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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Spatio-temporal variability and light absorption property of carbonaceous aerosol in a typical glacierization region of the Tibetan Plateau" *by* Hewen Niu et al.

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Response to referees' comments Journal: ACP Title: Seasonal variation and light absorption property of carbonaceous aerosol in a typical glacier region of the southeastern Tibetan Plateau Author(s): Hewen Niu et al. MS No.: acp-2017-865 MS Type: Research article Iteration: Correction Special Issue: Study of ozone, aerosols and radiation over the Tibetan Plateau (SOAR-TP) (ACP/AMT inter-journal SI)

We thank the referees for their careful review and insightful comments and suggestions, which greatly helped us to improve the manuscript. Below are our point-by-point responses and detailed corrections/revisions made to the manuscript accordingly.

RC 1 Anonymous Referee #2

This paper reports two-years filter-based measurements of carbonaceous aerosols at two sites a typical glacierization region of the Tibetan Plateau. The dataset provided by this manuscript is important because of the unique location of the experimental site and the profound implication of carbonaceous aerosols deposition on glacier melting. However, I found that some of the conclusions were not solid and lack of sufficient data analysis (or at least relevant reference) support; many of the discussions were superficial and need to be revised. I suggest this manuscript may be accepted for a publication in ACP after the authors could address the following comments. Major concerns: 1. Despite Mt. Yulong and Ganhaizi are special sampling sites, I have not seen much interesting or unique scientific findings. Except providing the basic dataset (numerous descriptive words like "high", "low", "important"), neither was the dataset analyzed and discussed enough, nor were the conclusions obtained in a solid way. Many statements lack sound evidence or support. Here are some examples: P8, line 2-4; P10, line 17-20; P10, line 28-30; P11, line 13-15; line 23-25; P12, line 2-8; line 21-23; P13, line 27-30; P14, line 2-5; P15, line 25-29.

Response: We have thoroughly revised the manuscript, particularly, to the places mentioned here, by providing solid evidence or support for each argument/statement. Detailed revisions to the paper for some arguments are discussed below in response to specific comments and we have now highlighted new findings of this work. After revision, we believe the manuscript is much improved and the interesting scientific findings are clear.

2. The authors claim that the sampling sites are in a typical glacier region of the Tibetan Plateau; that is not true. The two sampling sites are very close to each other: The Mt. Yulong site is likely a mountaintop site and the Ganhaizi site is located on the foot of Mt. Yulong. In this regard, the title of this MS is apparently overstated because two ground-based measurement sites are not enough to capture the characteristics of "spatio-temporal variability of carbonaceous aerosol". Two sites are "... separated

from the urban area, but is an increasingly popular tourist destination, with a geological museum and a golf course. Moreover, there is a spacious parking lot and a tourist dining-center in GHZ." Local sources may be important as already stated in the text.

Response: We agree with the referee that the two sampling sites are close to each other and cannot be used to characterize "spatial variability". However, there is an obvious elevation difference between the two sampling sites. The sampling site in Mt. Yulong (4510 m asl) is above the terminal of the Baishui Glacier (4430 m asl), while GHZ (3054 m asl) is a flat basin and nearly at the foot of Baishui Glacier. Both of the two sampling sites are in glacier region. We have revised the title of this manuscript accordingly to "Seasonal variation and light absorption property of carbonaceous aerosol in a typical glacier region of the southeastern Tibetan Plateau".

3. I appreciate the efforts of a global aerosol-climate model used in the study to quantify the source attribution of BC. However, the model simulation didn't cover the sampling period of this study, which dramatically diminish the reliability and importance of their modeling work. The authors found that there was a significant inter-annual variation of carbonaceous aerosols induced by emission reduction, I don't think they a climate model with outdated emission input can be used to infer the sources of carbonaceous aerosols measured in the current work.

Response: We agree that it would be more desirable to have the model simulation cover the same time as the sampling period for observations. However, as we mentioned in the manuscript, the emissions are only available up to year 2014. Even if we had an up-to-date global emission inventory available, which usually doesn't have a good estimate of small-scale local sources, a coarse-resolution climate model wouldn't be able to capture the quantitative source attributions of the measured BC at the sampling sites. Therefore, the model experiment was not designed to simulate the two-year observations of aerosols in the sampling area, but rather for a recent time period (2010-2014) to estimate the climatological mean source attributions and seasonal variations of near-surface BC concentrations, which has approved to be useful to help under-

stand the observational analysis in the TP region (e.g., Wang et al., 2015). We did find a significant difference in measured carbonaceous aerosols between 2015 and 2016, which was partly attributed to emission changes based on some emission data of Yunnan province rather than model results. We agree that it is speculative and the model results cannot directly support this hypothesis. We have now clarified this in the paper and added precipitation difference as another possible driver of the difference in the measured aerosols (see Fig. 11a, b) as follows:

"The annual mean concentrations of carbonaceous matter collected in 2016 were lower than those in 2015 for the two sites (Fig 11a, b), which could be partly due to the strict mitigation measures that improved the local atmospheric environment in the Mt. Yulong region. For example, the amount of soot emissions (2.44 Tg C) in 2016 were reduced by 21.76% compared to 2015 in Yunnan province (http://www.zhb.gov.cn/), where the Mt. Yulong locates. Other factors likely contributed to the inter-annual difference as well. The amount of precipitation is important to determine aerosol wet removal from the atmosphere during the transport (e.g., Wang et al., 2015). The stronger precipitation in 2016 than in 2015 at Mt. Yulong and GHZ (Fig. 11a, b) partly explains the smaller carbonaceous aerosols mass concentrations in 2016."

4. The current form of the abstract is informative but frequently distract my focus. Please show the readers your most important and exciting findings.

Response: We have significantly reduced the length of the abstract to just highlight the most important results and findings. Below is the revised abstract: "Deposition and accumulation of light-absorbing carbonaceous matters on glacier surfaces can alter the energy balance of glaciers. In this study, two years (December 2014 to December 2016) of continuous observations of carbonaceous aerosols in glacierized region of Mt. Yulong and Ganhaizi (GHZ) basin are analyzed. The average black carbon (BC) and organic carbon (OC) concentrations were 1.51$\pm$0.93 and 2.57$\pm$1.32 g m-3, respectively. The average secondary OC (SOC) concentration, quantified using BC-tracer method, was 1.67$\pm$1.15 g m-3. Although the annual mean OC/BC ratio was 2.45$\pm$1.96, monthly

mean BC concentrations during the post-monsoon season were even higher than OC in the high altitudes (approximately 5000 m asl) of Mt. Yulong. Strong photochemical reactions and local tourism activities were likely the main factors inducing high OC/BC ratios in the Mt. Yulong region during the monsoon season. The mean mass absorption efficiency (MAE) of BC, measured for the first time in Mt. Yulong, at 632 nm with a thermal-optical carbon analyzer under the filter-based method, was 6.82±0.73 m2 g-1, comparable with the results from other studies. Strong seasonal and spatial variations of BC MAE were largely related to the OC and SOC abundance. Source attribution analysis using a global aerosol-climate model, equipped with a BC source tagging technique, suggests that East Asia emissions, including local sources, have the dominant contribution (over 50%) to annual mean near-surface BC in the Mt. Yulong area. There is also a strong seasonal variation in the regional source apportionment. South Asia has the largest contribution in BC emission during the pre-monsoon season, while East Asia dominates the monsoon season and post-monsoon season. Results in this study have great implications for accurately evaluating the influences of carbonaceous matter on glacial melting and water resource supply in glacierization areas."

Specific comments: 1. Title: overstated as pointed out above.

Response: We have changed the title of this manuscript to "Seasonal variation and light absorption property of carbonaceous aerosol in a typical glacier region of the southeastern Tibetan Plateau".

2. P3, line 8: missing reference.

Response: We have added at least one reference for each point as follows. "... visibility (Park et al., 2003), atmospheric radiative balance (Bond et al., 2013; Schuckmann et al., 2016), and the surface albedo of snow and ice (Gertler et al., 2016; Kaspari et al., 2014; Niu et al., 2017a, b)."

3. P4, line 1: specify the “absorbing aerosols”.

Response: The "absorbing aerosols" denote aerosols containing BC, brown carbon, and/or dust that can absorb solar radiation. Below is the revised text. “... absorbing aerosols (e.g., BC, dust, and brown carbon)”

4. Line 7: partially correct.

Response: We have revised this sentence to the following: EC is usually denoted by BC (e. g., Cheng et al., 2011a; Ming et al., 2013; Xu et al., 2009b), which is assumed to have a nearly elemental composition. This is most suitable to diesel soot or lamp black, where BC has a chemical composition close to EC (Andreae and Gelencsér, 2006).

5. Line 13: overstated.

Response: We revised the previous sentence to the following: “This is closely related to water resources for a large population of local habitants in South Asia.”

6. Line 17: For BC particles,

Response: We have added "," after "For BC particles" in the revised manuscript.

7. P6, line 27: specify the number of samples collected at the two sites.

Response: We added the number of samples collected at the two sites in the current submitted manuscript. "The number of aerosol samples collected at Mt. Yulong and GHZ was 117 and 120, respectively."

8. P7, line 3-5: this is nonsense if you have no attempt to give more details.

Response: We agree that this sentence doesn’t add much useful information to the paper, so we have decided to remove this sentence.

9. Line 9: specify the temperature protocol you adopted.

We adopted the improved U.S. Interagency Monitoring of Protected Visual Environments (IMPROVE)-A thermal/optical reflectance protocol (Niu et al., 2017b). The res-

idence time of each heating step was determined by the stabilization of the carbon signal. This has now been added to the text.

10. Line 20: the authors suggested different values of (OC/BC)min should be used to estimate SOC for different dataset (pre- or after-monsoon season), but I didn't see the discussion about data processing.

Response: We have now added a discussion on different values of (OC/BC)min in section 3.1 of the revised manuscript as follows. "Note that the minimum value of OC/BC ratio is used in the SOC calculation, as described in Eqs. (1) and (2). It varies greatly among different seasons (i.e., 0.38, 0.71, 0.42 and 0.35 for winter, pre-monsoon, monsoon, and post-monsoon in Mt. Yulong, respectively), so seasonal minimum values are used to estimate SOC concentrations in the corresponding seasons."

11. P8, line 1-4: given that "there is no standard or more perfect method of OCsec estimation (Yu et al., 2007)", how can you claim that "BC-tracer method employed here is reliable in determining SOC concentrations."

Response: We have now revised the statement to the following: "The semi-empirical (BC-tracer technique) estimates of OCsec after screening for outliers are comparable to the empirical estimates, and sometimes are even superior to empirical estimates. Thus, we believe the BC-tracer method employed here is acceptable in determining SOC concentrations."

12. P10, line 29: needs solid evidence.

Response: We have now provided the following evidence to support the statement in the revised manuscript. "An obviously higher BC and OC concentrations were found in the post-monsoon season at the Mt. Yulong site and in winter season at the GHZ site (Figs. 4 and 5) when wet removal by precipitation is inefficient. This suggests the importance of seasonal changes in sources (Carrico et al., 2003; Cong et al., 2015b; Wan et al., 2017). In addition, OC/BC ratio was usually employed to evaluate the

combustion fuel sources. Previous studies reported that the global mean of OC/BC by biomass burning was higher than fossil fuel burning (Bond et al., 2004; Cao et al., 2010; Liousse et al., 1996). Vehicle emissions from numerous touring buses in the GHZ basin play an important role in the variations of OC/BC ratios."

13. P11, line 9: NCO-P?

Response: NCO-P stands for Nepal Climate Observatory-Pyramid (NCO-P). We have noted this in the revised manuscript.

14. Line 13-15: needs solid evidence.

Response: This was based on conclusions of previous studies that are mentioned later in the text. We have decided to remove this statement here.

15. Line 17: keep a constant significance digit.

Response: Done as suggested for the entire manuscript.

16. Line 17-20: lacking of logic.

Response: We have revised the statement and provided solid evidence to support our argument in the revised manuscript. "It is quite likely that frequent rainfall events with occasional dust (e.g., Dong et al., 2011; Niu et al., 2014) from anthropogenic activities (Shrestha et al., 2000) during the monsoon in 2015 are responsible for this unusual phenomenon (i.e. relatively high content in monsoon season in 2015)."

17. Line 21: the annual average concentrations of OC and SOC.

Response: This sentence has been revised as follows. ". . . the annual average concentrations of OC and SOC, 3.50 $\pm$ 1.50 ïAęg m-3 and 2.20 $\pm$ 1.40 ïAęg m-3, respectively, . . ."

18. Line 23-25: any solid evidence?

Response: We don't have solid evidence, but we revised the text to reflect our hypothe-

sis/assumption on this: “Remote sources are likely to have a similar impact on aerosols over the two sites that are fairly close to each other. Therefore, the additional OC and SOC in the GHZ basin was more likely contributed by local sources such as fossil fuel (vehicle) emissions associated with the frequent and intense tourism activities, which is totally different from that of Mt. Yulong.”

19. P12, line3-5: missing reference or supporting evidence.

Response: We have rewritten this sentence to actually support the previous hypothesis on the impact of local vehicle emissions. “. . . which is consistent with the assumption that the extensive vehicle emissions contributed to SOC formation in the GHZ basin. The monsoon season is also the peak tourism season, and the strong solar radiation in summer (Fig. 2) can enhance the yield of SOC through photochemical reactions (Antony et al., 2011; Schneidemesser et al., 2009; Wan et al., 2017; Niu et al., 2017b, c).”

20. Line 6: “somewhat” should be avoided as much as possible in a scientific paper.

Response: We deleted the word “somewhat” in the revised manuscript.

21. Line 5-8: needs solid evidence.

Response: We have now made it clear in the revised manuscript. “. . .The seasonal variation of carbonaceous aerosols found in the GHZ basin was different from that found in Mt. Yulong. It is likely related to the distinct elevation difference (nearly 1,500 m) and different amount of local human activities (here mainly referring to tourism related activities) between the Mt. Yulong and the GHZ sampling sites. The GHZ site location is close to a parking lot for private vehicles and touring buses and a visitor service center that involves food cooking. These tourism activities can contribute to local emissions of carbonaceous aerosols and precursor gases for SOC (Borrego et al., 2000; Cong et al., 2015a; Shi et al., 2017). However, we don’t have direct observational evidence to support this.”

22. Line 21-26: needs solid evidence.

Response: Please see our response to the previous comments on similar issues. There is no solid evidence for the statement, so we have removed it here.

23. P13, line 18: any reference?

Response: We have added two references to the statement. “In addition, brown carbon (BrC) appearing in the particle mixture can decrease the BC MAE (Jeong et al., 2004; Hecobian et al., 2010).

24. Line 25: “apparent spatial discrepancy”?, I don’t think so.

Response: We have rephrased it to "a discernable small-scale spatial variation".

25. Line 28-30: unconvinced evidence. Any reference?

Response: We agree that the evidence is not convincing. The statement has been rewritten as “In addition to the elevation difference, other potential factors such as the more tourism activities near GHZ than at Mt. Yulong could partly account for the difference, but there is no observational evidence to support this.”

26. P14, line 2-5: I suggest the authors to explore the effects of precipitation on the difference of carbonaceous aerosols mass loadings measured in different years.

Response: Following the great suggestion, we analyzed the annual total precipitation amount (mm) in 2015 and 2016 observed at the Mt. Yulong and GHZ sites and added it to Fig. 11a, b. We have also included a discussion on the possible impact of precipitation on the difference of carbonaceous aerosols mass loading between 2015 and 2016 at the Mt. Yulong and GHZ sites as follows: “Other factors likely contributed to the inter-annual difference as well. The amount of precipitation is important to determine aerosol wet removal from the atmosphere during the transport (e.g., Wang et al., 2015). The stronger precipitation in 2016 than in 2015 at Mt. Yulong (Fig. 11a) and GHZ (Fig. 11b) partly explains the smaller carbonaceous aerosols mass concentrations in 2016.”

Fig. 11 (a) and (b)

27. Line 12-16: I am not sure if these results in previous studies were derived from TSP samples.

Response: We confirmed that the results in previous studies were derived from TSP samples.

28. P15, line 25-27: needs more solid evidences.

Response: This statement is based on the climate model simulation that used the 2010-2014 emissions. As discussed in our response to the major comment #3, we don't mean to attribute sources for BC measured at the sampling sites during 2015-2016, but rather to characterize the seasonal variation in the climatological mean source attribution. We have revised the sentence to clarify a little more. "According to our climate model results, emissions (2010-2014) from East Asia, including local sources accounted in the emission dataset, have a dominant contribution to the near-surface BC in the Mt. Yulong area during the monsoon (78%) and post-monsoon (68%) seasons, as well as the winter season (53%)."

References Andreae, M. O. and Gelencsér, A.: Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols, Atmos. Chem. Phys., 6, 3131-3148, doi:10.5194/acp-6-3131-2006, 2006. Antony, R., Mahalinganathan, K., Thamban, M.: Organic carbon in Antarctic snow: spatial trends and possible sources, Environ. Sci. Technol., 45, 9944-9950, 2011. Bond, T. C., Streets, D. G., Yarber, K. F.: A technology-based global inventory of black and organic carbon emissions from combustion, J. Geophys. Res-Atmos., 109(14), D14203. DOI:10.1029/2003JD003697, 2004. Bond, T. C., et al.: Bounding the role of black carbon in the climate system: A scientific assessment, J. Geophys. Res. Atmos., 118, 5380-5552, doi:10.1002/jgrd.50171, 2013. Borrego, C., Tchepel, O., Barros, N., Miranda, A.: Impact of road traffic emissions on air quality of the Lisbon region, Atmos. Environ., 34, 4683-4690, 2000. Cao, J. J., Tie, X., Xu, B. Q., Zhao, Z., Zhu, C., Li G., Liu S.: Measuring and modeling black

carbon (BC) contamination in the SE Tibetan Plateau, J. Atmos. Chem., 67, 45-60, 2010. Carrico, C. J., Bergin, M. H., Shrestha, A. B.: The importance of carbon and mineral dust to seasonal aerosol properties in the Nepal Himalaya, Atmos. Environ., 37, 2811-2824, 2003. Cheng, Y., He, K. B., Zheng, M., Duan, F. K., Du, Z. Y., Ma, Y. L., Tan, J. H.: Mass absorption efficiency of elemental carbon and water-soluble organic carbon in Beijing, China, Atmos. Chem. Phys., 11, 11497-11510, 2011a. Cong, Z. Y., Kawamura, K., Kang, S. C., et al.: Penetration of biomass-burning emissions from South Asia through the Himalayas: new insights from atmospheric organic acids, Scientific Report, 5, 9580, Doi:10.1038/srep09580, 2015a. Cong, Z. Y., Kang, S. C., Kawamura, K.: Carbonaceous aerosols on the south edge of the Tibetan Plateau: concentrations, seasonality and sources, Atmos. Chem. Phys., 15, 1573-1584, 2015b. Dong, Z. W., Li, Z. Q., Ross, E., Wu, L. H., Zhou, P.: Temporal characteristics of mineral dust particles in precipitation of Urumqi River Valley in Tian Shan, China: a comparison of alpine site and rural site, Atmos. Res., 101, 294-306, 2011. Gertler, C. G., Puppala, S. P., Panday, A., Stumm, D., Shea, J.: Black carbon and the Himalayan cryosphere: a review. Atmos. Environ., 125, 404-417, 2016. Hecobian, A., Zhang, X., Zheng, M., Frank, N., Edgerton, E. S., Weber, R. J.: Water-soluble organic aerosol material and the light-absorption characteristics of aqueous extracts measured over the Southeastern United States, Atmos. Chem. Phys., 10, 5965-5977, 2010. Jeong, C. H., Hopke, P. K., Kim, E., and Lee, D. W.: The comparison between thermal-optical transmittance elemental carbon and Aethalometer black carbon measured at multiple monitoring sites, Atmos. Environ., 38, 5193-5204, 2004. Kaspari, S. D., Painter, T. H., Gysel, M., Skiles, S. M., Schwikowski, M.: Seasonal and elevational variations of black carbon and dust in snow and ice in the Solu-Khumbu, Nepal and estimated radiative forcings, Atmos. Chem. Phys., 14, 8089-8103, 2014. Liousse, C., Penner, J. E., Chuang, C.: A global three-dimensional study of carbonaceous aerosols, J. Geophys. Res-Atmos., 101(D14), 19411-19432, 1996. Ming, J., Wang, P. L., Zhao, S. Y., Chen, P. F.: Disturbance of light-absorbing aerosols on the albedo in a winter snowpack of Central Tibet, J. of Environ. Sci., 25(8), 1601-1607, 2013. Niu, H. W., He, Y. Q., Lu, X.

X., Shen, J.: Chemical composition of rainwater in the Yulong Snow Mountain region, Southwestern China, Atmos. Res., 144, 195-206, 2014. Niu, H. W., Kang, S. C., Shi, X. F.: In-situ measurements of light-absorbing impurities in snow of glacier on Mt. Yulong and implications for radiative forcing estimates, Sci. of the Tot. Environ., 581-582, 848-856, 2017a. Niu, H. W., Kang, S. C., Zhang, Y. L., Shi, X. F.: Distribution of light-absorbing impurities in snow of glacier on Mt. Yulong, southeastern Tibetan Plateau, Atmos. Res., 197, 474-484, 2017b. Niu, H. W., Kang, S. C., Shi, X. F.: Water-soluble elements in snow and ice on Mt. Yulong, Sci. of the Tot. Environ., 574, 889-900, 2017c. Park, R. J., Jacob, D. J., Chin, M., Martin, R. V.: Sources of carbonaceous aerosols over the United States and implications for natural visibility, J. Geophys. Res., 108(D12), 4355, doi:10.1029/2002JD003190, 2003. Schneidemesser, E. V., Schauer, J. J., Hagler, G. S. W.: Concentrations and sources of carbonaceous aerosol in the atmosphere of Summit, Greenland, Atmos. Environ., 43, 4155-4162, 2009. Schuckmann, K. V., Palmer, M. D., Trenberth, K. E.: An imperative to monitor Earth's energy imbalance, Nature Climate Change, 6, DOI: 10.1038/NCLIMATE2876, 2016. Shi, X. F., Niu, H. W., He, Y. Q.: Characteristics of rainwater chemistry in Lijiang-Yulong Snow Mountain, Environmental Chemistry 36(5), 994-1002, 2017. (in Chinese with English abstract) Shrestha, A. B., Wake, C. P., Dibb, J. E., Mayewski, P., Whitlow, S.: Seasonal variations in aerosol concentrations and compositions in the Nepal Himalaya, Atmos. Environ. 34, 3349-3363, 2000. Wan, X., Kang, S. C., Li, Q. L., Rupakhetiet, D., Zhang, Q. G., Guo, J. M., Chen, P. F.: Organic molecular tracers in the atmospheric aerosols from Lumbini, Nepal, in the northern Indo-Gangetic Plain: Influence of biomass burning. Atmos, Chem. Phys., 17, 8867-8885, 2017. Wang, M., Xu, B. Q., Cao, J. J., et al.: Carbonaceous aerosols recorded in a southeastern Tibetan glacier: analysis of temporal variations and model estimates of sources and radiative forcing, Atmos. Chem. Phys., 15, 1191-1204, 2015. Xu, B. Q., Wang, M., Joswiak, D. R., Cao, J. J., Yao, T. D., Wu, G. J., Yang, W., Zhao, H. B.: Deposition of anthropogenic aerosols in a southeastern Tibetan glacier, J. Geophys. Res., 114, D17209, doi:10.1029/2008JD011510, 2009b.

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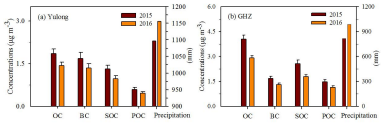

Fig. 11 (a) and (b)

Fig. 1. Fig. 11 (a) (b)