



Jun. 10, 2021

Dear Prof. Ivan Kourtchev,

Thanks for your kind handling our manuscript!

Here we uploaded our revised manuscript (ACP-2021-83) for consideration to be published on ACP:

Title: Saccharide composition in atmospheric fine particulate matter during spring at the remote sites of Southwest China and estimates of source contributions

Authors: Zhenzhen Wang, Di Wu, Zhuoyu Li, Xiaona Shang, Qing Li, Xiang Li, Renjie Chen, Haidong Kan, Jianmin Chen

Special Issue: The role of fire in the Earth system: understanding interactions with the land, atmosphere, and society (ESD/ACP/BG/GMD/NHESS inter-journal SI)

Corresponding author: Jianmin Chen; Address: Department of Environmental Science & Engineering, Fudan University, Shanghai 200433, China; Email: jmchen@fudan.edu.cn.

Thank you for your kind reminder. To better answer the questions from the reviewer #3, we replied to all questions, and also added some sentences reflecting the comments (including questions 1 and 2) to the revised paper.

We appreciate the positive comments and suggestions about the manuscript. We are willing to categorize our manuscript into “Measurement Reports” if necessary.

We acknowledge the comments of three reviewers. The suggestions of the Reviewers gave us great help to improve our manuscript. We have updated the manuscript on the basis of the Reviewers’ comments. Below is our response to the comments resulting in a number of clarifications. A marked file in the PDF format was also uploaded so that the reviewers could easily check our update. We expect this manuscript to be published on Atmospheric Chemistry and Physics.

Sincerely yours,
Jianmin Chen

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

36 sources based on correlation and ratio of species, the authors attempt a source-apportionment of
37 the different saccharide using a Non-Negative matrix Factorization (NMF) method and
38 successfully identify 5 different factors of saccharides.

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

42

43 **Reply:**

44 Dear Prof. Samuel Weber,

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

47

48 **Specific comments:**

49 1 Samake et al. (2019) highlight that the different polyols are mostly in the coarse fraction of
50 the PM. Also, it has been hypothesis that the different size distribution of polyols may be a
51 proxy of the different microbiota. Did the authors have also sampled the PM₁₀ fraction and
52 could provide the size distribution of the different saccharides?

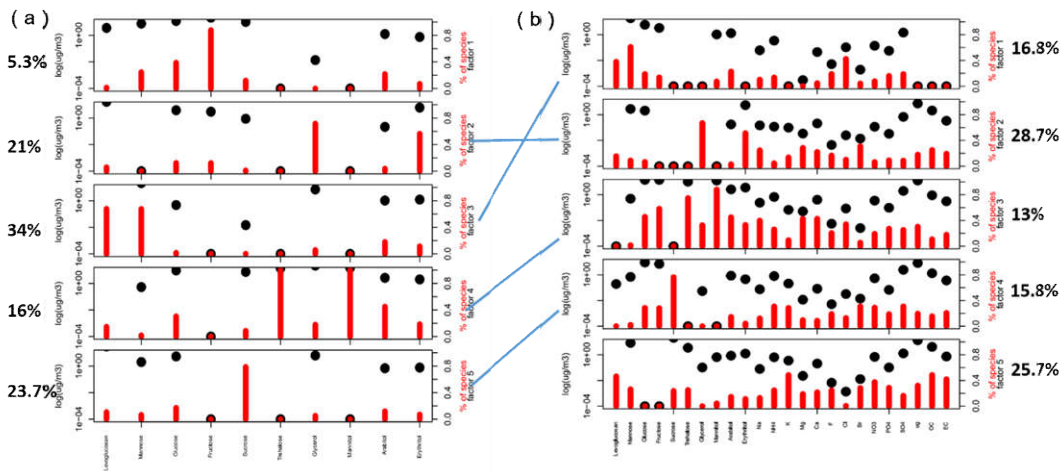
53 **Reply:** Thank for the reviewer's suggestion. Indeed, previous results have indicated that
54 polyols (especially mannitol and arabitol) and glucose were prevalent existed in the coarse
55 fraction (Fu et al., 2012; Fuzzi et al., 2007; Pio et al., 2008; Yttri et al., 2007), and were
56 mainly associated with the coarse PM fraction (Samaké et al., 2019). But PM₁₀ fraction was
57 not collected due to some practical difficulties, we can't provide the size distribution of the
58 saccharides in this study.

59 We've cited a reference and rephrased the sentence in line 440-442. "The contribution of
60 fungal spores might be underestimated because previous results had indicated that mannitol
61 and arabitol were mainly associated with the coarse PM fraction (Samaké et al., 2019)."

62

63 2 The source apportionment (SA) is a very interesting part, although it lacks of important
64 information that should be reported: Why didn't you included the whole species available in
65 the SA? It could help identify more robustly BB, but also saccharides from soil resuspension
66 (with Ca²⁺), and moreover quantify the apportionment of the different factors to the total
67 PM_{2.5} mass.

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



72
73 Figure 1. The factor profile obtained by NMF analysis based on the saccharide components (a)
74 and the factor profile based on all the species (b).

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

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

90
91 3 It is stated that the SA is still uncertain, but no estimation of the uncertainties is given. It
92 would be of great interest to report the species uncertainties, for instance with bootstrapping
93 your input data.

94 **Reply:** We only have 91 samples in total, so we cannot carry out resampled runs for many
95 times. The analytical uncertainty was high in present study due to the limited sample number
96 by using the currently used formula in PMF model. We used 0.3 plus the analytical detection
97 limit for estimating uncertainty according to the method of Xie et al. (1999). The constant 0.3
98 corresponding to the log(Geometric Standard Deviation, GSD) was calculated from the

99 normalized concentrations for all measured species, and was used to represent the variation of
100 measurements. The use of GSD was suitable for our measurement set in a small sample size.

101
102 4 The timeserie contribution would also be of great interest. Even if the authors did not include
103 a total variable (namely, PM_{2.5}), the timeserie of the total saccharide for the 5 factors would
104 be informative.

105 **Reply:** We agree with the reviewer's view of the importance on the timeserie contribution.
106 The timeserie of the total saccharide for the 5 factors are shown in Figure S5. We've rewritten
107 the relevant content from Line 536. "During the sampling periods, daily variations on
108 proportion of the five factors are shown in Figure S5. Factor 2 soil microbiota emissions
109 could be associated to soil reclamation and cultivation of farming periods. Factors 3 plant
110 senescence and factor 5 plant detritus could be associated to harvesting of vegetation or crop.
111 During the observation period of a month, along with the weather warming as sunshine
112 enhanced, human left two obvious traces of cultivated soil during 9-17 March and 27 March-
113 8 April and a trace of vegetation or crop harvest during 17-30 March. The stronger pollen
114 discharge occurred in March, probably due to the flowering of certain plants. The BB
115 emissions peaked on 9, 16 March, and 31 March-1 April were more prone to be open
116 burnings. Therein, the BB during 31 March-1 April was probably from the burning of ghost
117 money during the Qingming Festival."

118
119 5 The "Soil microbiota" factor, identified mainly by the presence of Trehalose and Mannitol
120 (and Arabitol) denotes with the finding of Samake et al. (2020) that found that Arabitol and
121 Mannitol are associated with fungi and bacteria from the leaves and not with the soil (even if
122 some mixing are probable). I would suggest naming it "Soil and leave microbiota".

123 **Reply:** We agree with the reviewer's suggestion, "Soil and leaves microbiota" is more specific.
124 We've named it "Soil and leave microbiota" and gave an explanation in line 514-522. "These
125 saccharide compounds had all been detected in the suspended soil particles and associated
126 microbiota (e.g., fungi, bacteria and algae) (Simoneit et al., 2004; Rogge et al., 2007). A recent
127 study found that leaves were a major source of saccharides-associated microbial taxa in a rural
128 area of France (Samaké et al., 2020). Hence, this factor was attributed to soil and leaves
129 microbiota."

130
131 6 Overall, the naming of the different factors identified is too rapidly explained, and more
132 detailed could be written to ease the interpretation of the different factors.

133 **Reply:** Since each type of sugar has been described in the text, the factors were resolved in a
134 little brief way. In the new version, the naming of the different factors have been more detailed
135 explained from Line 509.

136 “As shown in Figure 6a, factor 1 was characterized by high level of levoglucosan (71.8%)
 137 and mannosan (78.7%), suggesting the source of BB (Simoneit et al., 1999; Nolte et al., 2001).
 138 Factor 2 was characterized by trehalose (99.9%) and mannitol (100.0%), and was enriched in
 139 the other saccharides components, i.e., arabitol (44.1%), glucose (29.6%), erythritol (18.2%),
 140 glycerol (17.8%), levoglucosan (14.7%), and sucrose (8.6%). These saccharide compounds had
 141 all been detected in the suspended soil particles and associated microbiota (e.g., fungi, bacteria
 142 and algae) (Simoneit et al., 2004; Rogge et al., 2007). A recent study found that leaves were a
 143 major source of saccharides-associated microbial taxa in a rural area of France (Samaké et al.,
 144 2020). Hence, this factor was attributed to soil and leaves microbiota. Factor 3 has high levels
 145 of glycerol (71.4%) and erythritol (58.2%), and showed loadings of glucose (12.8%) and
 146 fructose (11.8%). Kang et al. (2018) reported that glycerol and erythritol presented larger
 147 amounts in winter and autumn, when the vegetation decomposed. This factor was thought as
 148 the sources from plant senescence and decay by microorganisms. Factor 4 exhibited a
 149 predominance of sucrose (78.7%), and showed loadings of glucose (17.2%), arabitol (11.8%).
 150 This factor was regarded as the source of airborne pollen, because pollen is the reproductive
 151 unit of plants and contains these saccharides and saccharide alcohols as nutritional components
 152 (Bielecki, 1995; Miguel et al., 2006; Fu et al., 2012). Factor 5 characterized by the dominance
 153 of fructose (88.2%) was resolved, and was enriched in glucose (38.2%) and arabitol (21.2%),
 154 thus it could be regarded as the source of plant detritus.”

155
 156 **Minor comment:**

157 1 Please provide the pie chart of Figure 6b in a non-3D way, as the relative proportion is much
 158 harder to see in 3D compare to regular 2D graph.

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

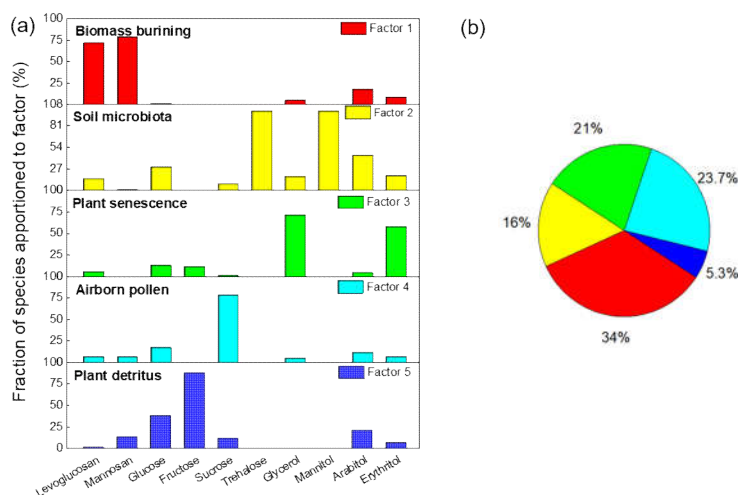


Figure 6. Factor profile obtained by NMF analysis (a). Source contribution of the five factors to the total saccharides in PM_{2.5} samples (b).

- 161
162 **References:**
163 Fuzzi, S., Decesari, S., Facchini, M. C., Cavalli, F., Emblico, L., Mircea, M., Andreae, M. O.,
164 Trebs, I., Hoffer, A., Guyon, P., Artaxo, P., Rizzo, L. V., Lara, L. L., Pauliquevis, T., Maenhaut,
165 W., Raes, N., Chi, X., Mayol-Bracero, O. L., Soto-García, L. L., Claeys, M., Kourtchev, I.,
166 Rissler, J., Swietlicki, E., Tagliavini, E., Schkolnik, G., Falkovich, A. H., Rudich, Y., Fisch, G.,
167 and Gatti, L. V.: Overview of the inorganic and organic composition of size-segregated aerosol in
168 Rondônia, Brazil, from the biomassburning period to the onset of the wet season, *J. Geophys.*
169 *Res.*, 112, D01201, <https://doi.org/10.1029/2005JD006741>, 2007.
170 Pio, C. A., Legrand, M., Alves, C. A., Oliveira, T., Afonso, J., Caseiro, A., Puxbaum, H.,
171 Sanchez-Ochoa, A., and Gelencsér, A.: Chemical composition of atmospheric aerosols during the
172 2003 summer intense forest fire period, *Atmos. Environ.*, 42, 7530– 7543,
173 <https://doi.org/10.1016/j.atmosenv.2008.05.032>, 2008.
174 Samaké, A., Jaffrezo, J.-L., Favez, O., Weber, S., Jacob, V., Albinet, A., Riffault, V., Perdrix, E.,
175 Waked, A., Golly, B., Salameh, D., Chevrier, F., Oliveira, D. M., Bonnaire, N., Besombes, J.-L.,
176 Martins, J. M. F., Conil, S., Guillaud, G., Mesbah, B., Rocq, B., Robic, P.-Y., Hulin, A., Meur, S.
177 L., Descheemaeker, M., Chretien, E., Marchand, N., and Uzu, G.: Polyols and glucose
178 particulate species as tracers of primary biogenic organic aerosols at 28 French sites, 19, 3357–
179 3374, <https://doi.org/10.5194/acp-19-3357-2019>, 2019.
180 Samaké, A., Bonin, A., Jaffrezo, J.-L., Taberlet, P., Weber, S., Uzu, G., Jacob, V., Conil, S., and
181 Martins, J. M. F.: High levels of primary biogenic organic aerosols are driven by only a few
182 plant-associated microbial taxa, 20, 5609–5628, <https://doi.org/10.5194/acp-20-5609-2020>, 2020.

184 **Comment 2#**

185 **General comments:**

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

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

198 **Major comments:**

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

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

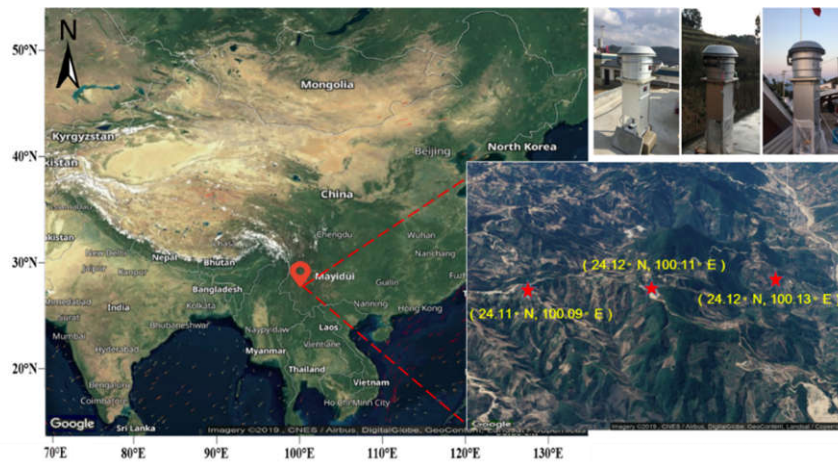


Figure S1. Map of sampling sites. The location of the sampling sites was marked with five-pointed star.

207
208 2. The source appointment is mainly based on the 72h backward trajectories of HYSPLIT
209 model. However, high uncertainty existent for the application of HYSPLIT model at high
210 elevation site because topographic relief. The frequencies of HYSPLIT or meteorological
211 analysis should provide more creditable results.

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

215 Herein, this sentence has been rewritten in line 472. "46.7% of air mass backward
216 trajectories were generally over 2000 meters, while 53.3% of them were below 2000
217 meters."

218 Some meteorological analysis has been added in line 486-492. "The southwest wind
219 from the Indian Ocean prevailed at Lincang all the year round. In spring, the southwest
220 wind was often affected by the low temperature downhill wind blowing from the snow-

221 covered Hengduan Mountains. The weather alternated between hot and cold frequently,
222 with unstable air pressure and strong wind. Therefore, the lower air could be diluted by
223 the relatively clean cold air over the plateau. The upper air mainly came from the
224 westerlies.”

225

226 **Minor comments:**

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

230 **Reply:** Thank for the reviewer’s suggestion. The title have been changed to “Saccharide
231 composition in atmospheric fine particulate matter during spring at the remote sites of
232 Southwest China and estimates of source contributions”.

233

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

235 **Reply:** “Wu et al., 2020” has been corrected to “Wu et al., 2021”. “(Wu et al., 2021)” has
236 been cited in Line 62 in the revised manuscript.

237

238 3. Line 71-72, “10.1-383.4 ng m⁻³ over the Tibetan Plateau (Li et al., 2019)”, the reference
239 Li et al., 2019, EP is glacier cryoconites not aerosol samples.

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

242

243 4. Line 75, Sichuan Basin, not “Chengdu Basin”.

244 **Reply:** “Chengdu basin” have been changed to “Sichuan Basin” in line 76.

245

246 5. Line 79-81, Levoglucosan emission of China is estimated by BB activities by Wu et al.,
247 2021, this sentence is not rigorous.

248 **Reply:** This sentence have been rewritten. “Recently study reported that total
249 levoglucosan emission of China exhibited a clear decreasing trend from 2014 (145.7 Gg)
250 to 2018 (80.9 Gg) (Wu et al., 2021), suggesting BB activities might reduce in China.

251

252 6. Line 109-112, you should better add some references.

253 **Reply:** In line 113, “Referring to the official website of Lincang Municipal People's
254 Government, the forest coverage rate of Lincang reaches to 65%.”

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256 7. Line 116, do you have samples over other period?

257 **Reply:** We only sampled at the Lincang sites for a period of about a month.

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8. Line 126-130, please add a figure for sample sites.
Reply: Line 138, 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: In line 294-298, "It was worth noting that the peak days during 31 March-1 April (L/M = 11.52 ± 1.34) neared the Qingming Festival. Therefore, another possibility of BB events was that people burned large quantities of ghost money, candles and firecrackers to sacrifice ancestor according to Chinese tradition. The main raw materials of ghost money are bamboo and wood."
 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 in line 472-473. "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.

295 **Reply:** Some sentences were added in line 486-492. “The southwest wind from the Indian
296 Ocean prevailed at Lincang all the year round. In spring, the southwest wind was often
297 affected by the low temperature downhill wind blowing from the snow-covered
298 Hengduan Mountains. The weather alternated between hot and cold frequently, with
299 unstable air pressure and strong wind. Therefore, the lower air could be diluted by the
300 relatively clean cold air over the plateau. The upper air mainly came from the westerlies.”
301

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

303 **Reply:** In line 561, “ $\mu\text{g m}^{-3}$ ” has been replaced by “ ng m^{-3} ”.

304
305 17. Line 521, only Myanmar.
306 **Reply:** In line 569-571, “The sampling sites suffered from both local emissions and BB
307 via long-range transport from Southeast Asia (Myanmar, Bangladesh) and the northern
308 Indian Peninsula.”
309

310 **Comment 3#**

311 **General comments:**

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

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

325
326 **Reply:** We appreciate the positive comments and suggestions about the manuscript. We agree
327 with the reviewer's comments, and have updated the manuscript on the basis of these suggestions.
328

329 **Specific comments:**

330 1. It is good to see the utilization of the Metrohm sugar columns (requiring substantially lower
331 eluent concentrations), instead of the usual CarboPak columns from Dionex used in most

332 other studies. Did the authors encounter any co-elution problems of certain sugar species with
333 this system?

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

338 For example, it was difficult to separate glycerol and sorbitol, the retention times of which
339 were respectively 5.82 and 5.97 under the condition of the method in this paper. Because
340 there could be a ~1% deviation of the peak location, data of sorbitol was not accurate and was
341 not included in this paper. When testing the outfield samples, the sorbitol peak might be
342 attributed to glycerol.

343 Under the same condition, we repeated the experiment many times to carefully identify the
344 peak location for every saccharide. The relative deviation of retention time and peak area
345 were less than 1%. When it showed a good linear relationship between peak area and
346 concentration value ($R^2 > 99.9\%$), the saccharides were selected to measure. We finally
347 decided to test thirteen kinds of saccharide compounds in this article. The selected
348 saccharides were inositol, glycerol, erythritol, arabitol, trehalose, manitol, mannose, glucose,
349 fructose, galactosan, levoglucosan, mannosan and sucrose, the retention times of which were
350 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,
351 respectively.

352 Some sentences were added in the section of 2.2 Measurements. "In the preliminary
353 experiment, some co-elution problems were encountered when using the Metrohm sugar
354 column. By changing the concentration of the eluent and the flow rate, the measurements of
355 every saccharide were repeated many times to ensure that the relative deviation of retention
356 time and peak area was less than 1% and the correlation between peak area and concentration
357 value was more than 99.9%."

358 .
359 2. Lines 276-278: Do the authors know what are the traditional burning practices during the
360 Qingming Festival, i.e., what types of biomass the local residents may be burning that are
361 special for that holiday or is it just enhanced cooking activity, perhaps with more outdoor
362 BBQ cooking?

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

368 This sentence has been rewritten in line 294-298. “It was worth noting that the peak days
369 during 31 March-1 April ($L/M = 11.52 \pm 1.34$) neared the Qingming Festival. Therefore,
370 another possibility of BB events was that people burned large quantities of ghost money,
371 candles and firecrackers to sacrifice ancestor according to Chinese tradition. The main raw
372 materials of ghost money are bamboo and wood.”

373
374 3. Lines 416-418: While erythritol may have been used as surrogate for the 2-methyltetrols, I
375 believe it was mainly for quantification of the 2-methyltetrol peaks when no authentic
376 standards were available, rather than representing the ambient 2-methyltetrol levels. Since the
377 2-methyltetrols can be separated by HPAEC-PAD, did the authors see any unidentified peaks
378 in the sugar alcohol region of the chromatogram that could potentially be attributed to the 2-
379 methyltetrols?

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

384
385 4. Lines 495-500: What are the typical crops that are planted in this region? And what kind of
386 burning practices do the local farmers have, e.g., post-harvest burning of straw or other
387 agricultural residues? Knowledge of these practices would be helpful for explaining the BB
388 patterns and specifically the anhydrosugar diagnostic ratios.

389 **Reply:** Thank for the reviewer’s suggestion. This region abounds with black tea, nuts, coffee
390 and sugar cane. The main crops in this region are rice, wheat and corn. Crop straw burning is
391 a common phenomenon after the harvest, including the indoor combustion and open burning.
392 We've put these information into the analysis from line 318.

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

401

402

403 **Technical corrections:**

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

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

408
409 2. Lines 144-145: Please, check the correct supplier of the DRI Model 2015 analyzer -- I don't
410 think that it is "Atmoslytic" anymore but "Magee" or "Aerosol"

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

413 Thank for the reviewer's correction. "Atmoslytic Inc." have been changed to "Aerosol
414 Inc." in line 152.

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