We deeply appreciate Reviewers for their constructive recommendation and the helpful suggestion. We have updated the manuscript following these comments and addressed all points raised. These comments are very helpful for improving our manuscript. Specific responses to each of the comments are provided below (review's comments in **black**, our responses in blue, details of the changes made to the manuscript in *blue font*). And the modifications in the revised manuscript with marks are marked in yellow. We are pleased to provide the revised manuscript and hope both Reviewers are satisfied with our responses.

Response to Reviewer #3

1) Reviewer's general comment:

Mo et al. investigated light absorption and the sources of extractable organic carbon (EX-OC), which encompasses water-insoluble carbon (WIOC), humic-like substances (HULIS-C), and hydrophilic OC (non-HULIS-C). The study revealed that WIOC constituted the majority of OC mass concentrations and light-absorbing efficiency at 365 nm. Additionally, the authors found that the radiative forcing effects of EX-OC were mainly contributed by relatively hydrophobic fractions. The authors also proposed a light-absorbing carbonaceous continuum, revealing that components enriched with fossil sources exhibit stronger light-absorbing capacity, higher aromatic levels, increased molecular weights, and greater atmospheric recalcitrance. This study is pivotal for comprehensively understanding the climate-forcing brown carbon and developing related mitigating strategies. However, before publication, the authors need to address the following concerns:

Response: We thank Reviewer#3 for the assessment of our manuscript and for providing suggestions to improve clarity and quality.

2) Reviewer's comment:

Line 25–27: Too many brackets in this sentence. Please simplify this sentence to create a concise abstract.

Response: Thanks for your suggestion. This sentence has been revised to *"On average, WIOC made up 33.4 ± 7.66% and 40.5 ± 9.73% of concentrations and light* *absorption at 365 nm (Abs365) of extractable OC (EX-OC), which includes relatively hydrophobic OC (WIOC and humic-like substances: HULIS-C) and hydrophilic OC (non-humic-like substances: non-HULIS-C).”* (**Lines 25 to 28**)

3) Reviewer’s comment:

Line 51: Please review and remove any duplicate abbreviation definitions found throughout the manuscript, such as OC, WIOC, etc.

Response: We have removed the duplicate abbreviation definitions in the revised manuscript.

4) Reviewer’s comment:

Lines 65-91: This paragraph lacks clarity in addressing the main questions concerning WIOC. While the author discusses the method of measuring light-absorbing OC and compares the light-absorbing properties of WIOC and WSOC, the discussion on the health effects and atmospheric lifetime of WIOC seems disconnected. Additionally, the motivation for this study is relatively vague. Therefore, I would suggest that the authors restructure the paragraph to clearly emphasize current scientific inquiries and provide a more cohesive rationale for their research.

Response: We appreciate the reviewer's insightful comments. In response, we have reconstructed this paragraph to emphasize current scientific inquiries and provide a more cohesive rationale for our research. Please see the revised manuscript

The manuscript is revised as follows:

Lines 66 to 81: *“According to water solubility, OC can be classified into two main categories: water-soluble OC (WSOC) and water-insoluble OC (WIOC). While WSOC has been extensively studied over the past decades, with investigations focusing on its sources, light-absorbing properties, and atmospheric processes (Bosch et al., 2014; Dasari et al., 2019; Mo et al., 2021; Wang et al., 2020; Wozniak et al., 2014). WIOC, which makes up large fraction of OC (~up to 80%) and a substantial portion of light* *absorption by BrC, has received comparatively less attention. WIOC exhibits a significantly higher light-absorbing capacity compared to WSOC, attributed to the enrichment of strong light-absorbing BrC chromophores in WIOC. For instance, certain strong BrC chromophores like polycyclic aromatic hydrocarbons (PAHs) and their derivatives, as well as high-molecular-weight oligomers, are water-insoluble (Huang et al., 2020; Kalberer et al., 2006; Xie et al., 2017). Indeed, Zhang et al. (2013) reported that the light absorption by methanol-extracted OC in Los Angeles was approximately 3 and 21 times higher than that by WSOC. Moreover, field observations indicate that WIOC exhibits greater recalcitrance during long-range transport processes compared to WSOC, leading to a longer lifetime for WIOC (Fellman et al., 2015; Kirillova et al., 2014; Wozniak et al., 2012). Given that WIOC represents a relatively long-lived OC component with a higher light-absorbing capacity, a comprehensive understanding of its sources and light-absorbing properties is imperative."*

5) **Reviewer's comment:**

Lines 145-147: Please review the two identical sentences and revise them accordingly.

Response: Thank you for your careful review. We have addressed the repeated sentences in the revised manuscript

6) **Reviewer's comment:**

Line 251: Figure 1c should be Figure 2c?

Response: Corrected.

7) **Reviewer's comment:**

Lines 262-304: A more comprehensive discussion would involve comparing the light-absorbing properties of EX-OC between areas with and without central heating.

Response: Thanks for your good suggestion. We have added more discussion on the spatial variation in light-absorbing properties of EX-OC in the revised manuscript. We found that a significant disparity in the MAE_{365} of WIOC between areas with and without central heating. Specifically, areas with central heating exhibited a notably higher MAE_{365} (1.75 ± 0.64 m^2/gC) compared to those without (1.48 ± 0.46 m^2/gC), with a statistically significant difference ($p < 0.01$). This disparity was more pronounced during colder seasons, with a 21.5% difference in MAE365 between areas with central heating (2.20 ± 0.51 m^2/gC) and those without (1.81 ± 0.28 m^2/gC), compared to a 10.3% difference during warmer seasons. We attribute this spatial variability primarily to coal combustion, given the considerably higher coal consumption for central/domestic heating in areas with central heating.

The manuscript is revised as follows:

Lines 301 to 308: *"Spatially, the MAE365 of WIOC was significantly higher in areas with central heating than without central heating (1.75 ± 0.64 m2/gC vs. 1.48 ± 0.46 m2/gC, p < 0.01). The difference in MAE365 of WIOC between the areas with and without central heating was more pronounced during colder seasons (2.20 ± 0.51 m2/gC vs. 1.81 ± 0.28 m2/gC, 21.5% difference) than warmer seasons (1.29 ± 0.37 m2/gC vs. 1.17 ± 0.26 m2/gC, 10.3% difference). Given that coal consumption for central/domestic heating is considerably higher in areas with central heating compared to those without, it is plausible that the spatial variability in MAE365 of WIOC is predominantly influenced by coal combustion."*

8) Reviewer's comment:

Lines 269-272: Given that the light absorption of BrC, as measured by solvent extraction, appears to be underestimated compared to under ambient aerosol conditions, it is imperative to determine whether the authors considered this factor when comparing with "tar ball" and "unextractable dark BrC".

Response: Indeed, the light absorption of BrC measured in solvent extracts often underestimates its absorption in ambient aerosol conditions. To address this disparity and predict the corresponding BrC absorption in ambient aerosols, it's essential to calibrate the absorption determined in solvent extracts using a correction factor. To date, the correction factor proposed by Liu et al. (2013), commonly set at 2, is widely used for this purpose. Even after applying this correction factor, we observed that the MAE550 of WIOC (0.28 ± 0.09 m^2/gC) remains an order of magnitude lower than that of "tar ball" (~3.6 to 4.1 m^2/g) and "unextractable dark BrC" (~1.2 m^2/g). In response, we have enriched the discussion on BrC light absorption correction in the revised manuscript.

The manuscript is revised as follows:

Lines 279 to 289: *"It should be noted that light absorption of BrC, as measured by solvent extraction, appears to be underestimated compared to under ambient aerosol conditions. To accurately derive the corresponding BrC absorption in ambient aerosols, it is necessary to calibrate the absorption determined in solvent extracts using a correction factor. Presently, the correction factor proposed by Liu et al. (2013), typically set at 2, is widely employed for this purpose. Despite WIOC being recognized as the most light-absorbing OC component, even after applying this correction factor, we observed that the MAE of WIOC at 550 nm(0.28 ± 0.09 m^2/gC) remains an order of magnitude lower than that of amorphous tar ball BrC (approximately 3.6 to 4.1 m^2/g) and unextractable "dark BrC" (approximately 1.2 m^2/g) as determined by transmission electron microscopy (Alexander et al., 2008; Chakrabarty et al., 2023), indicating the light-absorbing capacity of the extractable OC is relatively weakly."*

9) Reviewer's comment:

Line 327: Please maintain consistency to ensure a unified description of E2/E3 in both the plot and text.

Response: Corrected

10) Reviewer's comment:

Lines 333-335 and 341-342: The authors observed a robust correlation between WIOC and Abs365, WIOC (r = 0.97, p < 0.01), indicating similar sources and

formation processes. However, they also concluded differences in sources and formation processes between WIOC and light-absorbing compounds during warm seasons. These findings may appear contradictory and confusing. Clarification is needed regarding the rationale behind analyzing the correlations between WIOC and Abs365, WIOC in individual warm and cold seasons.

Response: Thank you for your constructive comments. We apologize for the lack of clarity in our previous sentences. Our intention was to show that while WIOC correlates well with Abs365, WIOC throughout the entire year ($r = 0.97$, $p < 0.01$), as indicated in Table S2, the correlations of WIOC with water-soluble ions vary notably between warm and cold seasons. This discrepancy suggests differences in sources and formation processes between WIOC and light-absorbing compounds during warm and cold seasons. We have revised these sentences to enhance clarity in the revised manuscript.

The manuscript is revised as follows:

Lines 356 to 360: *"Although the WIOC exhibited strong correlation with Abas$_{365, WIOC}$ ($r = 0.97$, $p < 0.01$) for the entire year, as listed in Table S2, the correlations between WIOC and Abs365, WIOC, as well as WIOC and water-soluble ions, differed notably between warm and cold seasons. This discrepancy suggests differences in sources and formation processes of WIOC and light-absorbing compounds between warm and cold season."*

11) Reviewer's comment:

Line 375: Figure 3b should be Figure 4b?

Response: Corrected.

12) Reviewer's comment:

Line 382: Please clarify the methodology employed by the authors to determine the source contributions to Abs365, WIOC.

Response: Thanks for your suggestion. We used the PMF model to resolve the sources contribution to $Abs_{365,\ WIOC}$. This methodology has been clarified in the revised manuscript.

The manuscript is revised as follows:

Lines 413 : *"Figure 4c shows the contributions of sources identified by PMF model to the $Abs_{365,\ WIOC}$."*

13) Reviewer's comment:

Lines 390-392: Could you elaborate on why biomass burning, rather than secondary sources, contributes more to the light absorption of BrC in summer? What distinguishes the conclusion of your research from the previous studies mentioned?

Response: Thank you for your questions. Indeed, higher solar radiation and temperatures in summer facilitate the secondary generation of BrC, with secondary BrC being more enriched in the WSOC. Previous studies have observed a significant contribution of secondary sources to WSBrC during summer (Du et al., 2014; Yan et al., 2017). Conversely, the WIOC tends to be more enriched with primary sources such as BB and coal combustion. As temperatures rise in summer, coal consumption typically declines. Consequently, during summer, $Abs_{365,WIOC}$ is predominantly contributed by BB. These points have been clarified in the revised manuscript

The manuscript is revised as follows:

Lines 421 to 427: *"In summer, the elevated solar radiation and temperatures can promote the secondary generation of BrC, with secondary BrC being more enriched in the WSOC. Previous studies have observed a significant contribution of secondary sources to WSBrC during summer (Du et al., 2014; Yan et al., 2017). However, the BrC within WIOC tends to be more enriched with primary sources such as BB and coal combustion (Figure 4c). As temperatures rise in summer, coal consumption typically declines. Consequently, during summer, $Abs_{365,\ WIOC}$ is predominantly contributed by*

BB (Figure 4d).”

14) Reviewer’s comment:

Lines 398-411: Was the correlation between biomass burning source contribution and the light absorption of WIOC discussed by the author? Was there a correlation similar to that observed with coal combustion?

Response: Thanks for your questions. We conducted correlation analysis between the MAE365 of WIOC and the contribution of biomass burning. As shown in Figure R1, unlike the strong positive correlation observed for coal combustion ($r = 0.72$, $p < 0.01$), the contribution of biomass burning exhibited a negative correlation with the MAE365 of WIOC ($r = -0.34$, $p = 0.46$). Thus, the light-absorbing compounds derived from coal combustion have a stronger light-absorbing capacity than BB, and enhanced the overall MAE365 of WIOC. In the revised manuscript, we have added more discussion on this point to provide further insights into the contrasting contributions of coal combustion and biomass burning to the light-absorbing properties of WIOC.

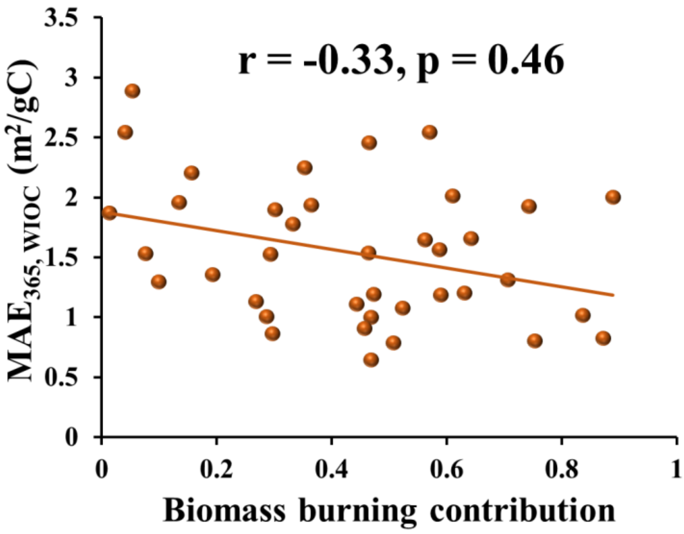

Figure R1. The relationship between the MAE_{365} of WIOC and relative contribution of biomass burning.

The manuscript is revised as follows:

Lines 433 to 440: *“Both BB and coal combustion are recognized as the sources* *of BrC with strong light-absorbing capacity. We found that contribution of BB exhibited a negative correlation with $MAE_{365,WIOC}$ (r = -0.34, p = 0.46, Figure S3b), whereas a strong positive correlation was observed for coal combustion (r = 0.72, p < 0.01, Figure S3a). These suggest that the light-absorbing compounds derived from coal combustion have a stronger light-absorbing capacity than BB, and enhanced the overall $MAE_{365,WIOC}$."*

15) Reviewer's comment:

Line 423: Figure 1c should be Figure 2c?

Response: Corrected.

16) Reviewer's comment:

Line 504: Figures 2b and c should be Figures 3b and c?

Response: Corrected

References:

Du, Z.Y., He, K.B., Cheng, Y., Duan, F.K., Ma, Y.L., Liu, J.M., Zhang, X.L., Zheng, M., Weber, R. (2014), A yearlong study of water-soluble organic carbon in Beijing II: Light absorption properties, *Atmospheric Environment*, *89*, 235-241.https://doi.org/10.1016/j.atmosenv.2014.02.022

Liu, J., Bergin, M., Guo, H., King, L., Kotra, N., Edgerton, E., Weber, R.J. (2013), Size-resolved measurements of brown carbon in water and methanol extracts and estimates of their contribution to ambient fine-particle light absorption, *Atmospheric Chemistry and Physics*, *13*(24), 12389-12404.https://doi.org/10.5194/acp-13-12389-2013

Yan, C., Zheng, M., Bosch, C., Andersson, A., Desyaterik, Y., Sullivan, A.P., Collett, J.L., Zhao, B., Wang, S., He, K., Gustafsson, O. (2017), Important fossil source contribution to brown carbon in Beijing during winter, *Scientific Reports*, *7*.https://doi.org/10.1038/srep43182