Responses to reviewers for the paper "Strong Primary Contribution to Brown Carbon Light Absorption in Tibet and Urban Areas: Insights based on in situ measurements"

By Wenhui Zhao et al.

We appreciate the two reviewers' comments and support for the publication of this manuscript after minor revisions. Following the reviewers' suggestions, we have carefully revised the manuscript. To facilitate the review process, we have copied the reviewer's comments in black text. Our responses are in regular blue font. We have responded to all the referee comments and made alterations to our paper (**using the revision mode**).

Anonymous Referee #2

General Comments

This manuscript presents concurrent field campaigns in the Qinghai–Tibet Plateau (Yangbajing) and urban Guangzhou during July 2022, combining multi-wavelength Aethalometer and aerosol mass spectrometer (AMS) measurements to quantify brown carbon (BrC) optical properties, identify their sources, and estimate radiative forcing. The authors apply positive matrix factorization (PMF) for source apportionment, multiple linear regression (MLR) to derive source-specific mass absorption cross-sections (MAC), and a simple forcing efficiency (SFE) approach to estimate BrC radiative effects. The study is well-motivated, addressing an important gap in quantifying BrC MAC specific for individual OA factors, in a high-altitude background environment, while providing a direct urban–background comparison under simultaneous meteorological conditions.

The dataset is valuable, especially for the Tibetan site where few high-time-resolution BrC source apportionments exist. The methodology is robust and well-documented, and the analyses are generally clear. The findings are important for both modeling and policy-relevant emission reduction strategies. The observation that fossil fuel–related BrC has comparable MAC to biomass burning–related BrC in these environments is noteworthy.

However, several aspects could be clarified or expanded to strengthen the manuscript's contribution. I recommend publication after minor revisions addressing the points below.

Major Issues

Global significance of Tibetan data

R2.1: The introduction should more clearly articulate the broader relevance of the central QTP measurements—particularly for regional climate forcing and glacier melt—and distinguish them from prior work on the QTP margins.

A2.1: We have added explanation regarding the significance of light-absorbing substances for the melting of central glaciers and their radiative forcing. Furthermore, we have clarified the differences between the central QTP measurements and those in the marginal QTP areas, and emphasized the importance of the central measurements.

Lines 103-120: "**Compared to the southern and northern regions of the QTP, anthropogenic BC is the dominant driver of glaciers melt in the central QTP, whose radiative forcing can be up to 17 times greater than that in the southern QTP (Li et al., 2017; Ming et al., 2013). Moreover, a portion of the glaciers in central QTP are situated in the headwaters of the Yangtze River, thus, the melting directly impacts the livelihoods of millions of people downstream. The melt sensitivity of these glaciers to light-absorbing impurities (such as OA, BC and mineral dust) is not only higher than that of their southern counterparts but also carries greater regional ecological and social significance (Li et al., 2017). Although BrC is an important light-absorbing OA, limited studies on its variation and sources have been conducted in the QTP, and most have focused on the edge of the southern region (Wang et al., 2019a; Zhang et al., 2021; Tian et al., 2023). …… However, OA in the central QTP not only originated from long-range transport (Xu et al., 2018), but anthropogenic activities also have a significant impact on the OA sources, such as local biomass burning and fossil fuels (Xiang et al., 2024). Field measurements in the central QTP can improve our understanding of how human activities in remote regions impact light-absorbing aerosols and, consequently, glacial melting and radiative forcing.**"

Clarification of reported forcing contribution

R2.2: Line 25: the authors state that BrC contribute to 12-50% of the total positive radiative forcing. It would improve clarity if they would specify whether it is the total human-induced radiative forcing (which is generally dominated by greenhouse gases), or the positive radiative forcing related just to atmospheric particles (I suspect this is what they meant).

A2.2: Here, total positive radiative forcing represents the positive radiative forcing related just to atmospheric aerosols, more precisely, it represents light-absorbing atmospheric aerosols. To improve clarity in the manuscript as suggested by the reviewer, we modified the sentences by adding "from light-absorbing atmospheric aerosols" as follows:

Revised sentence (Lines 34-36): "**Current global model simulation studies have demonstrated that the impact of BrC can contribute 12–50 % of the total positive radiative forcing from light-absorbing atmospheric aerosols and can be regionally**

different, …"

Terminology

R2.3: Line 31: Clarify "non-fossil biomass burning," which is not a standard term. Rephrase the following sentence so it does not imply that fossil-fuel OA lacks any absorptive properties.

A2.3: Thanks for the reviewer's suggestion. We replaced "non-fossil biomass burning" with "biomass burning". And we replaced the statement of "and the OA from fossil fuel is non–absorbing" with "and the absorption capacity of BrC from fossil fuel is often neglected (Saleh, 2020)".

Revised sentence (Lines 40-42): "**Over the past decade, primary BrC was found to be dominated by biomass burning, and the absorption capacity of BrC from fossil fuel is often neglected (Saleh, 2020).**"

Consistency in site naming and values

R2.4: Line 127: Confirm whether Guangzhou should be labeled (GIG) rather than (GZ).

A2.4: GIG is the sampling site, GZ is the city where the campaign was carried out. We replaced "GZ" with "GIG" as the reviewer's suggestion in lines 160. The revised sentence is shown in the following:

Line 160: "**Figure 1. (a) Map of the locations of Yangbajing (YBJ) and Guangzhou (GIG) sites (yellow triangles).**"

R2.5: Lines 545–549: Resolve the discrepancy between the reported values of 21.4 W g^{-1} and 19.2 W g^{-1} for the YBJ site.

A2.5: The integrated total simple forcing efficiency (SFE) of BrC across the 370–660 nm wavelength range was 21.4 W g^{-1} at the YBJ site, while the SFE values for POA were 19.2 W g^{-1}. Specifically, 21.4 W g^{-1} for the YBJ site represents the total SFE values (from total OA absorption), including BBOA, HOA, LO-OOA and MOOOA, while 19.2 W g^{-1} represents the SFE values of POA, only including BBOA and HOA. To avoid any misunderstandings regarding of the total SFE values, we have added the following explanations:

Line 602 "**In contrast, the integrated total SFE (from total OA absorption) across the 370–660 nm wavelength range was 21.4 W g^{-1} at the YBJ site …**"

Figures

R2.6: Figure 5e: Clearly explain the notation "×6." If it indicates scaling for LO-OOA and MO-OOA, state this explicitly. A logarithmic axis could be considered as an alternative.

A2.6: The notation "×6." in the Figure 5e indeed indicates scaling for LO-OOA and

MO-OOA in order to better show the trend of diurnal patterns of LO-OOA and MO-OOA. We have added the following explanation to the caption of Figure 5 in the original manuscript:

Lines 436-437: "**The "x6" indicates the light absorption of LO-OOA and MO-OOA at 370 nm expanded by 6 times to better guide the eyes for the trend of diurnal patterns.**"

R2.7: Figure 8: Explain why LO-OOA and MO-OOA are absent from panel (b), both in the legend and the text.

A2.7: For GIG site, LO-OOA and MO-OOA are absent in Fig. 8b, which was because their BrC light absorption at GIG site was not resolved out in the GIG dataset via multiple linear regression (MLR) analysis, as shown in Fig. 5d. We have considered the contribution of the intercept, i.e., the "Unidentified" in the panel were in the original manuscript. We already provided a detailed discussion on the selection of the MLR scheme and the uncertainty assessment process in the methodology section (Section 2.5.1)."

Editorial Suggestions

Rephrase for clarity and grammar at Lines 25, 44–45, 66, 93–94, 96, 264–265, 286, 356, 357, 425–426, 442, 513, 529, and 589–591.

R2.8: Line 25: "can contributing" – the authors should rephrase this.

A2.8: We modified the sentences by replacing "can contributing" with "can contribute" as follows:

Revised sentence (Lines 34-36): "**Current global model simulation studies have demonstrated that the impact of BrC can contribute 12–50 % of the total positive radiative forcing from light-absorbing atmospheric aerosols and can be regionally different, …**"

R2.9: Lines 44-45: "also found to be important BrC sources as well" – the authors should rephrase this in order to avoid repetition (also – as well).

A2.9: We modified the sentences by deleting "as well" as follows:

Lines 57-59: "**Aqueous reactions of carbonyl groups with reduced nitrogenous organic compounds, such as organic amines and ammonium, are also found to be important BrC sources (Powelson et al., 2014; Tang et al., 2022).**"

R2.10: Line 66: The term BC was not defined yet (I assume it is black carbon).

A2.10: Yes, the term BC is black carbon, we have added the definition as follows:

Line 82: **"Due to the complexity and diversity of BrC, in conjunction with black carbon (BC), …**"

R2.11: Lines 93-94: "despite the online BrC data was reported…" – check grammars, probably need to rephrase it.

A2.11: We replaced "despite" with "although" and replaced "seldomly" with "seldom" as follows:

Lines 126-127: "**In addition, although online BrC data have been reported for the Tibet region, the dynamic variation of BrC and its source contributions are seldom shown.**"

R2.12: Line 96: "investigation on the dynamic…" – check grammars, probably need to rephrase it.

A2.12: We replaced "investigation on" with "investigation of" as follows:

Lines 129-130: "**The investigation of the dynamic variation of BrC, e.g., diurnal variation, can greatly promote the understanding of BrC fate in the ambient air, which shall be further studied.**"

R2.13: Lines 264-265: "We used a […] can be expressed as follows" – needs rephrasing.

A2.13: We reorganized the sentence structure to make the logic clearer.

Revised sentence (Lines 305-307): "**In this study, we used a modified version of the wavelength-dependent SFE without considering mass scattering and expressed it as follows…**"

R2.14: Line 286: "The campaign-averaged of…" – needs rephrasing.

A2.14: "Campaign-averaged" is an adjective, which should directly modify nouns and cannot be followed by "of". We modified the sentences by deleting "of" as follows:

Lines 328-331: "**The campaign-averaged BrC light absorption coefficients (Abs_{BrC}) and their contributions to total aerosol light absorption increased with decreasing wavelength at both sites (AAE of BrC is 2.6 for YBJ and 3.2 for GIG; Figs. 2c, 3b, and 3d), indicating strong BrC light absorption at shorter wavelengths.**"

R2.15: Lines 311-312: Provide reference.

A2.15: Figure 2 can explain this sentence and the corresponding references can be found in Table S4. We have added figure citation "(Fig. 2d)" as follows:

Lines 357-358: "**In general, BrC light absorption in urban areas tends to be higher or comparable to that in QTP regions due to typically higher OA mass concentrations (Fig. 2d).**"

R2.16: Line 356: "where enhanced at 10:00" – needs rephrasing.

A2.16: We check the entire sentences in the manuscript, and found the clause

introduced by "where" lacks a subject, and the usage of "enhanced" in this context is also not quite appropriate. We have reorganized the entire sentence and made the following revisions.

Revised sentence (Lines 401-404): **"The diurnal patterns of $Abs_{BC,370}$ and $Abs_{BrC,370}$ at the GIG site, which showed an enhancement at 10:00 and a stronger peak at 21:00, were slightly different from those at the YBJ site. This difference suggests distinct source emissions or secondary formation patterns between the two locations"**

R2.17: Line 357: "for each study"? – aren`t these two sites part of this same study? Maybe the authors meant something else?

A2.17: The statement of "for each study" is indeed misleading because the two measurements were carried out simultaneously at different sites in the same study. We replaced "for each study" with "between the two locations" The revised sentence is shown in the following. The A2.16 and A2.17 are shown in the same revised sentences.

Revised sentence (Lines 401-404): **"The diurnal patterns of $Abs_{BC,370}$ and $Abs_{BrC,370}$ at the GIG site, which showed an enhancement at 10:00 and a stronger peak at 21:00, were slightly different from those at the YBJ site. This difference suggests distinct source emissions or secondary formation patterns between the two locations"**

R2.18: Lines 425-426: This sentence seems incomplete, and there is a dot in the middle.

A2.18: I have noticed several obvious issues in this sentence. We modified the sentences as follows:

Lines 475-477: "**Especially for the Guangzhou samples, BBOA from regional transport was likely subjected to a longer oxidation process, which led to a lower MAC at the GIG site.**"

R2.19: Line 442: "from the direct distance of the…" – needs rephrasing

A2.19: We restructured the sentence to make the expression more accurate as follows:

Lines 494-495: "**Our observation site is located less than 1 km (straight-line distance) from the G6 Beijing–Tibet Expressway.**"

R2.20: Line 513: "due to soluble Brc was applied" – needs rephrasing

A2.20: We changed "soluble BrC was applied" to be "the application of soluble BrC", the revised sentences now read as:

Lines 568-569: "**Note that the offline studies may underestimate the light absorption contribution of POA due to the application of soluble BrC…"**

R2.21: Line 529: "and a better validate the model simulation" – needs rephrasing

A2.21: We revised the sentence as follows:

Lines 583-585: "**…, it is essential to conduct region–specific and season–specific observational research for a better understanding of the BrC sources and to better validate model simulations.**"

R2.22: Line 552: For the "BrC-specific" SFE, maybe? Because BC seems to be the dominant factor, making it impossible for BBOA and HOA to represent 80% of the overall SFE

A2.22: Thanks for the reviewer's suggestion. Specifying "BrC-specific SFE" will indeed be more accurate. We replaced "SFE" with "BrC-specific SFE" as follows:

Lines 608-609: "**POA, including BBOA and HOA, contributed significantly to the total BrC light absorption, accounting for over 80 % of the BrC-specific SFE in this study.**"

R2.23: Lines 589-591: "necessitating… shall be considered…" – needs rephrasing

A2.23: We revised the sentence as follows:

Lines 645-648: "**Notably, traffic-related BrC contributions remain substantial in Guangzhou and even in the Tibet background, which necessitates that explicit light absorption parameters for fossil-derived BrC (e.g., from vehicle emissions) be considered in model simulations."**

Anonymous Referee #3

General Comments

This manuscript presents interesting field measurements in the Qinghai–Tibet Plateau (Yangbajing) and urban Guangzhou during July 2022 with established measurement techniques including multi-wavelength Aethalometer and online aerosol mass spectrometer (AMS and ACSM) measurements to quantify black carbon (BC), brown carbon (BrC), and organic aerosols. Source apportionment (positive matrix factorization, PMF) and multilinear regression analysis were conducted to provide further insights on BrC source-specific mass absorption cross-sections (MAC), and a simple forcing efficiency (SFE) for individual OA factors. This work clearly falls within the scope of ACP, focusing on aerosol light absorption properties, radiative forcing, and aerosol composition. In addition, the manuscript provides a valuable comparison between a polluted city environment and a "pristine" environment (i.e., YBJ site), for which very few datasets have been reported so far.

However, the Method sections should include more technical details to further support the results and interpretation. And some results should be discussed further to extract more value from the dataset hence to improve the manuscript's relevance and importance. I recommend publication after the points outlined below are addressed.

R3.1: A few general comments. It would be useful to provide a broader discussion of the impact of BrC on radiative forcing in both regional and global contexts. There are some comments in the introduction and conclusion sections but they are rather scattered. This could be difficult for readers not familiar with aerosol speciation and its connection to regional BrC.

A3.1: Follow reviewer's suggestions, we provided a more detailed description of light absorbing aerosols, and also supplemented the impact of BrC on regional and global radiative forcing in the introduction.

Lines 22-31 "**Light absorbing components of atmospheric aerosols comprise black carbon (BC) and light absorbing organic aerosols, known as brown carbon (BrC). The BrC exhibits significant absorption in the near-ultraviolet (300–400 nm) and visible ranges (Andreae and Gelencsér, 2006; Kirchstetter and Thatcher, 2012; Laskin et al., 2015), but it remains largely overlooked in most climate models (Chung et al., 2012). Feng et al. (2013) found that accounting for the strong absorption of BrC in model could shift the global mean direct radiative forcing of OA at the top of the atmosphere from a cooling of −0.08 Wm^{-2} to a warming of +0.025 Wm^{-2}, emphasizing the significant role of BrC in global and regional direct radiative forcing of carbonaceous aerosols. Wang et al. (2025) revealed that dark BrC, emitted by wildfires and agricultural burning, strongly absorbs solar radiation, thereby generating a radiative effect of +0.02 to +0.68 Wm^{-2}, which contributed comparable warming as BC.**"

R3.2: SP-AMS was referred in Section 2.3 (L154) and SI. However, insufficient operational information was provided. With the results and information provided by the SI, I would assume the instrument was operated with tungsten vaporizer. In that regard, the instrument is typically referred to as the "high-resolution aerosol mass spectrometer (HR-AMS or HR-ToF-AMS)". If dual vaporizer mode was used, please provide the relevant details, as this would also affect the interpretation of the PMF results (e.g., refractory BC should be included in the PMF analysis).

A3.2: In this study, the SP-AMS was operated alternately in two sampling modes: tungsten-only vaporizer (TV) or dual-vaporizer (DV). However, the data for the PMF analysis in this study originated exclusively from the tungsten-only vaporizer mode. And the data for dual-vaporizer mode did not provide more useful information on the source apportionment of OA for current analysis, which can be found more details in the response to R3.4. We added brief operational information in the manuscript and more text in the supporting information (Text S1) to explain the AMS operation mode applied in this study.

Lines (195-197): "**For SP-AMS, dual vaporization mode and tungsten-only V mode were alternatively applied every 4 minutes. In this study, tungsten-only mode, which resembled the traditional HR-ToF-AMS, was applied.**"

Supporting information (text S1): "**SP-AMS is based on the design of a high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) (Decarlo et al., 2006), while SP-AMS has an additional Nd-YAG intra-cavity infrared (IR) laser module at the wavelength of 1064nm (Onasch et al., 2012). In this study, the SP-AMS at YBJ site was operated at 4 min alternating intervals between tungsten-only vaporizer mode (TV mode) and dual-vaporizer mode (DV mode), implemented by alternately activating (IR laser-on, i.e., dual vaporizers) and deactivating the laser vaporizer (IR laser-off, i.e., tungsten vaporizer only). In TV mode, the SP-AMS performs identically to the HR-ToF-AMS, providing continuous high-resolution mass concentration and mass spectral data for non-refractory submicron particulate matter (NR-PM_1). In DV mode, simultaneous use of both vaporizers enables real-time quantification of refractory black carbon (rBC) and NR-PM_1. rBC absorbs strongly at 1064nm and consequently heats up to ~ 4000 °C for rBC vaporization (Onasch et al., 2012). The term rBC is operationally defined, referring to BC particles detected by SP-AMS. However, the data for the PMF analysis in this study originated exclusively from the tungsten-vaporizer mode (the data for dual-vaporizer mode will be discussed in Liang et al. in preparation). Thus, the positive matrix factorization (PMF) was performed on the high-resolution mass spectra of organic aerosol (OA) of the SP-AMS operated with tungsten-only vaporizer at the YBJ site.**"

For AMS and ACSM PMF analysis, only limited justification is provided in the SI and I have several questions:

R3.3(a): For the AMS PMF, In Fig. S2a, the Q/Q_{exp} dropped below 1.0 after the 2-factor solution. Could you provide more justification for factor separation?

A3.3(a): Figure S2 is the standard determination panel for separating the PMF factors, as suggested by (Zhang et al., 2011). The sharp decrease of Q/Qexp suggest there are mass was not resolved out when 2-factor solutions were chosen. Values of $Q/Q_{exp} >> 1$ indicate either an underestimation of the errors or the presence of variability in the factor mass spectra that cannot be adequately modeled by the sum of the given number of factors. $Q/Q_{exp} << 1$ indicates overestimation of the errors of the input data (Ulbrich et al., 2009). In this study, $Q/Q_{exp} = 0.82$ under the final PMF result of 5-factor solution is within an acceptable range. To provide more justification of the factor separation, we added a table (Table S1), Fig. A3.3(a) and Fig. A3.3(b) to demonstrate how the PMF factor was resolved in the YBJ dataset. In general, the 5-factor solution is the optimum solution. For 4-factor solution, we found mixed factor between BBOA and Biofuel OA (Fig. A3.3(a)). For 6-10 factor solution, we found there is an extra factor with unexplained peak (m/z 77) in their spectrum as shown in Fig. A3.3(b). We have provided the justification for factor separation in the supporting information (text S1) as follows:

Supporting information (text S1): "**The sharp decrease of Q/Q_{exp} suggest there are mass was not resolved out when 2-factor solutions were chosen. Values of $Q/Q_{exp} >> 1$ indicate either an underestimation of the errors or the presence of variability in the factor mass spectra that cannot be adequately modeled by the sum of the given number of factors. $Q/Q_{exp} << 1$ indicates overestimation of the errors of the input data (Ulbrich et al., 2009). In this study, Q/Q_{exp} =0.82 under the final PMF result of 5-factor solution is within an acceptable range. Table S1 summarized the descriptions of PMF solutions. In general, the 5-factor solution is the optimum solution. For 4-factor solution, we found mixed factor between BBOA and Biofuel OA. For 6-10 factor solution, we found there is an extra factor with unexplained peak (m/z 77) in their spectrum.**"

Table S1. Descriptions of PMF solutions obtained at YBJ site and GIG site.

Factor number	**Fpeak**	**Q/Q_{exp}**	**Solution Description from Free PMF (YBJ site)**
1-3	0	1.4 to 0.93	Too few factors, large residuals at time periods and key m/z's. Q/Q_{exp} decreases very fast.
4	0	0.9	Except MO-OOA, LO-OOA and HOA, the characteristics of one factor is not clear. It seems that BBOA mixed with Biofuel OA.
5	**0**	**0.82**	**Optimum choices for PMF factors (MO-OOA, LO-OOA, HOA, BBOA and Biofuel OA). Time series and diurnal variations of PMF factors are consistent with the external tracers. The spectra of 5 factors are consistent with the source spectra in AMS spectra database.**
6	0	0.8	showed over-split factors without an explicit physical meaning
5	-1 to 1	0.873 to 0.892	Under the 5-factor solution, the factor contributions obtained by different fpeak are relatively stable, clarifying the stability and interpretability of the five-factor solution.
Factor number	**Fpeak**	**Q/Q_{exp}**	**Solution Description from free PMF (GIG site)**
1-3	0	1.96-1.21	Too few factors, large residuals at time periods and key m/z's. Q/Q_{exp} decreases very fast.
4	0	1.15	Too few factors. Factors are mixed based on the time series and spectra.
5	0	1.1	Relatively reasonable profiles and time series under a fully unconstrained condition. However, BBOA was still mixed in other factors.
6	0	1.01	Factor split, e.g., COA was split into

			two factors with similar spectra, however, different time series.
5	-1 to 1	1.104 to 1.112	Under the 5-factor solution, factor MS and time series are nearly identical and the factor contributions obtained by different fpeaks are relatively stable, clarifying the stability and interpretability of the five-factor solution.
Factor number	**a value**	**Q/Q_{exp}**	**Solution Description from ME2 (GIG site)**
5	**a value: 0**	**1.05**	**Optimum choices for PMF factors (MO-OOA, LO-OOA, HOA, BBOA and COA) resolved by ME2. Time series and diurnal variations of PMF factors are consistent with the external tracers. The spectra of 5 factors are consistent with the source spectra in AMS spectra database.**
5	a value: 0 to 0.8	1.03 to 1.05	Under the 5-factor solution, factor MS and time series are nearly identical under different a value (Fig. S3)

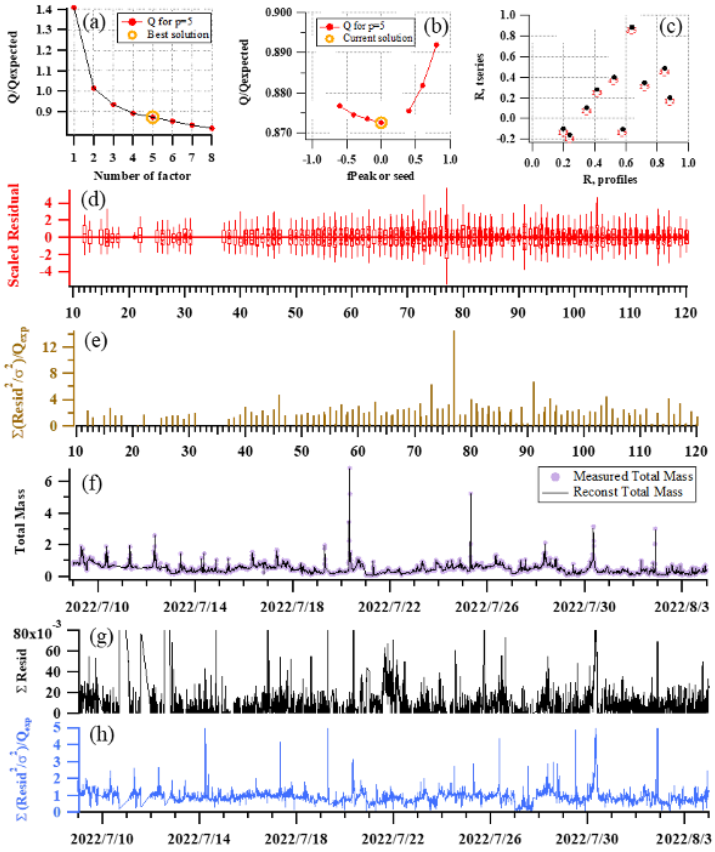


Figure S2. Diagnostics plots of factor selections in the unconstrained PMF at the YBJ site.

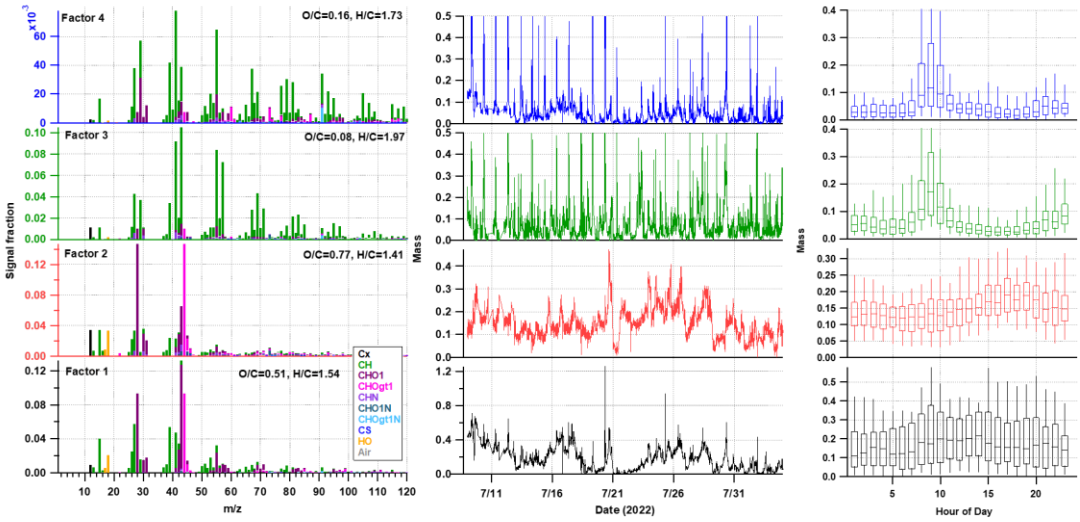


Figure A3.3(a). PMF result of 4 factors at YBJ site.

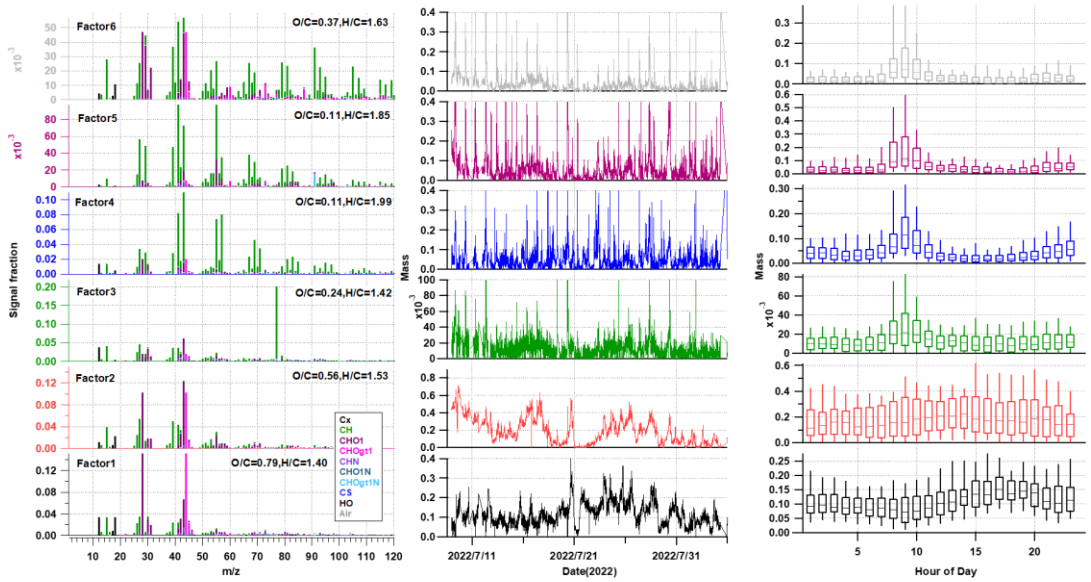


Figure A3.3(b). PMF result of 6 factors at YBJ site.

R3.3(b) All of your measurements peak at 0800 but not 2200 LT while the above 3 factors have similar diurnal profile, and some overlapping spectra. Could you provide more evidence?

A3.3(b): Following the above question, we regard reviewer ask us to provide more evidences for separating the three POA factors in 5-factor solution. We show evidences below:

1) For the diurnal profiles: Despite the three POA factors, i.e., HOA, BBOA, and Biofuel OA show similar peak in the morning, their night peaks are different with HOA peaked later an hour than BBOA and Biofuel OA, supporting their different origins based on their time series (e.g., the Pearson correlation coefficient between BBOA and HOA is only 0.41 (Fig. S6a)).

2) For the mass spectra: A) scatter plots showed correlation coefficients between BBOA and HOA, Biofuel-OA and HOA, BBOA and Biofuel-OA were 0.52, 0.85 and 0.64, respectively (Fig. S5), suggesting the BBOA spectra shows distinct differences with HOA and biofuel-OA, however, biofuel-OA and HOA have similarity; B) despite the similarity between biofuel-OA and HOA, we can still distinguish them based on their relatively unique and abundant "tracer ions". We added the Table S2 to show the main mass spectra characteristics of PMF factors. The most obvious difference between Biofuel-OA and HOA lies in the fact that Biofuel-OA has an obvious higher ratio of m/z 55/57 (2.6) than that of HOA (with m/z 55/57 = 0.9) (Zhang et al., 2011), as well as the higher m/z 60 ($C_2H_4O_2^+$) and 73 ($C_3H_5O_2^+$) signals. Different mass spectra characteristics prove that HOA and biofuel OA have significantly different sources. We also have added more information in the Supporting information (text S1) as follows:

Supporting information (text S1): "**Despite the three POA factors, i.e., HOA, BBOA, and Biofuel OA show similar peak in the morning, their night peaks are different with HOA peaked later an hour than BBOA and Biofuel OA, supporting their different origins based on their time series (e.g., the Pearson correlation coefficient between BBOA and HOA is only 0.41 (Fig. S6a)).**

For the POA factors mass spectra, scatter plots showed correlation coefficients between BBOA and HOA, Biofuel-OA and HOA, BBOA and Biofuel-OA were 0.52, 0.85 and 0.64, respectively (Fig. S5), suggesting the BBOA spectra shows distinct differences with HOA and biofuel-OA, however, biofuel-OA and HOA have similarity. Despite the similarity between biofuel-OA and HOA, we can still distinguish them based on their relatively unique and abundant "tracer ions". As shown in Table S2, the most obvious difference between Biofuel-OA and HOA lies in the fact that Biofuel-OA has an obvious higher ratio of m/z 55/57 (2.6) than that of HOA (with m/z 55/57 = 0.9) (Zhang et al., 2011), as well as the higher m/z 60 ($C_2H_4O_2^+$) and 73 ($C_3H_5O_2^+$) signals. Different mass spectra characteristics prove that HOA and biofuel OA have significantly different sources."

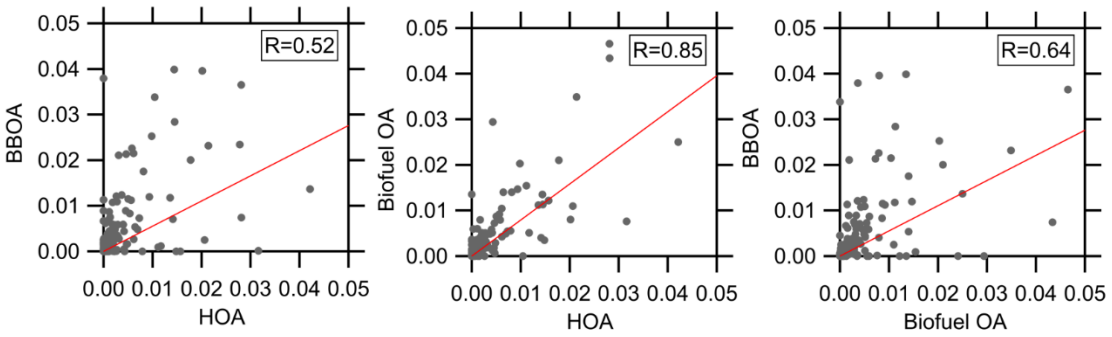


Figure S9. Mass spectra scatter plots among BBOA, HOA and Biofuel-OA at YBJ site.

Table S2. The main mass spectra characteristics of PMF factors at YBJ site and

GIG site.

Factor	Main mass spectra characteristics (YBJ site)
HOA	dominated by alkyl fragments ($C_nH_{2n+1}^+$ and $C_nH_{2n-1}^+$). The O/C ratio of HOA in this study was 0.11, suggesting its fresh property. The tight correlation between HOA versus BC and $C_4H_9^+$ (R = 0.53 and 0.92; Fig. S6a) also indicated that the source of HOA was from traffic emissions.
BBOA	characterized by an obvious peak at m/z 60 ($C_2H_4O_2^+$) and 73 ($C_3H_5O_2^+$) signals in the MS (Fig. S4), which are usually considered as a recognized tracer emitted from biomass burning.
Biofuel OA	characterized by the highest m/z 55 ($C_3H_3O^+$) signal and a higher ratio of m/z 55/57 (2.6) compared with HOA (0.9) (Zhang et al., 2011), as well as the m/z 73 ($C_3H_5O_2^+$) signal. The time series of biofuel OA component showed a close correlation with emissions of tracking ions fragments $C_3H_3O^+$ (R=0.8) and $C_6H_{10}O^+$ (R=0.93) (Fig. S6), which were also highly correlated with the emissions of biomass burning fragments $C_2H_4O_2^+$ (R=0.93). These findings demonstrated that this factor was associated with emissions of biomass burning and cooking.
LO-OOA	characterized by high peaks at m/z 44 (mostly CO_2^+) and had a larger peak at m/z 43 (mostly $C_2H_3O^+$) than MO-OOA as well. LO-OOA had a lower O/C (0.55) than MO-OOA. There is a good correlation between the LO-OOA factor time series and the characteristic ion fragments $C_2H_3O^+$ and $C_3H_3O^+$ with R of 0.95 and 0.75 (Fig. S6a), respectively.
MO-OOA	characterized by high peaks at m/z 44 (mostly CO_2^+) and the time series was consistent with sulfate (R=0.74; Fig. S6a). MO-OOA had the highest O/C (0.78) and the lowest H/C (1.40), indicating a high oxidation degree of this factor

Factor	Mass spectra characteristics (GIG site)
HOA	dominated by alkyl fragments ($C_nH_{2n+1}^+$ and $C_nH_{2n-1}^+$). HOA displayed a bimodal distribution with a moderate morning peak at 07:00 and a stronger evening peak at 20:00 (Fig. S4), consistent with typical urban HOA diurnal patterns.
BBOA	characterized by an obvious peak at m/z 60 and 73 signals and showed different diurnal variation with enhanced afternoon and nighttime peaks

	likely reflect regional transport of biomass burning emissions from agricultural activities in the Pearl River Delta region during summer.
COA	characterized by the highest m/z 55 signal and a higher ratio of m/z 55/57 compared with HOA. The diurnal variation showed three notable peaks (7:00, 12:00 and 19:00), corresponding to breakfast time, lunchtime, and dinner time.
LO-OOA	characterized by a high m/z 44 signal but lower than that of MO-OOA, indicating its relatively fresh features.
MO-OOA	characterized by high peaks at m/z 44. The concentration of MO-OOA showed a better correlation with that of sulfate (R = 0.87) than with nitrate (R = 0.7) (Fig. S6b), which is likely attributed to similarly high oxidation degrees of both MO-OOA and sulfate.

R3.4: Following the above question, rBC and metal detected by the dual vaporization mode can help separate sources further (Bibi et al., 2021; Ma et al., 2025; Rivellini et al., 2020). It is relevant for the context since the HOA and BBOA have very similar diurnal profile. If the instrument measured with dual vaporization mode, PMF should be evaluated with refractory components.

A3.4: We have performed the PMF analysis of OA, rBC, and metals (Na^+、Mg^+、K^+、Cu^+、Zn^+、Rb^+) of dual vaporization measurements (DV-mode) (Ma et al., 2025; Bibi et al., 2021; Török et al., 2018). 6-factors were resolved out under DV-mode, including Biofuel OA (equivalent to BBOA + Biofuel OA of tungsten vaporizer mode), HOA, LO-OOA and MO-OOA, as shown Fig. A3.4(a). In addition to the 5-factor resolved out in the conventional V-mode of tungsten vaporizer, the extra two factors were referred as metal and rBC factors. The left 5 OA factors show exactly the same time series and mass spectrum to the conventional V-mode ($R > 0.75$ on time series; $R > 0.94$ for mass spectrum, as shown in Fig. A3.4(b)). This suggests the data of DV mode cannot help to resolve more OA sources. The DV-mode data also confirm the validity of 5-factor applied with traditional only-V mode data. The validity of separation among HOA, BBOA and biofuel OA factors are already explained in A3.3, which will not be repeated here.

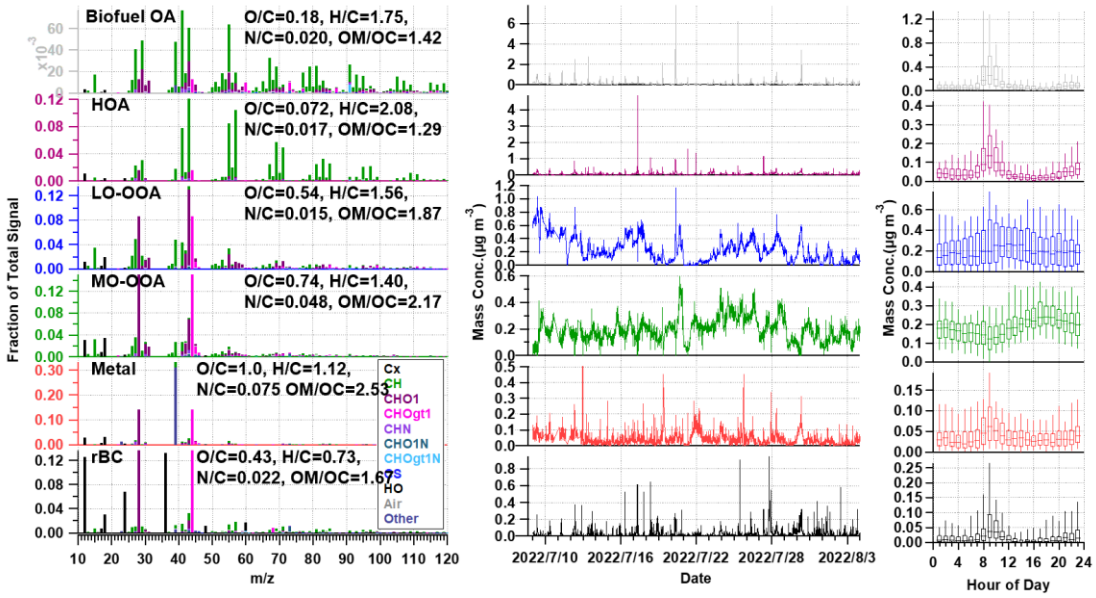


Figure. A3.4(a): PMF Result (6 factors) of OA, rBC, and metals of dual vaporization measurements at YBJ site.

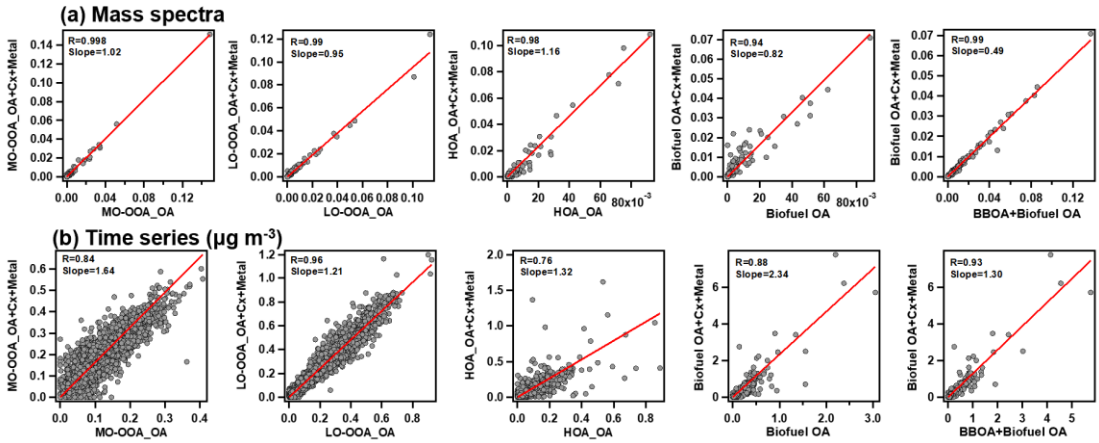


Figure. A3.4(b): Time series and mass spectra scatter plots of each PMF factor in DV mode and TV mode.

R3.5: For Fig. S4a, it is somewhat confusing to label peaks using m/z values since they are high-resolution PMF results.

A3.5: The mass spectrum was plotted in the form of UMR. We used the m/z values to label peaks in order to more quickly locate the m/z position of the mass spectra and intuitively distinguish the characteristic ions for each factor. And each peak was colored on the basis of the contributions of 8 ion categories: Cx, $CxHy^+$, $CxHyO1^+$, $CxHyOz^+$, $CxHyNp^+$, $CxHyO1Np^+$, $CxHyOzNp^+$, HxO^+, which conforms to the traditional AMS mass spectrum standards (Canagaratna et al., 2007; Ng et al., 2011; Ng et al., 2010; Sun et al., 2011). To enhance readability, we have added the following explanations to the caption of Fig. S4 as follows:

Revised caption: "**Figure S8. The final optimum solution for OA sources at the**

YBJ site and the GIG site. The mass spectrum was plotted in the form of UMR, and each peak was colored on the basis of the contributions of 8 ion categories: C_x^+, $C_xH_y^+$, $C_xH_yO_1^+$, $C_xH_yO_z^+$, $C_xH_yN_p^+$, $C_xH_yO_1N_p^+$, $C_xH_yO_zN_p^+$, H_xO^+."

R3.6: For the ACSM PMF, please provide the rationale for constraining BBOA.

A3.6: For UMR spectrum, multiple studies show that the BBOA and COA is hard to separate with unconstrained-PMF (Zhang et al., 2019). Especially, when the BBOA contribution is less than 10%, their good separation in the unconstrained-PMF is unachievable. However, we know that the GIG sampling site was impacted by the biomass burning from surrounding areas based the results from previous study (Cai et al., 2023; Jiang et al., 2021; Jiang et al., 2022**)**. In addition, in Fig. 1 we show fire spots data during our observation period. The back trajectory data indicates our observation site might impacted by the fire plumes from surrounding. To resolve out the contribution of BBOA, ME-2 was applied here. Previous study proved that using the standard BBOA spectrum, the BBOA can be efficiently resolved out in the UMR dataset (Crippa et al., 2014; Lanz et al., 2007). We have added explanations in the Supporting information (text S1) as follows:

Supporting information (text S1): "**For UMR spectrum, multiple studies show that the BBOA and COA is hard to separate with unconstrained-PMF (Zhang et al., 2019). Especially, when the BBOA contribution is less than 10%, their good separation in the unconstrained-PMF is unachievable. However, we know that the Guangzhou sampling site (GIG site) was impacted by the biomass burning from surrounding areas based the results from previous study (Cai et al., 2023; Jiang et al., 2021; Jiang et al., 2022). In addition, in Fig. 1 we show fire spots data during our observation period. The back trajectory data indicates our observation site might impacted by the fire plumes from surrounding. To resolve out the contribution of BBOA, Multilinear Engine 2 (ME-2; SoFi 6.8) (Canonaco et al., 2013) was applied here. Previous study proved that using the standard BBOA spectrum, the BBOA can be efficiently resolved out in the UMR dataset (Crippa et al., 2014; Lanz et al., 2007).**"

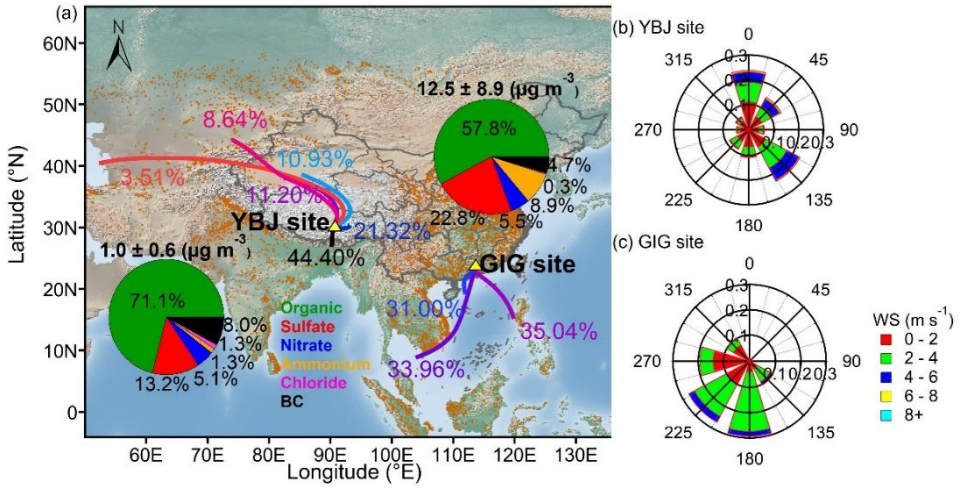


Figure 1. (a) Map of the locations of Yangbajing (YBJ) and Guangzhou (GIG) sites (yellow triangles). Solid colored lines represent the average back trajectory clusters during the whole campaign and the corresponding contributions plotted using the MeteoInfo version 2.2.6 developed by Wang (2019) (download from *http://www.meteothink.org*, last access time: 23 June 2025). The orange dots on the map indicate the location of the fire spot (download from *Active Fire Data Earthdata (nasa.gov)*, last access time: 14 December 2023). The pie charts represent the chemical compositions of submicron particulate matter (PM_1) along with their contributions at both sites during this campaign. The rose plots colored by wind speed (WS) at (b) the YBJ site and (c) the GIG site are also shown.

R3.7: Changes in final PMF factors will affect MLR analysis.

A3.7: Yes, changes in final PMF factors will affect MLR analysis. There are two conditions on this: (1) changing the PMF factor number and types will lead to variation of MLR result; (2) the multicollinearity among PMF factors time series also will affect the MLR analysis. We already considered this in the section 2.5.1 and Text S2. We considered multiple schemes and evaluated the uncertainty of the MLR model brought by each scheme. Eventually, we selected PMF factors with low multicollinearity and strong correlation with BrC light absorption as the input data for the MLR analysis.

R3.8: For the assumption of AAE_{BC} value, could you elaborate more on the decision to use 0.8-1.2 range? To be specific, L203 stated "In this study, the value of AAE 1.4 lead to most of the BrC light absorption coefficient to be negative values…" However, some studies have shown that thermal denuded BC_{AAE} values can be higher than 1.4 (e.g., Török et al., 2018). I may miss the justification of BrC existence at both sites.

A3.8: Although Török et al. (2018) found AAEs of soot particles were between 1 and 3.5, the high AAE was mainly due to the high mass-fractions of organic carbon (OC) and pyrolytic carbon (PC), which mixed with black carbon (BC). In other words, the AAE of soot particles (mixture of BC and OC), cannot reflect the AAE values for pure BC (AAE_{BC}, BC has no contribution from OC). For pure BC, AAE_{BC} was suggested as

0.8–1.4 (Lack and Langridge, 2013; Kasthuriarachchi et al., 2020; Zhai et al., 2022; Corr et al., 2012). We summarized the ambient AAE_{BC} values from the literatures, as shown in Table S3, and we could find all the actually used values are between 1 – 1.18 (Sun et al., 2021; Kasthuriarachchi et al., 2020; Tian et al., 2023; Wang et al., 2019b; Xie et al., 2019; Zhang et al., 2021; Zhang et al., 2022). E.g., within one campaign (Kasthuriarachchi et al., 2020), 93% of directly measured AAE from ambient core BC in Singapore was found to be less than 1.1, as shown in Fig. A3.8(a). In addition, Lack and Langridge (2013) reported that the analysis of a range of atmospheric measurements of the AAE for aerosol sourced from fresh fossil fuel burning and urban pollution (where the dominant absorber was BC) shows an average value for the AAE of 1.1 ± 0.3 (1σ) and declare that use of an AAE = 1 is the common default. AAE_{BC} of 1.4 represents a typical high value, considering the mixing of non-absorbing material with BC in the extreme case (Lack and Langridge, 2013). For our study, we found ~82% of data was negative value when the AAE_{BC} of 1.4 was applied (Fig. A3.8 (b)), which is unrealistic to use in this study. Our total aerosol light absorption coefficients from 370 to 950nm is already low due to lower $PM_{2.5}$ mass concentrations at Yangbajing (YBJ site). Thus, the extreme value of 1.4 will not be used here. The AAE_{BC} of 0.8 to 1.2 was finally applied, which is also consistent with the summarized results applied in other field studies (1-1.18). We have made relevant additions to the manuscript as follows:

Revised sentence (Lines 238-243): "**We summarized the AAE_{BC} values from the literatures in Table S3, and found all the actually used values are between 1 – 1.18. E.g., within one campaign (Kasthuriarachchi et al., 2020), 93% of directly measured AAE from ambient core BC in Singapore was found to be less than 1.1. An AAE_{BC} of 1.4 represents a typical high value, considering the mixing of non-absorbing material with BC in the extreme case (Lack and Langridge, 2013). For our study, we found ~82% of data was negative value when the AAE_{BC} of 1.4 was applied, which is unrealistic to apply here.**"

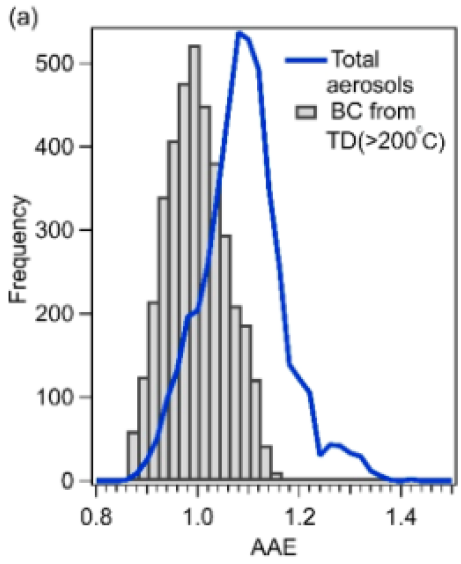


Figure A3.8(a): Histograms of AAE of thermal-denuded (TD) BC and total aerosols (Kasthuriarachchi et al., 2020).

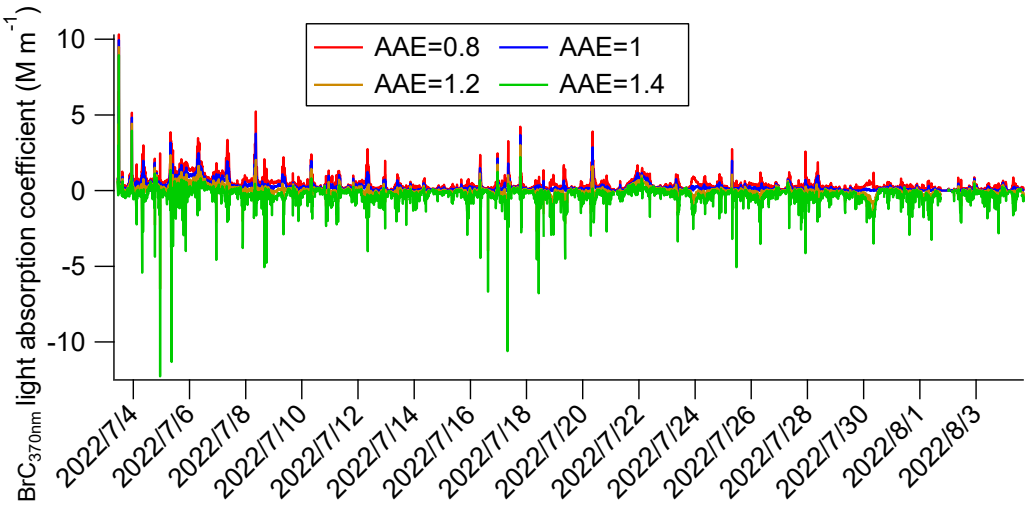


Figure A3.8 (b): Light absorption coefficients of BrC at 370nm using different AAE_{BC} at YBJ site.

Table S3. The AAE_{BC} values used to separate BC and BrC light absorption in other studies.

Observation sites	**AAE_{BC}**	**Reference**
Qomolangma Station	1.187	(Zhang et al., 2021)
Nam Co Station	1.086	
Waliguan Station	1.042	
Lhasa	1	(Zhu et al., 2017)

Lulang	1	
Gaomeigu	1.1	(Tian et al., 2023)
Xianghe	1.1	(Wang et al., 2019b)
Beijing 2016	1	(Xie et al., 2019)
Beijing 2020	1	(Sun et al., 2021)
Gucheng	1	
Xian	1	(Zhang et al., 2020)
Hong Kong	1	
Singapore	0.99 to 1.04	(Kasthuriarachchi et al., 2020)

Minor comments

R3.9: A few sentences could be reorganized to improve clarity, particularly at L38-41, L87-90, and L289-292.

A3.9: We have reorganized the sentences as follows:

Lines 49-52: "**This finding raises the question of how much primary fossil and non-fossil sources contribute to ambient BrC. Regarding secondary BrC, its formation involves complex gaseous, particulate, and liquid-phase reactions from diverse precursors (Laskin et al., 2015).**"

Lines 113-115: "**We found that cross-border transport of biomass burning from South Asia was responsible for a significantly higher BrC light absorption contribution in the southern QTP than in the central and northeastern regions.**"

Lines 331-335: "**At 370 nm, where BrC contributes the most to the total light absorption, Abs_{BrC} in Tibet (0.2 ± 0.3 M m^{-1}) was found to be a factor of 13 lower than that in Guangzhou (2.9 ± 2 M m^{-1}). This finding is consistent with the much lower OA mass concentrations (by a factor of ~10) at the YBJ site (OA: 0.7 ± 0.4 µg m^{-3} vs 6.9 ± 5.8 µg m^{-3} at GIG; Figs. 2b and 2c).**"

R3.10: L289: revise to … "where BrC contributes the most"

A3.10: We have revised the sentence as shown in A3.9 (Lines 331-335).

R3.11(a): L 445-448. Do you mean some of the HOA is transported? It is interesting the se, at YBJ site, HOA increased at night time while NO stay flat or decreased.

A3.11(a): Yes, the HOA source during the night was more regional than that in the morning, which was supported by the fact that the NO mass concentration was only enhanced during the morning but not during the night, while an obvious NO_2 peak at both periods was observed (Fig. 4a). During the observation period at the YBJ site, both the wind speed and wind direction showed regular changes throughout the day (Figs. S11 and S13). Combining the Rose plots and Bivariate polar plots (Fig. S12), HOA was affected by the wind direction and wind speed, with the morning peak affected by the north and southeast traffic emissions plumes, while the evening peak was affected by the southeast and northeast traffic emissions plumes with stronger wind speed. Combining with the high wind speed and the low NO concentration, we speculate that the source of HOA at night has a more regional nature. We have added additional information in the manuscript as follows:

Lines 501-503: "**Combining the Rose plots and Bivariate polar plots (Fig. S12), the evening peak of HOA was also affected by the southeast and northeast traffic emissions plumes with stronger wind speed, which also supported the regional nature of evening HOA peak**"

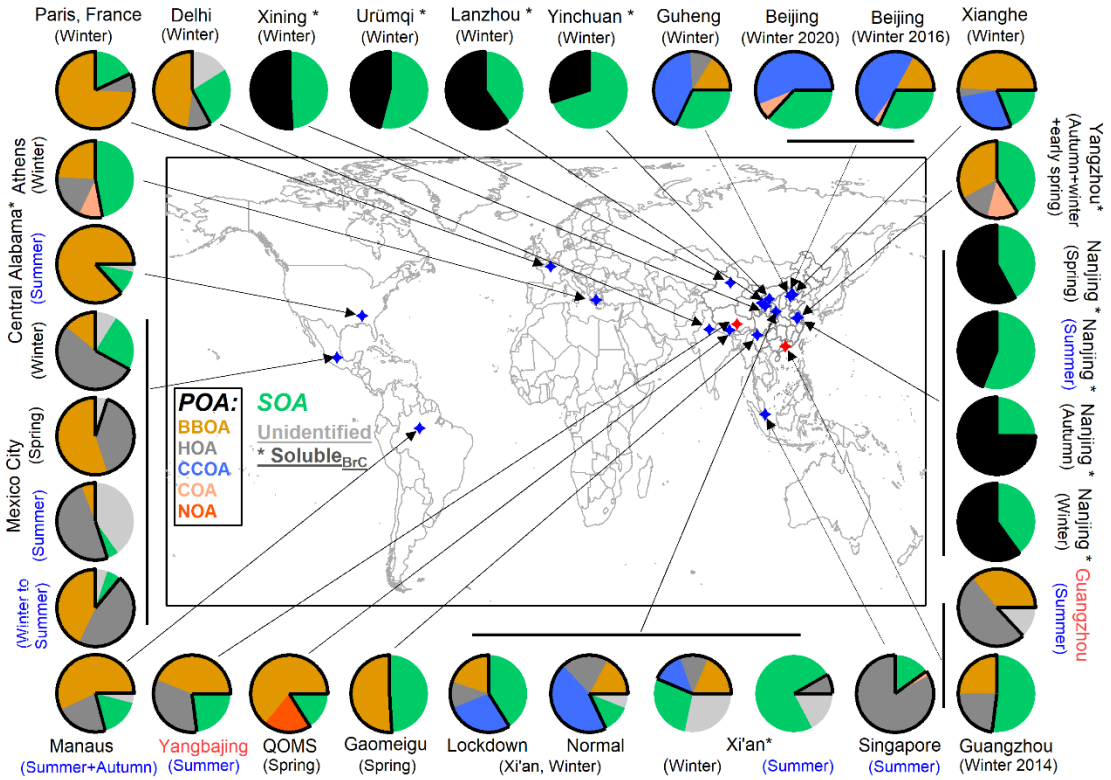
R3.11(b): What are the sources to the strong morning NO spike at YBJ and GIG sites?

A3.11(b): The strong peak of NO occurred simultaneously with NO_2 and CO, which were due to the local anthropogenic emissions (such as vehicle emissions). This is consistent with the concurrent peaks in traffic-related aerosol components such as HOA (Fig. 4).

R3.12: L525: … "Singapore during winter" is misleading as Singapore do not have winter season based on temperature.

A3.12: Indeed, the average daily temperature in Singapore remains consistently between 26°C and 32°C throughout the year. There is no clear division into distinct seasons; only summer exists. We also rechecked the observation period of this study, which was from May 14th to June 9th, 2017 (Kasthuriarachchi et al., 2020). We modified the sentence and the Figure. 7 as follows:

Revised sentence (Lines 580-581): "**…, HOA shows extremely high light absorption contribution in Singapore during summer (83 %), and in Mexico in both winter (54 %) and summer (49 %), …**"



"Figure 7. The summary of the contribution to BrC light absorption at 370 nm from different sources using the PMF–MLR method. The asterisks (*) represent the light absorption contributions of soluble BrC from different sources at 365nm. The sources include POA: biomass burning OA (BBOA), hydrocarbon‐like OA (HOA), coal combustion OA(CCOA), cooking‐related OA(COA), nitrogen‐containing OA(NOA); The total SOA from oxygenated OA was used here. In addition, the unidentified fraction from the intercept of the MLR method was also shown. The detailed information on each pie is provided in Table S9."

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