Comment 1#

General comments:

In this study the authors reported measurement of $PM_{2.5}$ component over 3 different sites in China during a sampling period of 1 month, during spring 2019. Different saccharides were measured, including biomass burning proxy such as levoglucosan, manossan and galactosan, as well as more uncommon mono(di)saccharide, aiming at tracing the primary biogenic and possibly secondary biogenic sources. After a discussion on the potential link between emissions sources based on correlation and ratio of species, the authors attempt a source-apportionment of the different saccharide using a Non-Negative matrix Factorization (NMF) method and successfully identify 5 different factors of saccharides.

This interesting study reports a comprehensive observational dataset (although not covering the full year) and gives useful insight concerning the sources of organic components thanks to the use of proxy species not-usually used in the literature.

Reply:

Dear Prof. Samuel Weber,

We appreciate the positive comments and suggestions about the manuscript. We agree with the reviewer's comments, and have updated the manuscript on the basis of these suggestions.

Specific comments:

- Samake et al. (2019) highlight that the different polyols are mostly in the coarse fraction of the PM. Also, it has been hypothesis that the different size distribution of polyols may be a proxy of the different microbiota. Did the authors have also sampled the PM_{10} fraction and could provide the size distribution of the different saccharides?

Reply: Thank for the reviewer's suggestion. Indeed, previous results have indicated that polyols (especially mannitol and arabitol) and glucose were prevalent existed in the coarse fraction (Fu et al., 2012; Fuzzi et al., 2007; Pio et al., 2008; Yttri et al., 2007), and were mainly associated with the coarse PM fraction (Samaké et al., 2019). But PM_{10} fraction was not collected due to some practical difficulties, we can't provide the size distribution of the saccharides in this study.

We've cited a reference and rephrased the sentence in line 428-430. **"**The contribution of fungal spores might be underestimated because previous results had indicated that mannitol and arabitol were mainly associated with the coarse PM fraction (Samaké et al., 2019).**"**

1. The source apportionment (SA) is a very interesting part, although it lacks of important information that should be reported: Why didn't you included the whole species available in the SA? It could help identify more robustly BB, but also

saccharides from soil resuspension (with Ca^{2+}), and moreover quantify the apportionment of the different factors to the total $PM_{2.5}$ mass.

Reply: The source apportionment including the other species could quantify the apportionment of the different factors to the total $PM_{2.5}$ mass. We have tried to include the whole species available in the source apportionment. To make the result be better correlate with the five sources of saccharides, we ran a five-factor NMF. The result is shown as below.

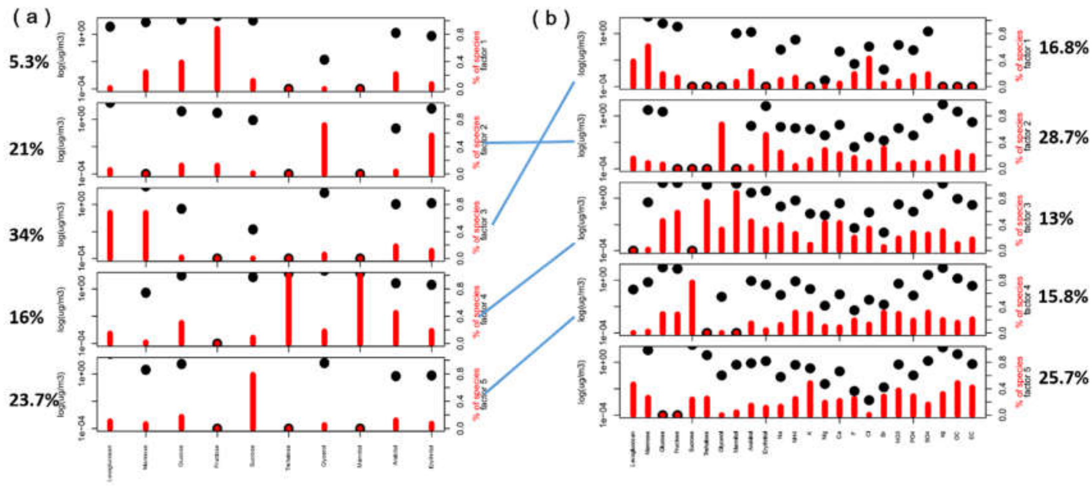

Figure 1. The factor profile obtained by NMF analysis based on the saccharide components (a) and the factor profile based on all the species (b).

In Figrue 1a, the sources of plant detritus (factor 1), plant senescence (factor 2), biomass burning (factor 3), soil microbiota (factor 4) and airborne pollen (factor 5) respectively contributed 5.3%, 21.0%, 34%, 16.0% and 23.7% to the total saccharides. We matched the factors one-to-one in the two figures according to the characteristic saccharide species. The other various species showed decentralized load on these factors. Based on the compositional data of saccharides, five factors associated to the total $PM_{2.5}$ mass were correspond one-to-one to the factors associated to the total saccharides. Factor 1-4 were correspond to the sources of biomass burning, soil microbiota, plant senescence and airborne pollen, respectively. Factor 5 was more appropriate to be thought as a mixed source.

Thus, in Figure 1b, the sources of biomass burning (factor 1), plant senescence (factor 2), soil microbiota (factor 3), airborne pollen (factor 4) and mix sources (factor 5) respectively contributed 16.8%, 28.7%, 13%, 15.8% and 25.7% to the total $PM_{2.5}$ mass. However, we think the naming of these factors associated to the total $PM_{2.5}$ mass are not accurate and comprehensive. In order to get more clear information about the sources and their contribution to the total saccharides, we decided to only report the source apportionment of saccharides.

2 It is stated that the SA is still uncertain, but no estimation of the uncertainties is given. It would be of great interest to report the species uncertainties, for instance with bootstraping your input data.

Reply: We only have 91 samples in total, so we cannot carry out resampled runs for many times. The analytical uncertainty was high in present study due to the limited sample number by using the currently used formula in PMF model. We used 0.3 plus the analytical detection limit for estimating uncertainty according to the method of Xie et al. (1999). The constant 0.3 corresponding to the log(Geometric Standard Deviation, GSD) was calculated from the normalized concentrations for all measured species, and was used to represent the variation of measurements. The use of GSD was suitable for our measurement set in a small sample size.

3 The timeserie contribution would also be of great interest. Even if the authors did not include a total variable (namely, $PM_{2.5}$), the timeserie of the total saccharide for the 5 factors would be informative.

Reply: We agree with the reviewer's view of the importance on the timeserie contribution. The timeserie of the total saccharide for the 5 factors are shown in Figure S5. We've rewritten the relevant content from Line 525. "During the sampling periods, daily variations on proportion of the five factors are shown in Figure S5. Factor 2 soil microbiota emissions could be associated to soil reclamation and cultivation of farming periods, and factors 3 plant senescence and factor 5 plant detritus could be associated to harvesting of vegetation or crop. During the observation period of a month, along with the weather warming as sunshine enhanced, human left two obvious traces of cultivated soil during 9-17 March and 27 March-8 April and a trace of vegetation or crop harvest during 17-30 March. The stronger pollen discharge occurred in March, probably due to the flowering of certain plants. The BB emissions peaked on 9, 16 March, and 1 April were more prone to be open burnings."

4 The "Soil microbiota" factor, identified mainly by the presence of Trehalose and Mannitol (and Arabitol) denotes with the finding of Samake et al. (2020) that found that Arabitol and Mannitol are associated with fungi and bacteria from the leaves and not with the soil (even if some mixing are probable). I would suggest naming it "Soil and leave microbiota".

Reply: We agree with the reviewer's suggestion, "Soil and leaves microbiota" is more specific. We've named it "Soil and leave microbiota" and gave an explanation in line 502-507. "These saccharide compounds had all been detected in the suspended soil particles and associated microbiota (e.g., fungi, bacteria and algae) (Simoneit et al., 2004; Rogge et al., 2007). A recent study found that leaves were a major source of saccharides-associated microbial taxa in a rural area of France (Samaké et al., 2020). Hence, this factor was attributed to soil and leaves microbiota."

5 Overall, the naming of the different factors identified is too rapidly explained, and more detailed could be written to ease the interpretation of the different factors.

Reply: Since each type of sugar has been described in the text, the factors were resolved in a little brief way. In the new version, the naming of the different factors have been more detailed explained from Line 497.

"As shown in Figure 6a, factor 1 was characterized by high level of levoglucosan (71.8%) and mannosan (78.7%), suggesting the source of BB (Simoneit et al., 1999; Nolte et al., 2001). Factor 2 was characterized by trehalose (99.9%) and mannitol (100.0%), and was enriched in the other saccharides components, i.e., arabitol (44.1%), glucose (29.6%), erythritol (18.2%), glycerol (17.8%), levoglucosan (14.7%), and sucrose (8.6%). These saccharide compounds had all been detected in the suspended soil particles and associated microbiota (e.g., fungi, bacteria and algae) (Simoneit et al., 2004; Rogge et al., 2007). A recent study found that leaves were a major source of saccharides-associated microbial taxa in a rural area of France (Samaké et al., 2020). Hence, this factor was attributed to soil and leaves microbiota. Factor 3 has high levels of glycerol (71.4%) and erythritol (58.2%), and showed loadings of glucose (12.8%) and fructose (11.8%). Kang et al. (2018) reported that glycerol and erythritol presented larger amounts in winter and autumn, when the vegetation decomposed. This factor was thought as the sources from plant senescence and decay by microorganisms. Factor 4 exhibited a predominance of sucrose (78.7%), and showed loadings of glucose (17.2%), arabitol (11.8%). This factor was regarded as the source of airborne pollen, because pollen is the reproductive unit of plants and contains these saccharides and saccharide alcohols as nutritional components (Bieleski, 1995; Miguel et al., 2006; Fu et al., 2012). Factor 5 characterized by the dominance of fructose (88.2%) was resolved, and was enriched in glucose (38.2%) and arabitol (21.2%), thus it could be regarded as the source of plant detritus."

Minor comment:

1 Please provide the pie chart of Figure 6b in a non-3D way, as the relative proportion is much harder to see in 3D compare to regular 2D graph.

Reply: We agree with the reviewer's comment. We've provided the pie chart of Figure 6b in a 2D way in the new version of manuscript.

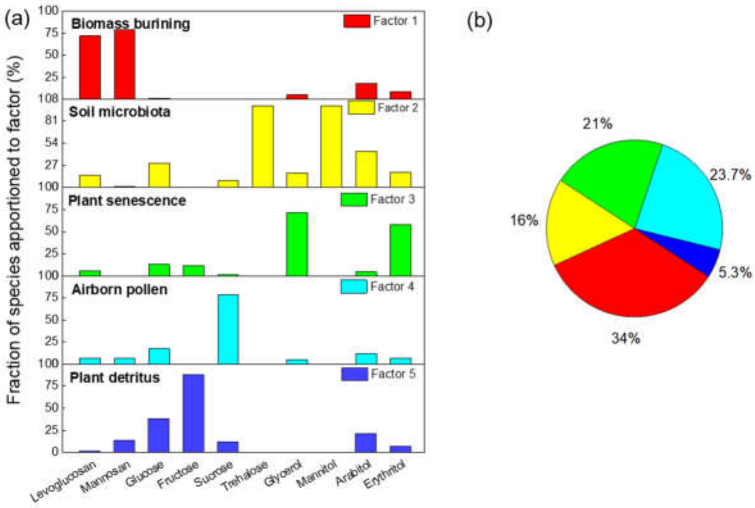

Figure 6. Factor profile obtained by NMF analysis (a). Source contribution of the five factors to the total saccharides in $PM_{2.5}$ samples (b).

References:

Fuzzi, S., Decesari, S., Facchini, M. C., Cavalli, F., Emblico, L., Mircea, M., Andreae, M. O., Trebs, I., Hoffer, A., Guyon, P., Artaxo, P., Rizzo, L. V., Lara, L. L., Pauliquevis, T., Maenhaut, W., Raes, N., Chi, X., Mayol-Bracero, O. L., Soto-García, L. L., Claeys, M., Kourtchev, I., Rissler, J., Swietlicki, E., Tagliavini, E., Schkolnik, G., Falkovich, A. H., Rudich, Y., Fisch, G., and Gatti, L. V.: Overview of the inorganic and organic composition of size-segregated aerosol in Rondônia, Brazil, from the biomassburning period to the onset of the wet season, J. Geophys. Res., 112, D01201, https://doi.org/10.1029/2005JD006741, 2007.

Pio, C. A., Legrand, M., Alves, C. A., Oliveira, T., Afonso, J., Caseiro, A., Puxbaum, H., Sanchez-Ochoa, A., and Gelencsér, 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.

Samaké, A., Jaffrezo, J.-L., Favez, O., Weber, S., Jacob, V., Albinet, A., Riffault, V., Perdrix, E., Waked, A., Golly, B., Salameh, D., Chevrier, F., Oliveira, D. M., Bonnaire, N., Besombes, J.-L., Martins, J. M. F., Conil, S., Guillaud, G., Mesbah, B., Rocq, B., Robic, P.-Y., Hulin, A., Meur, S. L., Descheemaecker, M., Chretien, E., Marchand, N., and Uzu, G.: Polyols and glucose particulate species as tracers of primary biogenic organic aerosols at 28 French sites, 19, 3357–3374, https://doi.org/10.5194/acp-19-3357-2019, 2019.

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Comment 2#

General comments:

The paper entitled "Saccharide composition in atmospheric fine particulate matter at the remote sites of Southwest China and estimates of source contributions" by Zhenzhen Wang and colleagues provide the characteristic of saccharides during spring 2019 at Lincang, a rural site in Southwest China. The authors reported molecule tracers including anhydrosugars, mono (di) saccharides and sugar alcohols, combined with statistical analysis and HYSPLIT model, they concluded that biofuel and open biomass burning (BB) activities could have a significant impact on ambient aerosol levels at Lincang. Overall, this paper is logically organized, and knowledge of this work is needed and helpful for better understanding air conditions in Southwest China. The topic of this paper is within the scope of the journal Atmospheric Physics and Chemistry. I would like to recommend this paper published after the following of my concerns be resolved.

Reply: We appreciate the positive comments and suggestions about the manuscript. We agree with the reviewer's comments, and have updated the manuscript on the basis of these suggestions.

Major comments:

1. The surrounding environmental condition is crucial for understanding the results, I strongly suggest the authors added a figure to show the sampling sites as Figure 1. This figure should include some necessary information about the topography, vegetation, residential area nearby Lincang, and photos of three sampling sites are also crucially needed.

   Reply: We've added Figure S1 for the location of the sampling sites in the Supporting Information. The number of all the Figures referring to the Supporting Information has been changed.

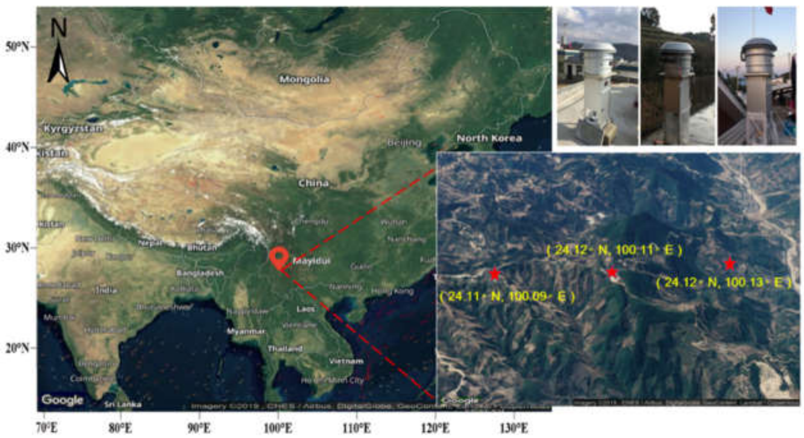

Figure S1. Map of sampling sites. The location of the sampling sites was marked with five-pointed star.

2. The source appointment is mainly based on the 72h backward trajectories of HYSPLIT model. However, high uncertainty existent for the application of HYSPLIT model at high elevation site because topographic relief. The frequencies of HYSPILT or meteorological analysis should provide more creditable results.

   Reply: Thank for the reviewer's suggestion. More detailed analyses on topography and meteorology, as well as the frequencies of HYSPILT backward trajectories are stated in the section 3.2 Sources and transport.

   Herein, this sentence has been rewritten. "46.7% of air mass backward trajectories were generally over 2000 meters, while 53.3% of them were below 2000 meters."

   "The southwest wind from the Indian Ocean prevailed at Lincang all the year round. In spring, the southwest wind was often affected by the low temperature downhill wind blowing from the snow-covered Hengduan Mountains. The weather alternated between hot and cold frequently, with unstable air pressure and strong wind. Therefore, the lower air could be diluted by the relatively clean cold air over the plateau. The upper air mainly came from the westerlies."

Minor comments:

1. The samples of this work are mainly in spring, the title should be changed to "Saccharide composition in atmospheric fine particulate matter during spring at the remote sites of Southwest China and estimates of source contributions".

   Reply: Thank for the reviewer's suggestion. The title have been changed to "Saccharide composition in atmospheric fine particulate matter during spring at the remote sites of Southwest China and estimates of source contributions".

2. Line 62, Wu et al., 2020 is not cited in references.

   Reply: Wu et al., 2020 have been cited in Line 62 in the revised manuscript.

3. Line 71-72, "10.1-383.4 ng m^{-3} over the Tibetan Plateau (Li et al., 2019)", the reference Li et al., 2019, EP is glacier cryoconites not aerosol samples.

Reply: “10.1-383.4 ng m^{-3} over the Tibetan Plateau (Li et al., 2019)” have been changed to “10.1-383.4 ng g^{-1} dry weight in cryoconites over the Tibetan Plateau (Li et al., 2019)”.

4. Line 75, Sichuan Basin, not “Chengdu Basin”.
   Reply: “Chengdu basin” have been changed to “Sichuan Basin”.

5. Line 79-81, Levoglucosan emission of China is estimated by BB activities by Wu et al., 2021, this sentence is not rigorous.
   Reply: This sentence have been rewritten. “Recently study reported that total levoglucosan emission of China exhibited a clear decreasing trend from 2014 (145.7 Gg) to 2018 (80.9 Gg) (Wu et al., 2021), suggesting BB activities might reduce in China.

6. Line 109-112, you should better add some references.
   Reply: “Referring to the official website of Lincang Municipal People's Government, the forest coverage rate of Lincang reaches to 65%.”

7. Line 116, do you have samples over other period?
   Reply: We only sampled at the Lincang sites for a period of about a month.

8. Line 126-130, please add a figure for sample sites.
   Reply: We’ve added Figure S1 for the location of the sampling sites in the Supporting Information.

9. Line 183, why do not use meteorological data at Lincang?
   Reply: The satellite data and Lincang meteorological website data were not exactly the same, but were overall similar. In order to obtain more complete data of all indicators, satellite data were used uniformly.

10. Line 231-233, “no distinct variation”, has statistical significance?
    Reply: Thank for the reviewer’s correction. This sentence is not completely accurate. In the revised manuscript, this sentence was deleted.

11. Line 239-248, samples in those references are not collected at the same period.

Reply: Indeed, the samples in these studies were collected at different times. So we presented the specific sampling time of each research. Even if not all samples were taken in the spring, it would be of great interest to report these information.

12. Line 276-277, how about the L/M for burned ghost money?

    Reply: "It was worth noting that the peak days during 31 March-1 April (L/M = 11.52 ± 1.34) neared the Qingming Festival. Another possibility of BB events was that people burned ghost money to sacrifice ancestor according to Chinese tradition."

13. Line 290-291, references for L/K^+?

    Reply: We've added the references "(Schkolnik et al., 2005; Lee et al., 2010)".

14. Line 431-441, Figure 4, only one air mass from Hengduan Mountain region. Maybe frequency is better for understanding air sources.

    Reply: Thank for the reviewer's suggestion. Herein, this sentence has been rewritten. "46.7% of air mass backward trajectories were generally over 2000 meters, while 53.3% of them were below 2000 meters."

15. Line 450-452, how about the atmospheric dynamics for aerosol transport from Southeast Asia to Lincang, especially for residential cooking and heating.

    Reply: Some sentences were added. "The southwest wind from the Indian Ocean prevailed at Lincang all the year round. In spring, the southwest wind was often affected by the low temperature downhill wind blowing from the snow-covered Hengduan Mountains. The weather alternated between hot and cold frequently, with unstable air pressure and strong wind. Therefore, the lower air could be diluted by the relatively clean cold air over the plateau. The upper air mainly came from the westerlies."

16. Line 512, ng m^{-3}?

    Reply: "µg m^{-3}" has been replaced by "ng m^{-3}".

17. Line 521, only Myanmar.

Reply: "The sampling sites suffered from both local emissions and BB via long-range transport from Southeast Asia (Myanmar, Bangladesh) and the northern Indian Peninsula."

Comment 3#

General comments:

This manuscript presents measurement results of particulate sugar compounds from a rural region in Southwest China. Individual sugar species concentrations, correlations among each other, as well as diagnostic ratios were utilized together with meteorological parameters, back trajectories, and fire counts to constrain the main emission sources, including biomass burning, microorganisms and plant emissions. Biomass burning emissions were the dominant contributor to the ambient $PM_{2.5}$, derived from both local burning activities and long-range transport from surrounding countries.

The results presented in this paper are interesting as they give insight into the sources of ambient aerosols in this part of China for which limited data have been reported. The results are based on a sound measurement approach, and include a large number of chemical PM components, while the measurement period is relatively short and doesn't show seasonal patterns. Overall, the manuscript is fairly well written and structured, and should therefore be published in ACP following minor revision based on the comments given below.

Reply: We appreciate the positive comments and suggestions about the manuscript. We agree with the reviewer's comments, and have updated the manuscript on the basis of these suggestions.

Specific comments:

1. It is good to see the utilization of the Metrohm sugar columns (requiring substantially lower eluent concentrations), instead of the usual CarboPak columns from Dionex used in most other studies. Did the authors encounter any co-elution problems of certain sugar species with this system?

   Reply: We have encountered some co-elution problems when using the Metrohm sugar column. At first, we prepared twenty standard saccharide compounds for the method test, and found that several saccharides co-eluted. By changing the concentration of the eluent and the flow rate, there were still some saccharides compounds that cannot be separated well.

For example, it was difficult to separate glycerol and sorbitol, the retention times of which were respectively 5.82 and 5.97 under the condition of the method in this paper. Because there could be a ~5% deviation of the peak location, data of sorbitol was not accurate and was not included in this paper. When testing the outfield samples, the sorbitol peak might be attributed to glycerol.

Under the same condition, we repeated the experiment many times to carefully identify the peak location for every saccharide. The relative deviation of retention time and peak area were less than 1%. When it showed a good linear relationship between peak area and concentration value (R^2>99.9%), the saccharides were selected to measure. We finally decided to test thirteen kinds of saccharide compounds in this article. The selected saccharides were inositol, glycerol, erythritol, arabitol, trehalose, manitol, mannose, glucose, fructose, galactosan, levoglucosan, mannosan and sucrose, the retention times of which were 4.88, 5.82, 6.22, 7.84, 8.96, 9.58, 10.93, 11.97, 14.59, 16.94, 17.96, 19.32 and 22.54, respectively.

2. Lines 276-278: Do the authors know what are the traditional burning practices during the Qingming Festival, i.e., what types of biomass the local residents may be burning that are special for that holiday or is it just enhanced cooking activity, perhaps with more outdoor BBQ cooking?

   Reply: The weather around Qingming Day is not very suitable for barbecue. We think the sudden increase in biomass burining may not be a significant cooking activity. The most likely activity is the sacrifice around the Tomb-Sweeping Day, during which large quantities of ghost money, candles and firecrackers were burned. The main raw materials of ghost money are bamboo and wood.

3. Lines 416-418: While erythritol may have been used as surrogate for the 2-methyltetrols, I believe it was mainly for quantification of the 2-methyltetrol peaks when no authentic standards were available, rather than representing the ambient 2-methyltetrol levels. Since the 2-methyltetrols can be separated by HPAEC-PAD, did the authors see any unidentified peaks in the sugar alcohol region of the chromatogram that could potentially be attributed to the 2-methyltetrols?

   Reply: The usage of erythritol was due to the lack of the standard 2-methyltetrols. The retention time of erythritol was very short when using the Metrohm sugar columns. The peak positions of erythritol and sorbitol were often overlapped, so it was difficult for us to find other substances in the peak location of the erythritol.

4. Lines 495-500: What are the typical crops that are planted in this region? And what kind of burning practices do the local farmers have, e.g., post-harvest burning of straw or other agricultural residues? Knowledge of these practices would be helpful for explaining the BB patterns and specifically the anhydrosugar diagnostic ratios.

   Reply: Thank for the reviewer's suggestion. This region abounds with black tea, nuts, coffee and sugar cane. The main crops in this region are rice, wheat and corn. Crop straw burning is a common phenomenon after the harvest, including the indoor combustion and open burning. We've put these information into the analysis.

   "Previous results showed the emissions from the combustion of crop residuals such as rice straw, wheat straw and corn straw exhibited comparable L/K^+ ratios, typically below 1.0. The averages of L/K^+ ratios in this study was 0.48 ± 0.20, which was higher than the ratio for wheat straw (0.10 ± 0.00) and corn straw (0.21 ± 0.08), but was lower than the ratio for Asian rice straw (0.62 ± 0.32) (Cheng et al., 2013). In this study, higher L/K^+ ratios were observed during 8-10 March (1.20 ± 0.19) than those during 31 March-1 April (0.40 ± 0.13), which suggested that the open fire event during 8-10 March was more possibly due to smoldering combustion of residues at low temperatures."

Technical corrections:

1. Throughout the manuscript, grammar and wording needs to be polished.

   Reply: Thank for the reviewer's correction. We'll try the best to polish the grammar and wording of this manuscript. The writing has been updated with the help of a colleague scientist whose native language is English.

2. Lines 144-145: Please, check the correct supplier of the DRI Model 2015 analyzer -- I don't think that it is "Atmoslytic" anymore but "Magee" or "Aerosol"

   Reply: We rechecked the relevant information and found that DRI Model 2015 analyzer was produced by the Aerosol Inc.

   Thank for the reviewer's correction. "Atmoslytic Inc." have been changed to "Aerosol Inc."