

**General Comments:** Brown carbon (BrC) is an important constituent of carbonaceous aerosols and significantly contributes to total solar light absorption of aerosols. The manuscript titled “Optical and structural properties of atmospheric water-soluble organic carbon in China: Insights from multi-site spectroscopic measurements” presents measurements of optical properties and structural characteristics of WSOC based on different spectroscopic techniques (absorbance, fluorescence and FTIR) from different regions of China. Overall, the study promotes a better understanding of the spatial heterogeneity of optical and structural properties of WSOC and their influencing factors (emission sources, aging processes, relative humidity (RH), etc.) in China and deepened the understanding of the contribution of WSOC fluorescence to its light absorption. However, the manuscript has many shortcomings in its current version. It needs through language editing and clarifications at many places throughout the manuscript. It also misses consistency while using different terminologies for the same parameter (for example, authors have used WSOC and WS-BrC interchangeably to refer to BrC). Yet, the study has relevance to the atmospheric research community and can be accepted for publication in the journal after major revision. The detailed comments are given below:

**Major comments**

1. Methodology and elsewhere: What do you mean by “regional site (rs)” in your manuscript? Do you mean “remote/rural site”? It’s confusing. Clarify.
2. Methodology (section 2.3): Equation S9 in text S3 is incorrect (could be a typo error). The equation should consist both mass scattering efficiency (MSE) and MAE. Recheck and correct it.
3. Methodology (section 2.3, text S4) and section 3.2 of results section: The author measured WSOC absorbance from 250-700 nm and used WSOC absorbance from 250 to 400 nm in PMF model for source apportionment. However, it is well known that WSOC absorbance < 340 nm is highly influenced by absorbance from nitrate aerosols. Did authors consider this aspect during PMF run? How this will impact the findings?
4. Results and Discussion (section 3.1): The authors observed a significant spatial variability in WSOC, OC, EC, etc. across ten sites. What could be the potential reasons (e.g., different sources, metrology, etc.) behind this variability, discuss briefly?
5. Results and Discussion (section 3.1): The authors compared Abs<sub>365</sub> and MAE<sub>365</sub> values between different regions (e.g., northwest China, southwest China, etc.). Did you carry out any significance test to check whether difference was significant or not?
6. Results and Discussion (section 3.1): The sampling durations were different at different sites representing different administrative regions (Table S1). Do you think the “day versus night variability” in optical properties could have also contributed to the inter-regional variability observed in optical properties of WS-BrC in your study.
7. Lines 208-210: The authors reported that light absorbing ability (SFE) of WS-BrC and mass concentration of WSOC are directly proportional (related). How did authors come with such conclusions? Please cite relevant studies in this context.
8. Fig. 6c-d: Is this the integrated absorbance from 250-400 nm, clarify?

9. Line 341-346: Please revisit this portion, especially, portion where authors mentioned that photochemical bleaching will be higher during severe pollution days. In fact, opposite is likely to be true as lower pollution levels mean higher visibility, resulting in higher availability of solar flux.

10. The study only discusses optical and structural properties of WS-BrC, which represents only 50-70% fraction of OC. What about water-insoluble OC? What are the implications of your findings on light-absorbing water-insoluble OC?

### Minor Comments

1. Line 30-34: “The light absorption factors.....impact on fluorophores.” Confusing sentence. Rewrite it.

2. Line 45-46: “BrC contributes up to 72% of the total light absorption of aerosols at 370 nm and the direct radiative effect of BrC (+0.048 W·m<sup>-2</sup>) is about 30% of black carbon (+0.17 W·m<sup>-2</sup>)”. Is this global average or only valid for China? Please clarify.

3. Line 51: Use either “commonly” or “widely”. One of them is redundant.

4. Line 51-53: Make it two sentences.

5. Line 55-59: Grammatical mistakes at many places. Some sentences are confusing. Revise it.

6. Line 61: Should be “Spectroscopy-based studies conducted...”

7. Line 81: “methods are often used separately in previous studies”?

8. Line 82-86: Difficult to follow as this is a very long sentence. I suggest to break it into smaller sentences.

9. Line 110: should be “0.45 μm pore-size PTFE syringe filter”.

10. Line 118-120: Break it into two sentences like “...calculated. More details can be found in Text S2.”

11. Line 152: “Additionally, the XGBoost model is also used to..” “also” is redundant in this sentence.

12. Line 169: “...mass concentrations of carbonaceous components at HS site are not that low...” Compared to what, clarify?

13. Line 157-158: “During the wintertime observation period, WSOC mass concentrations exhibit a significant spatial variation across the ten sites ( $p < 0.05$ ) (see Figure 1 and Table S2).” Which test did you use to derive significance level?

14. Line 308-309: The sentence is confusing. Rewrite it.

15. Line 341: Change “great” to “large”.

16. Line 378: Typo? “WOSC” should be “WSOC”

17. “In contrast, the relationships between MAE365 values and functional groups may differ from Abs365. Similarly, C=C, O-H and R-ONO<sub>2</sub> exhibit the strongest correlations with MAE365 at most sites (e.g., TJ, QD, SH, TS, and HS).” Similar to what? These sentences are confusing. Rewrite them.

18. Line 413: “discrepancies” doesn’t seem to be the write word here. Replace it.