Review of "Measurement report: Quantifying source contribution and radiative forcing of fossil fuel and biomass burning black carbon aerosol in the southeastern margin of Tibetan Plateau" (ACPD-2020-408, Liu et al)

General comment

This paper reports on measurements and modeling regarding the contribution of fossil fuel and biomass burning sources to black carbon (BC) aerosols abundance and radiative forcing at a site south-east of the Tibetan Plateau. Methods used in the study are robust, and the results are sound. However, it is difficult to ascertain the novelty and actual contribution to the overall understanding of, for instance, how BC aerosols are affecting the Tibetan Plateau. In my opinion, authors may turn this study into a relevant one if they would consider using the data at hand by better explaining the reasons that make these data important for improved understanding. In its present form, detailed measurements and modeling are more suited for a technical report not suitable, in my opinion, for this prestigious journal.

Response: The authors appreciate the reviewer's valuable comments and suggestions, and we believe that the revised manuscript has been significantly improved after considering them. Below are point-to-point responses, and the modifications to the manuscript are marked.

Specific comments

(1) The text would improve in clarity and possibly be shorten if reviewed by a native English writer/speaker. Also, some results could be summarized in tables improving the readability of the text.

Response: We have had this manuscript polished by a native English speaking scientist. In addition, some results also are summarized in Table R1 (also see Table 1 in the revised manuscript) as suggested.

Table R1 Derived AAE, MAC and source contribution of BC from difference sources at the sampling site

	AAE	MAC (m^2 g^{-1})	Mass concentration (μg m^{-3})	Contribution ratio
$BC_{biomass}$	1.7	10.4	0.4 ± 0.3	57%
$BC_{traffic}$	0.8	9.1		
BC_{coal}	1.1	15.5		

BC_{fossil}	0.9	12.3	0.3 ± 0.2	43%

(2) Abstract: Re-write according to suggestions below. What do we learn from this study? In what context is this useful? What is the novelty?

Response: As suggested, we have rewritten the abstract and clearly state the purpose, importance and novelty of the study. It now reads as follows:

"Anthropogenic emissions of black carbon (BC) aerosol are transported from Southeast Asia to the southwestern Tibetan Plateau (TP) during the pre-monsoon; however, the quantities of BC from different anthropogenic sources and the transport mechanisms are still not well constrained because there have been no high-time-resolution BC source apportionments. Intensive measurements were taken in a transport channel for pollutants from Southeast Asia to the southeastern TP during the pre-monsoon to investigate the influences of fossil fuels and biomass burning on BC. A receptor model coupled multi-wavelength absorption with aerosol species concentrations was used to retrieve site-specific Ångström exponents (AAE) and mass absorption cross-sections (MAC) for BC. An 'aethalometer model' that used those values showed that biomass burning had a larger contribution to BC mass than fossil fuels ($BC_{biomass}$ = 57% versus BC_{fossil} = 43%). The potential source contribution function indicated that $BC_{biomass}$ was transported to the site from northeastern India and northern Burma. The Weather Research and Forecasting model coupled with chemistry (WRF-Chem) model indicated that 40% of the $BC_{biomass}$ originated from Southeast Asia, while the highest BC_{fossil} was transported from the southwest of the sampling site. A radiative transfer model indicated that the average atmospheric direct radiative effects (DRE) of BC were +4.6 ± 2.4 W m^{-2} with +2.5 ± 1.8 W m^{-2} from $BC_{biomass}$ and +2.1 ± 0.9 W m^{-2} from BC_{fossil}. The DRE of $BC_{biomass}$ and BC_{fossil} produced heating rates of 0.07 ± 0.05 and 0.06 ± 0.02 K day^{-1}, respectively. This study provides insights into sources of BC over a transport channel to the southeastern TP and the influence of the cross-border transportation of biomass burning emissions from Southeast Asia during the pre-monsoon."

• *Introduction*

(3) Page 2, lines 13-14. In addition to characterizing source regions and their contributions to aerosol burden downwind it is also important to assess the timing in which this impact occur, how is the aging process, etc.

Response: We agree with the reviewer that timing and aging are also importation factors, so we have re-written this sentence in the revised manuscript. It now reads as follows:

"Nonetheless, assessments of regional transport of bulk BC aerosol have not fully revealed the impacts of different BC emission sources because the optical properties and radiative effects of BC not only can vary among sources in complex ways but also can be affected by aging during transport (Tian et al., 2019; Zhang et al., 2019). Therefore, quantitative information on the contributions of different sources of BC over the TP is lacking, but it is critically needed for a better understanding the influence of anthropogenic emissions on its environment and climate."

(4) *Page 2, lines 22-24. Uncertainties in modeling studies not only depend on uncertain emission estimates but also on how well chemistry, transport and deposition processes are represented, initial/boundary conditions, etc. It appears necessary to review other studies to get an idea of the uncertainty when using models to simulate long-range transport, particularly over complex terrain.*

Response: We agree that the uncertainties of modeling method can be caused by the factors mentioned by reviewer and likely others. The complex terrain has impact on simulation of meteorological field over the Tibetan Plateau. We followed the reviewer's suggestion and have rewritten the relevant paragraph in the revised manuscript; it now reads as follows:

"On the other hand, the accuracy of model simulations is dependent on many factors, including uncertainties associated with initial particle parameters, aging processes, the accuracy of emission inventory, meteorological fields over the complex terrain, and the modules for chemistry and planetary boundary layer (PBL) dynamics, etc. (Koch et al., 2009; Madala et al., 2014; Vignati et al., 2010)."

(5) *Page 3, lines 1-24. This is a lengthy discussion about distinguishing between biomass and fossil fuel black carbon according to multiple observational and methodological studies. Rather than listing the pros and cons of the different methods, it would be good to have a clearer idea of which is the method fit for purpose to be discussed in the work. For that, it is key to establish a clear purpose, and how this will help improving understanding of a given phenomenon.*

Response: Following the reviewer's suggestion, we have rewritten the introduction in the revised manuscript and presented a clear explanation as to why we chose the online method, including why and how we optimized the methods. To clarify the purpose of this research, we re-organized the last two paragraphs of the introduction. They now read as follows:

"To make up for the deficiencies of filter-based analysis, BC source apportionments based on high-time resolution online data has been conducted in many locations but for the TP are limited. An 'aethalometer model' based on multi-wavelength absorption data is one of efficient approaches for distinguishing between BC from fossil fuel and biomass burning sources (Sandradewi et. al., 2008). Although it has been widely used elsewhere, this approach has not been applied to the TP. The accuracy of the 'aethalometer model' relies on the input parameters, including the Ångström exponents (AAE) and BC mass absorption cross-sections (MAC_{BC}) of different sources (Zotter et al., 2017). Limited information on site specific AAEs and MAC_{BC}s, lead most studies to rely on values taken from measurements made in other locations (e.g., Healy et al., 2017; Zhu et al., 2017). This results in unquantified uncertainties because the AAEs and MAC_{BC}s vary with specific fuel subtypes and combustion conditions (Wang et al., 2018; Tian et al., 2019). Therefore, site-dependent AAE and MAC_{BC} are essential for improving the reliability of BC source apportionment by the 'aethalometer model'.

In this study, field measurements of BC were taken on the southeastern margin of the TP during the pre-monsoon. This region connects the high altitude TP with the low altitude Yungui Plateau and forms a transport channel for pollutants from Southeast Asia (Wang et al., 2019a), and it is an ideal region for investigating the impact of pollutant transport to the southeastern TP. A receptor model that combined multi-wavelength absorption with aerosol species concentrations was used to retrieve site-dependent AAEs and MAC_{BC}s. This was done to improve the 'aethalometer model' with the goal of obtaining a more accurate BC source apportionment. The primary objectives of this study were to (1) quantify the mass concentrations of BC from fossil fuel and biomass burning sources; (2) determine the impact of regional transport on source-specific BC; and (3) assess the radiative effects caused by BC from different sources. This study provides insights into the BC sources on southeastern TP and an assessment of their radiative effects during the pre-monsoon."

(6) Page 4, lines 1-6. You state that previous studies have dealt with radiative impacts of bulk BC, no distinguishing BC sources. Furthermore, you state that this study would be unique as it provides the first estimate of BC radiative forcing split by source regions. However, you estimate the instantaneous forcing over one site which is, by definition, locally representative, and not necessarily climatically important. Other studies may have estimated bulk BC forcing but over much longer periods of time, and over large areas, including the Himalayan cryosphere. Hence, I urge the authors to make their study unique by better establishing the purpose of it.

Response: We appreciate this comment. Taking into consideration Comments 1–5, we have re-written the introduction to explain the purpose of our study and we deleted this paragraph in the revised manuscript. BC is an important atmospheric light-absorbing material that can have significant radiative effects. The SBDART model has been widely used to estimate the instantaneous radiative effects of BC based on the ground observations (Gharibzadeh et al., 2017; Rajesh et al., 2018; Panicker et al., 2010). Although data from only one site on the southeastern TP were collected (because of practical limitations in personnel, equipment, logistics, etc.), we believe that the unique geographic location of sampling site (i.e., transport channel) on TP make our results of considerable interest. In addition, due to paucity of studies that have separately quantified BC mass from biomass burning and fossil fuels on TP, we think that it is important to understand their radiative effects and potential influences on climate.

References:

Gharibzadeh M , Alam K , Abedini Y , et al. Monthly and seasonal variations of aerosol optical properties and direct radiative forcing over Zanjan, Iran, J. Atoms. Sol-Terr. Phy.,164,268-275, dx.doi.org/10.1016/j.jastp.2017.09.006, 2017

Panicker, A. S., Pandithurai, G., Safai, P. D., Dipu, S., and Lee, D.-I.: On the contribution of black carbon to the composite aerosol radiative forcing over an urban environment, Atmos. Environ., 44, 3066-3070, 10.1016/j.atmosenv.2010.04.047, 2010.

Rajesh, T. A., and Ramachandran, S.: Black carbon aerosols over urban and high altitude remote regions: Characteristics and radiative implications, Atmos. Environ., 194, 110-122, 10.1016/j.atmosenv.2018.09.023, 2018.

• *Methodology*

(7) Page 4, line 17. Improve the precision of attitude and longitude to allow a proper location of the site.

Response: Following the reviewer's suggestion, we have specified the attitude and longitude of the sampling site in the revised manuscript. It now reads as follows:

> "Intensive field measurements were made at the rooftop of a building (~10 m above the ground) at the Lijiang Astronomical Station, Chinese Academy of Sciences (3260 m above sea level, 100°1'48"E, 26°41'24"N), Gaomeigu County, Yunnan Province, China (Fig. 1) from 14 March to 13 May 2018. (Fig. 1)."

(8) Page 4, lines 18-19. As per your reference, Wang et al (2019a), your observation site is located

along a "transportation channel". Describe the overall transport patterns affect. Is the period of observations representative of which transport/circulation pattern? An overall meteorological description is missing.

Response: In the pre-monsoon season when the southeastern margin of the TP is influenced by the westerly winds (Chan et al 2017; Niu et al., 2017), a pathway for the cross border transport of emissions from southeast Asia to the TP. The sampling period for this study was from March to May and therefore in the pre-monsoon. In the revised manuscript, we have added some information about the 'transport channel'. It now reads as follows:

> "During the campaign, westerly winds created a potential pathway for cross border transport from southeast Asia to southwest China. During the study, the average relative humidity and temperature were 80% ± 20% and 7.6 ± 3.2℃, respectively; the mean wind speed near surface was 5.4 ± 2.1 m s^{-1}, and the winds were mainly from the west and southwest."

References:

Chan, C. Y., Wong, K. H., Li, Y. S., Chan, Y., and Zhang, X. D.: The effects of Southeast Asia fire activities on tropospheric ozone, trace gases and aerosols at a remote site over the Tibetan Plateau of Southwest China, Tellus B , 58B, 310-318, 10.1111/j.1600-0889.2006.00187.x, 2017

Niu H., Kang., S., Zhang, Y., Shi, X., Shi., X., Wang S., Li, G., Yan, X., Pu, T. He, Y., Distribution of light-absorbing impurities in snow of glacier on Mt. Yulong, southeastern Tibetan Plateau, Atmos. Res., 197, 474-484, 10.1016/j.atmosres.2017.07.004, 2017.

(9) Page 4, lines 19-20. You say that the population surrounding your observational site is small. Small compared to what? Then you go onto establishing that limited anthropogenic activities are found there. However, your results show a non-negligible contribution. The site should be better described, including a brief description of aerosol sources.

Response: At the sampling site, the influence of anthropogenic activities is limited due to the low population density and lack of industries. The local emissions have small effects on the BC source apportionment results compared with the contributions of fossil fuel BC from two highways (5.5 km from the sampling site) and transport from the border with Burma. Following the reviewer's suggestion, we have added some information about the possible anthropogenic emissions in the surrounding area of the sampling site. The revised manuscript now reads as follows:

“The sampling site is 3–5 km from Gaomeigu village, which has 27 households and 110 residents. Villagers there rely on farming for their livelihoods, and biomass is the primarily residential fuel (Li et al, 2016). There are no large industries near the village and traffic is light. However, two highways (Hangzhou-Ruili Expressway and Dali-Nujiang Expressway) are located ~5.5 km to the west of the sampling site.”

(10) Page 7, section 2.5. HYSPLIT can be used with large-scale (synoptic) meteorological fields. Do you have an assessment of how well this approach works over complex terrain?

Response: The meteorological data used in this study were obtained from the Global Data Assimilation System (GDAS) and had a spatial resolution of 1° × 1°. The HYSPLIT model converted the vertical layers from the original coordinate system into its own terrain-following coordinate system (sigma) and directly used the data contained in meteorological files for the trajectory calculations (Draxler and Hess, 1997). The surface in the terrain-following coordinate system is consistent with the ground, and that solves the problem of modelling near mountainous areas (Phillips, 1965). Furthermore, this method has been used over complex terrain with various meteorological data in a number of studies (Burley and Bytnerowicz 2011; Wang et al., 2015; Wang et al.,2019; Qu et al 2015 and Khan et al., 2010).

To determine if the trajectory would be impacted by the surface rising, we also ran calculations at heights of 150m and 1000m in addition to 500m (Figure R1). The results showed that directions were similar, particularly between the results at150m and at 500m. We finally decided to use the 500m results because greater heights be higher than the height at which the samples were collected, and 500m is generally representative of the average planetary boundary height at the site (~600m).

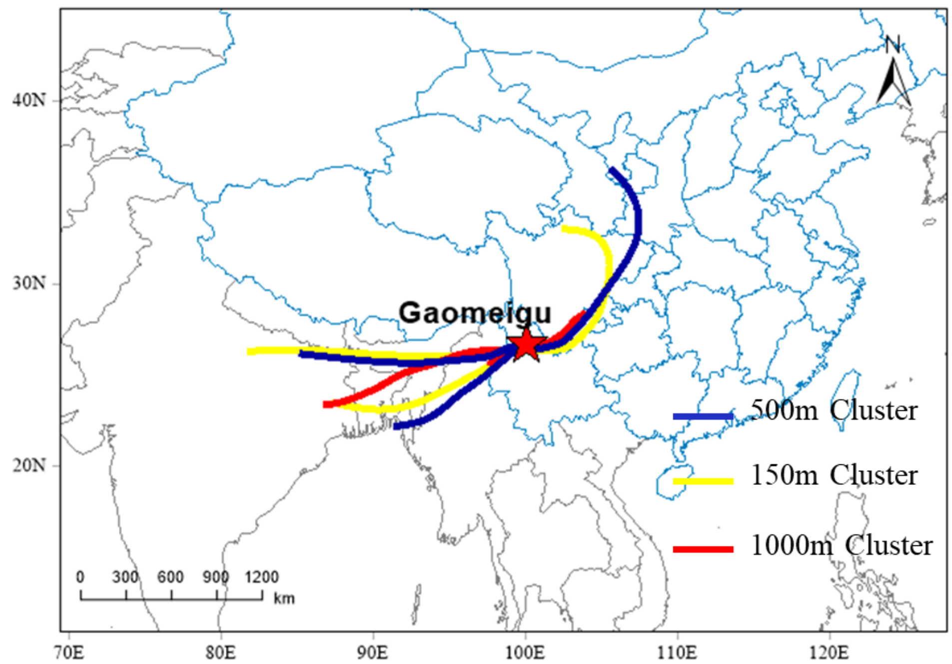

Figure R1. the blue lines represent the clusters at 500m height, the yellow lines represent the clusters at 150m height, the red lines represent the cluster at 1000m height.

References:

Burley J D , Bytnerowicz A . Surface ozone in the White Mountains of California, Atmospheric Environment, 45, 4591-4602, 10.1016/j.atmosenv.2011.05.062, 2011.

Chengkai Qu, Xinli Xing, Stefano Albanese, et al. Spatial and seasonal variations of atmospheric organochlorine pesticides along the plain-mountain transect in central China: Regional source vs. long-range transport and air－soil exchange, Atmos. Environ. 122, 31-40, 10.1016/j.atmosenv.2015.09.008, 2015.

Draxler, R., and Hess, G.: An overview of the HYSPLIT_4 modelling system for trajectories, Aust. Meteorol. Mag., 47, 1998.

Khan A J , Li J , Dutkiewicz V A , et al. Elemental carbon and sulfate aerosols over a rural mountain site in the northeastern United States: Regional emissions and implications for climate change, Atmos. Environ., 44, 2364-2371, 10.1016/j.atmosenv.2010.03.025, 2010.

Phillips N.A. a coordinate system having some special advantages for numerical forecasting, Shorter contributions, J. Atmos. Sci., 14, 184-185, 1957.

Wang, Q. Y., Huang, R. J., Cao, J. J., Tie, X. X., Ni, H. Y., Zhou, Y. Q., Han, Y. M., Hu, T. F., Zhu, C. S., Feng, T., Li, N., and Li, J. D.: Black carbon aerosol in winter northeastern Qinghai–Tibetan Plateau, China: the source, mixing state and optical property, Atmos. Chem. Phys., 15, 13059-13069, 10.5194/acp-15-13059-2015, 2015.

Wang, Q., Han, Y., Ye, J., Liu, S., Pongpiachan, S., Zhang, N., Han, Y., Tian, J., Wu, C., Long, X., Zhang, Q., Zhang, W., Zhao, Z., and Cao, J.: High Contribution of Secondary Brown Carbon to Aerosol Light Absorption in the Southeastern Margin of Tibetan Plateau, Geophys. Res. Lett., 46, 4962-4970, 10.1029/2019gl082731, 2019.

(11) Why do you choose 3-day back trajectories instead of 2 or 5 days?

Response: Studies have indicated that BC lifetime varies from 3.3 to 12 days in the atmosphere (Liu et al., 2011 and references therein). The BC lifetime depends on many factors such as morphology, size, mixing state, aging condition, and meteorological conditions. It is not possible to know the exact lifetime of the BC sampled at the study site, and therefore, we chose the lowest value for the BC lifetimes to minimize the effects of BC deposition during transport to the site. In addition, the 3-day backward trajectories also have been widely used in previous studies to investigate BC transport pathways (e.g., Wang et al., 2018; Verma et al., 2010).

References:

Liu, J., Fan, S., Horowitz, W. L., Levy, H., 116, Evaluation of factors controlling long‐range transport of black carbon to the Arctic, J. Geophys. Res., doi:10.1029/2010JD015145, 2011.

Verma, R. L., Sahu L. K., Kondo, Y., Takegawa, N., Han, S., Jung, J. S., Kim, Y., J., Fan, S., Sugimoto, N., Shammaa, M. H., Zhang, Y., H., and Zhao, Y.: Temporal variations of black carbon in Guangzhou, China, in summer 2006, Atmos. Chem. Phys., 10, 6471–6485, 10.5194/acp-10-6471-2010, 2010.

Wang, Q., Cao, J., Han, Y., Tian, J., Zhu, C., Zhang, Y., Zhang, N., Shen, Z., Ni, H., Zhao, S., and Wu, J.: Sources and physicochemical characteristics of black carbon aerosol from the southeastern Tibetan Plateau: internal mixing enhances light absorption, Atmos. Chem. Phys., 18, 4639-4656, 10.5194/acp-18-4639-2018, 2018.

• *Results and discussion*

(12) Page 11, lines 20-25. Your BC aerosol appears to have aged. Can't you use your WRF-

Chem simulations to attempt providing further insights on this issue?

Response: We thank the reviewer providing us with this suggestion. However, BC is usually considered as chemically inert in the atmosphere (Bond et al., 2013), and the WRF-Chem model only accounts for physical processes of BC in the atmosphere, such as the wet and dry deposition. Although we concluded that BC underwent substantial aging, the objective of the study was to apportion the BC sources, and that did not include contributions from materials coating the BC. Moreover, due to the limitation of the measurement methods in this study, we could not obtain quantitative information regarding BC aging, which would have been the best way to constrain the model simulation. Therefore, the aging of BC is something that would be better addressed in future studies.

References:

Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G., and Zender, C. S.: Bounding the role of black carbon in the climate system: A scientific assessment, J. Geophys. Res.-Atmos., 118, 5380-5552, 10.1002/jgrd.50171, 2013.

○ *Page 12, section 3.2.*

(13) *Some of your results could be better appreciated if summarized in a table.*

Response: As suggested, results have been summarized in to a table. Please see the response of comment 1 above.

(14) *You make multiple references to FigS3. Maybe it is better to bring it to the main manuscript. If so, it could be useful to split the graphs for daytime and nighttime periods as it would better fit with Figure 2.*

Response: The mass concentrations of levoglucosan and benzothiazolone were obtained from 24 h filter samples, and so we cannot compare daytime versus nighttime periods. Note that the online

BC data were integrated to match each filter sampling times. We did follow the reviewer's suggestion and combined Fig. S3 with Fig. 2 in the revised manuscript (also see Fig. R2 below).

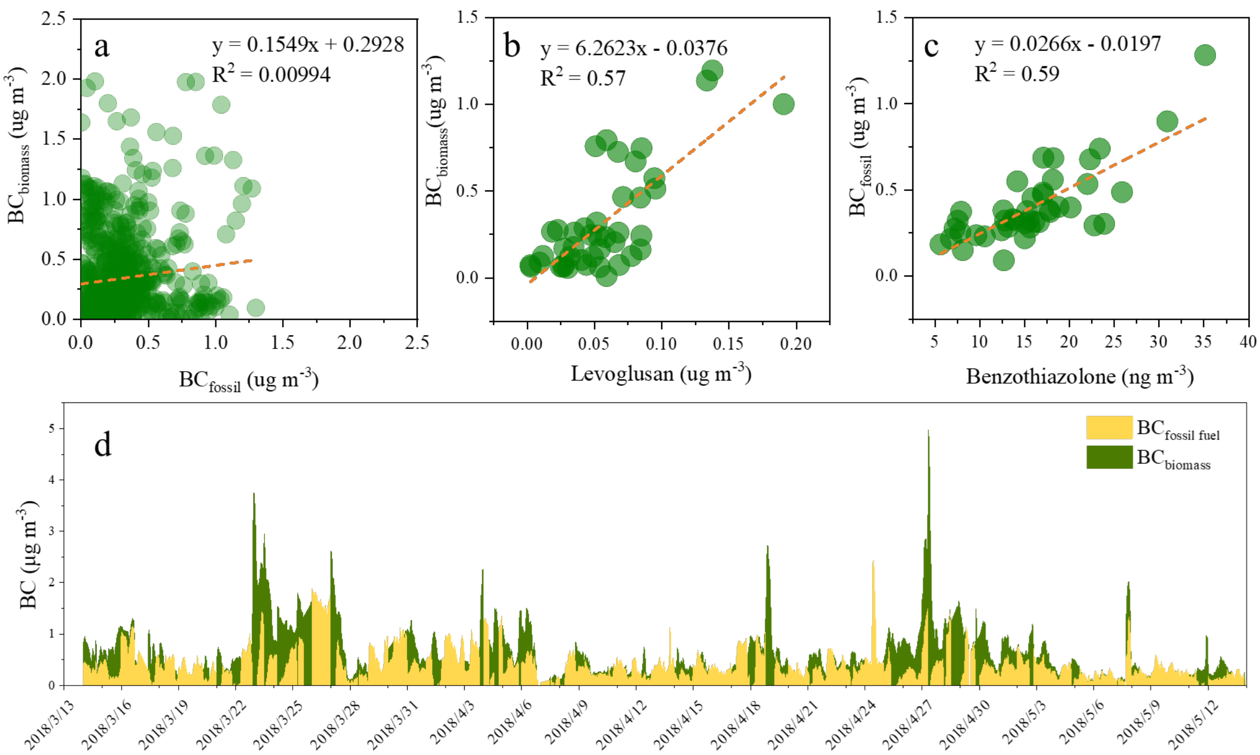

Figure R2. Scatter plots of (a) biomass burning BC ($BC_{biomass}$) versus fossil fuel combustion BC (BC_{fossil}), (b) $BC_{biomass}$ versus levoglucosan, and (c) BC_{fossil} versus benzothiazolone. $BC_{biomass}$ and BC_{fossil} represent black carbon aerosol contributed by biomass burning and fossil fuel sources, respectively. (d)Time series of hourly averaged mass concentrations of black carbon (BC) aerosol from biomass bunirng ($BC_{biomass}$) and fossil fuel sources (BC_{fossil}) during the campaign.

• *Conclusion*

(15) Stress the novelty, and make it explicit that the period studied correspond to a given set of transport/circulation patterns.

Response: We followed the reviewer's suggestion and rewrote the conclusions in the revised manuscript. It now reads as follows:

> "This study quantified the source contributions of BC aerosol from fossil fuel and biomass burning at a site on the southeastern margin of the TP that represents a regional transport channel for air pollution. The study was conducted during pre-monsoon when the

southeastern TP was heavily influenced by the air mass from southeast Asia. To reduce the uncertainties caused by interferences in absorption measurements (i.e. secondary absorption and dust) and assumptions relative to AAE and MAC_{BC}, the traditional 'aethalometer model' was optimized in two aspects. First, a BC-tracer method coupled with a minimum R-squared approach was applied to separate secondary absorption from the total absorption, and as a result, the interferences of absorption from secondary aerosols have been eliminated. Then, an optical source apportionment model that used primary multi-wavelength absorption and chemical species as inputs was used to derive site-dependent AAE and MAC_{BC} values—these minimize the uncertainties associated with prior assumptions on these parameters. The AAE (MAC_{BC}) calculated in this way was 0.9 (12.3 m^2 g^{-1}) for the fossil fuel source and 1.7 (10.4 m^2 g^{-1}) for biomass burning. The results of 'aethalometer model' that used these values showed that the average mass concentration of BC was 0.7 ± 0.5 μg m^{-3} of which 57% was from biomass burning and 43% from fossil fuels. Trajectory analysis showed that the $BC_{biomass}$ over the site was mainly driven by regional transport from northeastern India and Burma, while BC_{fossil} was primarily influenced by traffic emissions from areas surrounding the sampling site. Moreover, the WRF-Chem model indicated that biomass burning in Southeast Asia contributed 40% of the BC loadings over the southeastern margin of the TP. The SBDART model showed that a DRE of +4.6 ± 2.4 W m^{-2} for the total $PM_{2.5}$ BC, of which +2.5 ± 1.8 W m^{-2} was from $BC_{biomass}$ and +2.1 ± 0.9 W m^{-2} from BC_{fossil}. The results of our study provide useful information concerning the sources of BC over an atmospheric transport channel to the southeastern TP, and they highlight the importance of the cross-border transport of biomass burning emissions from Southeast Asia on the region during the pre-monsoon."