An itemized response (**blue words**) to reviewers' comments and suggestions

Dear Editor,

Thank you for your useful comments and suggestions on our manuscript (Manuscript Number: bg-2022-140). The manuscript has been carefully revised according to the reviewers' comments. The following are the reviewer's comments related to the manuscript and how we have addressed each of reviewer's concerns (**blue words**). Changes have been marked as **blue** in the manuscript.

Marine DOM is important in the carbon cycle and the ecosystem functioning. Sea-surface microlayer (SML) is an important interface between the atmosphere and the seawater, and had distinct biogeochemical properties from the subsurface water. To date, there is little information on the optical properties of DOM in the SML. This study examined the changes and underlying mechanisms of DOM in the SML of Yellow Sea and East China Sea, using measurements of multiple proxies. The authors revealed an overall enrichment of DOM in SML with evident variability for both different regions and seasons and for different chemical species. They also tested the influences of environmental factors and photo-degradation based on field observations and incubation experiments. Overall, this study is focused on an interesting and novel issue with a large comprehensive dataset. The results from this study would make a nice contribution to the field of marine DOM study.

Thanks for the reviewer's positive comment. According to the reviewer's suggestions, we have made the revision in the revised manuscript.

Major comments:

1. Line 21 and Line 288-290: Please note that marine DOM usually has higher absorption slope than terrestrial DOM, and the higher slope (no matter it is $S_{320-412}$ or $S_{275-295}$) means higher absorption coefficient at shorter wavelength (not longer wavelength). In addition, it is not appropriate to assign $S_{275-295}$ to terrestrial.

   We agree with the reviewer's viewpoint. Although marine DOM usually has higher absorption slope than terrestrial DOM, the higher slope (no matter it is $S_{320-412}$ or $S_{275-295}$) means higher absorption difference between different wavelength (from higher wavelength to lower wavelength) in marine CDOM. Sasaki et al. (2005) reported that the contribution of a(440) was ~50% of total absorption, except for the bloom. In addition, the visible fluorescence signal in ocean waters has two components: one emitting in the region of 400 nm and another at 440 nm (Jørgensen et al., 2011; Kowalczuk et al., 2013; Yamashita et al., 2010). Therefore, marine production of DOM had the larger influence on the CDOM absorption properties in the longer wavelength range (Danhiez et al., 2017). (Line 295-298)

Previous studies have reported $S_{275–295}$ values in the range of 0.020–0.030 nm^{-2} and 0.010–0.020 nm^{-2} for ocean and coastal waters respectively (Del Vecchio and Blough 2002), 0.014–0.018 nm^{-2} for wetlands (Helms et al., 2008), and 0.012–0.023 nm^{-2} for terrestrial systems (Spencer et al., 2012). Therefore, marine production of DOM had the higher $S_{275-295}$ value.

2. Line 254: The fluorescence at Em 310 nm can be assigned to tyrosine-like component, but that at Em 375 nm can not. In addition, Em is described as 310 (375) nm in the text, but is 375 (310) nm (microbial or marine humic-like) in Table 1. The number in the parenthesis means the position of the secondary peak. Please double check.

   Thanks for the reviewer's suggestions. We agree with the reviewer's viewpoint and have confirmed that the secondary peak is the tyrosine-like component (255 nm/310 (375) nm) in the revised manuscript. (Line 261; Table 1)

   According to the reviewer's suggestion, we have made the revision in the revised manuscript.

   C2 exhibited Ex/Em maxima at 255 nm/310 (375) nm, which could be considered tyrosine-like fluorescence (Stedmon et al., 2003) and attributed to autochthonous and/or microbial FDOM. (Line 261-262)

3. Line 215-216: Please show the absorption spectra in the supplementary file. I am wondering if there is any shoulder peak that is reported for algal DOM? If

so, it would be needed to use the absorption coefficient at longer wavelength for CDOM level.

Thanks for the reviewer's suggestion. We agree with the reviewer's viewpoint and have shown the absorption spectra in the supplementary files (Fig. S1.). We observed that there is not any shoulder peak that is reported for algal DOM.

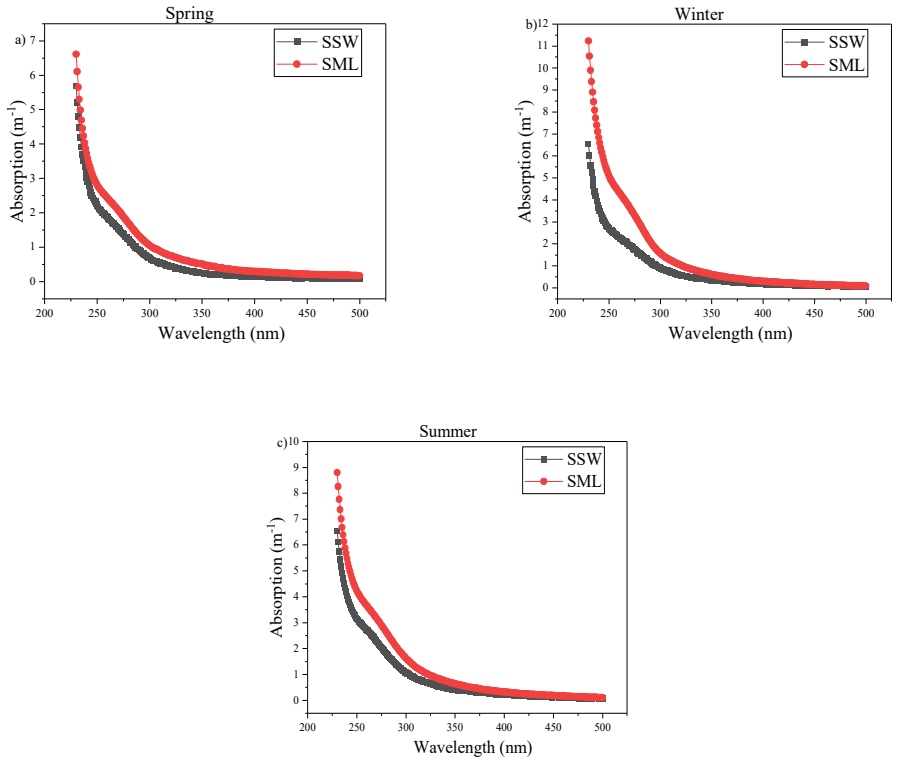


Fig. S1. Absorption spectra averaged by seawater samples between 230 to 500 nm in spring (a), winter (b), and summer (d).

4. 6: Some DOC data exceed 100%, please give some explanation (e.g., contamination or analytical errors?).

Thanks for the reviewer's comments. Although photodegradation causes CDOM absorption to decrease, DOC is not sensitive to photodegradation in our photodegradation experiments, implying that the light exposure preferentially removed the colored DOM rather than the non-colored DOM (Moran et al., 2000; Bittar et al., 2015; Vähätalo et al., 2004). We didn't contaminate all samples. Therefore, both measurement and analytical errors will let DOC data exceed 100%.

5. I can not find the supplementary files. Please double-check if the supplementary figures and tables were uploaded.

Thanks for the reviewer's comment. According to the reviewer's suggestion, we have added the supplementary files in the revised manuscript.

Minor comments:

1. Line 93-95: Five or four cruises? Please double-check.

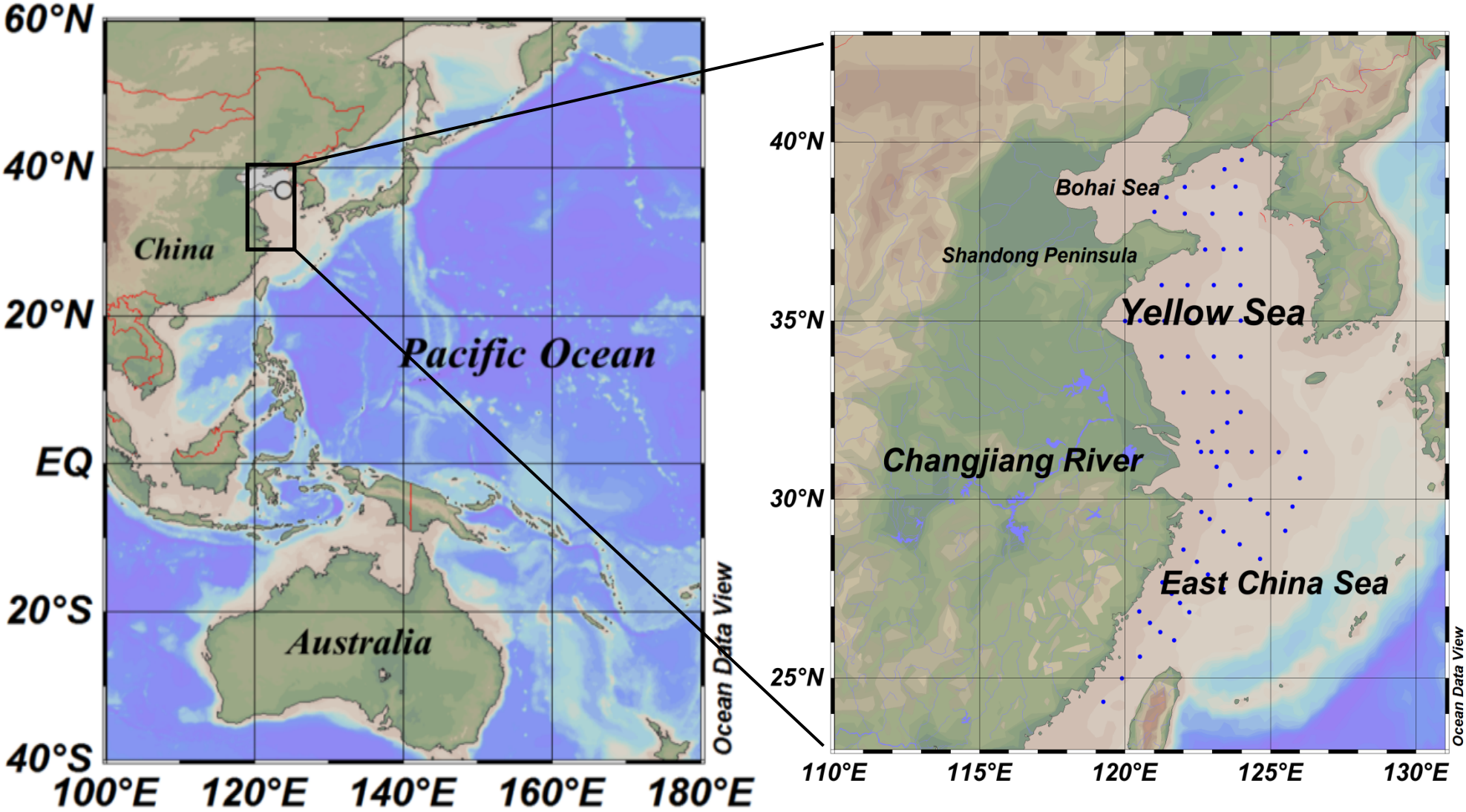
Thanks for the reviewer's comment. We agree with the reviewer's viewpoint and have made the revision in the revised manuscript.

"Four cruises were conducted during the four seasons, specifically, from: 27 March to 15 April 2017 (R/V "*Dong Fang Hong 2*"), 26 June to 19 July 2018 (R/V "*Dong Fang Hong 2*"), March 2019 (R/V "*Zheyu No. 2*"), and 28 December 2019 to 16 January 2020 (R/V "*Dong Fang Hong 3*")." (Line 93-95)

2. Line 96: Please move the sampling map from the supplementary file to the main text, if there is no limit on the number of figures.

   Thanks for the reviewer's suggestion.

   According to the reviewer’s suggestion, we have moved the sampling map to the main text. (Fig. 1)


60°N
40°N
20°N
EQ
20°S
40°S
100°E
120°E
140°E
160°E
180°E
China
Pacific Ocean
Australia
Ocean Data View
40°N
35°N
30°N
25°N
110°E
115°E
120°E
125°E
130°E
Bohai Sea
Shandong Peninsula
Yellow Sea
Changjiang River
East China Sea
Ocean Data View

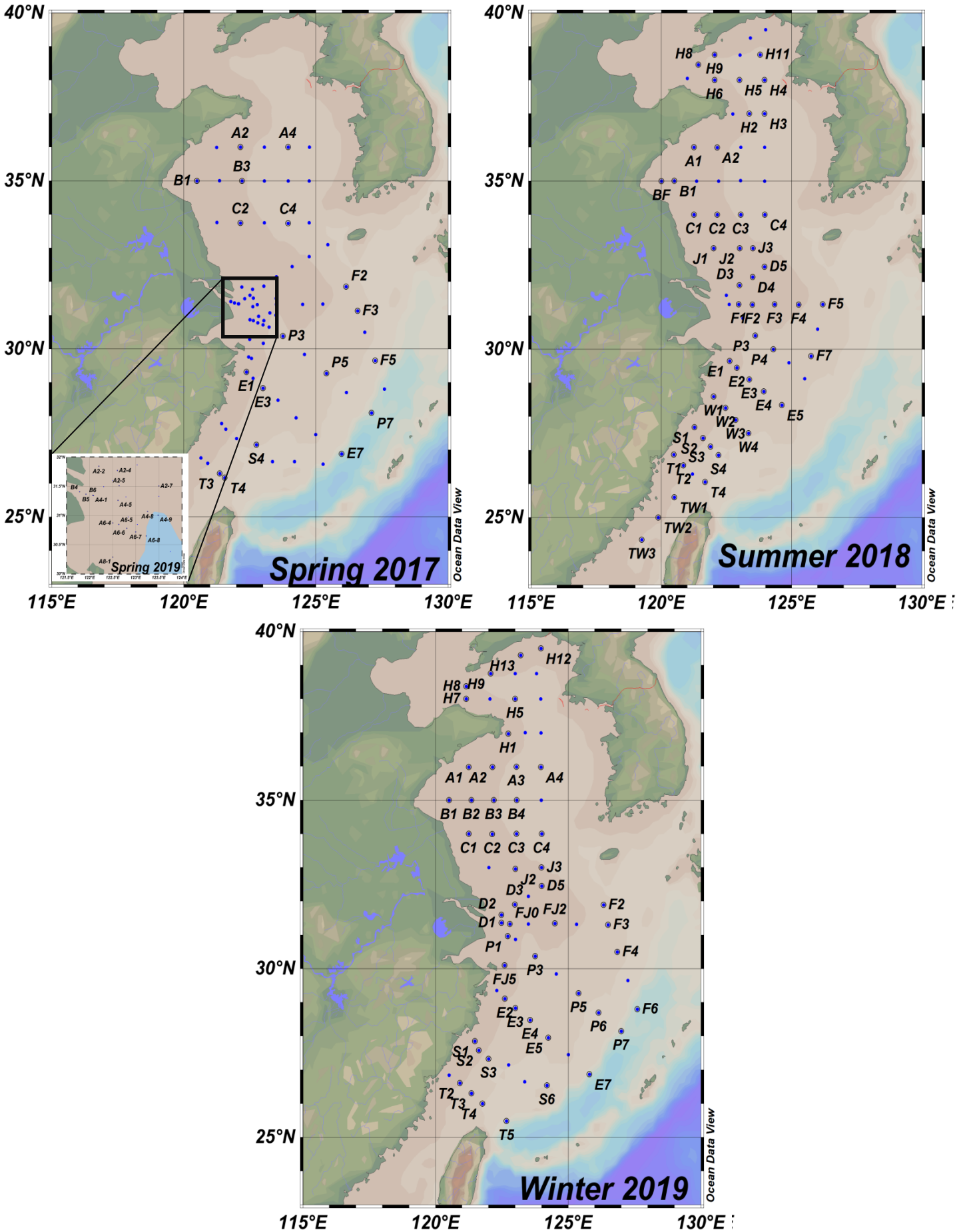


Fig. S1 Map of sampling stations.

3. Line 104: Please show a photo for the sampler in the supplementary file.

Thanks for the reviewer's suggestion.

According to the reviewer's suggestion, we have shown the sampler in the supplementary file. (Fig. S7.)

Fig. S7. The Screen Sampler

4. Line 105-109: Please clarify the thickness of the SML sampled.

Thanks for the reviewer's suggestions. According to the reviewer's suggestion, we have made the revision in the revised manuscript.

Repeated dipping was conducted until the desired volume was collected (11 times, 600 ml; the thickness of the SML sample is nearly 300 um). (Line 113)

5. Line 121-123: Were the quartz tubes placed in the water bath? If so, at which water depth?

Thanks for the reviewer's suggestions, we have made the revision in the revised manuscript.

"The quartz tubes were positioned on their sides under the irradiation source to maximize the exposure of the sample; the water depth in each tube was 5 cm (i.e. the diameter of the tube)." (Line 128)

6. Line 133-136: Did you carry out the baseline correction (e.g., subtracting the mean absorbance over 700-800 nm)?

Thanks for the reviewer's suggestions, we have made the revision in the revised manuscript.

Yes, we did, we have subtracted the mean absorbance over 700-800 nm.

7. Line 150-152: Please note that $SUVA_{254}$ is calculated as dividing the absorbance at 254 nm (not the absorption coefficient) by DOC.

Thanks for the reviewer's suggestion.

According to the reviewer's suggestion, we have made the revision in the revised manuscript.

"$SUVA_{254}$ is calculated as dividing the absorbance at 254 nm by DOC." (Line 159)

8. Line 260: "increasing DO level", please show it in the figure or supplementary file. Please check DO or AOU is used?

Thanks for the reviewer's suggestion.

According to the reviewer’s suggestion, we have shown it in the supplementary file. (Table S1)