Response

The manuscript by Wang et al. provides a comprehensive analysis of the physicochemical properties of black carbon (BC)-containing particles over the Tibetan Plateau, with an emphasis on the impacts of anthropogenic emissions. The authors conduct detailed field observations, which contribute valuable data to this field of study. Their findings represent a noteworthy advancement in elucidating the impact of anthropogenic emissions on the properties of BC, especially within the environmentally sensitive Tibetan Plateau region. See my detailed comments below.

We appreciate the reviewer's kind effort and insightful comments. The amendment and modification have finished followed by all constructive comments in the revised manuscript and supporting information. Please kindly find our point-by-point responses listed below. The reviewer's comments are in blue font followed by our responses and revisions in the manuscript (in Italic).

Major Comments:

1. **Addressing Seasonality and Expanding Temporal Scope:** The manuscript effectively outlines how regional transport influences the BC characteristics over the plateau. This is particularly evident in the comparison between the northeast and southeast regions. Yet, the study seems to focus predominantly on the spring season (Table 1). It would be valuable if the authors could discuss the potential seasonality of these findings or provide reasons for the focus on this particular season, including how the results might vary in other seasons.

Response 1

Thanks so much for your foresighted suggestions. Due to the limitation of experimental condition, it is very hard to conduct long-term continuous observation at these sites in TP. Therefore, we did intensive observation focusing on the specific targets and we will conduct field campaign in more seasons in the future to explore the seasonal variation of BC characteristics.

The Xihai observation was designed to study the transportation of stronger anthropogenic emission from lower altitude regions in northwestern China. As Fig. 7a shows, Xihai was under relatively strong influence of transported anthropogenic emission from lower altitude regions during this period.

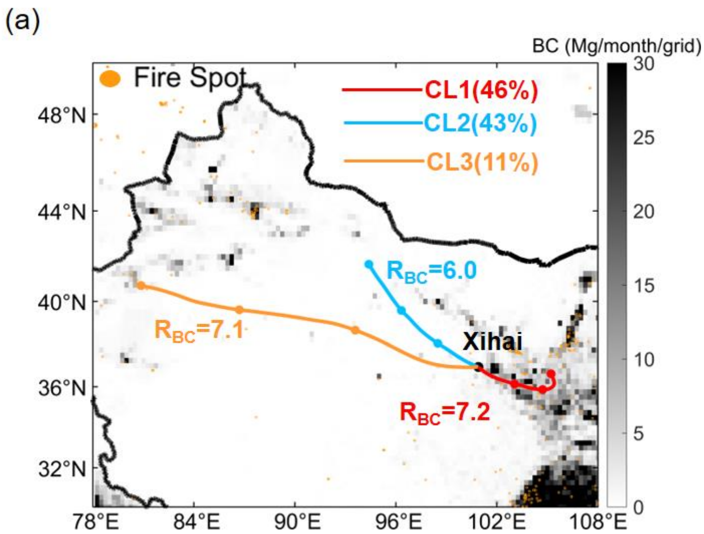

Figure 7 (a): The maps show the backward trajectories in different clusters of Xihai.

The Lulang observation was focused on studying the impact of biomass burning (BB) on BC characteristics over Southeastern TP. The average level of BC emissions from wildfires in surrounding area of Lulang was much higher during the observation period (93381 kg d^{-1}) than other periods (33877 kg d^{-1}), which provide us ideal condition to investigate the impacts of BB on BC physical and chemical properties.

2. **Lack of Detailed Model Evaluation：** The manuscript utilizes the WRF-Chem model to simulate the atmospheric processes and black carbon (BC) characteristics over the Tibetan Plateau. However, there seems to be a lack of detailed evaluation or validation of the model simulations against observational data. Without proper validation, the reliability of the model results and the subsequent conclusions drawn from them may be questionable.

Response 2

Thanks so much for your suggestions about the validation of model in our manuscript. We have added the evaluation and validation of model simulations into the revised SI in Line 20-32:

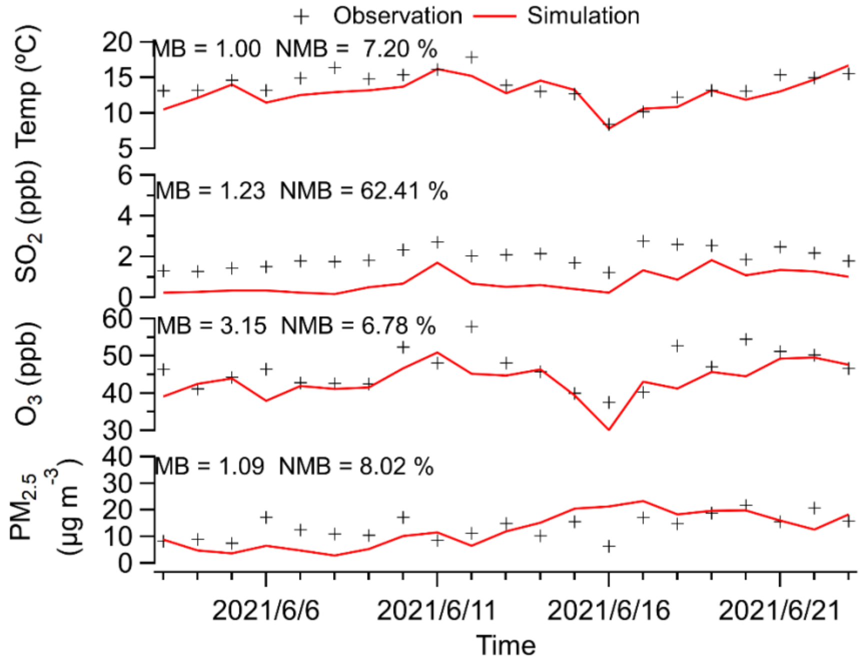

Figure S1: The time series of the near-surface air temperature, sulfur dioxide (SO_2), ozone (O_3) and mass concentration of fine particulate matter ($PM_{2.5}$) in the Xihai and surrounding area. The line and marker represent the results of ambient measurement and modelling respectively. The MB and NMB are mean bias and normalized mean bias of each parameter.

In this study, the air temperature at 2 m were evaluated based on the measurement data from our measurement and publicly available meteorological datasets of the University of Wyoming (http://www.weather.uwyo.edu/surface/). The air temperature was pretty close between the modelling and measurement, and the mean bias was +1.00 ℃. It was shown that the model had a good performance in the simulation of meteorological fields.

The air quality dataset at Xihai and the monitoring stations near to Xihai (https://quotsoft.net/air/) were used to evaluate the WRF-Chem model in simulating the air pollution. There were overall good agreement and small bias between model-simulated and observed concentration values of gaseous pollutant (SO_2, O_3) and particulate matter. The modelled SO_2 concentration level is relatively low, however, it is not the pollutants of major concern in this study.

Minor Comments:

3.Line 155: According to my understanding, the study by Cui et al. (2022) only provides BC concentrations in urban areas of Shanghai, and does not directly provide the number 25%. Please directly provide the BC concentrations mentioned in Cui et al. (2022)'s study and explain how the calculation for obtaining 25% is done. In addition, although EC is sometimes used as a substitute for BC in some cases due to the lack of BC observations, it should not be said that they are equivalent.

Response 3

Thanks so much for your suggestions, and we have modified the relevant description.

Firstly, the 25% ratio was calculated based on Cui et al. (2022) and our study. In Cui et al. (2022), the campaign-average rBC concentration measured by SP-AMS was 0.92 ± 0.81 μg m^{-3} in Shanghai which is a typical urbanized area. In our study, the average rBC concentration was 0.17 ± 0.17, 0.24 ± 0.20 μg m^{-3} in Lulang and Xihai, respectively. Lulang and Xihai represent the general environment in different regions of the Tibetan Plateau.

Since these two studies used the same instrument, the measured concentration level can be compared directly. The ratio of rBC concentration in Xihai and Shanghai was approximately 25%, and for Lulang, the average rBC concentration was 18% to that in Shanghai. We have modified the expression in the manuscript to make the meaning of this ratio clearer:

Line 176-178: *Compared to measurements using the same instrument in a metropolitan area (Cui et al., 2022), the rBC concentration of TP (0.24 ± 0.20 μg m^{-3}) was approximately 25% or less of Shanghai (0.92 ± 0.81 μg m^{-3}).*

Secondly, thanks you for reminding us to add the necessary clarification about the differences in measurement methods for black carbon. "BC (EC)" here is not very precise. The term "black carbon (BC)" has not been used rigorously or consistently throughout all previous modelling and measurement literature. Similar terms including "rBC", "eBC", and "EC" has also been widely used corresponding to different measurement techniques. We have corrected the heading of Table 1 as "BC concentration" and modified the Line 170-176 of manuscript:

Note that, the term "black carbon (BC)" has not been used rigorously or consistently throughout all previous modelling and measurement literature (Bond et al., 2013). Similar terms including "rBC", "eBC", and "EC" has also been widely used corresponding to different measurement techniques. BC measured by laser-induced techniques is often referred as "rBC", and measured BC using light absorption (e.g. Aethalometer, AE) and thermal/optical methods are normally named as "the equivalent BC (eBC)" and "elemental carbon (EC)", respectively. In Table 1, BC concentrations in TP measured by several common techniques were collected and grouped according to the methods to make clearer comparison.

4.Line 158: The sentence suggests that the intermediate position of the concentration within the TP region may be due to anthropogenic emissions in the surrounding area. However, it does not explain why it is not in a high concentration position.

Response 4

Thanks for your comments. The expression here is not very precise. As mentioned in Response 3, it is better to compare the rBC concentration measured by same measuring techniques. Hence, we have modified the description in the Line 178-180 in revised manuscript and further discussed the anthropogenic emissions in the subsequent sections:

The rBC concentration in Xihai was relatively high compared to southeastern and central TP measured using same technique (Table 1). This was potentially attributed to the strong BC emissions in surrounding area of northeast TP (Fig. 1).

5.Line 175: Is this difference statistically significant? Are there any indicators for testing the significance of differences that can be reported?

Response 5

Thank you very much for reminding us to confirm the significance of the statistical results. The t-test (α=0.05, n=51) results proved that the rBC, PM_{BC} and R_{BC} were significantly different between Xihai and Lulang, and we have added the t value in the revised manuscript in Line 194:

the difference (t_{rBC}=2.8, t_{PMBC}=2.1) between the two sites was proved by the t-test (α=0.05, ν=50).

and Line 199-200:

The difference on mixing states of PM_{BC} was also demonstrated by the t-test (t_{RBC}=2.4).

6.Line 192: The sentence could be improved by providing citations to support the claim that C2H3O+ is a typical biomass burning (BB) tracer.

Response 6

Thanks so much for your suggestion, we have added the citations in Line 221-222:

*The POA factor in Lulang had higher fraction of signal of $C_2H_3O^+$ (m/z 60) ion (f_$C_2H_3O^+$) in mass spectrum, which is the fragment of levoglucosan mainly from BB **(Lee et al., 2010)**.*

7.Line 222: 17.3% cannot be reflected in Fig. 5b, but only in Fig. 5a, and there is no particular reason to switch from two decimal places to one decimal place in the figure.

Response 7

Thanks so much for your carefully reviewing, we have corrected this sentence and standardized decimal places in the revised manuscript:

Line 245: *Besides MO-OOA, NO_3^- (17%) and HOA (35%) al**so made large contribution on BC coating (Fig. 5a) and coated OA (Fig. 5b) in Xihai compared to Lulang.***

8.Line 240: The statement "The abundant NO3- was closely associated with anthropogenic sources" is mentioned here, but it should be referenced in line 222 to support the conclusion that "indicating that anthropogenic..."

Response 8

Thanks so much for your suggestion. We have modified the sentence following your suggestion to make the statements appearing in the more appropriate position that is more relevant to the viewpoints:

Line 246-250: ***The HOA and NO_3^- were both closely associated with anthropogenic sources because the anthropogenic sources emitted the HOA (Zhang et al., 2005) and precursors of NO_3^- largely (Dall'osto et al., 2009; Richter et al., 2005; Sun et al., 2018).** It indicated that anthropogenic emissions have a strong influence on coating process of PM_{BC} in northeast TP, which is quite different from southeast TP.*

Reference

Cui, S. J., Huang, D. D., Wu, Y. Z., Wang, J. F., Shen, F. Z., Xian, J. K., Zhang, Y. J., Wang, H. L., Huang, C., Liao, H., and Ge, X. L.: Chemical properties, sources and size-resolved hygroscopicity of submicron black-carbon-containing aerosols in urban Shanghai, Atmospheric Chemistry and Physics, 22, 8073-8096, 10.5194/acp-22-8073-2022, 2022.

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Lee, T., Sullivan, A. P., Mack, L., Jimenez, J. L., Kreidenweis, S. M., Onasch, T. B., Worsnop, D. R., Malm, W., Wold, C. E., Hao, W. M., and Collett, J. L.: Chemical Smoke Marker Emissions During Flaming and Smoldering Phases of Laboratory Open Burning of Wildland Fuels, Aerosol Science and Technology, 44, I-V, 10.1080/02786826.2010.499884, 2010.

Richter, A., Burrows, J. P., Nüss, H., Granier, C., and Niemeier, U.: Increase in tropospheric nitrogen dioxide over China observed from space, Nature, 437, 129-132, 10.1038/nature04092, 2005.

Sun, P., Nie, W., Chi, X., Xie, Y., Huang, X., Xu, Z., Qi, X., Xu, Z., Wang, L., Wang, T., Zhang, Q., and Ding, A.: Two years of online measurement of fine particulate nitrate in the western Yangtze River Delta: influences of thermodynamics and N2O5 hydrolysis, Atmospheric Chemistry and Physics, 18, 17177-17190, 10.5194/acp-18-17177-2018, 2018.

Zhang, Q., Worsnop, D. R., Canagaratna, M. R., and Jimenez, J. L.: Hydrocarbon-like and oxygenated organic aerosols in Pittsburgh: insights into sources and processes of organic aerosols, Atmospheric Chemistry and Physics, 5, 3289-3311, 10.5194/acp-5-3289-2005, 2005.