Interactive comment on "Measurement report: Quantifying source contribution and radiative forcing of fossil fuel and biomass burning black carbon aerosol in the southeastern margin of Tibetan Plateau" by Huikun Liu et al.

Anonymous Referee #1

General comments: In this manuscript, the sources of BC aerosols over the Tibetan Plateau and their radiative effects were investigated. BC aerosols were distinguished into fossil fuel combustion source and biomass burning source. Regional transport of source-specific BC was further explored by models. On this basis, the radiative effects caused by BC from different sources were evaluated. Overall, the manuscript is well structured, the methods are technically sound, and the main findings presented seem to be reasonable and be of general interests to the Tibetan Plateau ecosystem and climate stability study. I think the topic fits within the scope of ACP. I would recommend acceptance of this manuscript for publication pending the following revisions:

Response: We sincerely thank the reviewer for the comments and suggestions, and we have revised the relevant text and content. Below are point-to-point responses, and the modifications to the manuscript are marked.

Specific comments:

1.Please spend time picking through the manuscript and check for spelling and grammatical errors, especially the tense, prepositions and articles. For example, 'BC on the TP' should be replaced with 'BC over the TP', 'transport to TP' should be replaced with 'transport to the TP'

Response: We have taken the suggestion to heart and have corrected the relevant mistakes as shown below. Also, the paper has been polished by a native English speaker.

> "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."

> "Nonetheless, these studies have been helpful for understanding the sources of BC over the TP."

2.Section 1, this part should introduce the research background and significance, current status, concealed problems, as well as research mentality and content of this study. Please highlight the innovation and importance from another angle and reduce describing the deficiencies of previous research appropriately.

Response: We have re-written the introduction to include more background material, to clarify the research focus, to explain why we were interested in study area and why we chose the methods we did. The revised introduction now reads:

"The Tibetan Plateau (TP) is an important regulator of climate change in the northern hemisphere, and it plays crucial roles in the functions of global ecosystems and climate stability (Liu et al., 2019b). The TP is covered by one of the largest ice masses on Earth, and it has been called the water tower of Asia (Liu et al., 2020). The glaciers on the TP are facing rapid retreat, however, and if unchecked, that could result in adverse effects on Asian hydrological cycle and the Asian monsoon (Luo et al., 2020). In spring, the glaciers on the TP begin to melt as part of the natural hydrological cycle, but the increasing quantities of black carbon (BC) aerosol transported to the TP has accelerated this process (Bond et al., 2013) by causing a warming effect in atmosphere over the TP and enhancing the absorption of radiation on the surface of the glaciers (Ming et al., 2009).

The southern part of the TP is bounded by South Asia where air pollution often is severe (Chan et al. 2017). Several studies have shown that pollutants (including BC) from South Asia can be transported to the south of the TP along mountain-valleys, especially during the pre-monsoon (March–May) when southwestly winds prevail (e.g., Cao et al., 2010; Xia et al., 2011 Zhu et al., 2017; Niu et al. 2017). For example, Xia et al., (2011) analyzed satellite data and air mass trajectories and found that the TP, particularly the southern TP, was affected by pollutants carried by southwesterly winds from nearby regions in South Asia. In addition, numerous studies have shown that the high bulk BC mass loadings and the associated regional influences on the TP are related to transport from South Asia (Liu et al., 2015; Han et al., 2020; Cong et al., 2015; Wang et al., 2015). 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.

Several studies have assessed the contributions of different BC sources through model simulations or isotopic methods. For example, Zhang et al. (2015) investigated BC sources for different parts of the TP by using a chemical transport model and a source tagging approach, and they found that the contributions of BC sources varied among regions and with the seasons. Li et al., (2016) used filter sampling and carbon isotopes ($\Delta^{14}C$ and $\delta^{13}C$) to determine the BC from fossil

fuels and biomass burning in several areas of the TP. A major disadvantage of filter-based measurements is they are constrained by low time resolution, and this makes it challenging to capture the detailed evolution of pollution events. 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). Nonetheless, these studies have been helpful for understanding the sources of BC over the TP.

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."

3.Please try to avoid expressions like 'our study', which seems not be objective. Technical corrections: 1.P1, Line 24, 'reveal' should be changed to 'revealed'.

Response: We have changed all "our study" in the manuscript into "this study". The verbal tenses have been correct in the following sentence:

> "The potential source contribution function indicated that $BC_{biomass}$ was transported to the site from northeastern India and northern Burma."

2.P1, Line 26, add 'which' before 'can explain'.

Response: We have changed the sentence to

> "The Weather Research and Forecasting model coupled with chemistry (WRF-Chem) model indicated that 40% of the $BC_{biomass}$ originated from Southeast Asia"

3.P2, Line 1, delete 'and' before 'heating rates of'.

Response: We have changed this sentence to

> "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."

4.P2, Line 1, 'The glaciers on the TP recently shows are rapidly retreating' should be revised.

Response: We have corrected this to

> "The glaciers on the TP are facing rapid retreat, however, and if unchecked, that could result in adverse effects on Asian hydrological cycle and the Asian monsoon (Luo et al., 2020)."

5.P2, Line 4-8, Please cite these literatures, doi:10.1093/nsr/nwz191, doi.org/10.1016/j.atmosenv.2020.117583, doi: 10.1016/j.atmosenv.2019.04.001.

Response: We cite those papers in the revised version.

> "The Tibetan Plateau (TP) is an important regulator of climate change in the northern hemisphere, and it plays crucial roles in the functions of global ecosystems and climate stability (Liu et al., 2019b). The TP is covered by one of the largest ice masses on Earth, and it has been called the water tower of Asia (Liu et al., 2020)."

6.P2, Line 8-10, Please cite these literatures, doi.org/10.3390/rs12020231, doi:10.1002/joc.6430.

Response: We now cited these references in the revised version.

"The glaciers on the TP are facing rapid retreat, however, and if unchecked, that could result in adverse effects on Asian hydrological cycle and the Asian monsoon (Luo et al., 2020)."

7.P2, Line 16, 'the atmospheric BC studies on the TP' should be changed to 'studies on the TP atmospheric BC'.

Response: We have revised the sentence

"In addition, numerous studies have shown that the high bulk BC mass loadings and the associated regional influences on the TP are related to transport from South Asia (Liu et al., 2015; Han et al., 2020; Cong et al., 2015; Wang et al., 2015)."

8.P2, Line 16-18, Please cite the literature, doi: 10.5194/acp-15-12581-2015.

Response: We have read that paper and now cite it. Please see it in the answer for the above question.

9.P2, Line 24, 'the other is based on the field observations to apportion BC into different sources through a certain data analytical method.' Please add appropriate literature.

Response: We have re-written this text, and now it reads as follows:

"Several studies have assessed the contributions of different BC sources through model simulations or isotopic methods. For example, Zhang et al. (2015) investigated BC sources for different parts of the TP by using a chemical transport model and a source tagging approach, and they found that the contributions of BC sources varied among regions and with the seasons. Li et al., (2016) used filter sampling and carbon isotopes ($\Delta^{14}C$ and $\delta^{13}C$) to determine the BC from fossil fuels and biomass burning in several areas of the TP."

10.P2, Line 25, 'transport to TP' should be changed to 'transport to the TP'.

Response: We have corrected and add the article "the" into TP in the revised version.

11.P3, Line 5, 'are advantageous to capturing' should be changed to 'are advantageous to capture'.

Response: We revised the sentence

"A major disadvantage of filter-based measurements is they are constrained by low time resolution, and this makes it challenging to capture the detailed evolution of pollution events."

12.P3, Line 10, 'cross-section (MAC) used in the model.' Please add appropriate literature.

Response: We have added the relevant literature:

> "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)."

13. P4, Line 11, 'on the rooftop of' should be changed to 'at on the rooftop of'.

Response: We have corrected "on" into "at". The sentence now reads

> "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."

14. P4, Line 15, 'the radiative effect' should be changed to 'the radiative effects'.

Response: We have changed all "radiative effect" into "radiative effects" in the revised version.

15. P5, Line 4, 'was resolved using' should be changed to 'was resolved by using'.

Response: We have changed the sentence as below:

> "A dual-spot technique for the aethalometer measurements was used to compensate for non-linearity, while a factor of 2.14 was used to correct for the artifacts caused by the quartz filters (Drinovec et al., 2015)."

16. P8, Line 10, 'the number of the endpoints' should be changed to 'the number of endpoints'.

Response: We have deleted "the" before endpoints in the sentence:

> "where m_{ij} is the number of endpoints associated with BC mass concentration higher than the set criterion;"

17. P9, Line 8, 'model is elaborated in Ricchiazzi and Yang, (1998)' should be changed to 'model was elaborated in Ricchiazzi and Yang (1998)'.

Response: We have corrected the tense as follows:

> "The direct radiative effects (DRE) of source-specific BC were estimated with the widely used Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model—a detailed description of which may be found in Ricchiazzi and Yang (1998)."

18. P10, Line 15, delete 'which is' before 'within a relative boarder range'. P10, Line 18, delete 'which was' before 'over two times'. In addition, it is necessary to pay attention to the tense errors, which often appear throughout the manuscript.

Response: Thanks for pointing out the error in tense. The paper has been edited and thoroughly revised, and the problems with tense should have been corrected. The sentences were changed to

> "That was within the relatively broad range of $AAE_{biomass}$ (1.2–3.5) determined by other methods (e.g., $\Delta^{14}C$ and organic tracers) in previous studies (Sandradewi et al., 2008; Helin et al., 2018; Harrison et al., 2012; Zotter et al., 2017). The estimated average $MAC_{BC}(880)_{biomass}$ was 10.4 $m^2\ g^{-1}$; this is more than twice the value for uncoated BC particles suggested by Bond and Bergstrom (2006) ($MAC_{BC}(880)_{uncoated}$ = 4.7 $m^2\ g^{-1}$, extrapolated from 550nm to 880 nm by assuming AAE_{BC} = 1.0)."

19. P10, Line 19, Line 85, the comma before '(2006)' should be deleted.

Response: We corrected this mistake:

> "The estimated average $MAC_{BC}(880)_{biomass}$ was 10.4 $m^2\ g^{-1}$; this is more than twice the value for uncoated BC particles suggested by Bond and Bergstrom (2006) ($MAC_{BC}(880)_{uncoated}$ = 4.7 $m^2\ g^{-1}$, extrapolated from 550nm to 880 nm by assuming AAE_{BC} = 1.0)."

20. P11, Line 5, 'may be relation with' should be changed to 'may be related with' or 'may have relation with'.

Response: We corrected this:

> "Although unleaded gasoline has been used extensively in China since 2005, a considerable fraction of the Pb in the environment is still associated with vehicle-related particles, especially from the wear of metal alloys (Hao et al., 2019)."

21. P12, Line 26, 'This suggests and' should be changed to 'This suggests that'.

Response: We have changed the sentence as following:

> "One can infer from this that biomass-burning emissions were responsible for the high BC-loading episode during the campaign."

22. P14, Line 12, 'the mainland China' should be changed to 'mainland China' or 'the mainland of China'.

Response: We have corrected this mistake in the revised manuscript:

“The air masses associated with Cluster 3 originated from the interior of China, and this group had the lowest BC mass concentrations of the three clusters, 0.4 ± 0.1 μg m^{-3}. This third cluster composed small fraction of total trajectories (2%), and none of them were identified as polluted, suggesting minor influences from mainland China during the campaign.”

“The transportation sector has grown rapidly in mainland China (Liu, 2019a), and the regional transport of motor vehicle emissions may have been the cause for the observed diurnal variations in BC_{fossil} for Cluster 3.”