General comments

This manuscript by Cheng et al. provided the detailed information of biomass burning markers, including levoglucosan, mannosan and water-soluble potassium (K^+), over Beijing as well as in source samples. The PMF model and the ratios of these markers were also applied to indicate the contribution of biomass burning. It is also very interesting that the authors tried to identify different types of biomass based on the comparison of the levoglucosan to K^+ ratio and the levoglucosan to mannosan ratio. The manuscript merits publication in ACP.

Major comments

(1) My major concern is the PMF model. Firstly, the authors used 7 species as input variables. However, these species are not independent. OC contains WSOC; and oxalate and levoglucosan contributes to OC and WSOC. Since there were 14 species measured in this study, authors are suggested to use independent species as input variables and re-run PMF model. Secondly, the factor explanation is not so robust and reasonable. As authors described, Factor 1 was "secondary nature" and Factor 2 was "not primary". However, levoglucosan and K^+ that have high loadings in the two factors are both primary tracers. Are there any secondary sources or processes leading to levoglucosan and K^+? Thirdly, as the PMF results showed, BB was the most important contributor to OC (50%), which was quite different from other studies in Beijing, such as by Wang (2009) and Zheng (2005). Authors should make a comparison of BB contribution to OC among different studies in Beijing and explain the potential reasons.

Our response: Three points were raised about the PMF model. Firstly, it is suggested that independent species should be used to run the PMF model, which is also mentioned by Referee #1. To our understanding, it is acceptable to use overlapping species as the input data for the PMF analysis. For example, WSOC, levoglucosan, oxalate were included in the PMF modeling in a study conducted in the southeastern US, although levoglucosan and oxalate are part of WSOC (Zhang et al., 2010, 2012). Moreover, both OC and organic tracers were used to run the PMF model in several previous studies, although these organic tracers constitute a fraction of OC (e.g., Jaeckels et al., 2007). In addition, as shown in the User Guide of PMF (US EPA, 2008), both OC and OM (organic matter) could be used as the input data for the PMF analysis.

The second point is about the explanation of factor 1 and 2. We agree with the

referee that the description that “factor 2 was likely not primary” is not suitable, since factor 2 has high loading of levoglucosan. In fact, we think factor 2 should be associated with condensation processes. We have revised the explanation of factor 2 to avoid any misunderstandings. On the other hand, high loading of K^+ in factor 1 and the secondary nature of factor 1 do not contradict each other, because K^+ is not only from primary sources such as biomass burning (e.g., gaseous acid can react with minerals to yield water-soluble potassium).

The third point is about the estimated biomass burning contribution to OC, which is quite different from Wang et al. (2009) and Zheng et al. (2005). A variety of factors could be responsible for this difference. (1) We notice that the measurement period during summer is quite different between Wang et al. (2009) and our study. The collection of summer samples were performed during June and July 2011 in our study, whereas in Wang et al. (2009), the summer samples were collected from 2 to 31 August 2005 and from 16 August to 10 September 2006. (2) We notice that Zheng et al. (2005) relied on a relative small set of samples. Though five sampling sites were included (four urban sites and one rural site), there were only five samples for each site during each season. We are not sure whether it is reliable to use five samples to represent a season, it is surprising that Zheng et al. (2005) showed the biomass burning contribution to OC was negligible during summer in Beijing, which does not agree with the considerable summertime levoglucosan concentrations measured by Zhang et al. (2008), Wang et al. (2009) and our study. (3) The study by Zheng et al. (2005) was conducted in the year 2000, more than ten years earlier comparing with our study. During the last decade, the Beijing Municipal Government has implemented numerous air pollution control strategies, such as relocating heavy-polluting industrial facilities, implementing more stringent local emission standards for coal-fired boilers, and replacing older vehicle fleets with newer and cleaner ones. Thus, the source of $PM_{2.5}$ in Beijing might have changed substantially during the last decade, introducing a lot of difficulties and complexities in the direct comparison of source apportionment results between our study and much earlier ones such as Zheng et al. (2005). (4) Source apportionment by Wang et al. (2009) and Zheng et al. (2005) was based on CMB, whereas our study relied on PMF. A comparison study based on the same data set showed that there were indeed some discrepancies in the source apportionment results between CMB and PMF (Jaeckels et al., 2007). However, the different source apportionment results observed by our study and the two earlier studies (i.e., Wang et al., 2009; Zheng et al., 2005) can

not be simply attributed to the methodology, because the samples used in our study and the two earlier ones are substantially different.

At present, we feel very difficult to provide reliable and robust explanation for the different source apportionment results observed by our study and the two earlier ones, mainly because there are too many possible factors that are very difficult to evaluate. Nevertheless, we agree with the reviewer that comparison of source apportionment studies are important. We think that it would be more useful if the compared studies were based on either the same source apportionment model or the same samples. Thus, in the manuscript, we only presented a brief comparison of biomass burning contribution to $PM_{2.5}$ estimated by our study and the previous ones which were also based on PMF.

Specific comments

(2) Page 15 line 20-22: The authors pointed out that K^+ and levoglucosan exhibited an exponential correlation during summer BB episode. However, there are only two points with extreme high values which in fact drive the correlation between levoglucosan and K^+. If removing these two points, the correlation should be linear.

Our response: We agree with the reviewer that if removing the two points with extremely high levoglucosan concentrations, levoglucosan and K^+ measured during the BB episode would exhibit linear correlation (slope=0.09±0.02, intercept=0.03±0.12, R^2=0.71). We have clarified this point in the revised manuscript.

(3) Page 18 line 18-23: PMF results showed that BB was not the major source of K^+, while levoglucosan was mainly from BB. If so, how to explain the good correlation between levoglucosan and K^+? Is it the transport process that resulted in the good correlation?

Our response: Biomass burning contribution to K^+ showed significant seasonal variations. During the typical summer period, biomass burning was not the major source of K^+, coinciding with the week correlation (R^2=0.34) between levoglucosan and K^+. During typical winter, biomass burning was the major source of K^+, consistent with the much stronger correlation (R^2=0.82) between levoglucosan and K^+. We have clarified this point in the revised manuscript.

(4) Page 19 line 25-30: The authors only present the average source contributions for the whole dataset. Since emission sources in Beijing are probably quite different in

summer and winter, particularly for biomass burning. The average source contributions during summer and winter are suggested to be separately discussed.

Our response: We agree with the reviewer that useful information would be provided if the source apportionment results were discussed separately between winter and summer. Now we are preparing another manuscript (Zhen-yu Du, co-author of the present manuscript, is the first author) based on the chemical composition (including levoglucosan as well as other components such as OC, EC, WSOC and water-soluble ions) of about 400 $PM_{2.5}$ samples collected at the same sampling site as the present study. We plan to discuss the seasonal variations of the source apportionment results in that manuscript. After carefully considering referee's suggestions, we provided the source apportionment results during summer and winter separately in the supplementary materials.

(5) Figure 8: The diagnose plot is interesting and informative. The authors are suggested to plot all BB source samples in this figure. Thus, the readers can easily see the individual region of each BB source and the potential overlap regions.

Our response: Figure 8 was used to explain the new source identification method developed by our study. We agree with the reviewer that plotting all the source samples in Figure 8 will help the readers see the individual region of each BB source and the potential overlap regions more easily. Only ambient samples were plotted in the current version of Figure 8; and we think the current version of Figure 8 is clear and is easy to read and understand. If adding all of the source samples as suggested, Figure 8 would be too complex for an illustration of the new source identification method. Therefore, we provided a new figure, by replacing the ambient samples in Figure 8 with the source samples, in the supplementary materials.

References

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