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.

Yang et al.

This paper present the spatialatemporal distribution, photoreactivity and environmental control of dissolved organic matter in the sea-surface microlayer of the eastern marginal seas of China. The paper has a good database and discussion, with only a few comments from my side.

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

Introduction: all citations in the introduction were outdated. Seem like you missed a lot of important CDOM studies in the SML. Please add an updated reference (within 5 years). I believe there are many studies on CDOM in the SML has been published recently.

Thanks for the reviewer's useful suggestions. According to the reviewer’s suggestions, we have added updated references in the revised manuscript (within 5 years).

“In addition, Mustaffa et al. (2018 and 2017) observed that FDOM enrichment in the SML in the coastal regions and open Atlantic Ocean, and FDOM is frequently enriched during upwelling events in the Baltic Sea.” (Line 73-76)

Line 42: The surface-active compounds were found to be enriched in the SML at a wind speed of 13 m/s. (<https://doi.org/10.1002/2017GL072988>**)**

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

“With a total thickness ranging between 1 µm and 1000 µm, the SML remains present in wind speeds of up to 13 m s-1 (Sabbaghzadeh et al., 2017).” (Line 41-42)

Line 47: Explain why the role of SML in oceanic emission is not well understood.

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

“The SML 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. In addition, another uncertainty is whether ﻿photochemical reactions on the SML affect the flux of volatile species at the air-sea interface (Blough, 1997).” (Line 46-50) Therefore, the role of the microlayer in oceanic emissions is not well understood and fundamental advance in understanding its properties are needed.

Line 50: The processes leading to the enrichment of DOM in the SML are not solely controlled by changes in the DOM concentration at the sea surface microlayer, but are more complex.

Mustaffa, N.I.H., Badewien, T.H., Ribas-Ribas, M. et al. High-resolution observations on enrichment processes in the sea-surface microlayer. Sci Rep**8,** 13122 (2018). <https://doi.org/10.1038/s41598-018-31465-8>

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

“The processes leading to the enrichment of DOM in the SML are not solely controlled by changes in the DOM concentration at the sea surface microlayer, but are more complex (Mustaffa et al., 2018). Because of its unique position at the air-sea interface, the biological and photochemical reactions of DOM in the SML could strongly impact the biogeochemical cycling of biologically important elements, for example, via the conversion of DOM into volatile species such as carbonyl sulfide (OCS), which influence the atmospheric chemistry and climate (Mopper et al., 2002).” (Line 52-57)

Line 66: The author mentioned recent studies but the citation was from 2017 and 2018. Please add updated citations.

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

“Recent studies have mainly focused on using the characteristics of CDOM as indicators of the sources and degradation states of DOM (Massicotte et al., 2017) in the SSW, and its vertical distribution in estuaries and open oceans (Yamashita et al., 2017; Margolin et al., 2018).” (Line 71-73)

Line 80: How do you define coastal and off-shore regions?

Thanks for the reviewer's suggestions.

The way that we define coastal and off-shore regions is basing on the variation of salinity.

There were significantly negative linear correlations between salinity and a(254) in all cruises in the SSW(p < 0.01, Fig. 2), especially in the ECS. (Line 236-237) Lower salinities were observed in the Changjiang Estuary and coastal waters. (Line 224-225)

Line 87: Since CDOM highly undergoes photobleaching, correlations to solar radiation and temperature other parameters would therefore be essential.

Line 350 – 374: How about solar radiation and temperature during sampling conditions? These parameters influence the enrichment of CDOM in SML than wind speed conditions.

Thanks for the reviewer's suggestions.

We are so sorry that we didn’t observe the significant relationship between temperature and the EF of CDOM value. (Fig. S6)



Fig. S6. Relationships between temperature and EFs of a(254), Chl-*a*, DOC, and four fluorescence components.

Additionally, we are so sorry that we didn’t record the solar radiation variation during the sampling period. In the future research, we will investigate the solar radiation variation in all sample stations. We will research the correlations between solar radiation and temperature and other parameters. Thanks for the reviewer's helps.

Line 103: What is the wind speed condition during sampling?

Line 350-352: Please add references on how you define wind regime.

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

Meteorological data (e.g., wind speed and air temperature) were recorded simultaneously by a ship-borne weather instrument (Li et al., 2019). (Line 126-127)

The wind speeds during our observations ranged from 0.2 to 14.9 m s-1. (Line 366)

Lines 99-104. I would like to have some more information about how the SML is sampled. For instance, I would like to know whether they made blank measurements of the sampling system and how were those blanks. Information about the instrument's detection limit, or the thickness of the SML that is sampled, would be also appreciated.

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

We agree with the reviewer’s viewpoint and 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 118-119)

However, we didn’t make blank measurements of the sampling system. Before all the sampling actions, we washed the screen sampler using Milli-Q water 5 times, in order to make the screen sampler clear.

Line 350: The author mentioned that the wind speed ranged between 0.2 to 14.9 m/s. However, previous studies only found that the SML only persist at the wind speed of 10 m/s. More recently, surface-active compounds were found to be enriched in the SML at a wind speed of 13 m/s. However, the full integrity of the SML sampled at high wind speeds in this study is concerning because the samples were taken directly from the ship's bow. The distance of the sampling point should be at least 500 m upwind from the vessel to avoid any disturbance of SML integrity produced by the ship’s movement and potential contamination.

Thanks for the reviewer's suggestion.

We agree with the reviewer’s viewpoint that surface-active compounds were found to be enriched in the SML at a wind speed of 13 m/s. The distance of the sampling point should be at least 500 m upwind from the vessel to avoid any disturbance of SML integrity produced by the ship’s movement and potential contamination.

However, our research included a large comprehensive dataset (four cruises), we need collect enough samples (location SML samples and the incubation experiments samples) during the limited time. Hence the samples were taken directly from the ship's bow when conditions were calm. In future research, we will away from the vessel to avoid any disturbance of SML integrity produced by the ship’s movement and potential contamination.

Line 366: Besides wind mixing, tidal mixing is an important factor, especially in the adjacent coastal water, but the manuscript was never mentioned or discussed. What is tidal type in your concerned Stations? Tidal variation during the sampling period should be investigated at some shallow water Stations.

Thanks for the reviewer's suggestion.

We agree with the reviewer’s viewpoint that tidal mixing is an important factor in SML research. We are so sorry that we didn’t record the tidal variation during the sampling period. In future research, we will investigate the tidal variation in all water stations, the influence of tidal mixing processes, and discuss the relationship between the tidal variation and EFs of CDOM and FDOM. Thanks for your helps.