We have already discussed about OC_{non-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}).