General comments

Photosensitized reaction occurring at the ocean surface is an important abiotic source of marine VOCs. This study utilized SFG to investigated a well-documented reaction (i.e. 4-BBA+NA), as previously reported by Tinel et al. (2016). By using SFG, the authors identified a novel SFG spectra band (assigned as aromatic signal) that not be reported by Tinel et al.'s study. Additionally, the authors evaluated the impact of light wavelengths, salinity and pH on this reaction, underscoring the importance for considering these parameters on the reaction's mechanism. It is important to study the effects of environmental parameters on photosensitized reaction mechanisms, however, in my view, the authors have not adequately demonstrated how these parameters influence the reaction mechanism in this study.

When reading the ABSTRACT, I assumed the authors had systematically evaluated the aforementioned parameters and quantified their impacts on the reaction. However, it appears only a range of light wavelengths was evaluated (e.g. Figure 5). Salinity, one of the key parameters investigated in the paper, as underlined in the ABSTRACT, was discussed in only two lines of words in the main text (Page 12 Line 1-2) and the authors stated "it is not the focus of this paper to discuss the details of the salinity….", and only two pHs were present in this work because of the interfere of carbonation on pH rather than intentionally designed. I suggest the authors revise the abstract to better reflect the study's core findings and objectives.

The literature cited in the INTRODUCTION is quite outdated, as all references are from before 2020, many recent related studies concerning the chemistry at SML have not been introduced in the INTRODUCTION.

In the CHEMICAL COMPOSITION section, the data comparison with Tinel et al. (2016) is missing, the comparison should be highlighted as this study built upon this previous work. Additionally, error bars only present in Figure 10, they are absent in Figures 7-9 and 13, which is critical for accurately representing variability and ensuring clarity in the results. I recommend that the authors integrate the "Results" and "Discussion" sections into one cohesive section. Solely describing the experimental results made it difficult for me to understand the intended purpose of the designed experiments. For example, paragraphs describing Figure 4&5 began by starting "To quantify the influence of the solar radiation…..", and then evaluated three different lights on the SFG signal of a solution with pH=4.5, it abruptly shifted to investigation on another solutions with pH=5.6 without drawing any insights from the data presented in Figure 4.. Separating these sections can reduce readability and make it challenging for readers to grasp the key points quickly derived from the experimental data, at the same time it can help in simplifying the paper because many contents are overlapped in these two sections.

In the DISCUSSION section, While I expected this section to provide deeper insights into the key findings and the key parameters influencing this reaction mechanism and how they compare to Tinel et al. (2016), the authors instead devoted two full pages (23-24) to explaining why the aromatic peak is absent in Figure 1b but appears in Figure 2b. Unfortunately, their speculative claims lack solid evidence or any supporting references.

Although it is interesting that the authors found that the shorter UV part of the solar spectrum is responsible for the studied reaction and the product abundances, this is a case study, and in my opinion, I question whether these findings could be generalized to other similar reactions (e.g. imidazole-2-carboxaldehyde or other chromophoric dissolved organic matter that serve as photosensitizers). As a result, the conclusions may have limited utility in refining photochemical

models.
In my view, this manuscript is poorly organized and has provided limited value-added insights to the community.

Specific comments

Page 3. Line 29: I would suggest the authors to define ppt as parts per thousand to avoid confusion with parts per trillion.
Page 5. Line 33: SI should be defined here.
Section 2.2: This section is actually describing the light source rather than the photochemical reactor.
Page 9 Line 10: I don't see the appearance of the new band at 3070 in Figure 2b, it is too weak to be seen, perhaps a zoom-in inset would improve clarity. This problem also applies to Figure 4&5, where I question how did the authors distinguish the signals around 3000 as C-H from aromatic compounds rather than background noise? To me, there signals appear nearly identical to the background. I reviewed the two cited studies (Gautam et al., 2000; Hardt et al., 2024) but didn't see a standard SFG spectra peak that assigned as aromatic compounds.
Page 11 Line 3: Please define the "UV part".
Page 11 Line 23-24: What are the so-called increase and decrease compared to? Based on my understanding from Figure S11, it should be except for 310nm, there is always an increase and then a decrease in the water bands with irradiation (compared to the dark condition).
Page 18 Line 5-7: A reference is needed here.
Page 18 Line 5-7: Why do not discuss in details here? Dividing the discussion into separate subsections will significantly reduce the readability of this paper.
Page 18 Line 13-16: What is the "UV part"? I would argue that the UV part above 300nm of the solar spectrum can reach the sea surface.
Page 18 Line 18-25: How do you see these? I strongly recommend that the authors combine the Discussion with the Results section. This integration will help to elaborate on the key findings more effectively while enhancing the overall flow and coherence of the paper
Page 18 Line 32: A reference is needed here.
Page 23 Line 1-19: References are needed to support the proposed two reaction pathways.
Page 24 Line 1-6: Green curve in Figure 1b is overall lower than others, not only the dangling HO band. How do you say it is because of benzaldehyde?

Technical corrections

Typing errors: Page 5. Line 25-26; Page 7 Line 19; Line 31: vapor-water → air-water (Maintain consistency in terminology);
Others: "SI" and "supporting information" were interchangeably throughout this paper. This inconsistency also applies to "Fig. X" and "Figure X", which should be unified throughout this paper.