We would like to thank the anonymous reviewer's helpful comments and suggestions which, we believe, have supported to improve the quality of the current manuscript. We have tried our best to incorporate the reviewer' comments in the manuscript. In the following responses, the reviewer' original comments are in black, authors' responses in blue and changes in the manuscript in red.

Responses to Referee #2

This manuscript presents measurement results from a year-long campaign at a site in the Himalayan-Tibetan Plateau. Ambient aerosol samples were subjected to chemical speciation, including molecular source tracers. Various methods were applied to identify emission sources and estimate the contributions from the individual sources. Primary and secondary organic aerosol components were assessed in the ambient aerosol, and the single largest source contribution was determined to be from biomass burning activities. The results presented here are important for better understanding the properties and emission sources of organic aerosols at such critical sites as the Himalayan-Tibetan Plateau, which have a profound influence on regional and even global climate. The paper, therefore, fits well within the scope of the journal, and is based on an extensive data set with adequate interpretation and discussions of the findings. Thus, I recommend publication of the manuscript in ACP, upon consideration of the comments and suggestions listed below.

Response: We thank the referee for the positive evaluation on our work. We have adopted most of the comments to improve the manuscript substantially.

Specific comments:

1. Lines 112-120: In the site description there is no mention of the airport (from which the met data were obtained, as stated in line 178) that is apparently in close proximity to the sampling site, and thus could have specific source influence on the collected samples. Please, add a brief statement regarding this potential impact, including the predominant wind patterns, i.e., during which periods the site is upwind and downwind

of the airport.

Response: We agree with the reviewer. The Tribhuvan international airport was located west of the site (~ 4 km from Bode). It may influence the organic aerosols via fossil fuel combustion. However, in our current study, we only report one toluene tracer, which cannot track the pollution from the airport. Regarding the wind patterns, as described in the Section 2.1, the local wind direction varied all the time during the whole day sampling, and the pollution sources are mixed. Thus, currently it is hard to reveal the effect of airport emission by analyzing the wind pattern. Now we add a sentence of "The Tribhuvan international airport in the west of Bode (~ 4 km) may have potential impacts when there is westerly wind" in lines 116-117.

2. Lines 264-166: The data plotted in this figure are apparently annual average values. It may be interesting to see the seasonal average numbers as well.

Response: We totally agree with the referee. However, due to the sampler breakdown, power interruption and maintaining, the sample distribution is not uniform, there were less samples during post-monsoon (sample number = 9) and winter (sample number = 9). In order to obtain the convinced relationship correlation, we choose to plot the figure using the annual average values.

3. Lines 447-450: Do the authors have a possible explanation for the association of biomass burning emissions with SOA formation from monoterpenes? Is there a predominance of coniferous trees in the area which might have been subject to burning?

Response: We now add the explanation as "The forests in the Kathmandu Valley consist of broad-leaved evergreen mixed forest of Schima castanopsis at the base, oak-laurel forest in the middle (1800 to 2400 m a. s. l.) and oak forest at the top, while the conifer tree species Pinus roxiburghii (Khote Salla) and Pinus wallichiana (Gobre Salla) are also found (Department of Plant Resources, 2015;Sarkar et al., 2016). Monoterpenes were chiefly emitted from needle leaf trees (coniferous trees) (Kang et al., 2018). The forests in the Kathmandu Valley consist of broad-leaved evergreen mixed forest of Schima castanopsis at the base, oak-laurel forest in the middle (1800

to 2400 m a. s. l.) and oak forest at the top, while the conifer tree species Pinus roxiburghii (Khote Salla) and Pinus wallichiana (Gobre Salla) are also found (Department of Plant Resources, 2015;Sarkar et al., 2016). Monoterpenes were chiefly emitted from needle leaf trees (coniferous trees) (Kang et al., 2018). Therefore, it suggested that biomass-burning activities have had a significant influence on the atmospheric composition over Kathmandu Valley, especially for SOA tracers" in lines 478-483.

4. Lines 467-488: This statement should be made with caution, as a good correlation may also be due to other dominant source emissions which coincided with the biomass burning emissions.

Response: What we want to do in lines 467-488 is to roughly discuss the influencing factors that can have an impact on the toluene- SOC concentration, thus shed light on further study to concentrate on the influencing factors concerning the SOA formation. Therefore, we use univariate analysis to see which factor may influence the apportioned SOC and see the correlation between the potential influencing factors and the apportioned SOC. The correlation between different parameters could at least enlighten us of the influencing factors for SOA formation in megacities such as Kathmandu under the complex air pollution conditions.

5. Lines 471-473: An additional source of the uncertainties is the lack of representative source profiles for the given study location.

Response: We changed the expression as "It should be noted here that tracer methods can provide a reasonable estimation, but uncertainties are introduced considering the site differences and the lack of representative source profiles for the given study location." Please check lines 502-503.

6. Line 504: Why would PBAP have a large contribution to the ambient PM at the sampling site?

Response: As discussed in Section 3.3.2 and 3.3.3, "Notably, the levels of PBAP discussed above were much higher than other sites in the world (Zhu et al., 2015;Chen

et al., 2013;Liang et al., 2016), indicating the strong fungal spore production in the Kathmandu Valley during the monsoon season". Therefore, for the contribution estimation, we infer that the PBAP may have a large contribution to the ambient PM at the study site.

7. Lines 514-517: It would be interesting to see a comparison here with measurements from other sites, reported in the literature.

Response: We appreciate for the referee's comments. We now add the comparison with measurements from other sites. "There are also some similar results from the literatures. For example, Zhu et al. (2016) reported the contribution of plant debris to OC was 5.6% in nighttime and 4.6% in daytime respectively from aerosols in a mid-latitudinal forest. Szidat et al. (2006) reported the plant debris contributed to 3.2% of OC during summer in urban aerosols collected in Zurich, Switzerland. Fungal-spore-derived OC was the biggest contributor to total OC of 3.1 % (0.03 %–19.8 %) in marine aerosols collected over the East China Sea during 18 May to 12 June 2014 (Kang et al., 2018). The study in the aerosols of Brazil urban site showed the mean contributions of fungal aerosol to OC was 8% (Emygdio et al., 2018). Liang et al. (2017) reported the contributions of fungal spores to OC of 1.2 ± 0.7% and 3.5 ± 3.7% in aerosols from an urban site and a rural site respectively during an entire year in Beijing, China. All above strengthened the importance of plant-debris and fungal spores to the aerosol burden in the atmosphere. Please see lines 552-561.

8. Lines 549-551: If the authors mention dicarboxylic acids (DCAs) as an additional OC fraction, this implies that they are not associated with any of the sources for which estimates were made. What other sources would the DCAs be derived from?

Response: Dicarboxylic acids (DCAs) can be emitted both from primary and secondary sources (Kawamura and Bikkina, 2016). There may be POC and SOC contribution to DCAs. However, in the current study, we didn't detect and consider DCAs, and the contribution from DCAs is difficult to be quantified. Legrand et al. (2013) reported mono- and di-carboxylic acids, originating from a broad range of primary organic compounds, could contribute 38–44 % of OC. Therefore, the others

may include the contribution from the DCAs. We rephrased the sentence as "Additionally, low molecular weight (LMW) dicarboxylic acids from both primary and secondary sources also constitute a significant fraction of atmospheric organic aerosols (Kawamura and Bikkina, 2016)" in lines 593-595.

Technical corrections:

1. Line 60: Omit "badly" before "poor".

Response: Corrected. Please see line 60.

2. Line 66: Delete the indefinite article "a" before "concern".

Response: Corrected. Please see line 66.

3. Lines 103 and 109: Delete the definite article "the" before "Kathmandu".

Response: Corrected. Please see lines 103 and 109.

4. Lines 108 and 109: Add the definite article "the" before "central-eastern", "Nagarkot", and "Bode".

Response: Corrected. Please see lines 108 and 109

5. Line 117: Add "of" before "a mix".

Response: Added. Please see lines 118.

6. Line 147: The sentence should start with "A trace gas chromatograph", and the name of the manufacturer is "Thermo Scientific".

Response: Corrected. Please see lines 155-156 as "A trace gas chromatography coupled to a Polaris Q mass spectrometry detector (GC-MS, Thermo Scientific) was used for analysis."

7. Line 153: The first part of the sentence is not complete and therefore needs to be reworded; especially the word "While" is not fitting here.

Response: We reorganized the expression as "For quantitative analysis, calibration curves were established by using authentic standards that were processed as described above. For the quantification of target compounds that were no available standards, they were estimated by the following surrogate compounds:" in lines 161-162.

8. Lines 160, 181, and 188: Add the definite article "the" before "current", "wet", and "Bode".

Response: Corrected. Please see lines 169, 204 and 211.

9. Line 162: Delete the definite article "the" before "artifacts".

Response: Deleted. Please see line 133.

10. Line 188: Change "are" to "is".

Response: Corrected. Please see lines 211.

11. Line 190: Add "tracers" or "products" at the end of the sentence.

Response: Added. Please see line 213.

12. Line 202: Add "were observed" at the end of the sentence.

Response: Added. Please see line 225.

13. Lines 224-227: Revise the sentence as follows: "This is consistent with the seasonal variation of the precursors NO_x, NO_2 and SO_2, which are mainly caused by automobile exhaust, household cooking, and operation of the typical biomass co-fired brick kilns ...".

Response: Revised as you suggested. Please see line 248-249.

14. Line 228: Change the sentence to "... run on the Kathmandu Valley roads ...".

Response: Changed. Now the sentence is "Currently, nearly 50% of the total motor vehicles in Nepal (approximately 2.33 million) run within on the Kathmandu Valley roads". Please see line 251.

15. Lines 229-231: Revise the sentence as follows: "Diesel- or gasoline-powered generators (producing higher NOx emissions) and garbage burning are other major sources ...".

Response: Changed. Now the sentence is "Diesel- or gasoline-powered generators (producing higher NOx emissions) and garbage burning are other major sources of air pollution in Nepal during the sampling period, which can also emit many aerosol precursors". Please see line 252-253.

16. Lines 256, 270 and 272: Change "ranged" to "ranging".

Response: Changed. Please see lines 279, 293 and 295.

17. Lines 270 and 271: Add "an" before "average".

Response: Added. Please see lines 293 and 294.

18. Line 281: Add a comma after "pollen".

Response: Corrected. Please see line 313.

19. Lines 295, 398, and 559: Add "being" after "while".

Response: Corrected. Please see lines 319, 422, and we change “being” to “their concentrations were” in line 604.

20. Line 304: Change "complicated" to "complex".

Response: Changed. Please see line 328.

21. Lines 326-327: Revise the sentence as follows: "In addition, the higher temperatures (Fig. S1a) were conducive for more active microbial activities."

Response: We changed the sentence as you suggested. Please see lines 351.

22. Line 357: Use consistent terms for anhydrosugars, i.e., change "dehydrated sugars" to "anhydrosugars".

Response: Corrected. Please see line 381.

23. Line 376: Change "are" to "occurs".

Response: Changed. Please see lines 400.

24. Line 455: Delete "in".

Response: Deleted. Please see lines 479.

25. Line 463: Change "the" to "a".

Response: Changed. Please see line 492.

26. Lines 545-548: These sentences need to be polished.

Response: We polished the sentences as “Nevertheless, there is still part of OC (55.5%) that we were not able to be attributed to any specific sources based on the

tracers analyzed in current study. There are partly uncertainties caused by the organic tracer analyses (estimation of measurement uncertainty was shown in Table S2)". Please check lines 589-592.

27. Line 574: Change "show" to "shows".

Response: Changed. Please see line 618.

Reference

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