# Response Letter to Review Comments

Dear Editor and Reviewers,

Thank you for the detailed review and valuable feedback provided on our review article entitled “Atmospheric Implications of Ocean-Atmosphere Physicochemical Interactions”.

We appreciate the time and effort invested in evaluating our manuscript. Below, we address each comment raised during the review process.

**Response to reviews**

Reviewer comments are in **bold**. Author responses are in plain text labeled with **[R]**. Line numbers in the responses correspond to those in the revised manuscript (the version with all changes accepted). Modifications to the manuscript are in *italics*.

**Reviewer #1**

**The manuscript provides a comprehensive review of the physico-chemical processes at the ocean-atmosphere interface. These processes are more important as they have an impact on atmospheric chemistry. The authors focus their article on sea surface microlayers and their impact on aerosol production. This review is particularly interesting and gives a state of the art of current research in this field. I suggest only a few minor changes before publication in ACP.**

[R0] We sincerely appreciate the constructive feedback and gratefully acknowledge the positive recognition of our work.

1) **The authors mention some papers in which the photochemical impact was studied under not-relevant wavelength conditions. For example, in line 219... the paper cited used experiments under UVC radiation which is far from the solar irradiation spectrum.**

[R1] We apologize for this confusion. In fact, Knipping et al. (2000) and Oum et al. (1998) investigated the reaction between hydroxyl radicals (OH) and sea salt particles (i.e. NaCl). The employed UVC irradiation to photolyze ozone ($O_3$), thereby generating OH radicals for their study. To clarify this point, we rewrote this paragraph as: "*Oum et al. (1998) and Knipping et al. (2000) investigated the reaction between hydroxyl radicals (OH) and sea salt particles, the results indicated that air-water interfacial reactions were essential to explain the observations, a process that may also occur on the ocean surface.*"

2) **Another article (line 231...) reports the effect of ozone photolysis at 254 nm and its effect on the deliquescence of sea salt particles. if publishers wish to cite these articles, it should be specified that the results were obtained at wavelengths below the solar spectrum.**

[R1] We consider these two studies seminal to the early understanding of interfacial reaction involving gaseous species and inorganic ions on aerosol particles and at the ocean-air interface. They provide crucial insights into the scientific evolution of this field. To address the reviewer's concern, we have revised the introduction of the Knipping et al. (2000) and Oum et al. (1998) studies to more clearly establish their foundational role in understanding interfacial reactions: "*Oum et al. (1998) and Knipping et al. (2000) investigated the reaction between hydroxyl radicals (OH) and sea salt particles, the results indicated that air-water interfacial reactions were essential to explain the observations, a process that may also occur on the ocean surface.*"

**Response to reviews**

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**Reviewer #2**

**Wang and Gligorovski report the review paper on the physical and chemical properties of the atmospheric-ocean interfaces or specifically the multiphase and heterogeneous processes of the sea surface microlayer. The text is generally written well and hence, is publishable after addressing the following minor issues.**

[R0] We sincerely appreciate the acknowledgment of our work, and the constructive insights provided.

**This text briefly mentions the deposition of gaseous species on the ocean surface. I am wondering if there are any effects of SML on the aerosol deposition onto the ocean surface, particularly for iron-containing aerosols. It would be great if the authors provided the current understanding.**

[R1] We thank the reviewer for the insightful and valuable comment on this issue. In response, we have addressed the current understanding of how aerosol-deposition perturbations impact SML's biota, and have emphasized the importance role of iron-containing aerosols. The expanded discussion in line 1111-1127 reads as follows: "*Phytoplankton communities in seawater have long been considered the primary source of reactive surfactants, such as fatty acids. Atmospheric deposition, in contrast, is typically viewed as a minor contributor, providing only supplementary nutrients. However, recent research reveals that wildfire smoke can directly deposit substantial quantities of microbes into the SML. This microbial input can be comparable to the estimated supply from seawater and may significantly diversify the SML's microbial community(Yue et al., 2025). Critically, biomass burning also transports biologically important trace metals particularly soluble iron (SFe), a well-established growth-limiting nutrient that can alter the growth rate and composition of the phytoplankton community(Bergas-Masso et al., 2025; Guieu et al., 2005; Mahowald et al., 2018). Model estimates indicate that rising SFe deposition, driven by increasing socio-economic activity, could boost phytoplankton productivity in the iron-limited North Atlantic by up to 20% annually(Bergas-Masso et al., 2025). Consequently, key questions arise: How does aerosol-induced modification of SML phytoplankton communities alter the production and abundance of reactive surfactants? Furthermore, how might these changes affect multiphase chemical reactions involving surfactants and atmospheric oxidants, with implications for atmospheric chemistry?*"

**For the section 5.1, can the authors provide a more general view about the photosensitized processes? The authors discussed several reaction systems individually, but there is a similarity in terms of processes involving the triplets.**

[R2] We revised the sentences from Line 743-763 as: "The organic compounds that exhibit photochemical activity in the SML are referred to as chromophoric dissolved organic matter (CDOM). Upon absorption of light *with sufficient energy*, a given CDOM molecule gets electronically excited *(electrons undergo excitation from their ground state to higher-energy unoccupied or singly occupied molecular orbitals)* and goes to an excited singlet state ($^1$CDOM*) which *then react immediately or quickly relax to the lower vibrational levels. The remaining excess energy is subsequently released via mechanisms including photochemical reactions and nonreactive processes such as fluorescence, internal conversion and* intersystem crossing *(ISC) where the singlet state decays to the lower-energy* excited triplet state ($^3$CDOM*). Both, $^1$CDOM* and $^3$CDOM* can induce formation of hydroxyl radical (OH), singlet oxygen ($^1O_2$), superoxide ($O_2^-$), $H_2O_2$, aqueous electron ($e^-_{aq}$), and organic radicals *through various mechanisms*(Sharpless and Blough, 2014; Mcneill and Canonica, 2016). *Photosensitized reaction represents a special subset of photochemical reactions. In this process, CDOM acts as a sensitizer, its absorption of light initiates chemical reactions involving a second species, resulting reforming CDOM and/or generating excited or ionized states of the second species and other species. This represents a key characteristic of photosensitized processes: they enable photoreactions at wavelengths where the "target" molecule itself does not absorb light*(George et al., 2015; Gomez Alvarez et al., 2012)."

In the corrected version, we have refined the illustration of the basic scheme of photochemical reactions and clarified the characteristics of photosensitized reaction among the photochemical reactions.

**Lines 891: The discussion about the ionic strength effect is quite limited in the present text, although the authors present the corresponding figure (Fig.4). The authors need to elaborate. What is the direct and indirect role of ionic strength?**

[R3] The ionic strength effect was discussed in section 1.1, where its presentation aligns with the relevant content. Since this review focuses on the physicochemical interactions at the air-ocean interface, a system where ionic strength remains relatively constant. A detail description about the direct and indirect if ionic strength effect is out of the scope of this paper, we thus provided only a brief overview of ionic strength's direct and indirect roles in line 240-257.

In addition, we elaborated the presented Figure 4 as the rephrased context in line 913-917: "Figure 4 illustrates the *photosensitized reaction initiated by sunlight-activated fluorene and dimethylsulfoxide (DMSO) in the presence of halide ions* $Cl^-$*,* $Br^-$ *and* $I^-$*.* The prompt formation of gas-phase methanesulfonic acid ($CH_3SO_3H$), methanesulfinic acid ($CH_3SO_2H$), hydroxymethanesulfonic acid ($CH_4O_4S$), and 2-hydroxyethenesulfonicacid ($C_2H_5O_4SH$) *was observed*."

**Minor comments**

**Please define "CLAW"**

[R] We defined the "CLAW" hypothesis in line 75 as "*The CLAW hypothesis, an acronym derived from the initials of its four authors, was proposed by Charlson et al. (1987)*".

# Reference:

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