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."