Dear reviewer:

Thank you very much for your valuable advice on our present research name as "Particle size-dependent fluorescence properties of water-soluble organic compounds (WSOC) and their atmospheric implications on the aging of WSOC", we have addressed all comments carefully, and the detailed corrections are described in the later paragraph. We also have sent our revised manuscript to professionals and native speakers to polish the language, we believe the language of the next version will be vastly improved. Thank you again for your time and patience.

Major Comments:

1. **Introduction:** Lacks organization and continuity? The reviewer suggests the revision of the introduction section to make it more organized and in tandem with the objective of the study.

Thank you very much for your valuable advice. We have reorganized the introduction section into four parts: Paragraph one is the general topic of WSOC; paragraphs two and three introduce several advanced analytical methods that have been used in recent research and refers their limitations, then the advantage of optical methods is proposed; paragraph four lists several recent research on size segregated WSOC; and the perspective of the present research is summarized in the end. The new introduction is shown as follows.

"The environmental, health and climate effect of aerosol particles has been reiterated for years (Pósfai and Buseck 2010; Burnett et al., 2018; Yan et al., 2020; Fan et al., 2020). WSOC is the active fraction of aerosol particles, comprises 10% to 80% of organic compounds (Qin et al., 2018; Almeida et al., 2020; Cai et al., 2020). Previous researchers have proved that WSOC plays a significant role in cloud formation, solar irradiation, and atmospheric chemistry (Asa-Awuku et al., 2009; Duarte et al., 2019). However, the majority of WSOC remains uncharacterized, with only 10% to 20% of the organic compounds structurally identified. Generally, WSOC mixture contains both aromatic nuclei and aliphatic chains (Decesari et al., 2001; Dasari et al., 2019), with functional groups or heteroatoms like hydroxyl, carboxyl, aldehyde, ketone, amino, and other nitrogen-containing groups (Duarte et al., 2007; Cai et al., 2020). Biomass burning and secondary transformations of organics were believed to be the main sources of WSOC (Park et al., 2017; Xiang et al., 2017).

Many sophisticated analytical techniques have been used to unveil the chemical structural information of WSOC (Johnston and Kerecman 2019). Nuclear magnetic resonance (NMR) are experts in obtaining structures of organics (Stark et al., 2013; Duarte et al., 2015, 2020; Chalbot et al., 2016). Mass spectrometry plays a crucial role in its high sensitivity and molecular specificity. The electrospray ionization with ultrahigh-resolution Fourier-transform ion cyclotron resonance mass spectrometry (ESI-FT-ICR-MS), and proton transfer reaction mass spectrometry (PTR-MS) sees increasing application because of the requirement of further insight into organics in particulate matter (Cai et al., 2020; Mayorga et al., 2021). Isotopic ratio mass spectroscopy (IRMS) and accelerator mass spectroscopy (AMS) are widely used to distinguish organic emissions from fossil combustion sources and biogenic sources using carbon isotopic characteristics (Masalaite et al., 2018; Zhao et al., 2019; Huang et al., 2020).

Although having various advantages, the expanding application of formerly mentioned instruments is limited by sampling requirements or expensive costs. In contrast, optical instruments like ultraviolet and fluorescence spectrophotometers are relatively low-cost and efficient. Moreover, the results of the optical method can provide quantitative and qualitative information simultaneously, which guaranteed their broad application on the research of organics like dissolved organic matter (DOM) in water and WSOC (Hecobian et al., 2010; Qin et al., 2018; Xiao et al., 2016). 3-Dimensional excitation-emission matrix (EEM) fluorescence spectroscopy is an informative optical method that has been used in atmospheric WSOC analysis (Duarte et al., 2004; Fu et al., 2014). Fluorescence analysis is helpful in investigating chromophoric organics like aromatics, protein, and other organic matters containing π-conjugated systems (Xiao et al., 2018; 2020). EEM spectrum is implemented to visualize the fluorescence regions and point out possible categories of WSOC by characteristic fluorescent regions in earlier studies (Duarte et al., 2004; Santos et al., 2009). It could reflect the aging of WSOC as well, by the red or blue shift of fluorescence peaks (Lee et al., 2013; Fu et al., 2015; Vione et al., 2019). Fluorescence indices are important subsidiary approaches to statistically analysis EEM data (Qin et al., 2018; Yue et al., 2019), which were determined by the chemical structure of pollutants (Andrade-Eiroa et al., 2013a).

Size distributions of WSOC have been explored for years (Deshmukh et al., 2016; Frka et al., 2018), and the optical properties of size segregated WSOC arouse increasing investigation recently. Generally, the mass concentrations of WSOC show bimodal distributions with dominant in accumulation mode (0.05-2μm) (Yu et al., 2004; Yu et al., 2016). Structural investigations on coal burning and biomass burning affected humic-like substances (a significant fraction of WSOC) of four particle sizes found that organic species of all samples were the same without size discrepancy, reversely, the absorption bands of aromatic groups were more intense compared to carboxylic groups in sub-3 μm fractions (Park et al., 2017; Voliotis et al., 2017). Jang et al., (2019) comprehensively analyzed the structures of size segregated humic-like substances during pre-heating, heating, and after heating periods, found that the chemical structure of HULIS changed with particle size. Liu et al., (2013) examined the light absorption properties of size-resolved BrC and methanol extracts in Georgia, results showed that chromophores were predominant in the accumulation mode with an aerodynamic mean diameter of 0.5 μm. More recently, even fluorescence properties for size segregated ambient WSOC and bioaerosols have been estimated in a coal burning city and a mountain site (Chen et al., 2019; Yue et al., 2019).

Yet comprehensively analyzing fluorescence properties for size-resolved aerosol is infrequent, enormous information is still hidden in the EEM spectra, not to mention the adjacent particle size bins. The perspective of the present research is to investigate the fluorescence properties of WSOC in different particle sizes and try to shed light on the size-dependent evolution of WSOC. 6 stage size segregated particles samples of winter and summer were collected in rural Beijing. The fast and efficient UV-Vis and fluorescence methods were applied in the present research, to obtain the light-absorbing and fluorescent properties of size segregated WSOC. A bunch of fluorescence indices, Stokes shift, and PARAFAC were performed to quantitatively disclosure the hidden connections and transformations of WSOC. Gary relational degree (GRD) is used to show the relations between particles."

2. Line 161: The size distribution of WSOC/OC and WSOC concentration doesn't follow similar trend. Although several studies in the past (Dasari et al., 2019 science advances; Choudhary et al., 2021 environmental pollution), as well as this study (in introduction), have stated that majority of WSOC are secondary (oxidized) in nature. The author can elaborate possible rationales briefly?

Thank you very much for the question. We are sorry for not discussing the size distribution of WSOC/OC ratios and WSOC. As a result, the size distribution of WSOC/OC and WSOC concentration doesn't follow a similar trend indeed. We can see in Figure A. below, that WSOC and OC show similar tendencies for both seasons, and the peaks of WSOC/OC show a delay (peaks around 1um) comparing to their concentrations. This may be because of an increased portion of WSOC or a decrease of OC, as we all know, WSOC is part of OC, thus, we prefer to believe that more OC oxidized to WSOC in particle sizes around 1um, since particles of those sizes can long-exist in the atmospheric environment.

Another possible reason might be that the difference in sources and transformation process of size segregated particles might lead to multiple WSOC/OC results. Ram et al. (2012) reported the WSOC/OC ratio of 0.47±0.11, and characteristically listed the former reported results in vehicle exhaust aerosol and biomass burning affected aerosol, and found that the ambient aerosol had a higher WSOC/OC value and concluded that WSOC/OC could serve as an indicator of secondary formation.

Reference:

Ram, K., Sarin, M. M., and Tripathi, S. N.: Temporal trends in atmospheric PM(2).(5), PM(1)(0), elemental carbon, organic carbon, water-soluble organic carbon, and optical properties: impact of biomass burning emissions in the Indo-Gangetic Plain, Environ Sci Technol, 46, 686-695, 10.1021/es202857w, 2012.

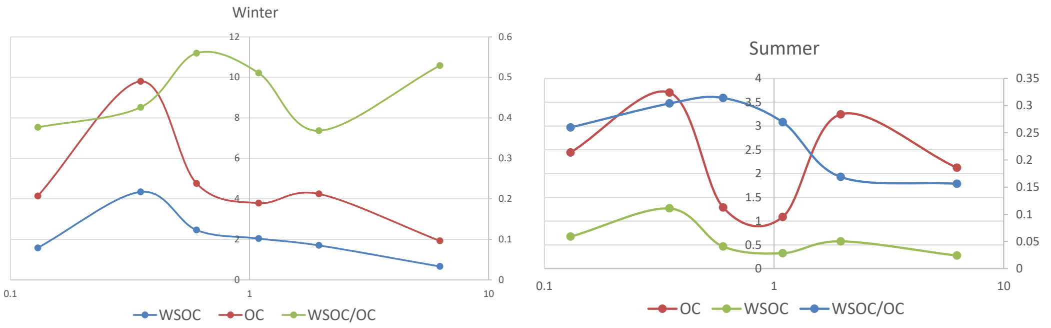

Figure A. size distributions of WSOC, OC and WSOC/OC

3. Line 174: Figure 1 is not discussed in the manuscript. Either delete it or add some relevant discussion about same?

Thank you very much for your valuable advice. We are sorry for not clarify the intention of putting Figure 1 in the manuscript. It is the AQI (air quality index) weighted 72h backward trajectories of our sampling period, providing the sampling information of present research. We have referred it in the manuscript section 2.1.

Lines 98-99: “The air quality index weighted 72h backward trajectories during the sampling period were exhibited in **错误!未找到引用源。**.” 69671251

4. Line 186-193 and 285-290: The author stated that "The AFI/WSOC ratios ranged from 0.22 to 0.57 in winter and 0.18 to 0.34 in summer, respectively." "Our unpublished research found that the AFI/WSOC ratios were lower than 0.2 for anthropogenic source samples, indicating that this ratio might be higher in oxidized fluorescent WSOC." If that is the case, size distribution of AFI/WSOC should have follow the distribution trend of WSOC/OC (a tracer for photochemical oxidation), but this is not the case in this paper (Figure 3). Explain the rationale/s behind this behaviour?

Thank you very much for your valuable advice. We also have noticed this inappropriate deduction during the revision period, thus the indication sentences of "..., indicating that this ratio might be higher in oxidized fluorescent WSOC." have been deleted in line 193. Because the phenomenon of low AFI/WSOC values in sources samples and $PM_{2.5}$ samples in a polluted city and relatively high values in ambient environment samples cannot strongly confirm this conjecture, and we provide some possible explanations instead in the Discussion section. The different size distributions of AFI/WSOC and WSOC/OC may be because that oxidization of WSOC causes fluorescence quenching, we mentioned this perspective in lines 295-298. We have reconstructed the sentences in lines 186-193 and 285-290 as follows.

Lines 184-187 "AFI/WSOC ratios could represent the overall average fluorescence density of WSOC (Xiao et al., 2016). Our unpublished research found that the AFI/WSOC ratios were lower than 0.2 for anthropogenic source samples. And in the present research, the AFI/WSOC ratios ranged from 0.22 to 0.57 in winter and 0.18 to 0.34 in summer, respectively, which were higher than that in source samples and the industrial city of Lanzhou (Qin et al., 2018)."

Lines 292-298 "Substitution and oxidation reactions of ambient organics might widen the delocalization of π electronics and reduce the excitation energy thereby resulting in a redshift of EEM spectra (Kalberer et al., 2004). The specific fluorescence area was widened in the ambient sample and thus having a higher AFI/WSOC ratio when WSOC concentrations were at a comparable level. However, continuously oxidation of organics may break up the π system of organics and extinct fluorescence (Zanca et al., 2017), and lead to a relatively low AFI/WSOC value for the particles having a long residence time, thus the AFI/WSOC values showed a wide variance for different particles."

5. Line 212: The author stated that Stokes shift (SS) of 1.2 μm-1 is an important border of hydrophobic and hydrophilic components. And later used Stokes shift of 1.1 to determine ratios of fluorescence intensity in high SS. Elaborate the possible reason/s?

Thank you very much for the question. We are sorry for not explaining the reason for using SS of 1.1um^{-1} as the border of hydrophobic and hydrophilic components. As a fact, Xiao's former research found that hydrophobic fractions tended to present fluorescence peaks at SS>1 um^{-1}, thus we use the average value of SS at 1.1 um^{-1} as the border, we add the description in line 224. The context is now shown as follows.

"Xiao et al., (2019) found that of stokes shift near 1.2 μm^{-1} is an important border of hydrophobic and hydrophilic components. Hydrophobic fractions tend to have higher intensity in stokes shifts >1.2, possibly as a result of the larger scale of the π conjugated system, versus, hydrophilic contents usually have ionogenic groups bond with fluorescent aromatics reduced the π-conjugated systems, hence, leading to high fluorescence intensities sitting on both sides of stokes shifts around 1.2. While their earlier research also reported that hydrophobic fractions tended to present fluorescence peaks at Stokes shift >1 (Xiao et al., 2016). Thus, the ratios of fluorescence intensity in high SS (SS>1.1) are calculated as the followed equation:"

6. Line 205: HIX (aromaticity) and WSOC/OC (oxidation) ratio following same size distribution trend. How come? This could be an important finding of the manuscript. Add some discussion about same in Discussion and Implication sections.

Thank you very much for your valuable advice. We have compared the size distributions of HIX and WSOC/OC ratio, find that they both show monomodal distributions and the peaks of WSOC/OC delay to larger particle sizes comparing to HIX. That means the HIX starts to decrease when WSOC/OC still increases. The decrease of HIX maybe because the fluorescent WSOC is oxidized to non-fluorescent organics during a long period of exposure in the ambient environment, as the fluorescence property requires conjugated systems in organics. While the long oxidation period also leads to more OC convert to WSOC at the same time, thus the WSOC/OC ratio keeps increase. We have added some discussion in lines 304-307 as follows.

Lines 304-307 "Besides, it was noticed that HIX and WSOC/OC showed similar size distributions except for larger peaking particle size of WSOC/OC values comparing to HIX. This might be because the fine particles with relatively large sizes could long exist in atmospheric oxidation environment, the WSOC/OC ratios increase gradually, however, the oxidation process could also cause fluorescence quenching and lead to the decrease of HIX (Vione et al., 2019)."

7. Line 209, 210, 243: The author categorized protein-like compounds into biogenic origin. But aerosols partitioned from VOCs (isoprene etc.) emitted from plants also categorized into biogenic aerosols. Does the author also incorporating these aerosols produced from VOCs in Protein-like compounds or it is just bioaerosols? Please clarify?
Thank you very much for your valuable advice. We are sorry for the unclear description of biogenic sources. In the present research, we have only considered protein-like compounds in the particulate organics as biogenic and neglected that biogenic VOC are biogenic aerosols as well. To avoid misunderstanding, we have changed the "biogenic sources" to "microbial related" throughout the article. The "biogenic oxygenated organics" in line 215 is a term of Huang's research, thus we keep it unchanged. The other corrections are as follows.
Line 214: "Peak T/Peak C peaked at coarse mode in both seasons indicating that fluorescent microbial related species were likely to exist in larger atmospheric particles."
Line 256: "demonstrated that microbial related WSOC were more likely to exist in large particles"

8. Line 256: Why did author used particles <0.26 μm as references for Grey relational analysis (GRA)? Why not use size bins where WSOC, UV and AFI are maximum?
Thank you very much for the question. We are sorry for not carefully explain the reasons for reference list selection. The minimum particle size is selected because it is assumed that the increase of particle size is an accumulation process, we are trying to find the connections between different particle sizes during the particle increase processes. We have added some explanations in Section 2.4.3, they are shown as follows.
Lines 154-157: "Firstly, considering the evolution of particle sizes as a changing system, larger particles might come from the accumulation and transformation of smaller particles, especially for ultrafine particles. By setting data of particles smaller than 0.26 μm (WSOC concentrations, AFI or UV) as references and particles larger than 0.26 μm as comparisons, their affinities were analyzed by GRA."

Minor Corrections:
In the Reviewer's opinion, the English language needs significant revision throughout the manuscript before acceptance. The some of English-related corrections and other minor comments

are suggested below:

We are sorry for the mistakes in the last version of the manuscript. We have carefully addressed all of the suggestions and corrected them, we also have checked through the article and modified them.

1. Line 33: Replace “mysterious” with either “Unknown” or "Uncharacterized".

Thank you very much for your valuable advice. We have changed “mysterious” to “Uncharacterized” in lines 31-32, they are now shown as follows.

“However, the majority of WSOC remains uncharacterized, with only 10% to 20% of the organic compounds structurally identified.”

2. Line 34-35: The sentence lacks continuity. Revise the sentence "incorporating with different highly oxidized functional groups or heteroatoms like" with may be something like "WSOC mixture contains both aromatic nuclei and aliphatic chains (Decesari et al., 2001; Dasari et al., 2019), with functional groups or heteroatoms like hydroxyl, carboxyl, aldehyde, ketone, amino, and other nitrogencontaining groups (Duarte et al., 2007; Cai et al., 2020)".

Thank you very much for your valuable advice. We have revised this sentence as follows.

Line 32-34 “Generally, WSOC mixture contains both aromatic nuclei and aliphatic chains (Decesari et al., 2001; Dasari et al., 2019), with functional groups or heteroatoms like hydroxyl, carboxyl, aldehyde, ketone, amino, and other nitrogen-containing groups (Duarte et al., 2007; Cai et al., 2020).”

3. Line 37: Is the reference “(ParkSeungShik et al., 2017)” is correctly cited and listed in the reference list (also see line 485).

Thank you very much for your valuable advice. We have checked the reference list and modified the citation format in the text. The citations are shown as follows.

In the text “(Park et al., 2017)”

In the reference list “Park S., Yu, J., Yu, G.-H. and Bae M. S.: Chemical and absorption characteristics of water-soluble organic carbon and humic-like substances in size segregated particles from biomass burning emissions, Asian J. Atmos. Environ., 11, 96-106, https://doi.org/10.5572/ajae.2017.11.2.096, 2017.”

4. Line 39: Revise "Nuclear magnetic resonance (NMR) and mass spectrometry (MS) are two remarkable analytical methods using to structurally unravel the complex WSOC (Duarte et al., 2020)."

Thank you very much for your valuable advice. We have reconsidered the context and modified the whole paragraph. This sentence is deleted now.

5. line 46: It is "Accelerator" not "accelerate".

Thank you very much for your valuable advice. Because we have reconstructed and modified the introduction section, this sentence is deleted now.

6. Line 46: Revise the sentence to something like "Isotopic ratio mass spectroscopy (IRMS) and accelerator mass spectroscopy (AMS) are widely used to distinguish organic emissions from fossil combustion sources and biogenic sources using carbon isotopic characteristics (Masalaite et al.,

2018; Zhao et al., 2019; Huang et al., 2020)."

Thank you very much for your valuable advice. We have corrected the sentence as suggested. They are now shown as follows in lines 42-44.

“Isotopic ratio mass spectroscopy (IRMS) and accelerator mass spectroscopy (AMS) are widely used to distinguish organic emissions from fossil combustion sources and biogenic sources using carbon isotopic characteristics (Masalaite et al., 2018; Zhao et al., 2019; Huang et al., 2020).”

7. Line 50-56: Whole paragraph lacks organization and continuity. The reviewer suggests the revision of the paragraph.

Thank you very much for your valuable advice. We have reconstructed the paragraph as follows.

“Although having various advantages, the expanding application of formerly mentioned instruments is limited by sampling requirements or expensive costs. In contrast, optical instruments like ultraviolet and fluorescence spectrophotometers are relatively low-cost and efficient. Moreover, the results of the optical method can provide quantitative and qualitative information simultaneously, which guaranteed their broad application on the research of organics like dissolved organic matter (DOM) in water and WSOC (Hecobian et al., 2010; Qin et al., 2018; Xiao et al., 2016). 3-Dimensional excitation-emission matrix (EEM) fluorescence spectroscopy is an informative optical method that has been used in atmospheric WSOC analysis (Duarte et al., 2004; Fu et al., 2014). Fluorescence analysis is helpful in investigating chromophoric organics like aromatics, protein, and other organic matters containing π-conjugated systems (Xiao et al., 2018; 2020). EEM spectrum is implemented to visualize the fluorescence regions and point out possible categories of WSOC by characteristic fluorescent regions in earlier studies (Duarte et al., 2004; Santos et al., 2009). It could reflect the aging of WSOC as well, by the red or blue shift of fluorescence peaks (Lee et al., 2013; Fu et al., 2015; Vione et al., 2019). Fluorescence indices are important subsidiary approaches to statistically analysis EEM data (Qin et al., 2018; Yue et al., 2019), which were determined by the chemical structure of pollutants (Andrade-Eiroa et al., 2013a).”

8. Line 57: Replace "3-Dimensional fluorescence of excitation-emission matrix (EEM)" to "3-Dimensional excitation–emission matrix (EEM) fluorescence spectroscopy"

Thank you very much for your valuable advice. We have corrected the phrase as suggested in lines 49-50.

“3-Dimensional excitation-emission matrix (EEM) fluorescence spectroscopy is an informative optical method that has been used in atmospheric WSOC analysis.”

9. Line 59: it should be "mainly helpful in investigating"

Thank you very much for your valuable advice. We have corrected the sentence in line 51.

“Fluorescence analysis is helpful in investigating chromophoric organics”

10. Line 62: what does author mean by "in early years"? Does author mean "earlier studies", if so, revise the sentense.

Thank you very much for your valuable advice. we have modified the phrase in lines 52-54 as follows.

“EEM spectrum is implemented to visualize the fluorescence regions and point out possible categories of WSOC by characteristic fluorescent regions in earlier studies (Duarte et al., 2004; Santos et al., 2009)"

11. Line 65: It should be "analyse" not "analysis"

Thank you very much for your valuable advice. Because we have reconstructed and modified the introduction section, this sentence is deleted now.

12. Line 69: "(great parts of WSOC)"? It should be something like "significant fraction of WSOC"

Thank you very much for your valuable advice. We have corrected the phrase accordingly as follows.

"Structural investigations on coal burning and biomass burning affected humic-like substances (a significant fraction of WSOC) of four particle sizes"

13. Line 70: "reversely"?

Thank you very much for your valuable advice. We have changed "but" to "reversely" in line 62, they are now shown as follows.

"reversely, the absorption bands of aromatic groups were more intense compared to carboxylic groups in sub-3 μm fractions (Park et al., 2017; Voliotis et al., 2017)."

14. Line 82: "neighbor particle sizes" should "adjacent particle size bins"

Thank you very much for your valuable advice. We have modified the phrase in line 70.

"Yet comprehensively analyzing fluorescence properties for size-resolved aerosol is infrequent, enormous information is still hidden in the EEM spectra, not to mention the adjacent particle size bins."

15. Line 83: The use of "But" is not perfect here. The reviewer suggests to use "and" instead.

Thank you very much for your valuable advice. Because we have reconstructed and modified the introduction section, this sentence is deleted now.

16. Line 94: confusing sentence "All samples were collected by quartz filters (Whatman) were prebaked for 5 hours (500°C) and wrapped by aluminum foil stored at -20°C after sampling." May be revised to "All samples collected on quartz filters (Whatman), prebaked for 5 hours (500°C) before sample collection, were wrapped by aluminum foil after sampling and stored at -20°C."

Thank you very much for your valuable advice. We have corrected the sentence as suggested. It is now shown as follows.

Line 82-83 "All samples collected on quartz filters (Whatman), prebaked for 5 hours (500°C) before sample collection, were wrapped by aluminum foil after sampling and stored at -20°C."

17. Line 95: Need clarification? Total 20 groups for 2 seasons or 20 groups each for every season?

Thank you very much for your valuable advice. We have added some descriptions of the group sets in line 84, they are now shown as follows.

"A total of 20 groups of 6 stage size segregated aerosol samples were collected at a rural site in Huairou Distinct, Beijing, from 14 November to 30 December 2016, and 30 June to 8 September

2017 for two seasons”

18. Line 106: Should be "The extract was then filtered through a 0.22 μm membrane filter to remove impurities."

Thank you very much for your valuable advice. We have corrected the sentence as suggested. It is now shown as follows.

Line 96 “The extracts were then filtered through a 0.22 μm membrane filter to remove impurities (Xiang et al., 2017).”

19. Line 113: Confusing? The sentence may be written like "The extraction procedure of samples subjected to fluorescence and ultraviolet-visible (UV-Vis) measurements were same as WSOC detection."

Thank you very much for your valuable advice. We have corrected the sentence as suggested. It is now shown as follows.

Lines 102-103 “The extraction procedures of samples subjected to fluorescence and ultraviolet-visible (UV-Vis) sampling were the same as WSOC detection.”

20. Line 117: Should be "Raman Unit"

Thank you very much for your valuable advice. We have capitalized “R” in the sentence, they are now shown as follows.

Line 108 “All EEM data in the present research were in the Raman unit (R.U.)”

21. Line 124: Revise the sentence "The EEM data were spectrally corrected by blank sample for instrument bias, inner filter effects, Rayleigh scattering, and most of Raman scatter had been removed" to "The EEM data were spectrally corrected by blank sample to remove interferences from instrument bias, inner filter effects, Rayleigh scattering, and Raman scatter."

Thank you very much for your valuable advice. Because we also have been asked to add some explanations of the data correction procedure, by considering both two suggestions, we have corrected the sentences as follows.

Lines 119-123 “All EEM data in the present research were in Raman unit (R.U.), the background signals, interfering signals (first- and second-order Rayleigh and Raman scatterings), and the inner-filter effect were removed by subtracting an EEM of blank, replace with a band of missing values or inserting zeros outside the data area, detailed procedures could be found in Bahram et al., (2006). Data correction and standardization followed procedures described in Xiao et al., (2016).”

22. Line 133-134: Equations number is not matching? Example: "equation (3)" should be "equation (2)" and "equation (4)" should be "equation (3)"

Thank you very much for your valuable advice. We are sorry for the unmatching equation number, they are now corrected properly as follows.

$$SS = \frac{1}{\lambda_{Ex}} - \frac{1}{\lambda_{Em}} \tag{2}$$

$$WH = 2(\frac{1}{\lambda_{Ex}} + \frac{1}{\lambda_{Em}})^{-1} \tag{3}$$

23. Line 218: Revise "On a large scale of a π-conjugated system, the...."

Thank you very much for your valuable advice. We have corrected the phrase as follows.

Line 219 “On a large π-conjugated system, the electron in the ground state needs relatively low excitation energy jumping to the excited state”

24. Line 222: "Supporting information Figure 3, and Figure 5(c)." shoud be Figure S3 and Figure S5(c). Do same thing for Figures S1, S2, S4 and Table S1, in Supporting Information.

Thank you very much for your valuable advice. We Have corrected the citation of supporting information accordingly as Figure S# or Table S#.

25. Line 87 and 228: The full form of PARAFAC is already mentioned on Line 87. No need to repeat it again. Follow same comment for others as well (e.g. GRA on line 249 etc.).

Thank you very much for your valuable advice. We have deleted the full forms of “PARAFAC” and “GRA” after firstly mentioned them in lines 20 and line 138.