Response

The manuscript provides observational evidence of the spatial discrepancies in the physicochemical properties of black carbon (BC)-containing particles within the Tibetan Plateau region. It emphases the significant impacts of elevated anthropogenic emissions from surrounding low-altitude areas on BC, altering its concentration and chemical composition, as well as enhancing its light absorption ability. The manuscript is well written, and the topic is of interest and fits the scope of ACP. I recommend a minor revision before publication. The detailed comments or suggestions are shown 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).

1.In this study, there are three factors identified using PMF, which is a little less than the usual number (normally 4 or 5 factors can be resolved by PMF for HR-AMS data). It is better to provide the explanation of why the 3-factor result is chosen in the main text or SI.

Response 1

Thanks so much for your suggestion. We have added relevant material in SI (Line 43-57, Figure S4-S7) to clarify why the 3-factors solutions were selected.

As the Fig. S4a shows, the reduction rate of PMF was small after the number of factors exceeds 3 and 4, and the measured and reconstructed signals matched well in 3-factors solutions (Fig. S4b). Hence, the 3-factors solution can reasonably analyze the source of OA in Lulang.

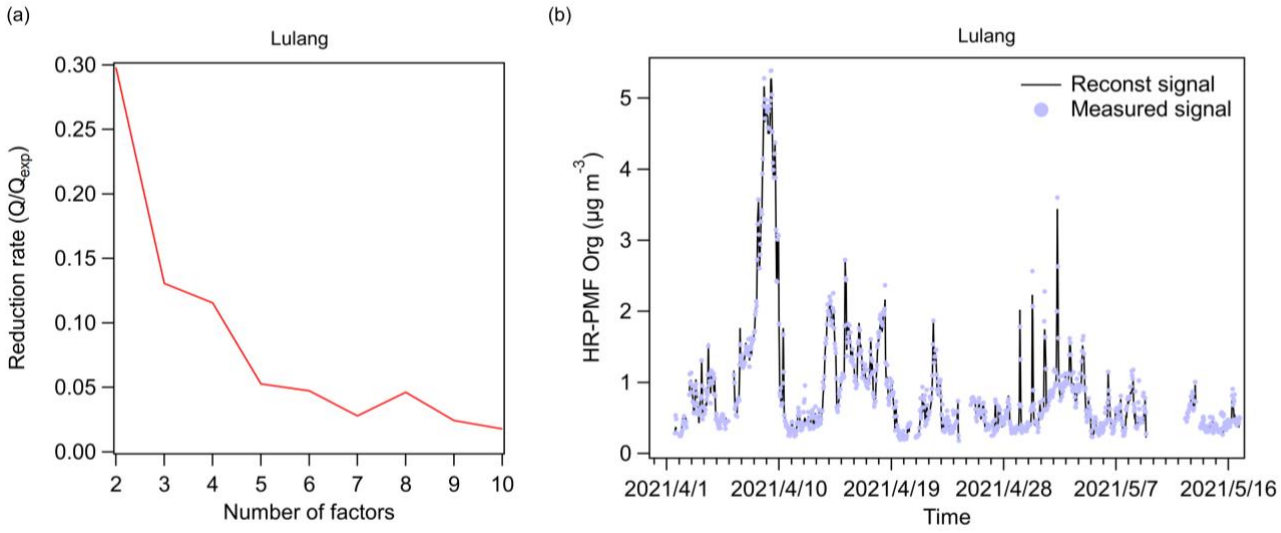

Figure S4:(a) The variation of the reduction rate of Q/Qexp with the number of factors in Lulang. (b) The time series of reconstructed signal and measured signal for 3-factors solution.

In addition, the Factor 3 in the 4-factors scheme was split primarily from LO-OOA and BBOA. This factor is strongly correlated with all other factors, predicting that this factor is not very independent and representative. It is likely to be a product of over-splitting.

The Factor 3 in the 4-factors scheme is characterized primarily by a strong $C_2H_3O^+$ signal (Fig. S5b), and the $C_2H_3O^+$ signal is usually a tracer ion for fresh SOA. It also has higher correlation to $C_2H_4O_2^+$ (r=0.81), meaning that it may be the aged BBOA. However, the BBOA factors in the 4-factors solution

also has a relatively high degree of oxidation compared to fresh BBOA. The meaning of the Factor 3 and BBOA is faintly repetitive, also indicating that the Factor 3 may the product of over-split.

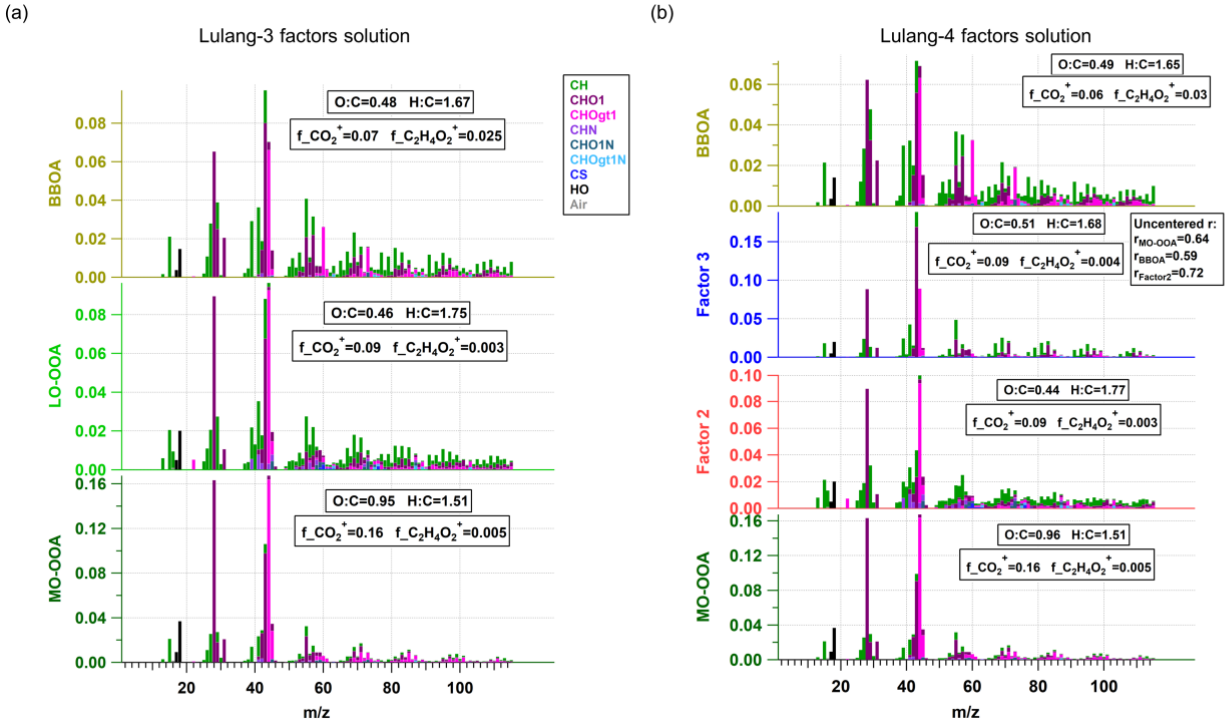

Figure S5: The mass spectrum of factors in the (a) 3-factors solution and (b) 4-factors solution.

Similar to Lulang, the 3-factors scheme reconstructs the OA concentrations well in Xihai (Fig. S6b). When the number of factors is high (>4), there is a small decrease in Q/Q_{exp} (Fig. S6a). For the 4-factors solution, the concentration of the new factor (Factor 4) appeared close to zero for most of the time (Fig. S7b), indicating that this factor did not represent a stable source of OA. The new Factor 4 only had a brief increase in concentration during the period of 18-20 June, with basically same variation as the LO-OOA. Considering above reasons, the Factor 4 is most likely a product of the excessive decomposition.

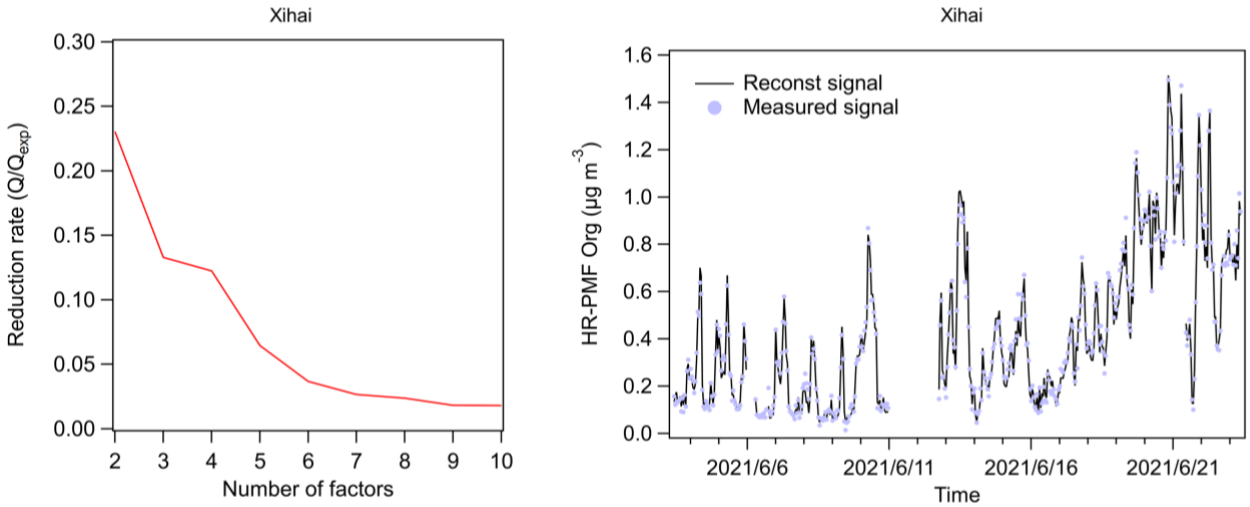

Figure S6:(a) The variation of the reduction rate of Q/Qexp with the number of factors in Xihai. (b) The time series of reconstructed signal and measured signal for 3-factors solution.

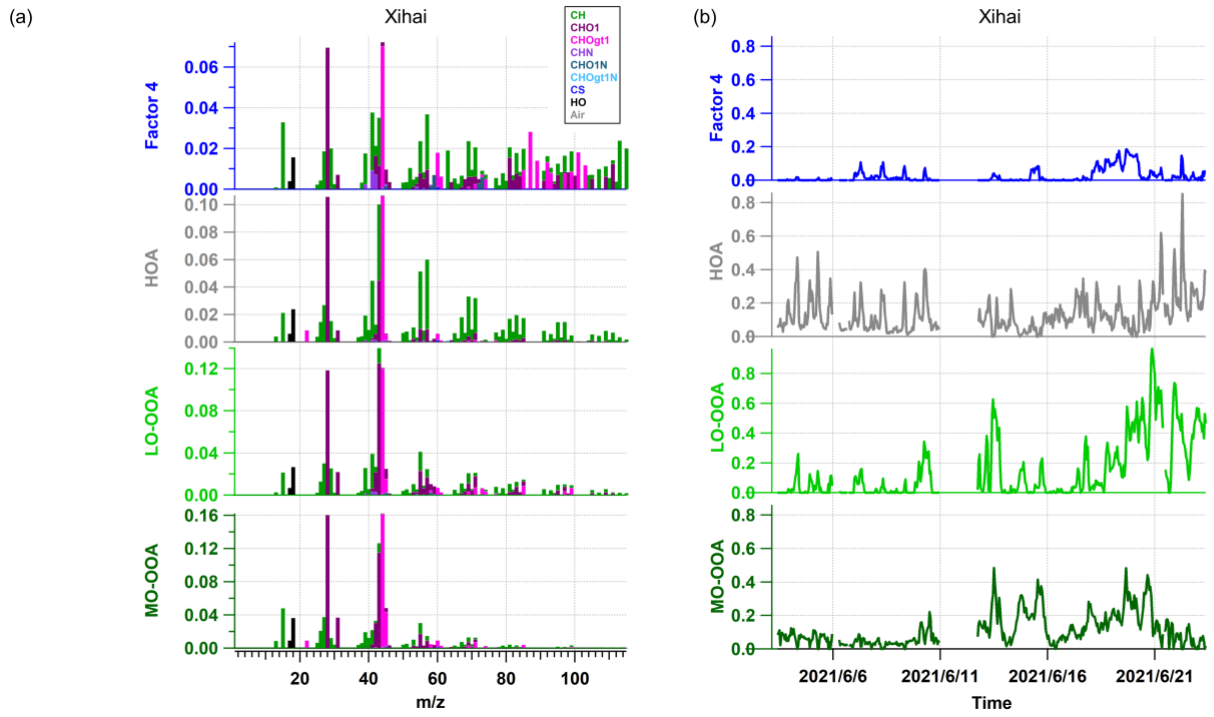

Figure S7: The (a) mass spectrum and (b) time series of factors in the 4-factors solution.

2.Line 131: what refractive index did you use for the core-shell Mie model? Please add the numbers you adopted and reference here.

Response 2

Thanks so much for your reminder, we have added the description of the refractive index we used in the revised manuscript (Line 145-146): *The refractive index was 1.95 - 0.79i for rBC (Bond and Bergstrom, 2006), and was 1.52 - 10^{-6}i (Pitchford et al., 2007) for BC coating at 550 nm wavelength.*

3.Line 154: It is suggested to clarify the different measuring instruments corresponding to different BC definitions before comparing BC concentrations in Table 1.

Response 3

Thank you for suggesting us to add the necessary clarification on the differences in measurement methods for black carbon (BC). This suggestion is important because the term "black carbon (BC)" has not been used rigorously or consistently throughout all previous modeling and measurement literature. Similar terms including "rBC", "eBC", and "EC" has also been widely used corresponding to different measurement techniques. We have revised the Table 1, and correspondingly modified the manuscript in Line 170-176:

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.In Table 1, the description “BC (EC)” is not very precise here. Please revise them.

Response 4

Thanks very much for your suggestion, we have corrected this imprecise expression, and the heading of Table 1 has been modified to “BC concentration”. We have also added the necessary clarification about the methods and definition of BC as Response 3 mentioned.

5. Table 1: Please add a note illustrating the meaning of the numbers in “BC concentration” column, i.e., is it a mean or median value? What does the range stand for in the parenthesis?

Response 5

Thank you for pointing out the lack of data description here. Followed by your advice, the meaning of data has been added in the caption of Table 1 (Line 166-167):

Table 1: Overview of the BC concentration (mean±1σ) at different sites of TP in existing studies. The minimum value and maximum value were shown in the parenthesis.

6.Line 106: please add abbreviation of light absorption coefficients (b_{abs}) here.

Response 6

Thanks so much for your notice, the abbreviation of light absorption coefficients (b_{abs}) has been added in Line 157 in revised manuscript:

PM_{BC} concentration and light absorption coefficients ***(b_{abs})*** *increased in the latter period of Xihai campaign.*

7.Figure 7: the color of CL2 in subplot (b) overlaps with the background map. It is better to change it to ensure the visibility.

Response 7

Thanks so much for your advice to improve the visibility of the figure, the color scheme of Fig. 7 has been modified as follow:

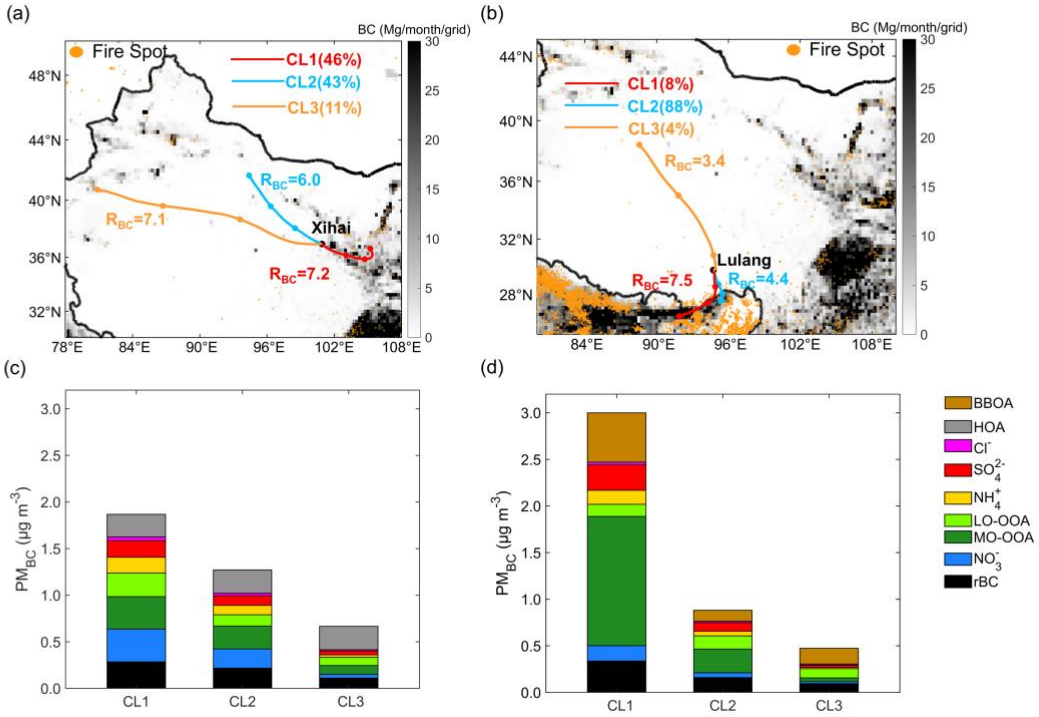

Figure 7: The maps show the backward trajectories in different clusters of (a) Xihai and (b) Lulang. Each circular marker along the trajectories denotes a 24-hour interval. The background shading represents the anthropogenic BC emission intensity and the orange spots represent the location of wildfire during the campaign in (a) and (b). The stacked bar plots show the mass concentration of coating components and rBC in (c) Xihai and (d) Lulang.

8.Please check and ensure that the number of significant digits are consistent throughout the manuscript.

Response 8

Thanks so much for your advice to improve the uniformity and preciseness of our article. The number of significant digits has been unified throughout the manuscript, and the modification was highlighted by blue font in the manuscript.

Reference

Bond, T. C. and Bergstrom, R. W.: Light absorption by carbonaceous particles: An investigative review, Aerosol Science and Technology, 40, 27-67, 10.1080/02786820500421521, 2006.

Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., et al.: Bounding the role of black carbon in the climate system: A scientific assessment. Journal of Geophysical Research: Atmospheres, 118(11), 5380–5552, 2013.

Pitchford, M., Malm, W., Schichtel, B., Kumar, N., Lowenthal, D., and Hand, J.: Revised algorithm for estimating light extinction from IMPROVE particle speciation data, Journal of the Air & Waste Management Association, 57, 1326-1336, 10.3155/1047-3289.57.11.1326, 2007.