Response to Comments by Review #2

Manuscript: egusphere-2024-2416

Title: Measurement Report: Optical and structural properties of atmospheric water-soluble organic carbon in China: Insights from multi-site spectroscopic measurements

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General Comments: *Brown carbon (BrC) is an important constituent of carbonaceous aerosols and significantly contributes to the 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.*

Response to General Comments: We thank the reviewer for the overall supportive comments. We also appreciate the reviewer's considerable efforts in reviewing the manuscript and providing valuable comments and suggestions for the improvements and clarifications. Based on the reviewer's comments and suggestions, the manuscript is thoroughly revised. In particular, we have carefully revised the language and inappropriate and unclear expressions throughout the manuscript, so as to interpret the data more accurately and reasonably, and improve the rigor and scientific nature of the discussion. Additionally, we have taken the reviewer's suggestion to use WSOC uniformly to ensure consistency in terminologies of the same parameter in the revised manuscript. Below, we detail our responses and resulting edits to all the comments. These are organized such that we first list the review comments in italics and blue, immediately followed by our responses in normal font. To make it clear, the contents in the revised manuscript are presented in quotes and italics, while the newly added contents in the revised manuscript are underlined.

Major Comments:

Comment #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.*

Response to Comment #1: We are sorry for the confusion. The "regional site" in this study does not refer to "remote/rural site". Regional sites are usually far away from urban built-up area and major pollution sources, and the distance from urban built-up areas and major pollution sources is more than 20 kilometers and less than 50 kilometers. In addition, regional site can be used as a representative receptor site for regional emissions within the region. That is, regional sites are not located in rural areas and are not as far away as remote sites.

In this study, the TS (1534 m a.s.l.) site, locating at the Taishan National Reference Climatological station at the summit of Mt. Tai and close to Taian City in Shandong Province, China, is less affected by anthropogenic emissions and has been widely used as a sampling site for researches on regional atmospheric pollution and atmospheric chemistry in Northern China (Chen et al., 2022; Jiang et al., 2020). Meanwhile, the HS site locates at the Atmospheric Super Monitoring Station in Jiangmen City in Guangdong Province, China and downwind of the Pearl River Delta (PRD), which has been used as a representative regional receptor site for the PRD region (He et al., 2019; Xu et al., 2022). It is worth noting that the distances between the sites and the cities are around 20-30 km. In summary, TS and HS can represent the atmospheric characteristics of the North China Plain and the PRD region to some extent, respectively, so we take them as regional sites in this study.

To make it clearer, we have made the following modification in the revised manuscript,

"The other two sites, Mt. Tai (TS) and Heshan (HS), are taken as regional sites in this study. TS (1534 m a.s.l.) locates at the Taishan National Reference Climatological Station at the summit of Mt. Tai and in the middle of the North China Plain, which is less affected by anthropogenic emissions and has been widely used as a sampling site for researches on regional atmospheric pollution and atmospheric chemistry in Northern China (Chen et al., 2022; Jiang et al., 2020). The HS site locates at the Atmospheric Environmental Monitoring Super-station in Guangdong, China and downwind of the Pearl River Delta (PRD), which is mainly surrounded by farmland protection areas and forest land with no obvious industrial or urban traffic pollution sources in the vicinity, and has been used as a representative regional receptor site for the PRD region (He et al., 2019; Xu et al., 2022)."

References:

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Comment #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.*

Response to Comment #2: We thank the reviewer for the careful editing. We have rechecked and corrected the equation S9 as follows,

$$\frac{dSFE}{d\lambda} = -\frac{1}{4}\frac{dS(\lambda)}{d\lambda}\,\tau_{atm}^{2}(\lambda)\,(1 - F_c)\left[2(1 - \alpha_s)^2\,\beta(\lambda)\,MSE(\lambda) - 4\alpha_s\,MAE(\lambda)\right] \qquad (S9)$$

"where $dS(\lambda)/d\lambda$ is the solar irradiance ($W{\cdot}m^{-2}{\cdot}nm^{-1}$) obtained from the ASTM G173-03 reference spectra, τ_{atm} is the atmospheric transmission (0.79), Fc is the cloud fraction (0.6), α_s is the surface albedo (average 0.19), β is the backscatter fraction, MSE and MAE are the mass scattering efficiency and mass absorption efficiency of WSOC, respectively. It should be noted that $\beta = 0$ and only light absorption is considered in the calculation in this study." (in the Supplement)

Comment #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?

Response to Comment #3: We thank the reviewer for raising this good question. We have checked the light absorption spectra of nitrate and nitrite solutions reported in previous studies and found that the absorption of nitrate is mainly concentrated below 250 nm (Ai, 2019; Afsana et al., 2022; Dong et al., 2022; Li et al., 2016; Pons et al., 2017; Zhang et al., 2021; Zhang et al., 2023) (see Figure R1). That means, the absorbance interference of nitrate aerosols should be mainly below 250 nm. In this study, WSOC light absorbance from 250 to 500 nm has been used as input in PMF model in the revised manuscript, and the results indicate that the absorption spectra of all light-absorbing factors resolved by PMF are close to those of previously identified BrC light-absorbing species. Therefore, the impacts by light absorption of nitrate may be negligible on the PMF results.

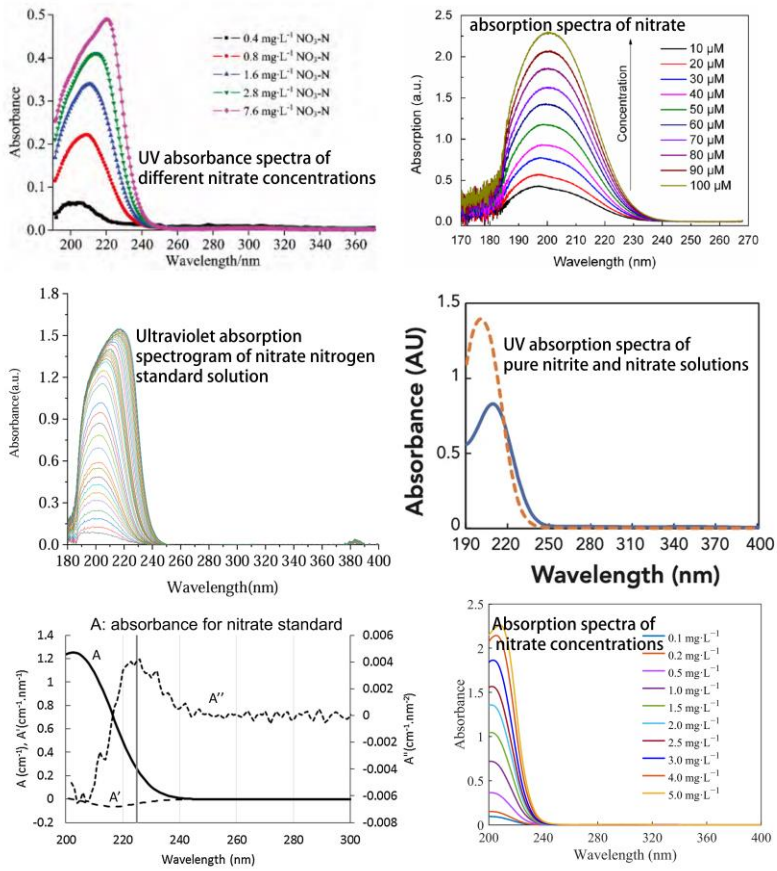

Figure R1. The light absorption spectra of nitrates measured in different studies.

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of forest aerosols in Hokkaido, Sci Rep, 12, 14379, https://doi.org/10.1038/s41598-022-18201-z, 2022.

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Comment #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?*

Response to Comment #4: We thank the reviewer for the constructive suggestion. In the revised manuscript, we have revised this section and briefly discussed the possible reasons for the spatial variation of carbonaceous components as follows,

"During the observation period, the mass concentrations of carbonaceous components (i.e., OC, EC and WSOC) increase with the increase of $PM_{2.5}$ concentration (see Figure S3), and exhibit significant spatial variations across the ten sites ($p < 0.05$). As shown in Figure S1 and Table S2, the average concentrations of OC, EC and WSOC observed at the ten sites ranged from 3.31 to 19.6 μg·m^{-3}, 0.35 to 2.86 μg·m^{-3}, and 1.97 to 10.6 μg·m^{-3}, respectively, with the highest mean mass concentrations of OC, EC and WSOC observed in CQ, XA, and HD, respectively, while the lowest values all observed in SH. Overall, the regional average carbonaceous component concentrations show

the spatial distribution trends of northwest China > southwest China > north China > east China > regional site ($p < 0.05$). This spatial variability may be attributed to differences in the sources of carbonaceous components and meteorological conditions. For example, the enhanced fuel (e.g., fossil fuels or biomass) combustion for winter heating activities may lead to increased emissions of carbonaceous components at sites in northern China (e.g., HD, XA, TJ and QD) (Wang et al., 2022a; Zhang et al., 2015). The unfavorable meteorological conditions such as low wind speeds may lead to the high concentration levels at sites in southwest China (e.g., CD: 1.16 ± 0.48 m·s^{-1}; CQ: 2.27 ± 0.89 m·s^{-1}). Moreover, concentrations of carbonaceous components in inland cities (i.e., HD, NJ, XA, CD, CQ) are much higher than those in coastal cities (i.e., TJ, QD, SH) ($p < 0.01$), which is consistent with that reported in previous studies that have shown that air masses from the ocean generally contain lower levels of aerosol content and carbonaceous components (Chen et al., 2023; Diesch et al., 2012; Mo et al., 2022; Zhang et al., 2022b). The 48-h backward air mass trajectory analysis shows that about 1/5 to 1/2 of the air masses that arrive at the coastal cities during the observation period pass through the ocean region, while the inland cities are predominately affected by continental air masses, which may contain a large number of anthropogenic aerosols (see Figure S4). Furthermore, it is worth noting that the regional site TS in NCP has a relatively low mass concentration of carbonaceous components compared to urban sites, which may be due to its high altitude (~1500 m) and low local anthropogenic activities (Jiang et al., 2020). In contrast, the mass concentrations of carbonaceous components at HS (another regional site) in the PRD region are relatively higher compared to TS site. The backward air mass trajectory analysis indicates that more than 80% of the air masses arriving at the HS site originated from the PRD region and are accompanied by low wind speeds (1.54 m·s^{-1} on average during the sampling period). This suggests that there may be high anthropogenic emissions in the PRD region during the winter sampling period."

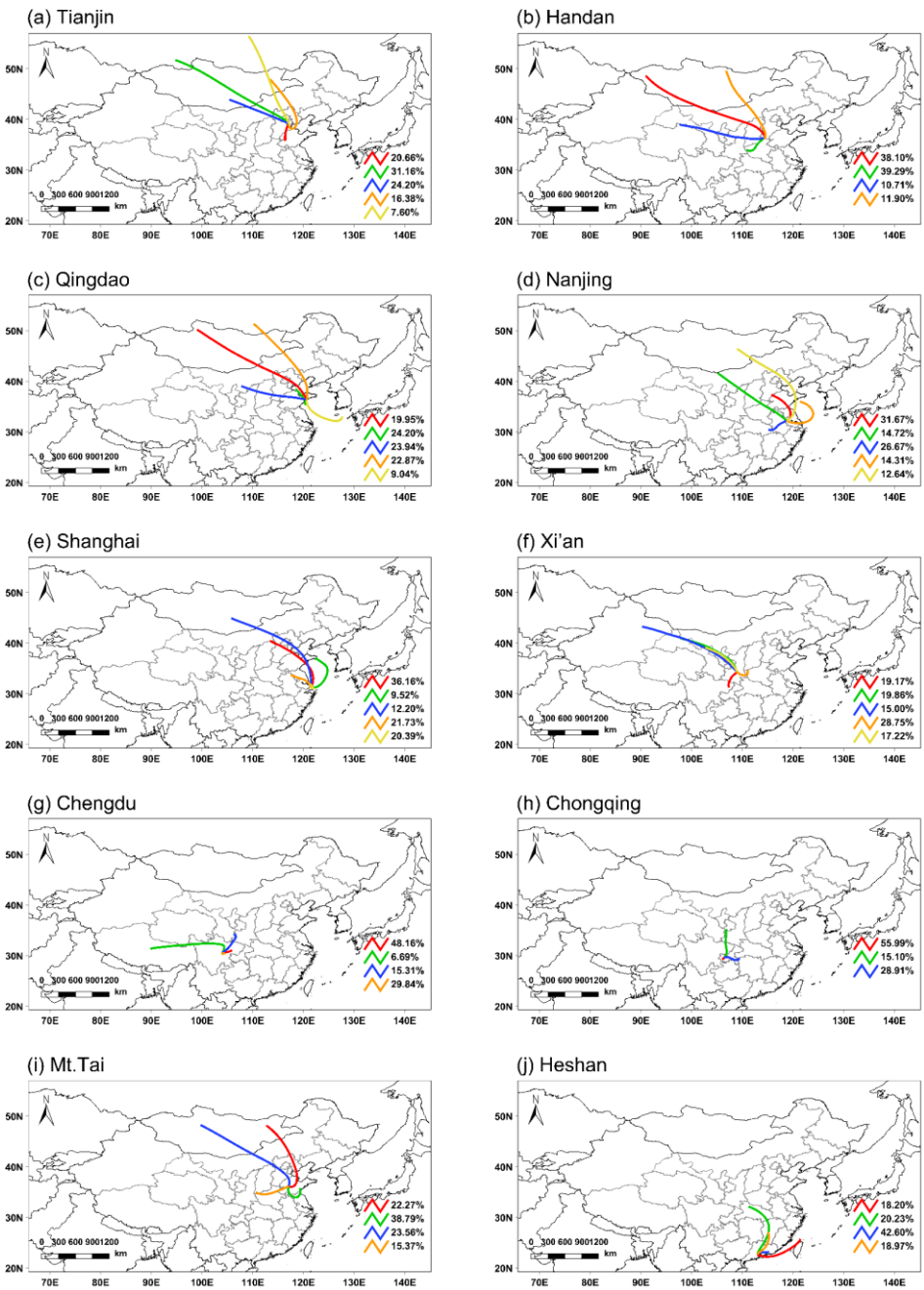

Figure S4. *Clusters of air masses derived from backward trajectory analysis at these ten sites. The 48-h backward trajectories at each site are calculated every 1 h and clustered at an ending height of 500 m above ground level based on the MeteoInfoMap software.*

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Comment #5: *Results and Discussion (section 3.1): The authors compared Abs_{365} and MAE_{365} 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?*

Response to Comment #5: We thank the reviewer for pointing this out. We have taken the reviewer's suggestion

and carried out a *t-test* (two-sample heteroskedasticity assumption) to check the significant differences in Abs_{365} and MAE_{365} in these regions, and made the following modifications in the revised manuscript,

"As shown in Table 1 and Figure S1, the average Abs_{365} (1.12 ± 0.53-13.1 ± 6.95 Mm^{-1}) and MAE_{365} (0.56 ± 0.11-1.26 ± 0.34 $m^2 \cdot g^{-1}$) of WSOC at the ten sites display significant spatial discrepancies ($p < 0.05$), with HD (SH) has the highest (lowest) average Abs_{365} (13.1 ± 6.95 Mm^{-1} (1.12 ± 0.53 Mm^{-1})) and MAE_{365} (1.26 ± 0.34 $m^2 \cdot g^{-1}$ (0.56 ± 0.11 $m^2 \cdot g^{-1}$)), respectively. MAE_{365} in SH, CD, TS and HS (0.56 ± 0.11-0.74 ± 0.24 $m^2 \cdot g^{-1}$ on average) are comparable to those reported in light-polluted areas such as in Guangzhou, Lulang, Waliguan, Urumqi in China and Los Angeles in the USA (0.48-0.81 $m^2 \cdot g^{-1}$) (Fan et al., 2016; Liu et al., 2018; Soleimanian et al., 2020; Wu et al., 2020; Xu et al., 2020; Zhong et al., 2023). However, they are all lower than those in TJ, HD, QD, NJ, XA and CQ (0.89 ± 0.22-1.26 ± 0.34 $m^2 \cdot g^{-1}$ on average), which are comparable to those reported in heavy pollution areas such as in Beijing, Xining, Yinchuan, Lanzhou, Taipei in China, and Patiala and Mohanpur in India (0.93-1.30 $m^2 \cdot g^{-1}$) (Cheng et al., 2016; Srinivas et al., 2016; Dey et al., 2021; Zhong et al., 2023; Ting et al., 2022). In this study, the light absorption of WSOC in different region is significantly different ($p < 0.05$), with the regional average Abs_{365} and MAE_{365} displaying as northwest China > southwest China > north China > east China > regional site. Moreover, the average Abs_{365} and MAE_{365} are higher in Northern China (including TJ, HD, QD, and XA, 7.34 ± 5.21 Mm^{-1} and 1.02 ± 0.29 $m^2 \cdot g^{-1}$, respectively) than in Southern China (including NJ, SH, CD, and CQ, 5.86 ± 3.91 Mm^{-1} and 0.78 ± 0.23 m^2 g^{-1}, respectively) and regional sites (e.g., TS and HS, 2.91 ± 1.38 Mm^{-1} and 0.72 ± 0.23 $m^2 \cdot g^{-1}$, respectively) ($p < 0.01$), and higher in inland (8.24 ± 4.75 Mm^{-1} and 0.91 ± 0.27 $m^2 \cdot g^{-1}$, respectively) than in coastal areas (4.37 ± 3.52 Mm^{-1} and 0.88 ± 0.32 $m^2 \cdot g^{-1}$, respectively) ($p < 0.01$, see Figure S5), which are consistent with the regional differences in carbonaceous component mass concentrations."

Comment #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.*

Response to Comment #6: Yes, we agree with the reviewer that "day versus night variability" in optical properties may contribute to inter-regional variability. However, it is not the case in this study and the effect of day versus night variability can be ignored in this study. The specific reasons are as follows:

(1) During the sampling period of this study, daytime and nighttime samples were only collected at three sites, including HD, NJ and TS. We compare Abs_{365} and MAE_{365} of daytime and nighttime WSOC at the three sites

and find that there is a small difference in light absorption between the daytime and nighttime WSOC (see Figure R2). Furthermore, the significance test of the daytime and nighttime data shows that the difference is not significant ($p > 0.05$, *t-test*).

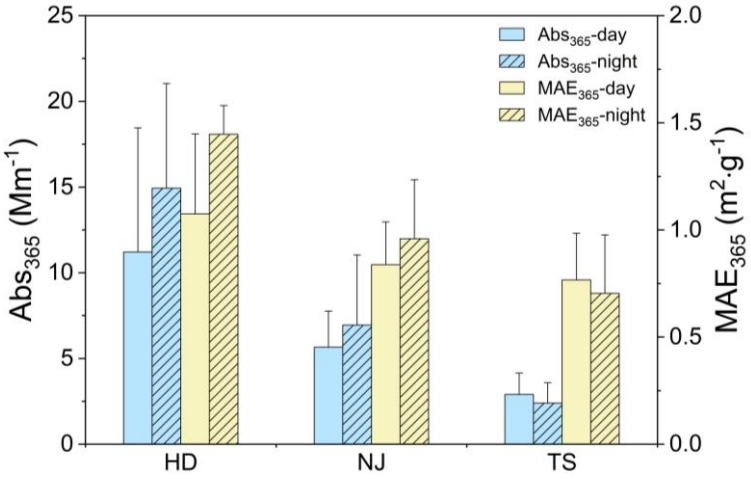

Figure R2. The average Abs_{365} and MAE_{365} values during daytime and nighttime at HD, NJ and TS.

(2) Since daytime and nighttime samples were not collected at all sites, in order to maintain consistency, we calculated the daily average values through daytime and nighttime samples during data processing, and then compared the differences of daily mean values among the ten sites, and did not compare the daytime and nighttime data in this study. Therefore, in the revised manuscript, the following sentence has been added.

"It is worth noting that in this study, the daily average of the parameters measured at each site is used for subsequent summary and comparison."

Comment #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.*

Response to Comment #7: Thanks for the reviewer's suggestion. In the revised manuscript, we have revised the statement and cited relevant references. The related paragraph has been revised as follows,

"Accordingly, the SFE within the wavelength range of 300-400 nm ($SFE_{300-400}$) and 300-700 nm ($SFE_{300-700}$) in different regions are calculated based on the measured MAE values. The dSFE/dλ spectra and integrated SFE values are shown in Figure S7. Overall, the integral SFE values exhibit significant spatial variations across the ten sites ($p < 0.01$), and the spatial distribution trend is similar to that of MAE_{365}. Among which, HD (SH) has the highest (lowest) $SFE_{300-400}$ (1.92 ± 0.51 $W \cdot g^{-1}$ (0.88 ± 0.17 $W \cdot g^{-1}$)) and $SFE_{300-700}$ values (4.50 ± 1.25 $W \cdot g^{-1}$ (1.82

± 0.38 W·g^{-1})). The similar variations in SFE and MAE values suggest that the stronger light-absorbing capacity of BrC may lead to an increase in its direct radiative forcing. This is consistent with previous studies, which suggest that more abundant BrC with stronger light-absorbing capacity may result in a remarkable increase in the direct radiative forcing of BrC (Deng et al., 2022; Zhang et al., 2020). It should be noted that $SFE_{300-400}$ accounts for more than 40% (38.6 ± 5.04%-48.9 ± 4.05% on average) of $SFE_{300-700}$ across the ten sites in this study, which is consistent with a previous study (Deng et al., 2022), indicating that the light absorption of WSOC plays a crucial role in the aerosol direct radiative forcing in the UV-Vis range. Notably, there is a significant negative correlation ($p < 0.01$) between SFE value and WSOC/OC at most sites (see Figure S7c and d). The WSOC/OC ratio has been used to infer the degree of secondary aerosol formation or aging of aerosols (Dasari et al., 2019; Ram et al., 2012), therefore, the reduction of SFE value of WSOC may be related to the secondary or aged organic aerosols. Previous studies have indicated that the light absorption capacity of secondary BrC is usually lower than that of primary BrC (Fan et al., 2018; Tang et al., 2020; Zhong and Jang, 2011). Furthermore, BrC chromophores with aromatic rings and nitro and phenolic groups may undergo photolysis or photochemical oxidation under sunlight, resulting in photo-bleaching and decrease of light absorption capacity and radiation effect of WSOC (Dasari et al., 2019). However, it is noted that this is only a speculation, and further research is needed in the future in conjunction with atmospheric chemical processes and WSOC component analysis."

References:

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Comment #8: *Fig. 6c-d: Is this the integrated absorbance from 250-400 nm, clarify?*

Response to Comment #8: We are sorry for the confusion. In the original draft, we calculated the averaged light absorption contribution with light absorbance at each wavelength. In the revised manuscript, we recalculated the average contribution based on the integrated absorbance from 250-500 nm of each factor.

For clarity, we have revised the title of the new Figure 5 (original Figure 6) in the revised manuscript as follows,

"***Figure 5.*** *The average light absorption spectra of the absorption factors resolved by PMF model at (a) East China sites (unimodal absorption spectral type) and (b) outside East China sites (bimodal absorption spectral type), as well as the average contribution by each factor calculated according to the integral absorbance from 250-500 nm at both types of sites (panel c and d).*"

Comment #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.*

Response to Comment #9: We thank the reviewer for pointing this out. In the revised manuscript, this paragraph has been revised as follows,

"To investigate the influence of air quality levels on the light absorption properties of WSOC, the sampling days are classified into five pollution levels including clean (0-35 μg·m^{-3}), relatively clean (35-75 μg·m^{-3}), slightly polluted (75-115 μg·m^{-3}), moderately polluted (115-150 μg·m^{-3}), and heavily polluted (> 150 μg·m^{-3}) according to the national ambient air quality daily Grade-II standard threshold values and ambient air quality indices. As shown in Figure 7a and b, Abs_{365} and MAE_{365} of WSOC both increase with the increase of pollution levels, in which Abs_{365} changes significantly ($p < 0.01$) while MAE_{365} changes relatively gently. The enhancement of WSOC light absorption under high pollution conditions may be related to the increase of WSOC concentration, light absorption capacity and light-absorbing species. Previous studies have reported that the mass fractions of oxidized organic aerosols increase significantly with the increase of $PM_{2.5}$ mass concentration, and the oxidized organic aerosols contain a large number of light-absorbing species such as nitroaromatics compounds (You et al., 2024). In this study, the relative abundances of O-H, C=C and R-ONO_2 functional groups, which are related to aromatic compounds and have a good positive correlation with the light absorption of WSOC, increase with $PM_{2.5}$ mass concentration (see Figure S14, and discussion in the next section). Additionally, the accumulation of anthropogenic emissions (especially those sources with strong light-absorbing BrC such as biomass burning and coal combustion sources) at high pollution levels will lead to an increase in BrC chromophore types and overall light absorption capacity (Li et al., 2020a; Tang et al., 2020; Wei et al., 2020)."

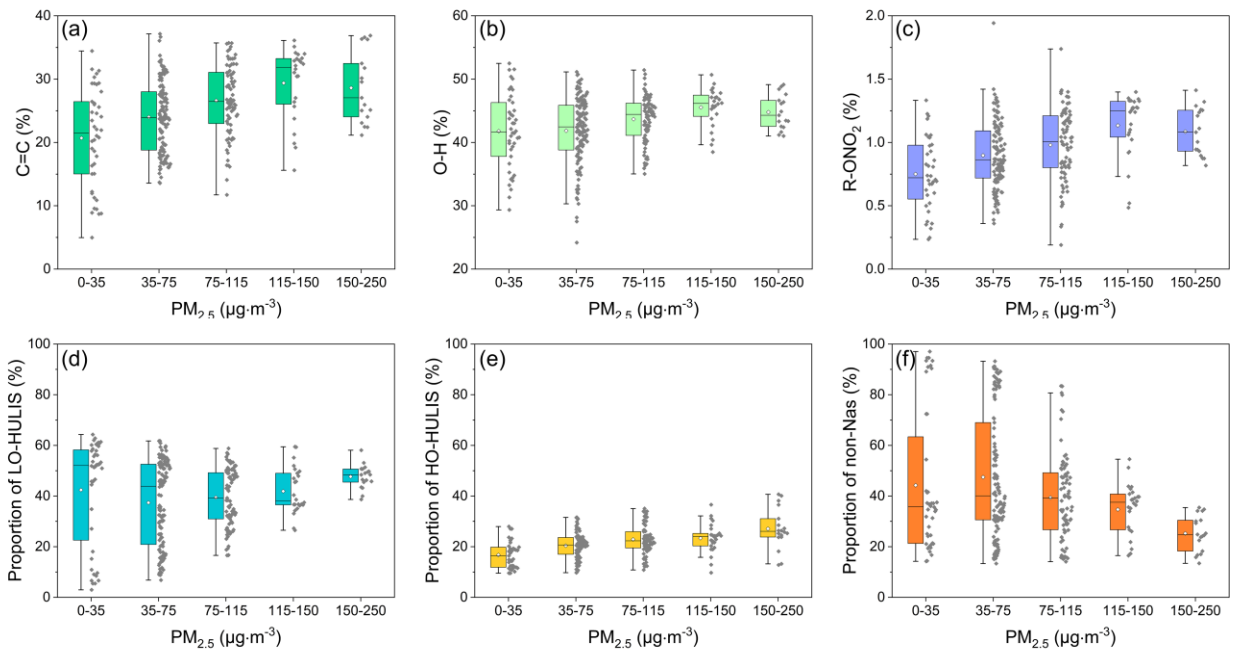

Figure S14. *Variations of the relative abundance of functional groups (a) C=C, (b) O-H and (c) R-ONO_2 and the proportion of (d) LO-HULIS, (e) HO-HULIS and (f) non-Nas fluorophores with $PM_{2.5}$ mass concentrations.*

References:

Li, J. J., Zhang, Q., Wang, G. H., Li, J., Wu, C., Liu, L., Wang, J. Y., Jiang, W. Q., Li, L. J., Ho, K. F., and Cao, J. J.: Optical properties and molecular compositions of water-soluble and water-insoluble brown carbon (BrC) aerosols in northwest China, Atmospheric Chemistry and Physics, 20, 4889-4904, https://doi.org/10.5194/acp-20-4889-2020, 2020.

Tang, J., Li, J., Su, T., Han, Y., Mo, Y. Z., Jiang, H. X., Cui, M., Jiang, B., Chen, Y. J., Tang, J. H., Song, J. Z., Peng, P. A., and Zhang, G.: Molecular compositions and optical properties of dissolved brown carbon in biomass burning, coal combustion, and vehicle emission aerosols illuminated by excitation-emission matrix spectroscopy and Fourier transform ion cyclotron resonance mass spectrometry analysis, Atmospheric Chemistry and Physics, 20, 2513-2532, https://doi.org/10.5194/acp-20-2513-2020, 2020.

Wei, Y., Chen, H., Sun, H., Zhang, F., Shang, X., Yao, L., Zheng, H., Li, Q., and Chen, J.: Nocturnal $PM_{2.5}$ explosive growth dominates severe haze in the rural North China Plain, Atmospheric Research, 242, https://doi.org/10.1016/j.atmosres.2020.105020, 2020.

You, B., Zhang, Z., Du, A., Li, Y., Sun, J., Li, Z., Chen, C., Zhou, W., Xu, W., Lei, L., Fu, P., Hou, S., Li, P., and Sun, Y.: Seasonal characterization of chemical and optical properties of water-soluble organic aerosol in Beijing, Science of The Total Environment, 930, 172508, https://doi.org/10.1016/j.scitotenv.2024.172508, 2024.

Comment #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?*

Response to Comment #10: We thank the reviewer for the good and enlightening question.

(1) Firstly, we have to admit that focusing only on WSOC is a shortcoming of this study. However, due to the limited filter area of the samples in this study and the uncertainty of the existing characterization methods for water-insoluble brown carbon, we only focused on the optical properties of WSOC and the relevant measurement of water-insoluble brown carbon was not carried out in this study.

(2) Secondarily, according to the OC and WSOC measurement data, WISOC (calculated as the difference between OC and WSOC) accounted for 26.7% to 46.6% of the OC masses across different sites, with WISOC/WSOC mass ratio about 0.39 to 0.92 (see Table R1). That means, the mass concentrations of WISOC were lower than that of WSOC.

Previous studies have reported that the mass absorption efficiency of WISOC was about 1.4-2.3 times and 1.6

times of that of WSOC in source samples and ambient samples, respectively (Yan et al., 2017; Yan et al., 2020). Therefore, we calculate the light absorption coefficient of WISOC ($Abs_{365, WISOC}$) using the factor of 1.6 for ambient samples, as well as the associated ratio of $Abs_{365, WISOC}$ to the light absorption coefficient of WSOC ($Abs_{365, WISOC}/Abs_{365, WSOC}$) at the ten sites in this study.

Accordingly, the $Abs_{365, WISOC}$ should be around 0.62 to 1.47 times of $Abs_{365, WSOC}$ among the ten sites. As shown in Table R1, the $Abs_{365, WISOC}$ at some sites (e.g., TJ, HD, QD, XA, CQ and TS) are higher than $Abs_{365, WSOC}$, which should be given special attention, while $Abs_{365, WSOC}$ is much stronger at other sites (e.g., NJ, SH, CD and HS). *However*, it is worth mentioning that among the ten sites, only the difference in light absorption between WSOC and WISOC in QD and CD is significant ($p < 0.05$, *t-test*), with the $Abs_{365, WISOC}$ at the two sites being 1.47 and 0.62 times that of $Abs_{365, WSOC}$, respectively.

Table R1. The mass concentrations and light absorption coefficients of WSOC and WISOC in this study.

Sites	WISOC/WSOC	WISOC/OC (%)	$Abs_{365, WSOC}$ (Mm^{-1})	$Abs_{365, WISOC}$ (Mm^{-1})	$Abs_{365, WISOC}/Abs_{365, WSOC}$
Tianjin (TJ)	0.74±0.36	40.1±11.8	5.57±3.83	7.08±7.15	1.18±0.58
Handan (HD)	0.90±0.41	44.8±13.3	13.1±6.95	18.3±7.57	1.45±0.65
Qingdao (QD)	0.92±0.55	44.9±11.6	4.80±3.09	7.31±5.30	1.47±0.89
Nanjing (NJ)	0.59±0.31	34.8±11.9	6.26±3.26	6.72±6.66	0.94±0.50
Shanghai (SH)	0.66±0.19	39.0±6.99	1.12±0.53	1.16±0.66	1.06±0.30
Xi'an (XA)	0.77±0.23	42.7±6.93	10.6±4.42	12.6±5.56	1.24±0.36
Chengdu (CD)	0.39±0.22	26.7±10.0	5.99±2.61	3.67±2.27	0.62±0.35
Chongqing (CQ)	0.89±0.18	46.6±4.94	10.6±4.10	15.0±6.15	1.42±0.29
Mt. Tai (TS)	0.73±0.69	34.9±19.6	2.66±1.22	3.51±3.95	1.16±1.11
Heshan (HS)	0.61±0.30	35.6±11.9	3.76±1.55	3.86±2.65	0.97±0.47

Taken together, although $MAE_{365, WISOC}$ and $Abs_{365, WISOC}$ may be stronger compared to $MAE_{365, WSOC}$ and $Abs_{365, WSOC}$, the estimated light absorption by WSOC and WISOC at the most sites during the study period show insignificant difference ($p > 0.05$, t-test). Relatively, the results of this study suggest that the light absorption contribution of WISOC in winter in northern cities is more noteworthy.

(3) Finally, the findings of this study show that combining multiple spectral results (light absorption spectrum, fluorescence spectrum and infrared spectrum) can provide a deeper understanding of the light-absorbing characteristics of WSOC. This study can also provide reference for further study of WISOC light absorption properties and its structural composition.

(4) Therefore, we have added a description of water-soluble and water-insoluble BrC in the revised manuscript, and added the limitations that this study only focuses on water-soluble BrC in the conclusion sections as follows,

① In the introduction section:

"Solvent-soluble organic carbon (e.g., water-soluble organic carbon, WSOC; methanol-soluble organic carbon, MSOC) is often used to act as a substitute of BrC. In particular, light absorption of WSOC has been extensively studied, due to its widespread presence and high atmospheric abundance in the atmosphere, as well as mature extraction methods, although some previous studies have indicated that water-insoluble OC (WISOC) contains more light-absorbing BrC (Cao et al., 2021; Chen et al., 2024b; Cheng et al., 2016; Yan et al., 2017). Absorption and fluorescence spectroscopy are two of the most widely used methods to reveal optical properties of WSOC (Wang et al., 2022b; Wu et al., 2021)."

"*In this study, $PM_{2.5}$ is collected from ten sites in different regions of China. The mass concentration, light absorption and fluorescent spectra, and functional group structures of WSOC (a substitute of water-soluble BrC) at the ten sites are analyzed using a unified method. The objectives of this study include: …Furthermore, this study can also provide reference for the future study of the light absorption properties and structural composition of WISOC.*"

② In the conclusion section:

"Additionally, it is important to note that this study only focused on WSOC. Since the WISOC may have stronger light absorption capacity, further research on light absorption, composition and structure of WISOC (especially in northern China in winter) and the correlation between them are also needed in the future."

References:

Cao, T., Li, M., Zou, C., Fan, X., Song, J., Jia, W., Yu, C., Yu, Z., and Peng, P. a.: Chemical composition, optical properties, and oxidative potential of water- and methanol-soluble organic compounds emitted from the combustion of biomass materials and coal, Atmospheric Chemistry and Physics, 21, 13187-13205, https://doi.org/10.5194/acp-21-13187-2021, 2021.

Chen, H., Zhou, R., Fang, L., Sun, H., Yang, Q., Niu, H., Liu, J., Tian, Y., Cui, M., and Yan, C.: Variations in optical properties of water- and methanol-soluble organic carbon in $PM_{2.5}$ in Tianjin and Handan over the wintertime of 2018-2020, Atmospheric Research, 303, 107332, https://doi.org/10.1016/j.atmosres.2024.107332, 2024b.

Cheng, Y., He, K. B., Du, Z. Y., Engling, G., Liu, J. M., Ma, Y. L., Zheng, M., and Weber, R. J.: The characteristics of brown carbon aerosol during winter in Beijing, Atmospheric Environment, 127, 355-364, https://doi.org/10.1016/j.atmosenv.2015.12.035, 2016.

Yan, C., Zheng, M., Bosch, C., Andersson, A., Desyaterik, Y., Sullivan, A.P., Collett, J.L., Zhao, B., Wang, S.X., He, K.B., Gustafsson, Ö. Important fossil source contribution to brown carbon in Beijing during winter. Scientific

Reports, 7, 43182, https://doi.org/10.1038/srep43182, 2017.

Yan, F., Kang, S., Sillanpaa, M., Hu, Z., Gao, S., Chen, P., Gautam, S., Reinikainen, S. P., and Li, C.: A new method for extraction of methanol-soluble brown carbon: Implications for investigation of its light absorption ability, Environ Pollut, 262, 114300, https://doi.org/10.1016/j.envpol.2020.114300, 2020.

Minor Comments:

Comment #11: *Line 30-34: "The light absorption factors…..impact on fluorophores." Confusing sentence. Rewrite it.*

Response to Comment #11: Thanks for pointing it out. This sentence has been rewritten as follows,

"The light absorption factors resolved by light absorption spectra-based positive matrix factorization model, and the abundance of aromatic O-H and C=C functional groups determined by FTIR, both indicate that aromatic compounds are significant light-absorbing substances in WSOC and have a significant impact on fluorophores."

Comment #12: *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 $W \cdot m^{-2}$) is about 30% of black carbon (+0.17 $W \cdot m^{-2}$)". Is this global average or only valid for China? Please clarify.*

Response to Comment #12: We thank the reviewer for pointing this out and apologize for the unclear statement. This statement is based on the simulation of global radiation effect of BrC by Wang et al. (2018), which is the global average. We have revised this statement as follows,

"It has been simulated that BrC contributes up to 72% of the total light absorption of aerosols at 370 nm globally and the global direct radiative effect of BrC (+0.048 $W \cdot m^{-2}$) is about 30% of black carbon (+0.17 $W \cdot m^{-2}$) (Wang et al., 2018)."

References:

Wang, X., Heald, C. L., Liu, J., Weber, R. J., Campuzano-Jost, P., Jimenez, J. L., Schwarz, J. P., and Perring, A. E.: Exploring the observational constraints on the simulation of brown carbon, Atmospheric Chemistry and Physics, 18, 635-653, https://doi.org/10.5194/acp-18-635-2018, 2018.

Comment #13: *Line 51: Use either "commonly" or "widely". One of them is redundant.*

Response to Comment #13: Thanks for the reviewer's kind reminder. The word "commonly" has been removed and the sentence has been revised as follows,

"Absorption and fluorescence spectroscopy are two of the most widely used methods to reveal optical properties of WSOC."

Comment #14: *Line 51-53: Make it two sentences.*

Response to Comment #14: We appreciate the reviewer's kind reminder. This sentence has been divided into two sentences in the revised manuscript, and the specific changes are as follows,

"Solvent-soluble organic carbon (e.g., water-soluble organic carbon, WSOC; methanol-soluble organic carbon, MSOC) is often used to act as a substitute of BrC. In particular, light absorption of WSOC has been extensively studied, due to its widespread presence and high atmospheric abundance in the atmosphere, as well as mature extraction methods, although some previous studies have indicated that water-insoluble OC (WISOC) contains more light-absorbing BrC (Cao et al., 2021; Chen et al., 2024b; Cheng et al., 2016; Yan et al., 2017). Absorption and fluorescence spectroscopy are two of the most widely used methods to reveal optical properties of WSOC (Wang et al., 2022b; Wu et al., 2021)"

References:

Cao, T., Li, M., Zou, C., Fan, X., Song, J., Jia, W., Yu, C., Yu, Z., and Peng, P. a.: Chemical composition, optical properties, and oxidative potential of water- and methanol-soluble organic compounds emitted from the combustion of biomass materials and coal, Atmospheric Chemistry and Physics, 21, 13187-13205, https://doi.org/10.5194/acp-21-13187-2021, 2021.

Chen, H., Zhou, R., Fang, L., Sun, H., Yang, Q., Niu, H., Liu, J., Tian, Y., Cui, M., and Yan, C.: Variations in optical properties of water- and methanol-soluble organic carbon in $PM_{2.5}$ in Tianjin and Handan over the wintertime of 2018-2020, Atmospheric Research, 303, 107332, https://doi.org/10.1016/j.atmosres.2024.107332, 2024b.

Cheng, Y., He, K. B., Du, Z. Y., Engling, G., Liu, J. M., Ma, Y. L., Zheng, M., and Weber, R. J.: The characteristics of brown carbon aerosol during winter in Beijing, Atmospheric Environment, 127, 355-364, https://doi.org/10.1016/j.atmosenv.2015.12.035, 2016.

Yan, C., Zheng, M., Bosch, C., Andersson, A., Desyaterik, Y., Sullivan, A.P., Collett, J.L., Zhao, B., Wang, S.X., He, K.B., Gustafsson, Ö. Important fossil source contribution to brown carbon in Beijing during winter. Scientific Reports, 7, 43182, https://doi.org/10.1038/srep43182, 2017.

Comment #15: *Line 55-59: Grammatical mistakes at many places. Some sentences are confusing. Revise it.*

Response to Comment #15: We apologize for grammatical mistakes and thank the reviewer for pointing it out. In the revised manuscript, this paragraph has been revised and these sentences have been revised as follows,

"By light-absorption spectroscopy analysis, light absorption characteristics and capabilities of WSOC from different sources or environments are usually characterized by the absorption coefficient or mass absorption efficiency of a specific wavelength over the range of 360-370 nm (average 365 nm) (Hecobian et al., 2010). And the direct radiative forcing of WSOC can be further estimated by simplified radiative forcing models combined with the measured absorption coefficient. The fluorescence spectra obtained by fluorescence spectroscopy measurement could be used to characterize fluorescence fingerprints of WSOC, and provide source-related information of WSOC according to fluorescent indices. In addition, the parallel factor analysis (PARAFAC) could be used to analyze fluorescence spectra and help to infer the chemical and structural characteristics of WSOC chromophores (Chen et al., 2020; Wu et al., 2021)."

Comment #16: *Line 61: Should be "Spectroscopy-based studies conducted…"*

Response to Comment #16: We thank the reviewer's careful editing and kind reminder. The sentence has been revised accordingly,

"Spectroscopy-based studies conducted in different countries or regions all over the world show that there are significant spatiotemporal differences in the light absorption characteristics of WSOC."

Comment #17: *Line 81: "methods are often used separately in previous studies"?*

Response to Comment #17: We apologize for the confusion. We want to express that the results of different spectral methods (e.g., absorption spectra, fluorescence spectra and FTIR spectra) were often discussed separately in previous studies, without delving into the relationship between these spectral results. Exploring the relationship between different spectra is one of the objectives of this study. To make it more accurate, we have revised this sentence as follows,

"However, in previous spectroscopy-based studies, the results of different spectral methods are often discussed separately, without in-depth discussion of the relationship between different spectra, which limits the full and comprehensive prediction of the optical and structural characteristics of BrC."

Comment #18: *Line 82-86: Difficult to follow as this is a very long sentence. I suggest to break it into smaller sentences.*

Response to Comment #18: We appreciate the reviewer's kind reminder and apologize for the bad feeling. We have taken the reviewer's suggestion and broken the long sentence into smaller ones.

"In this study, $PM_{2.5}$ is collected from ten sites in different regions of China. The mass concentration, light absorption and fluorescent spectra, and functional group structures of WSOC (a substitute of water-soluble BrC) at the ten sites are analyzed using a unified method. The objectives of this study include: (1) to explore the spatial heterogeneity of optical properties of WSOC in different regions of China and its influencing factors, (2) to reveal the relationship between light absorption, fluorescence and functional group structure of WSOC, and (3) to quantify the contribution of fluorescent chromophores to the light absorption of WSOC on the basis of multi-site spectral datasets."

Comment #19: *Line 110: should be "0.45 μm pore-size PTFE syringe filter".*

Response to Comment #19: Thanks for the reviewer's kind reminder. The sentence has been revised accordingly.

"A portion of each filter (about 6 cm^2) is extracted with ultrapure water (>18.2 MΩ·cm, 25 °C, Direct-Q, Millipore) by ultra-sonication for 30 min, and then the extract is filtered through a 0.45 μm pore-size PTFE syringe filter (Pall, USA) to remove water-insoluble materials."

Comment #20: *Line 118-120: Break it into two sentences like "…:calculated. More details can be found in Text S2."*

Response to Comment #20: We take the reviewer's suggestion and break it into two sentences accordingly.

"Light absorption parameters such as light absorption coefficients at 365 nm (Abs_{365}), mass absorption efficiency at 365 nm and 405 nm (MAE_{365} and MAE_{405}), Ångström exponent over 300-500 nm ($AAE_{300-500}$) are calculated. More details on the calculation methods can be found in Text S2."

Comment #21: *Line 152: "Additionally, the XGBoost model is also used to…" "also" is redundant in this sentence.*

Response to Comment #21: Thanks for the kind reminder. "also" has been removed from this sentence accordingly, and this sentence has been revised to,

"Additionally, the XGBoost model is used to evaluate the influence of conventional gas parameters (e.g., CO, SO_2, O_3, NO_2) on the light absorption of WSOC."

Comment #22: *Line 169: "…:mass concentrations of carbonaceous components at HS site are not that low…" Compared to what, clarify?*

Response to Comment #22: We are sorry for the confusion. The sentence has been revised as follows,

"Furthermore, it is worth noting that the regional site TS in NCP has a relatively low mass concentration of carbonaceous components compared to urban sites, which may be due to its high altitude (~1500 m) and low local anthropogenic activities (Jiang et al., 2020). In contrast, the mass concentrations of carbonaceous components at HS (another regional site) in the PRD region are relatively higher compared to TS site. The backward air mass trajectory analysis indicates that more than 80% of the air masses arriving at the HS site originated from the PRD region and are accompanied by low wind speeds (1.54 $m \cdot s^{-1}$ on average during the sampling period). This suggests that there may be high anthropogenic emissions in the PRD region during the winter sampling period."

References:

Jiang, Y., Xue, L., Gu, R., Jia, M., Zhang, Y., Wen, L., Zheng, P., Chen, T., Li, H., Shan, Y., Zhao, Y., Guo, Z., Bi, Y., Liu, H., Ding, A., Zhang, Q., and Wang, W.: Sources of nitrous acid (HONO) in the upper boundary layer and lower free troposphere of the North China Plain: Insights from the Mount Tai Observatory, Atmospheric Chemistry and Physics, 20, 12115-12131, https://doi.org/10.5194/acp-20-12115-2020, 2020.

Comment #23: *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?*

Response to Comment #23: Thanks for the good question. In this study, a two-sample t-test under heteroscedasticity with the 95% confidence level was used to evaluate the significance level of data differences. In the section "*2.6 Relationship and influencing factor analysis*" of the revised manuscript, we have added the following explanation of the test method used in this study.

"The calculation is mainly carried out through SPSS software (IBM SPSS Statistics 23). Notably, t-test (two-sample testing under heteroscedasticity, at the 95% confidence level) is conducted to evaluate the significance level of data differences in this study."

Comment #24: *Line 308-309: The sentence is confusing. Rewrite it.*

Response to Comment #24: We are sorry for the confusion. The sentence has been revised as follows,

"The contributions by different light absorption factors vary significantly at different wavelengths (see Figure S12)."

Comment #25: *Line 341: Change "great" to "large".*

Response to Comment #25: Thanks. The relevant sentence has been deleted in the revised manuscript.

Comment #26: *Line 378: Typo? "WOSC" should be "WSOC"*

Response to Comment #26: We apologize for the typo and thank the reviewer for the correction. "WOSC" has been replaced by "WSOC" in the revised manuscript.

"…indicating that the fluorescent components may contribute to WSOC light absorption to some extent."

Comment #27: *"In contrast, the relationships between MAE_{365} values and functional groups may differ from Abs_{365}. Similarly, C=C, O-H and R-ONO_2 exhibit the strongest correlations with MAE_{365} at most sites (e.g., TJ, QD, SH, TS, and HS)." Similar to what? These sentences are confusing. Rewrite them.*

Response to Comment #27: We apologize for the confusion. The sentence has been rewritten as follows,

"In contrast, the relationships of functional groups with MAE_{365} are slightly different from that with Abs_{365}. Overall, C=C, O-H and R-ONO_2 exhibit the strongest correlations with MAE_{365} at most sites (e.g., TJ, QD, SH, TS, and HS). C=O and C-O, which may be related to carboxylic acids, phenols and esters, show positive correlations with MAE_{365} at some sites (e.g., HD, NJ, XA and CQ)."

Comment #28: *Line 413: "discrepancies" doesn't seem to be the write word here. Replace it.*

Response to Comment #28: We thank the reviewer's kind reminder. The "discrepancies" has been replaced by "spatial variations", and the sentence has been revised as follows,

"Based on the same measurement methods and data processing processes, light absorption, fluorescence and FTIR spectra analysis are combined to investigate the optical properties and functional group characteristics of WSOC at ten sites in different regions of China. The spatial variations at various sites and the relationships between

absorbance, fluorescence, and functional groups of WSOC are revealed."