

# Response

Black carbon (BC) is one of the most important aerosol species affecting climate, glaciers and hydrology in Tibetan Plateau (TP). However, large uncertainties still exist in the estimation of BC DRF over the TP, which is related to the mixing states of BC. This study presents multi-point observations of BC mixing states, especially the chemical composition of BC-containing particles, and combines model simulation to reveal the causes of spatial differences and the impacts of transported emission sources. It provides valuable results which may support the future evaluation of BC climate and environmental effects over the TP region. The coupling mechanism between the planetary boundary layer, topography, and pollution mentioned in the text is also interesting. Overall, it is a well-organized manuscript. Thus, I suggest publishing after minor revisions. The detail comments 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. My main suggestion is that the model configuration and validation need to be more detailed, which can be described in 2.3 part. Additional figures of model validation can be added into the SI (e.g. meteorological parameters, gaseous pollutants). Furthermore, a brief setup of the chemical transport model should be elucidated within the main text, in addition to its inclusion in the Supplementary Information.

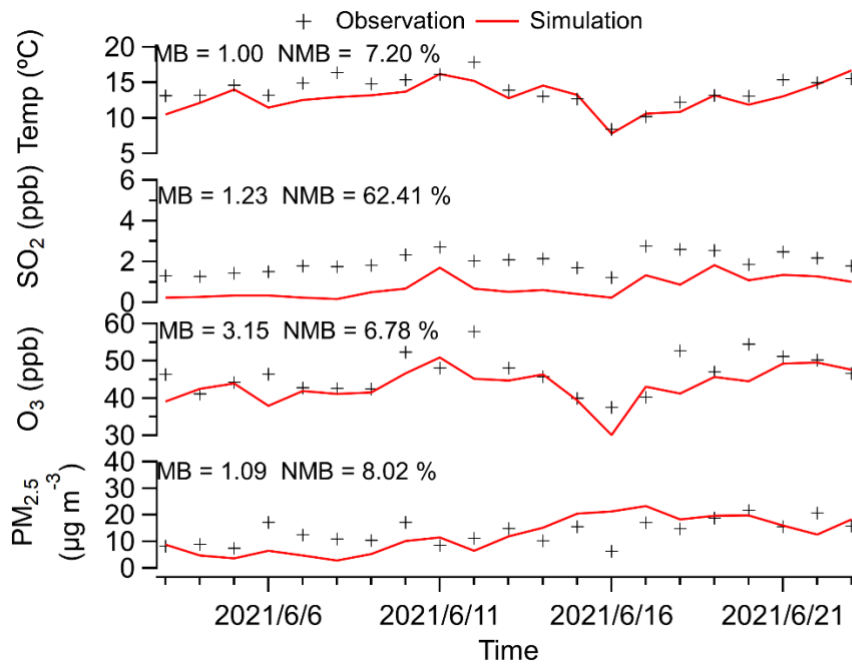
## Response 1

Thank you very much for your advice on the introduction of modelling methods, and we have added more details and validation of the modelling in the main text and the SI.

We have modified the main text as shown below:

1. Line 115-116: *The simulation was conducted for the longer period including the times of whole campaign from 3 June to 11 June 2021;*
2. Line 122-131: *The Yonsei University planetary boundary layer (YSU PBL) scheme was used to parameterize boundary layer processes (Hong et al., 2006). Other essential physical parameterization options included the unified Noah land surface model (Ek et al., 2003), the Lin microphysics scheme (Lin et al., 1983), and the Grell-Freitas cumulus parameterization scheme (Grell and Freitas, 2014). For representing atmospheric chemistry numerically, we utilized the Carbon-Bond Mechanism version Z photochemical mechanism along with the Model for Simulating Aerosol Interactions and Chemistry aerosol module (Zaveri and Peters, 1999; Zaveri et al., 2008). Both natural and anthropogenic emissions were considered in this regional WRF-Chem modeling study. Anthropogenic emissions were derived from the Multi-resolution Emission Inventory for China (MEIC), which includes emissions from power plants, residential combustion, industrial processes, on-road mobile sources, and agricultural activities (Li et al., 2017). Biogenic emissions were calculated online using the Model of Emissions of Gases and Aerosols from Nature (MEGAN), encompassing more than 20 biogenic species (Guenther et al., 2006)*

We have modified the SI in Line 20-32:



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Figure S1: The time series of the near-surface air temperature, sulfur dioxide (SO<sub>2</sub>), ozone (O<sub>3</sub>) and mass concentration of fine particulate matter (PM<sub>2.5</sub>) 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 (NMB) of each parameter.

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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 °C. It was shown that the model had a good performance in the simulation of meteorological fields.

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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<sub>2</sub>, O<sub>3</sub>) and particulate matter. The modelled SO<sub>2</sub> concentration level is relatively low, however, it is not the pollutants of major concern in this study.

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2.As I commented earlier in the discussion process, the authors should pay careful attention to the use of significant figures in both the main text and figures. For instance, in Figure 5, the fractions should be reported with 2 significant digits, not 4. Additionally, it is important that the number of significant figures is consistent across all panels in Figures 3 and 7.

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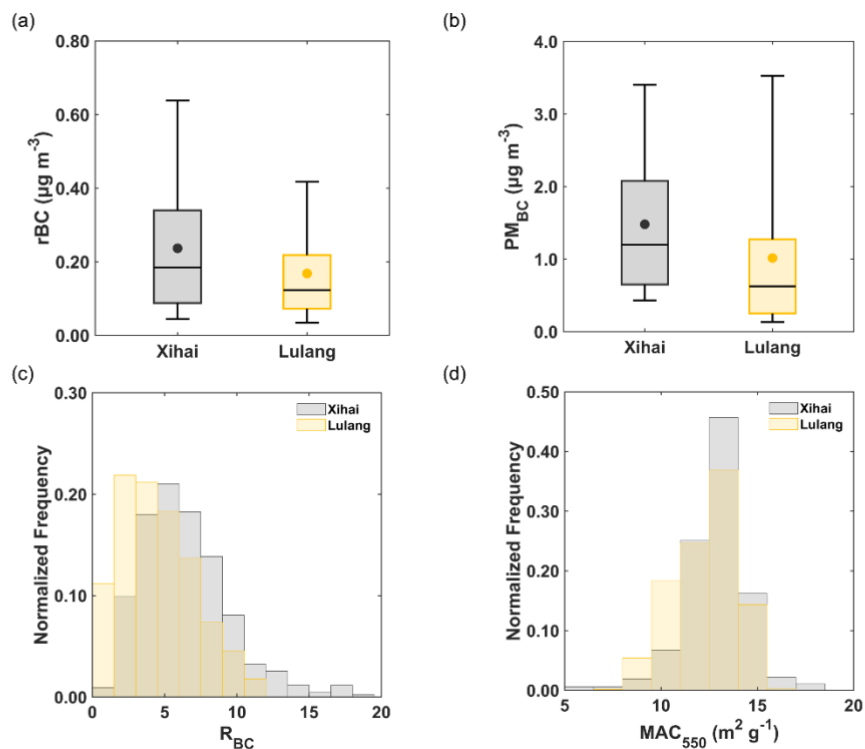
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### Response 2

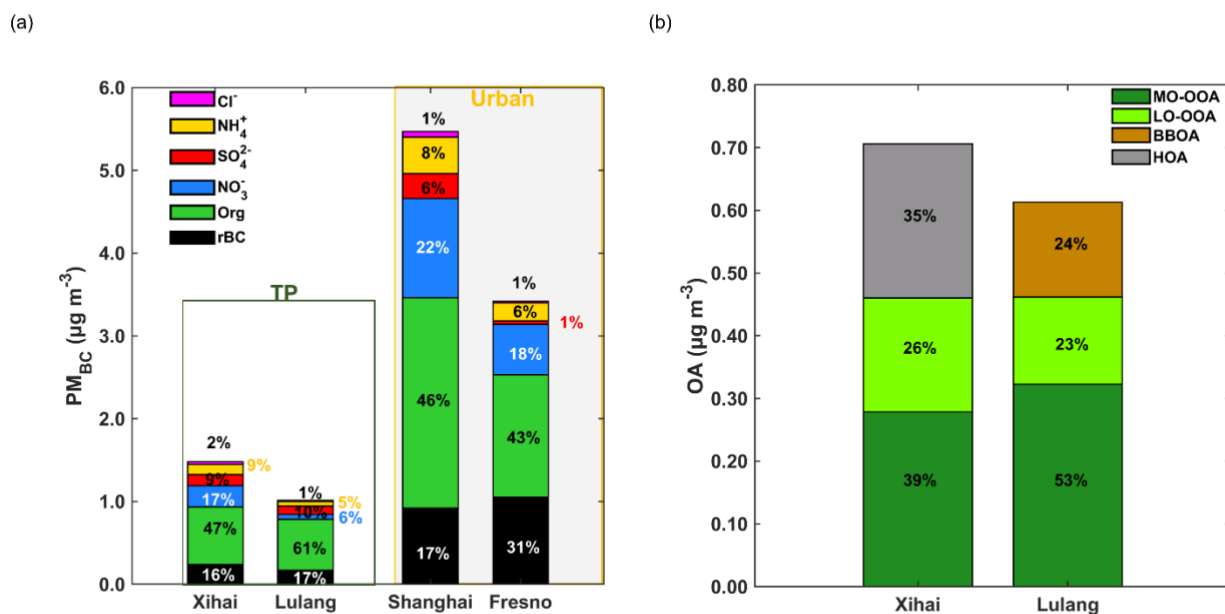
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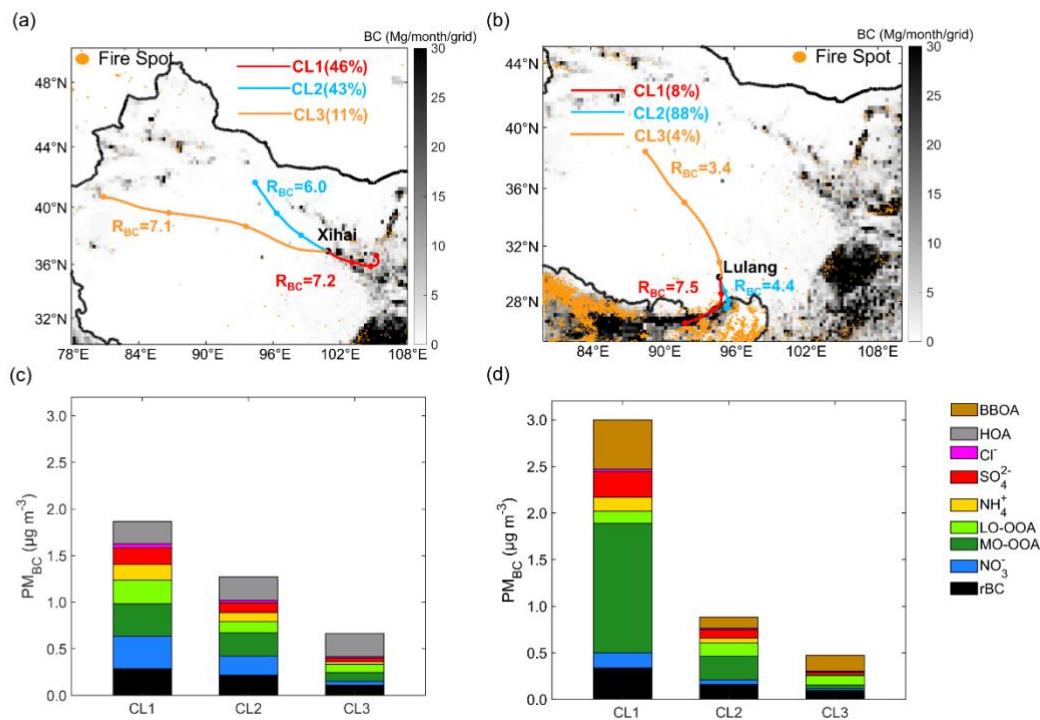
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 66 Figure 3: The box plots of (a) rBC and (b) BC-containing particles mass concentrations in Xihai and Lulang, the lower and upper lines of  
 67 box plot represent the 25<sup>th</sup> and 75<sup>th</sup> percentiles and the whiskers stand for 5<sup>th</sup> and 95<sup>th</sup> values. The charts of normalized frequency distribution  
 68 show (c) mass ratio of coating substance to rBC core ( $R_{\text{BC}}$ ) and (d) mass absorption cross-section (MAC). Only 1.15% of the  $R_{\text{BC}}$  exceeded  
 69 the maximum value of bin (19.5) in Xihai, and no  $R_{\text{BC}}$  exceeded the maximum value of bin in Lulang.



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 71 Figure 5: The stacked bars represent mass concentrations of (a) different species in BC-containing particles ( $\text{PM}_{\text{BC}}$ ), and (b) different factors  
 72 of organic aerosol in BC-containing particles. The numbers on the plot show the percentage of different species and organic factors. In  
 73 subplot (a),  $\text{PM}_{\text{BC}}$  in the TP (this study) was compared to  $\text{PM}_{\text{BC}}$  in urban regions (Collier et al., 2018; Cui et al., 2022).



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

79 3.Line 27: the term “self-elevated” is typically associated with plume uplift due to the absorption of  
 80 solar radiation by black carbon. However, as the uplift in question may not be solely attributed to this  
 81 mechanism, it is recommended to replace the term.

82 **Response 3**

83 Thanks so much for your advice. The relative content has been modified in the Line 26-28 in abstract:  
 84 *In contrast to Xihai, the thickly coated BC in Lulang was mainly caused by **elevation and***  
 85 ***transportation of biomass burning plume** from the South Asia.*

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 87 4.Line 36: The expression “2,500,000 km<sup>2</sup>” appears twice in a sentence. Please remove one instance  
 88 to ensure clarity and conciseness.

89 **Response 4**

90 Thanks so much for pointing out the repetition here. The Line 36 has been modified as followed:  
 91 *The Tibetan Plateau (TP) is the largest plateau of the world, covering approximately 2.5 million km<sup>2</sup>.*

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 93 5.Line 150, too many digitals for wind speed and gaseous pollutant concentrations

94 **Response 5**

95 Thanks for your suggestion. We have modified the decimal fraction of number in Line 156, 164 and  
 96 165.

97 Line 156: *The mass concentration of rBC shows large temporal variation at both sites, with ranges*  
 98 *of 0.02–1.28  $\mu\text{g m}^{-3}$  in Xihai and 0.02-2.22  $\mu\text{g m}^{-3}$  in Lulang.*

99 Line 164-165: *with mean value of 1.8  $\pm$  1.2 m s<sup>-1</sup> and 1.5  $\pm$  1.2 m s<sup>-1</sup>, respectively. In terms of gaseous*  
 100 *pollutants, higher levels of NO<sub>x</sub> and O<sub>3</sub> were observed in Xihai (5.3  $\pm$  3.4 and 48  $\pm$  13 ppb) than in Lulang*

101 *(4.0 ± 2.5 and 35 ± 15 ppb).*

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103 6.Line 175, a t-test is needed when comparing RBC between Xinhai and Lulang

104 **Response 6**

105 Thank you very much for reminding us to show the significance of our statistical results. The t-test  
106 ( $\alpha=0.05$ ,  $v=50$ ) results have proved that the difference of RBC in two sites was significant, and we have  
107 modified our manuscript in Line 199-200:

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

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110 7.Line 199-200, “MO-OOA had very high O:C (0.84), while the O:C of LO-OOA was only 0.49.” It  
111 is better to be rephrased to in Xinhai “MO-OOA exhibited higher O:C ratio (0.84) than LO-OOA (0.49)”

112 **Response 7**

113 Thanks so much for the suggestion. We have modified our manuscript following your suggestion:

114 Line 220-221: *MO-OOA exhibited higher O:C ratio (0.84) than LO-OOA (0.49).*

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116 8.Line 201 “Therefore, this POA factor was identified as biomass burning OA (BBOA) in Lulang.”.  
117 Since  $f_{60}$  from this factor is not as high as those from biomass burning source test, likely caused by aging  
118 process, it is better to state that this factor is likely associated with biomass burning activities.

119 **Response 8**

120 Thanks so much for your attention that the BBOA in Lulang was slightly different from the fresher  
121 BBOA. According to your advice, we have added some clarification about BBOA to make it clearer:

122 Line 223-225: *Moreover, the  $f_{CO_2^+}$  and  $f_{C_2H_4O_2^+}$  (0.07 versus 0.025) of this factor were also*  
123 *within the triangle area in previous BBOA study (Cubison et al., 2011), and the  $f_{C_2H_4O_2^+}$  was lower*  
124 *than the fresh BBOA, indicating that this factor was influenced by biomass burning activities and aging*  
125 *processes collectively.*

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127 9. Figure 6, in the figure caption, it is better to explain how the grid of x-axis is calculated since RBC  
128 equals 0 meaning externally mixed BC.

129 **Response 9**

130 Thanks so much for your suggestion. We agree that this may cause the misunderstanding. The RBC  
131 was calculated averagely in different bins from 0 to 20 in Xihai, and the width of bin is 1.5. In Lulang,  
132 the bin of RBC was from 0 to 12 with the same bin width. The coating composition at RBC=0 was average  
133 composition of BC coating with RBC between 0 and 1.5 in original figure. We have revised the Fig. 6. The  
134 x-axis is corresponding to the median value of each RBC bin, rather than the boundary value of each RBC  
135 bin.

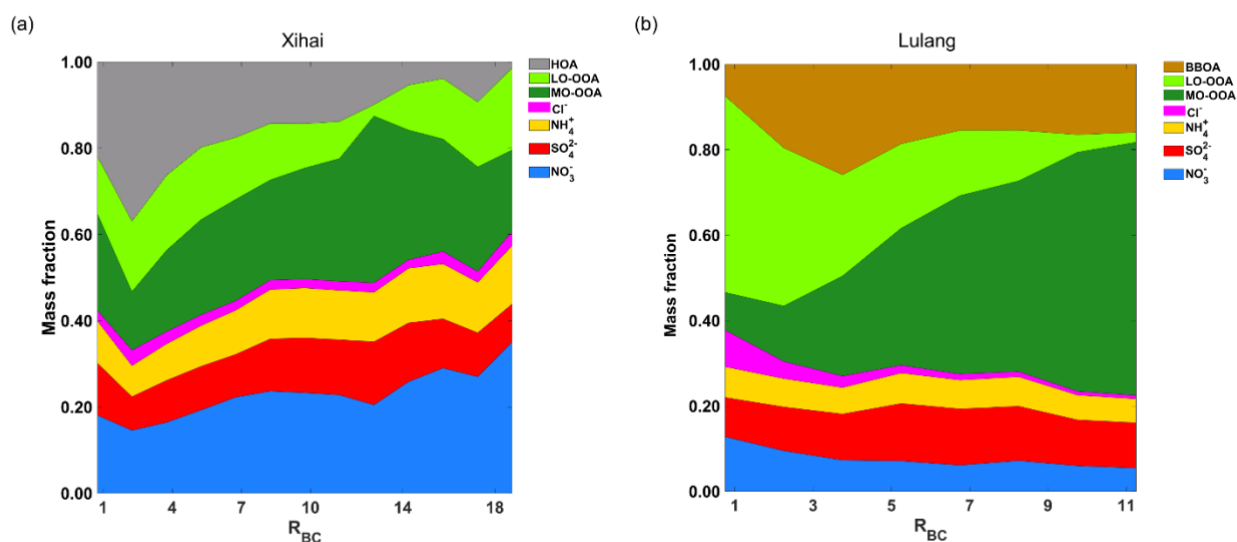


Figure 6: The variation of BC coating composition with  $R_{BC}$  between (a) Xihai and (b) Lulang. The x-axis represents the mass ratio of BC coating components and rBC cores ( $R_{BC}$ ), and the y-axis represents the mass fractions of BC coating components coated on rBC. The mass fraction of components was averaged in each bin of  $R_{BC}$  (bin width: 1.5).

10.Line 191: “The POA factor had higher signal of  $C_4H_7^+$  and  $C_4H_9^+$  in its mass spectrum...”. Please explain the major sources of  $C_4H_7^+$  and  $C_4H_9^+$  and add proper references here.

**Response 10**

Thanks so much for your reminder, we have explained the major sources of the two important ions and added the references in the revised manuscript.

Line 211-212: *The POA factor had higher signal of  $C_4H_7^+$  and  $C_4H_9^+$ , which is the important alkyl fragments from primary sources (Hu et al., 2016).*

11. Line 214: “OA was the dominant component of BC coating (Fig. 5b) at both sites”. Here it should refer to Fig. 5a rather than Fig. 5b.

**Response 11**

Thanks so much for your carefully reviewing and attention, we have corrected this image number in the revised manuscript.

Line 238: *OA was the dominant component of BC coating (Fig. 5a) at both sites.*

12. Line 219: “The dominance of MO-OOA in BC coating was resulted from strong atmospheric oxidizing capacity in TP and fast aging process during transport.”. Based on Fig. 5, I don’t think there is information about the atmospheric oxidation capacity and aging rates during transport. If this discussion is located in the latter part of the article, it is suggested to place this sentence in a more appropriate location.

**Response 12**

Thank you very much for the advice that makes the figures more relevant to the viewpoints. We have modified the article followed by your suggestion:

Line 242-245: *The BC coating was dominated by MO-OOA which was importantly affected by*



163 atmospheric oxidizing process. The concentration of  $O_3$  highly relative to atmospheric oxidizing capacity  
164 improved significantly in afternoon (Fig. S8), and the enhanced oxidizing capacity could cause increase  
165 of MO-OOA in BC coating in both Xihai and Lulang.

167 13. Line 305: is there any observed MAC in TP region? If yes, it is recommended to compare the  
168 results in this study with previous observations (references need to be added accordingly).

### 169 **Response 13**

170 Thanks so much, your suggestion is important to demonstrate the representativeness of our  
171 observations results. The manuscript has been modified as shown below:

172 Line 331: *The MAC were all relatively high in three clusters of airmasses of Xihai, with distribution*  
173 *peaked between 12 and 14  $m^2 g^{-1}$  that numerically comparable to previous studies (Wang et al., 2015).*

174 Line 336: *The MAC in Lulang was also comparable to previous studies (Wang et al., 2018) that the*  
175 *peak of MAC distribution was 7.6  $m^2 g^{-1}$  at 870 nm (12.0  $m^2 g^{-1}$  at 550 nm if the Absorption Ångström*  
176 *Exponent of BC is 1.0).*

177 Line 338: *In CL1 airmasses of Lulang, MAC mainly distributed at the bin between 12 and 14  $m^2 g^{-1}$*   
178 *that is close to MAC (13.1  $m^2 g^{-1}$  at 550 nm) at other TP sites affected by biomass burning plume (Tan*  
179 *et al., 2021).*

181 14. The abbreviations of CL 1,2,3 should be defined in the main text.

### 182 **Response 14**

183 Thanks so much for your carefully reviewing, the abbreviations of the three clusters of the airmasses  
184 have been clarified in Line 278-279:

185 *In Xihai, the airmasses were dominantly from eastern region outside of TP, as indicated by airmasses*  
186 *cluster1 (CL1), followed by the airmasses of cluster2 (CL2) from the northwest of Xihai, and the*  
187 *airmasses of cluster3 (CL3) from west of Xihai (Fig. 7a).*

### 189 **Reference**

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