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 #1

1) Reviewer's general comment:

The manuscript provided a detailed study on the light-absorbing characteristics, potential sources, and radiative forcing effects of WIOC across ten cities in China. The authors compared the characteristics of extractable OC (EX-OC) with different polarities (WIOC, HULIS, and non-HULIS). They discovered that biomass burning and coal combustion are the primary sources of WIOC in China and radiative forcing of EX-OC is mainly due to WIOC and HULIS. Furthermore, this study proposed a hypothesis of a light-absorbing carbonaceous continuum, demonstrating that carbonaceous components from fossil sources tend to have a stronger light-absorbing capacity, higher aromatic levels, and greater recalcitrance in the atmosphere. Overall, the paper is well-structured and well-written, but there are still some areas that require improvement. Please see my comments below.

Response: We thank the Reviewer#1 for carefully reviewing our manuscript and for the insightful comments and the constructive feedback! Please find our answers to each individual point below:

2) Reviewer's comment:

Major Comments:

In this study, the authors divide China into areas with and without central heating, which is a meaningful approach. However, the division based on a simple line in Figure 1 lacks convincing evidence. Are there data to support this division?

Moreover, the sources and light absorption properties of WIOC are expected to differ significantly between areas with and without central heating. However, the article does not adequately discuss the spatial differences in WIOC sources and light absorption capabilities. The authors should address this aspect.

Response: Thank you for your helpful suggestions. The areas with central heating typically experience an average annual temperature below 15°C, while areas without central heating generally have higher average annual temperatures, often exceeding 15°C. Notably, the differentiation between areas with and without central heating in our study closely aligns with the well-recognized Qinling Mountains-Huaihe River line, which serves as the north-south boundary in China (Figure R1). We have incorporated additional details and descriptions for Figure 1 to provide further clarity.

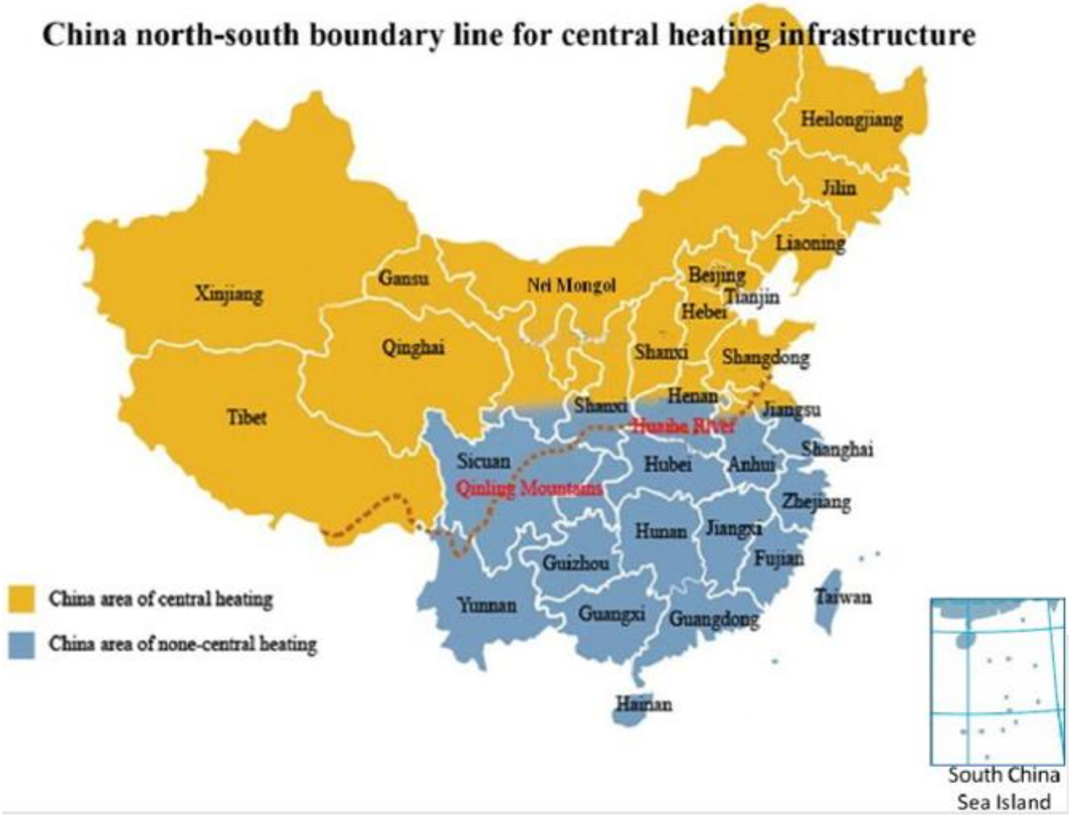

Figure R1. The central heating north–south boundary in China.

We have incorporated additional discussions on the spatial differences in the light-absorbing capacity and sources of WIOC in the revised manuscript, as reviewer suggested. In terms of light-absorbing capacity, our findings reveal 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 MAE_{365} 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.

Regarding the sources of WIOC, while the contribution of coal combustion was comparable to that of BB, both exhibited distinct spatial and seasonal variations. During winter, coal combustion emerged as the dominant source, constituting 48.4% of the total WIOC, likely due to increased coal usage in areas with central heating. Notably, coal combustion accounted for 56.2% of WIOC in areas with central heating during cold seasons, whereas in areas without central heating, BB surpassed coal combustion significantly (54.2% vs. 17.3%). Thus, coal combustion and BB were identified as the predominant sources of WIOC in areas with and without central heating, respectively, during cold seasons.

Moreover, during summer, with rising temperatures and solar radiation intensity, contributions from aging processes and BB increased to 39.3% and 41.3%, respectively. In spring, a significant fraction of WIOC was associated with dust/soil, reaching up to 28.8%, particularly pronounced in areas with central heating. This observation aligns with the influence of sandstorms from the Gobi desert, which border China and Mongolia, affecting the air quality of Northern China during springtime (Filonchyk et al., 2024).

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."*

Lines 390 to 411: *"Although the contribution of coal combustion to WIOC was comparable to that of BB, both exhibited distinct spatial and seasonal variations (Figure 4a). Specifically, during winter, coal combustion emerged as the dominant source of WIOC, accounting for 48.4% of the total, likely driven by increased coal usage in areas with central heating. Indeed, coal combustion constituted the primary source of WIOC in areas with central heating during cold seasons (56.2%). In contrast, in areas without central heating, the contribution of BB surpassed that of coal combustion significantly (54.2% vs. 17.3%). Therefore, coal combustion and BB were identified as the predominant sources of WIOC in areas with and without central heating, respectively, during cold seasons. Compared to primary emissions sources, the contributions of the sources related to aging processes and nitrogen-induced secondary formation were relatively lower, accounting for 18.2% and 5.2% of the WIOC, respectively. That may be due to these two secondary sources are more enriched in water-soluble components (HULIS-C + non-HULIS-C). Actually, although the uncertainties of sources contribution of HULIS-C and non-HULIS-C resolved by PMF model may be high, the aging processes and nitrogen-related secondary formation contributed 10.1% and 20.2% to HULIS-C, and 18.3% and 21.6% to non-HULIS-C, respectively. In addition, during the summer, when both temperature and solar radiation intensity rise, the contributions from aging processes and BB increased to 39.3% and 41.3%, respectively. In spring, a significant fraction of WIOC was associated with dust/soil, reaching up to 28.8%. Specially, the dust/soil contribution was much higher in the aeras with central hearing than those without central heating. This is consistent with the fact that sandstorms from the Gobi desert that borders China and Mongolia ride springtime winds to affect the air quality of Northern China*

(Filonchyk et al., 2024).”

3) Reviewer’s comment:

The method for extracting WIOC involves methanol extraction, concentrated, and analysis using an OC/EC analyzer, while WSOC is determined using a liquid TOC analyzer. The analysis mechanism of OC/EC analyzers and TOC analyzers differ, including the different catalysts and detectors they employ. Comparing measurement results from these two methods may be challenging. Additionally, the authors did not provide the blank value for OC/EC determination of WIOC, which is crucial given methanol's high propensity for extracting atmospheric organic matter.

Response: Thanks for the good questions! As highlighted by the reviewer, it's important to note that the analytical procedure and mechanism for determining carbon content differ between the OC/EC analyzer and the TOC analyzer. In our study, the WIOC is extracted and dissolved in methanol, making direct determination of its carbon content by the TOC analyzer unfeasible. Consequently, comparisons between the determined WIOC content using these two methods are not possible. However, it is important to note that both instruments involve converting carbon in the sample to CO2 and detecting CO2 using a nondispersive infrared detector (NDIR) or a flame ionization detector (FID) after CO2 conversion to methane. Thus, the analytical mechanisms are similar. Indeed, previous studies have systematically compared these two methods for determining WSOC in aerosols. The results have demonstrated no significant differences between the measurements obtained from the two methods(Fan et al., 2012). Therefore, the carbon content of various carbonaceous components determined by these two methods should be comparable. We have elaborated further on this point in the revised manuscript.

The field blanks for the WSOC, HULIS-C and WIOC were 0.39 ± 0.16, 0.66 ± 0.21, and 1.75 ± 0.48 ug C/cm2, respectively. All WSOC, HULIS, and WIOC concentrations presented in this study were corrected with field blanks.

The manuscript is revised as follows:

Lines 152 to 162: *"The analysis mechanism of OC/EC analyzers and TOC analyzers differ, including the different catalysts and detectors they employ. However, it is important to note that both OC involve converting carbon in the sample to CO_2 and detecting CO_2 using a nondispersive infrared detector (NDIR) or a flame ionization detector (FID) after CO_2 conversion to methane. Thus, the analytical mechanisms are similar. Indeed, previous studies have systematically compared these two methods for determining WSOC in aerosols (Yu et al., 2002). The results have demonstrated no significant differences between the measurements obtained from the two methods. Therefore, the carbon content of various carbonaceous components determined by these two methods should be comparable. Based on extractable OC (EX-OC) polarity, the EX-OC was separated into WIOC, HULIS-C, and non-HULIS-C. All WSOC, HULIS-C, and WIOC concentrations presented in this study were corrected with field blanks (0.39 ± 0.16, 0.66 ± 0.21, and 1.75 ± 0.48 $\mu gC/cm^2$, respectively)."*

4) Reviewer's comment:

The authors primarily utilize the PMF model to quantify the sources of WIOC. The authors suggest that the increase in coal combustion led to an increase in Abs365 in overall EX-OC (line 407-411), which seems plausible. However, correlating the quantitative results from coal combustion predicted by PMF with the Abs365 of overall EX-OC (Figure S3c) is not appropriate. The authors quantified the sources of WSOC using dual carbon isotopes (δ13C and Δ14C) in their previous study. Comparing the PMF model with source apportionment results based on dual carbon isotopes presents challenges. While 14C can accurately distinguish fossil sources from non-fossil sources, without chemical markers, it's difficult to quantitatively analyze sources related to atmospheric processes (such as secondary sources). On the other hand, although PMF can theoretically incorporate various chemical markers and include secondary sources in quantitative analysis, its results involve more human interpretation factors and

may not be as precise as 14C. Thus, caution is warranted when comparing source apportionment results obtained from PMF and dual carbon isotopes.

Response: Thank you for raising this important point. We fully concur that attempting to establish a correlation between the quantitative results of WIOC from coal combustion predicted by PMF and the Abs_{365} of overall EX-OC is inappropriate. In our previous study, we utilized dual carbon isotopes to quantify the source of WSOC, while the sources of WIOC were determined using the PMF model. As noted by the reviewer, 14C isotopic analysis offers a precise means of distinguishing between fossil and non-fossil sources. Conversely, while the PMF model theoretically has the capacity to resolve a broader range of source types than 14C isotopic analysis, its results are subject to more human interpretation factors and may not be as definitive as those obtained through 14C analysis. In light of this insight, we have removed the discussion on the relationship between Abs_{365} of EX-OC and coal combustion contribution predicted by the PMF model in the revised manuscript.

5) Reviewer’s comment:

In Section 3.4, the authors propose a concept of a light-absorbing carbonaceous aerosol continuum, which I find intriguing. It's important to note that in this study, WIOC, HULIS, and non-HULIS are well-defined based on their polarity. These components indeed exhibit significant differences in both sources and light absorption properties. However, regarding BC, I contend that BC is the strongest light-absorbing component. Although this assertion isn't reflected in Figure 6, the authors further subdivide BC into char and soot in this section without providing a clear definition. In reality, char and soot are defined differently across various environmental matrices (https://doi.org/10.1038/s43017-022-00316-6). For aerosols, biomass burning and coal combustion emit large amounts of large molecular weight soluble compounds, which may char and produce false char EC signals (artifacts). Additionally, there are overlaps between char and BrC. These issues warrant attention and clarification from the authors.

Response: We appreciate the insightful comments provided by the reviewer. It is indeed acknowledged that the definitions of Char and Soot vary across different environmental matrices (Coppola et al., 2022). In our study, the distinction between char-BC and soot-BC within carbonaceous aerosols is primarily based on operational definitions derived from the Interagency Monitoring of Protected Visual Environments (IMPROVE) protocol and the thermal-optical reflectance (TOR) method for OC and EC analysis, as referenced in previous studies (Cai et al., 2023; Han et al., 2010).

The carbonaceous aerosols encompass a wide array of diverse components, exhibiting a continuum of physical and chemical properties. The differentiation between these components relies on conceptual and operational definitions, which may not necessarily correspond to clear boundaries in reality. In our work, WIOC, HULIS-C, and non-HULIS-C are defined based on their polarity, while the definition of BC, including char-BC and soot-BC, is primarily related to thermal and optical properties. However, it is important to acknowledge that these operational definitions may result in overlaps between different carbonaceous components. For instance, carbonaceous emissions from BB and coal combustion often contain large molecular weight soluble compounds, such as HULIS, which can char and produce false char EC signals in the TOR analysis (Yu et al., 2002). Additionally, certain fractions of char-BC may exhibit chemical and physical behaviors resembling those of high-molecular-weight OC compounds, potentially overlapping with BrC. Therefore, delineating clear boundaries for carbonaceous components remains challenging.

The manuscript is revised as follows:

Lines 497 to 502: *"Char and soot are defined differently across various environmental matrices (Coppola et al., 2022). For the carbonaceous aerosols, the char-BC and soot-BC are widely and operationally defined by different temperatures in the Interagency Monitoring of Protected Visual Environments (IMPROVE) protocol and the thermal-optical reflectance (TOR) method for OC and EC analysis (Cai et al., 2023; Han et al., 2010). Generally, the light-absorbing capacity of soot-BC is higher* *than char-BC (Andreae and Gelencser, 2006; Corbin et al., 2019; Schnaiter et al., 2003).”*

Lines 605 to 617: *“It is important to acknowledge that carbonaceous aerosols encompass a wide array of diverse components, exhibiting a continuum of physical and chemical properties. The distinction between these carbonaceous components, as discussed above, is primarily based on conceptual and operational definitions, rather than clear boundaries in reality. In other words, the classification of carbonaceous components in aerosols is highly dependent on operational criteria. In this study, on the one hand, the WIOC, HULIS-C, and non-HULIS-C are well-defined based on their polarity. On the other hand, the definition of BC, which includes char- and soot-BC, is more closely associated with thermal and optical properties. These operational definitions may lead to overlaps between different carbonaceous components. For instance. BB and coal combustion emit large amounts of large molecular weight soluble compounds, such as HULIS (e.g., HULIS), which may char and produce false char EC signals in the TOT analysis (Yu et al., 2002). Additionally, certain portions of char-BC may exhibit chemical and physical behaviors akin to high-molecular-weight OC compounds, thereby overlapping with BrC. Therefore, there is no a clear boundary for the carbonaceous components.”*

6) Reviewer’s comment:
Minor Comments:
Line 20: “water-insoluble carbon” change to “water-insoluble organic carbon”

Response: Corrected

7) Reviewer’s comment:
Line 145 -147, the same phrase appears twice.

Response: Thanks for your careful work. We have deleted the identical sentences.

8) Reviewer’s comment:

Line 148, the authors mention using SPE for HULIS extraction. Given the various methods available for HULIS isolation (as referenced in https://doi.org/10.1016/j.envpol.2013.05.055), it would be helpful to provide reasons for choosing SPE for HULIS isolation.

Response: Thanks for your comment. The HULIS is an operational definition, and there are many methods to isolate the HULIS. The reason we select the HLB-SPE column for HULIS isolation is because the HLB-SPE column showed excellent reproducibility and high recovery yield (Fan et al., 2012; Lin et al., 2010).

The manuscript is revised as follows:

Lines 139 to 147: *"HULIS are operationally defined by the procedure used for isolation from bulk WSOC by removing low molecular weight organic acids and inorganic ions. The HLB (Oasis, 30 μm, 60 mg/cartridge, Waters, USA)-SPE method is most widely used to isolate HULIS due to its excellent reproducibility and high recovery yield (Fan et al., 2012; Lin et al., 2010b). Therefore, we used an HLB-SPE column to isolate the HULIS"*

9) Reviewer's comment:

Line 271-272, "BrC" changed to "extractable OC". There is no accurate method for extracting BrC.

Response: Corrected.

10) Reviewer's comment:

Line 388, it should be "carbon mass contribution"

Response: Corrected.

11) Reviewer's comment:

Figure 6. I suggest emphasizing in the caption that WIOC tends to be OC soluble in methanol but not in water. The WIOC in this work is not real water-insoluble organic carbon.

Response: Good suggestion! We have emphasized that *"The WIOC in this study does not strictly denote OC that is insoluble in water, it is more likely to be OC that is insoluble in water but soluble in methanol."* in the Figure 6.

References:

Andreae, M.O., Gelencser, A. (2006), Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols, *Atmospheric Chemistry and Physics*, *6*, 3131-3148.https://doi.org/10.5194/acp-6-3131-2006

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