Review of "Role of sea spray aerosol at the air-sea interface in transporting aromatic acids to the atmosphere" by Yaru Song *et al.*

Summary

Yaru Song *et al.* conducted a study to investigate the potential transport of organic acids, such as benzoic acids, from the ocean to the atmosphere via sea spray aerosols. The authors generated nascent sea spray aerosols in a laboratory setting using a sea spray simulation chamber equipped with a plunging water jet, which is a well-established method.

The authors aimed to determine the enrichment of the target organic acids in the aerosols compared to a sea spray tracer ion, sodium. Although the experimental principle aligns with previous successful studies (e.g., Johansson et al., 2019; Sha et al., 2021), the current manuscript lacks crucial details about the experimental procedures and quality control. This omission hinders the ability to assess the measurement quality.

The manuscript fails to provide the necessary information to evaluate the reasonableness of the results and the value of the upscaled estimates of global emissions. Consequently, it lacks scientific significance in its present form. Therefore, I recommend rejecting the manuscript for publication in ACP.

To enhance the manuscript's scientific rigor, I suggest the authors make a series of major improvements which I have outlined below. Subsequently, I outline some more minor points for improvement.

Major points

- In their introduction, the authors discuss the concept that the aromatic acids at the centre of their study can be of both natural and anthropogenic origin. Although this is an interesting and important discussion, the authors could expand on the relative importance of the different sources. As it stands, it is unclear whether most of the acids considered in this study are of natural or anthropogenic origin or whether they differ for the different acids investigated. A table summarising this information could help clarify the discussion.

- The description of the process of sea spray aerosol formation could be improved. For example, the initial description on line 57 is rather cumbersome and lacks accuracy. The authors can draw inspiration from the wealth of literature available on this topic to improve this section.

- The authors used a common approach to mimic the process of sea spray aerosol formation in the laboratory, namely a sea spray simulation chamber. Although such systems have

become quite common, there is diversity among the systems in use to the extent that it is important to be very precise in describing the particulars of the system in use. Since the characteristics of the different systems used can influence the properties of the aerosols generated, certain information must be conveyed to the reader. For example, the authors used a plunging jet-type system. It is important to include all the relevant details of the system used, such as the diameter of the nozzle through which the seawater flowed into the chamber, the type of pump used to generate the plunging jet, the distance between the exit of the nozzle and the surface of the seawater in the chamber, and the material from which the chamber itself was fabricated.

- It is also important to note that seawater temperature has been shown to influence the process of sea spray aerosol formation. Therefore, it would be helpful to know whether the seawater sample temperature in the chamber was controlled or monitored. Additionally, it would be useful to know how the chamber was cleaned between experiments. Furthermore, in the schematic shown in Figure S1, a 'sweep flow' of particle-free air into the chamber is shown, while the only outlet was to the SMPS." The authors did not specify where the excess air went. Did they not operate with an "overflow" exit to ensure that the chamber was always operated at atmospheric pressure? Additionally, was the Dekati LPI connected to the same outlet as the SMPS system or was it connected to another outlet so that both measurements were conducted simultaneously?

- When evaluating data from sea spray simulation systems, it is important to consider the size of the bubble plume generated by the plunging jet compared to the chamber itself. For instance, did the bubble plume reach the bottom of the chamber? If the surface foam patch is also relatively large in comparison to the surface area of the water, then "wall effects" can occur whereby bubbles reach the side and burst faster than they would if the sides had not affected the bubbles. Therefore, some insight here (perhaps some photographs) would add value to the manuscript.

- Regarding the use of a Dekati LPI and a TEM sampler to obtain offline samples for subsequent analysis, the authors did not mention obtaining blank samples to check for handling impacts. It is unclear whether blank substrates were loaded into the samplers and then analysed in the same way as the samples.

- When it comes to the results of their experiments, the authors first report the impact of different organic acids on surface tension. However, they did not provide enough information to discern how the experiments were carried out. It is assumed that the authors added or "spiked" a known concentration of the different compounds to the seawater samples they collected from the Yellow Sea. However, the concentration of the different compounds used, whether the seawater samples already contained some or all of the compounds, and the variability of the concentration of the analytes of interest in these seawater samples are not mentioned. Additionally, the presence of other surfactants in the form of organic matter derived from natural and anthropogenic sources, and how they could affect the results, is not discussed. All of this information is critical to helping the reader discern the implications of the results. Since natural seawater has variable amounts of these compounds, it is suggested that the authors could have "spiked" the analytes of interest into artificial seawater to try to negate this effect.

- The authors then report the impact of different acids on the size and number of particles that are emitted as SSA. It is unclear what the concentrations of the different analytes in these experiments were. It is also unclear whether the same seawater was used for all experiments or whether different seawater samples were spiked. The concentration of the analytes in the seawater sample and the level of organic matter in the seawater are not mentioned.

- Following this, the authors present enrichment factors of the different organic acids on the nascent sea spray aerosol they generate. However, many details are again missing, which makes it impossible to evaluate the results. For example, it is not clear to me whether the presented results are the average over all stages of the impactor or if they only represent a single stage. Furthermore, the authors did not mention how they quantified the sodium ion and the concentration of the organic acids in the same sample. Did the authors extract the substrates in ultrapure water and then subsample these extracts for the different analyses, or did they use a different approach? To generate their enrichment factor estimates, the authors will have used a concentration value for each of the acids in the seawater used to generate the aerosols. Was this value quantified, or did the authors simply use the "spike" concentration they aimed for? This is important given that surface active species, such as the organic acids used, have a tendency to stick to the walls of chambers and the actual concentration of the organic acids in the seawater may well have been well below the "spike" value the authors aimed for. Along the same lines, assuming that the authors did quantify the actual concentration of the organic acids in the seawater, it would be good to know at what point of the experiment they obtained these samples. For example, the authors state that the LPI was run for 5 hours - were water samples taken at the start or end of this period? In similar experiments using perfluoroalkyl acids, Johannsson et al. (2019) observed that the concentration of the substances in the seawater from which they were generating aerosol during their experiment decreased as these substances were "lost" to the aerosol.

- On Line 84, the authors introduce the idea of upscaling their measurements using literature values for the seawater concentration of these compounds. However, the reader requires more information at this juncture. Questions arise, such as: What are the typical seawater concentrations of these compounds? How much do they vary globally? How extensive is the dataset of such measurements? Incorporating this information into the previously introduced table (major point 1) would be beneficial. While the authors eventually provide some literature values in section 3.4 of the manuscript, it remains unclear which specific values were utilised. Was it an average of the literature values or a different approach? Considering the likely high spatial (and potentially temporal) variability in these concentrations, it is crucial to understand how the authors addressed this variability. Presently, it is nearly impossible to evaluate the magnitude of the emissions proposed by the authors due to the uncertainty in the conducted measurements.

Minor points

- Line 30: I don't think "captured" is the right word here. Perhaps "taken up" would be better.

- Line 31: Would read better as: "...leading to their enrichment within these organisms or transportation to remote areas (Fu et al.,..."
- Line 31: I would also break this sentence to aid readability, e.g.: "This process poses health risks to the endocrine system of aquatic organisms and the overall marine ecosystem (Saha et al., 2006)."
- Line 35: Would read better as "Previous studies suggest that these organic acids can alter the composition of SSA, subsequently influencing atmospheric processes such as cloud condensation nuclei (CCN) or ice nuclei (IN) activities.
- Line 36: Needs rephrasing. It is not the "organic acids" that are the source of the aerosol. Rather, it is SSA that may contain organic acids that play a role in Earth's climate.
- Line 50: Would read better as "...the identification of phthalic acid in organisms raises the possibility of its origin from the ingestion of marine plastics."
- Line 53: Would read better as "This phenomenon has prompted discussions about the concept of "missing aromatic acids."
- Line 54: Would read better as "Nevertheless, the currently available data do not provide a conclusive explanation for the existence of these "missing aromatic acids."
- Line 55: This paragraph could be better linked to the previous discussion. For example, "One possible reason for the disappearance of these aromatic acids is their release into the atmosphere. Existing datasets obtained from remote marine areas offer evidence of the presence of these compounds in the atmosphere (Fu et al., 2010)."
- Line 66: Would read better: "While field studies have demonstrated the presence of aromatic acids in both the ocean and atmosphere (Boreddy et al., 2017), the specific mechanisms influencing their transport at the air-sea interface require further investigation."
- Line 69: Here the authors introduce the air-sea transport of perfluoroalkyl acids (PFAAs) via sea spray aerosol. Although this is an interesting topic, the link to the work carried out in this study is unclear. I urge the authors to better describe this link or remove this reference.
- Line 74: Would read better as "In a previous study, we observed that the transfer of short-chain organic acids between the air and sea through SSA may be influenced by seawater surface tension. This factor could, in turn, impact the enrichment behaviour of organic acids (Song et al., 2022)."
- Line 77: The authors state "...other factors were discussed in our recent review..." What are the other factors? If this discussion is important, then the authors should do it justice and include all relevant information. Otherwise, I see no need to mention this.
- Line 78: There should be a new paragraph here: "In this study..."

References

Johansson, Jana H., et al. "Global transport of perfluoroalkyl acids via sea spray aerosol." Environmental Science: Processes & Impacts 21.4 (2019): 635-649.

Sha, B., Johansson, J. H., Benskin, J. P., Cousins, I. T., and Salter, M. E.: Influence of water concentrations of perfluoroalkyl acids (PFAAs) on their size-resolved enrichment in nascent sea spray aerosols, Environ. Sci. Technol., 55, 9489–9497, 10.1021/acs.est.0c03804, 2021