Comments:

I appreciate the author's responses and their efforts to address the questions in the revised manuscript. However, I regret to say that I still think some of the discussions in the result part are not clear enough. I have some extra questions as listed below:

Re: Thanks for your valuable comments, which is of great help to improve the quality of the manuscript. According to your comments, we have carefully revised the manuscript, and responded to all comments point by point, and explained how the reviewers' comments and suggestions are handled in the current manuscript.

General comments:

1. In section 2.1, line 128-129, samples were collected on the quartz fiber filters and the $PM_{2.5}$ concentrations were determined by weighing the filters before and after collection. According to past experience, quartz fiber filters are very fragile and flaky; the loss of fibers from the filter during the handling and sampling will be inevitable and thus make this type of filter VERY challenging for mass weighing, and probably impossible for the accuracy of 0.01 mg claimed here. Therefore, it is not clear to me how the accuracy of the $PM_{2.5}$ concentrations can be guaranteed in this study.

Re: Thanks. We agreed with your comments that "quartz fiber filters are very fragile and flaky; the loss of fibers from the filter during the handling and sampling will be inevitable and thus make this type of filter VERY challenging for mass weighing, and probably impossible for the accuracy of 0.01 mg claimed here". Based on your comments, we conducted a group of experiments to test the losing of fibers during the sample collection. In our experiment, the sample filters were put in the air sample holder for 5 min without pumping air, then the filters were wrapped in prebaked aluminum foil. Before and after sampling, the filters were weighed at 25°C and 50% RH on a microbalance. This treatment was repeated for three times. As indicated in

the following Table A, the weights of two PM$_{2.5}$ samples were 41.2±0.5 mg and 34.7±0.7 mg, respectively. No significant losing of fibers were observed.

The 0.01 mg is the Readability of the microbalance in this study, which is not the accuracy of the weighting of filter samples. We are sorry for this error in the original manuscript. In the current manuscript, we have revised that to "The mass accuracy achieved was < 2% based on triplicate analyses of filter samples". Please see Lines 128-129.

Table A: The weights of PM$_{2.5}$ filter samples determined at three times.

	Sample 1 (mg)	Sample 2 (mg)
Original sample	41.3	34.3
1	41.7	35.2
2	41.2	35.4
3	40.5	34.0
Average	41.2	34.7
Standard derivation (SD)	0.5	0.7
Relative standard derivation (RSD)	1.2	2.0

2. Related to the first comment, the claimed accuracy is 0.01mg, which is 10 µg; however, the reported masses of PM$_{2.5}$ are data like 9-35 µg, 18+-3.3 µg, etc. This seems a bit strange to me.

Re: Thank for your question. In this study, the mass of PM$_{2.5}$ were shown as PM$_{2.5}$ concentration in air (µg m^{-3}). They were calculated based on the following equation:

$$C = \frac{W \times 1000}{V \times t}$$

Where, C is the concentration of PM$_{2.5}$ in air (µg m^{-3}), W is the weight of PM$_{2.5}$ sample (mg), V is the flow rate of PM$_{2.5}$ sampler (1.0 m^3 min^{-1}), t is the sampling time (min). In this study, the weight of PM$_{2.5}$ samples collected were 13−166 mg, the calculated PM$_{2.5}$ concentrations were 9.3−115 µg m^{-3}. In the manuscript, the data such as "9-35, 18+-3.3", etc, is the PM$_{2.5}$ concentration (µg m^{-3}) rather than the PM$_{2.5}$ mass weight (mg). Therefore, it is scientifically reasonable.

3. Line 254-255: This added sentence is just iterating sentences 251-253 in a different format (i.e., a higher AAE value means a stronger wavelength dependence). To me, this is not explaining (i.e., higher wavelength dependence could result from higher molecular weights, since the HULIS fraction could isolate most of the large molecules from the WSOC; or it could also mean a higher degree of conjugation in molecular structure in HULIS, as they are the hydrophobic fraction, etc). in the first round of review, the previous suggestion of a more specific explanation of the results does not mean simply explaining the meaning of the factor itself.

Re: Thanks. We are apologized for our insufficient revision in the manuscript. In this study, AAE is a measure of the wavelength dependence of BrC light absorption, which appear to be related to the chemical composition of chromophores and the fitting wavelength ranges (Zhang et al., 2013; Chen et al., 2016; Park et al., 2018). Since the light absorption of BrC mostly occurs in near-UV and visible wavelengths, AAE (330-400 nm) was usually reported in many previous studies and also used in this study. As shown in Figure 1i, the AAE values for HULIS were obviously higher than those for WSOC in the same sample, indicating that light absorption of HULIS is more wavelength-dependent than that of WSOC. Similar results were also observed in previous studies (Park et al., 2018; Jiang et al., 2020; Cao et al., 2021). We think that this difference could be associated with the differences in chemical composition of chromophores in WSOC and HULIS. As shown in Table S2, the E_{250}/E_{365} values of HULIS (5.3–5.6) all higher than that (4.4–5.1) of WSOC, suggested that the light-absorbing species in HULIS may have relative lower aromaticity and/or lower molecular weight than those in WSOC (Chen et al., 2016; Li and Hur, 2017). This difference may be attributed to a fraction of higher MW species remained in the HLB column due to irreversible adsorption and/or incomplete elution (Fan et al., 2012). As the results, the light-absorbing organic species in the HULIS fractions have relative higher absorption at UV and short visible wavelengths and relative lower absorption at long visible wavelengths, which resulting in relative higher AAE values. We have

revised that in the current manuscript. Please see Lines 249-260.

Table S2. Average values of Abs_{365}, MAE_{365}, AAE, and E_{250}/E_{365} of WSOC and HULIS in $PM_{2.5}$ samples.

		Overall	Clean-I	Haze-I	Haze-II	Clean-II
Abs_{365}	WSOC	2.5 ± 2.0	0.76 ± 0.18	4.3 ± 2.0	3.9 ± 1.1	0.89 ± 0.35
	HULIS	1.8 ± 1.6	0.55 ± 0.06	3.4 ± 1.5	2.6 ± 0.85	0.64 ± 0.32
MAE_{365}	WSOC	1.0 ± 0.21	1.1 ± 0.16	0.76 ± 0.05	0.83 ± 0.07	1.1 ± 0.14
	HULIS	1.1 ± 0.27	1.3 ± 0.22	0.91 ± 0.03	0.95 ± 0.11	1.3 ± 0.27
AAE	WSOC	5.2 ± 0.34	4.9 ± 0.55	5.0 ± 0.63	4.4 ± 0.20	4.7 ± 0.33
	HULIS	6.2 ± 0.20	6.2 ± 0.16	6.1 ± 0.09	6.2 ± 0.18	6.1 ± 0.28
E_{250}/E_{365}	WSOC	4.8 ± 0.49	5.1 ± 0.70	4.9 ± 0.43	4.4 ± 0.15	4.8 ± 0.31
	HULIS	5.4 ± 0.35	5.6 ± 0.36	5.3 ± 0.15	5.4 ± 0.29	5.4 ± 0.52

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4. I found it interesting that the authors saw the AAE of HULIS fractions from all four stages are similar (line 256-257), whereas later in the manuscript, the analyses showed that HULIS collected from haze days have a slightly higher molecular weight and also higher oxidation of aromatics than those in clean days. Potentially, these two could lead to different trends in AAE: higher MW could lead to the shifting to a more humic-like structure (as stated in cited paper as Wong et al and Di Lorenzo et al), which might lead to a higher AAE; whereas the oxidation of aromatics could reduce the degree of conjugation, thus a less wavelength dependence. It would be better for the readers if the authors make this clearer.

Re: Thanks for your good comments. In this study, the AAE values of HULIS fractions from all four stages are similar, which could be related with the evolution of HULIS chromophores in different stages (Huang et al., 2018; Jiang et al., 2020; Deng et al., 2022). At first, the enhanced oxidation of aromatic species in haze days could lead to the bleaching or degradation of BrC chromophores, thus a less wavelength dependence (Forrister et al., 2015；Zhan et al., 2022). In contrast, the outburst of secondary organic aerosols and the photolysis of organic aerosols in haze days both tended to have higher AAE values (Saleh et al., 2013; Dasari et al., 2019). Consequently, the different trends in AAE were counterbalanced during the haze days, which resulting in no significant AAE variations were observed for HULIS fractions

from the four stages. This is also consistent with the trends of the E_{250}/E_{365} ratios of HULIS in the four stages (Table S2). We have revised that in the current manuscript. Please see Lines 261-271.

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5. The author might need to be careful in comparing the molecular weight (MW) with previous studies. For the size exclusion chromatography used in the cited papers, the analytical range of the column is usually from 250 Da ~ 75K Da. Considering the large range and the uncertainty associated with SEC working principle, (i.e., the MW is estimated from the log MW using a calibration curve), most of the MW reported (< 300 Da) here should be considered as "small fractions". Furthermore, in some cited papers (e.g., Di Lorenzo et al), the larger fraction of BrC is defined as MW larger than 500 Da, which means the analyzed fractions here are definitely "small" components.

Re: Thanks. We agreed with your comments that it should be very careful in comparing of the molecular weight (MW) with previous studies (e.g., Di Lorenzo et al, Wong et al., etc). In this study, the MW values of HULIS were determined with ESI FT-ICR MS. However, the MW measurements used in those papers are very different, in which the MW values were estimated by the SEC column retention time, and it's highly dependent on the column, the mobile phase, and the sample itself (e.g., polarity, aggregation, etc). Obviously, the theory of MW determination and the ranges of MW values are very different. Therefore, it is unreasonable for comparing the MW of HULIS with those previous studies (i.e., SEC method). Accordingly, we have removed that in the current manuscript. Please see Lines 347-348.