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.

Dear Authors:

Thank you for revising your manuscript considering all reviewer comments and suggestions. Your manuscript can be published after another careful revision that would be required to improve the following editorial points:

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

- Line (L) 19 in the track-changed manuscript: Please separate the long (and grammatically incorrect) sentence, like "In addition, photoexposure experiments were designed to compare...".

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

"In addition, photoexposure experiments were designed to compare photochemical degradation processes of DOM between the SML and the SSW." (Line 19)

- L 26: Please delete the unnecessary phrase "We find that".

- L 27 "is photobleached less": Didn't you use 'photobleaching' in the sense of 'photodegradation'? If it was the case, I think you need to change the phrase as follows (and in the past tense): "was photobleached (photodegraded) more". Please correct me and clarify the sentence, if I misunderstood it. Thanks for the reviewer's comment, we have made the revision in the revised manuscript.

"Higher EFs were generally observed in the SML in the off-shore regions than in the coastal regions, and CDOM in the SML was photobleached more after relatively strong irradiation, as also indicated by the lower percentages of humic-like DOM and lower specific UV absorbance values (SUVA₂₅₄) in the SML than the subsurface water (SSW)." (Line 26)

- L 31: The concluding sentence is still difficult to follow and hence requires further refinement. Did you mean something like "Our results revealed a new enrichment model for exploring the airsea interface environment, which can explain the more autochthonous properties of DOM in the SML than the SSW."

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

"Our results revealed a new enrichment model for exploring the air-sea interface environment, which can explain the more autochthonous properties of DOM in the SML than the SSW." (Line 31)

- L 48-50 "The is a very dynamic interface (Cunliffe et al., 2013), the impact of changes in UV radiation on air-sea fluxes in the SML of important trace gases will need to be assessed.": This and some other sentences are incomplete lacking conjunction. Please conduct a careful proofreading and grammar check-up of the revised manuscript.

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

"The SML is a very dynamic interface (Cunliffe et al., 2013), moreover, the impact of changes in UV radiation on air-sea fluxes in the SML of important trace gases need to be assessed." (Line 48)

- L 120-122: Repeated dipping was conducted "in the SML up to the depth of 1000 μ m (Is this the actual depth for your sampling?)" until the desired volume was collected (11 times, 600 ml).

Thanks for the reviewer's comment, $300 \sim 1000$ um is the actual depth for our sampling, and we have made the revision in the revised manuscript.

"Repeated dipping (11 times, 600 ml) was conducted until the desired volume was collected (the depth of the SML sample is nearly 300 ~ 1000 um)." (Line 122)

- L 196-199: Please provide the actual accuracy and precision information, for instance something like % error and CV of repeated reference measurements.

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

"Two forms of reference water have been developed for DOC analysis. Deep-ocean water, collected at 2600 m in the Sargasso Sea and containing biologically refractory DOC, as well as low carbon water for testing instrument blanks are available to the U.S. and international communities of aquatic chemists (Hansell, 2013; measurement and analytical errors < 19%)." (Line 197)

- L 219: Did you reflect changes in your analytical analyses in this section?

Thanks for the reviewer's suggestion.

This section is the statistical analyses measurement, and we didn't reflect changes in our analytical analyses.

2.5 Statistical analyses

The correlation coefficient (R) and probability (P) values were used to evaluate the goodnessof-fit. The correlation matrix, analysis of variance, and principal components analysis were conducted with SPSS version 18.0 (SPSS Inc., Chicago, IL, USA) to determine the possible relationships between the DOM parameters and environmental factors. A P-value ≤ 0.05 was considered significant. Regression analyses between the optical parameters of DOM and several biogeochemical parameters in the SSW and the SML samples were performed in the Table S1 and the Table S2, respectively.

- L 309 and Fig. 5 (and other revised figures): Please use 'r' for correlation and 'r2' for regression. The difference between simple correlation (r) and causal relationship (r2) needs to be checked for other figures and associated descriptions.

Thanks for the reviewer's comment, we have made the revision in the revised manuscript and we use 'r' for correlation in Fig. 5.









Fig. 5. Correlations between the microlayer CDOM, DOC, Chl-a, four fluorescence components concentrations, cyanobacteria, phytoplankton biomass, nutrients and bacterial abundance, and their subsurface water concentrations. The dashed lines correspond to the 1:1 lines, and the full lines are the regression models. (All DOM spectroscopic parameters sample were analyzed in spring, summer and winter; Chl-a was determined in spring, summer, and summer; cyanobacteria, phytoplankton biomass, nutrients and bacterial abundance were determined in spring and summer.).

- L 423-427 "All incubation samples were not contaminated, both measurement and analytical errors will let DOC data exceed 100%.": This is an awkward sentence: very difficult to understand its meaning. Please rewrite it.

Thanks for the reviewer's comment, we have deleted this sentence in the revised manuscript.

"Although photodegradation causes CDOM absorption to decrease, DOC is not sensitive to photodegradation in our photodegradation experiments (Fig. 7), implying that the light exposure preferentially removed the colored DOM rather than the non-colored DOM (Bittar et al., 2015)." (Line 424)

- Fig. 3: You still have an r value in one graph. Another issue is the dependent variable: Is salinity dependent or independent variable? If you wanted to explain absorption based on salinity, you need to switch x and y axes. Or if you just wanted to show correlation, please use r instead of r2.

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





Fig. 3. Relationships between a(254) and salinity in the subsurface water (SSW) in the East China Sea (ECS) and the Yellow Sea (YS) during spring, summer and winter.