

We thank the Reviewers for the insightful comments. We have revised our manuscript according to the suggestions of the Reviewers' comments and our responses to the comments are as follows: Reviewers' comments are in black, authors' responses are in blue, and changes to the manuscript are in red color text.

Reviewer #2:

While the authors have tried to enhance their manuscript, in my assessment, the work still falls short of meeting the standards required for ACP. Numerous issues persist, including poorly written text, absence of critical information, and inadequately explained figures, making it impossible for readers to verify the authors' conclusions. Below are examples highlighting the manuscript's shortcomings:

1) Writing Quality:

The manuscript exhibits poor writing quality, hindering the comprehension of the authors' ideas. Despite the authors' likely investment in study design and high-quality data collection, the narrative fails to match the same level of quality. For instance, the first paragraph lacks definitive articles and has verb mismatches and subject-verb disagreements throughout. The same issues are repeated throughout the manuscript. Additionally, sentence structure issues, such as awkward phrasing and lack of coherence, persist throughout the manuscript. An example is the use of acronyms (SSA, CCN, and IN) without prior explanation, which hampers clarity.

Author reply:

1) We have revisited our use of definite articles and verbs and performed further changes in the revised manuscript to improve its readability.

Lines 29–30:

They are ubiquitous in seawater and the atmosphere, and have an important influence on the marine and atmospheric environment.

Lines 33–35:

Other than the influence on marine systems, they have also been reported to have negative impacts on human health, bioaccumulation, environmental persistence, and climate change (Lu et al., 2021).

Lines 41–43:

Therefore, due to their importance in upper ocean biogeochemistry and their environmental risk, the transport processes and accumulation potential of aromatic acids at the air-sea interface

require a comprehensive understanding.

Lines 51–55:

This is consistent with previous studies that phthalic acid is primarily derived from anthropogenic sources, such as plasticizer, biomass burning and fossil fuel combustion (Boreddy et al., 2022, Ding et al., 2021, Ren et al., 2023, Sanjuan et al., 2023, Shumilina et al., 2023, Yang et al., 2020, Zhu et al., 2022), whereas hydroxybenzoic acid has both anthropogenic and natural sources (Castillo et al., 2023, Liao et al., 2019, Lu et al., 2021, Lu et al., 2023, Zhao et al., 2019).

Lines 81–83:

To this end, we developed a sea spray aerosol simulation chamber using the plunging jet method, providing the closest proxy to natural SSA currently available in a controlled laboratory monitoring environment, and used it to probe the transport process of aromatic acids.

Lines 187–189:

The surface tension of seawater was examined using a surface tensiometer (Sigma 700, Biolin Scientific, Sweden) equipped with a Wilhelmy plate, calibrated at 25 °C with 30 mL of ultrapure water.

Lines 200–201:

Seawater containing p-phthalic acid ($73.92 \pm 0.14 \text{ mN m}^{-1}$) exhibited the highest surface tension among the isomers of benzene dicarboxylic acids.

Lines 227–229:

The number concentration of SSA particles with added benzene dicarboxylic acids was much higher than that with added benzoic acids.

Lines 241–244:

The distributions show a decrease in the particle number concentrations when adding hydroxybenzoic acids, compared with the system with added benzoic acids, leading to the conclusion that hydroxybenzoic acids, acting as surfactants, could inhibit SSA production (Fig. 2C).

Lines 247–249:

Importantly, Fig. 2D shows that the particle number concentration decreased proportionally with the increase of the number of methoxy groups, providing further ground that organic matter with hydrophobic functional groups have preferential atomization ability.

Lines 286–287:

Notably, it is evident that the core morphology of salt particles underwent a significant change, with the cubic structure being transformed into a sphere structure.

Lines 301–303:

Figs. 4B show the morphology images of SSA particles containing hydroxybenzoic acids, where the core maintained the crystalline phase of sea salt with its cubic structure and the coatings mainly consisted of hydroxybenzoic acids.

Lines 323–324:

The EFs order of benzene dicarboxylic acids is opposite to that of surfactants, probably due to the amphiphilicity of *o*-phthalic acid.

2) In the Introduction section, the abbreviations are explained.

Lines 63–65:

Sea spray aerosols (SSAs), generated by breaking waves and bubble bursting, are one of the major sources of atmospheric particles (Andreae and Rosenfeld, 2008, Angle et al., 2021, Hasenecz et al., 2020, Malfatti et al., 2019).

Lines 36–40:

It has been shown that the addition of oleic acid as well as surfactants in artificial seawater can significantly enhance cloud condensation nuclei (CCN) activity (Moore et al., 2011). Previous studies suggested that these organic acids can alter the composition of SSA, subsequently influencing atmospheric processes such as CCN or ice nuclei (IN) activities (Moore et al., 2011, Zhu et al., 2019).

2) Structural Improvements:

The overall structure of the writing requires significant improvement. Taking the first paragraph as an example, the complexity of the topic results in a dense paragraph that is challenging to digest. Scientific writing often benefits from a one-idea-per-paragraph approach. Breaking down this paragraph into smaller, focused sections could enhance readability without sacrificing the depth of information. For instance, dividing it into two paragraphs—one emphasising the impact of acids on the environment and humans, and the other focusing on their potential climate impacts—would improve overall clarity. This approach should be taken throughout the manuscript

Author reply:

We thank the Reviewer for the very constructive comment. We have divided some of the paragraphs into two to improve the clarity. Lines 36–43, 116–126, and 224–238 were divided into three new paragraphs, each representing the influence of other factors on climate, the measurement of experimental basic parameters, and the effect of functional group position on SSA generation, respectively.

3) Figure Presentation:

The presentation of figures, such as Figure 7, falls below the expected standard. Figures should ideally be standalone, conveying information without heavy reliance on captions. In this case, the significance of the colour bars is unclear from both the plot and the caption, indicating a need for improved figure design and clarity. Readers need to understand the visual elements independently to enhance the overall effectiveness of the figures.

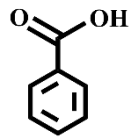
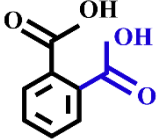
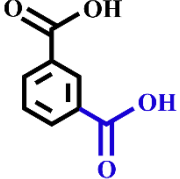
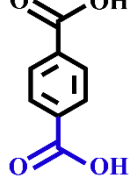
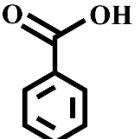
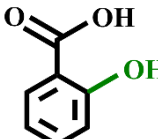
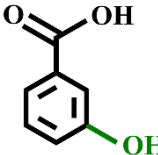
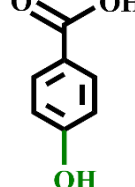
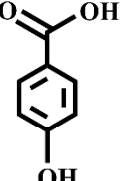
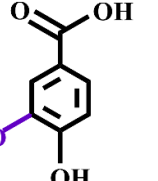
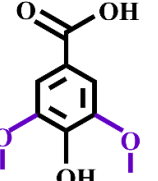
Author reply:

Thanks for the insightful comments on the figure presentation. Firstly, we have added Table 1 in an attempt to enhance the logical structure of the paper, providing readers with a clearer understanding of the structure of the article.

Lines 91–94:

Nine aromatic acids, including benzoic acid, phthalic acids (*o*-, *m*-, and *p*-isomers), hydroxybenzoic acids (*o*-, *m*-, and *p*-isomers), vanillic acid, and syringic acid, were investigated to determine the influence of functional group position and quantity on the transmission of aromatic acids at the sea-air interface (Table 1).

Table 1. Summary of aromatic acids used in experiments.

Aromatic acids				
	benzoic acid	<i>o</i> -phthalic acid	<i>m</i> -phthalic acid	<i>p</i> -phthalic acid
Position of -COOH				
	benzoic acid	<i>o</i> -hydroxybenzoic acid	<i>m</i> -hydroxybenzoic acid	<i>p</i> -hydroxybenzoic acid
Position of -OH				
	<i>p</i> -hydroxybenzoic acid	vanillic acid	syringic acid	
Number of -CH ₃ O				

Furthermore, we have modified the corresponding figures in the manuscript. Firstly, we modified the TEM images to succinctly illustrate the influence of functional group positions and quantities on the coating of organic layers.

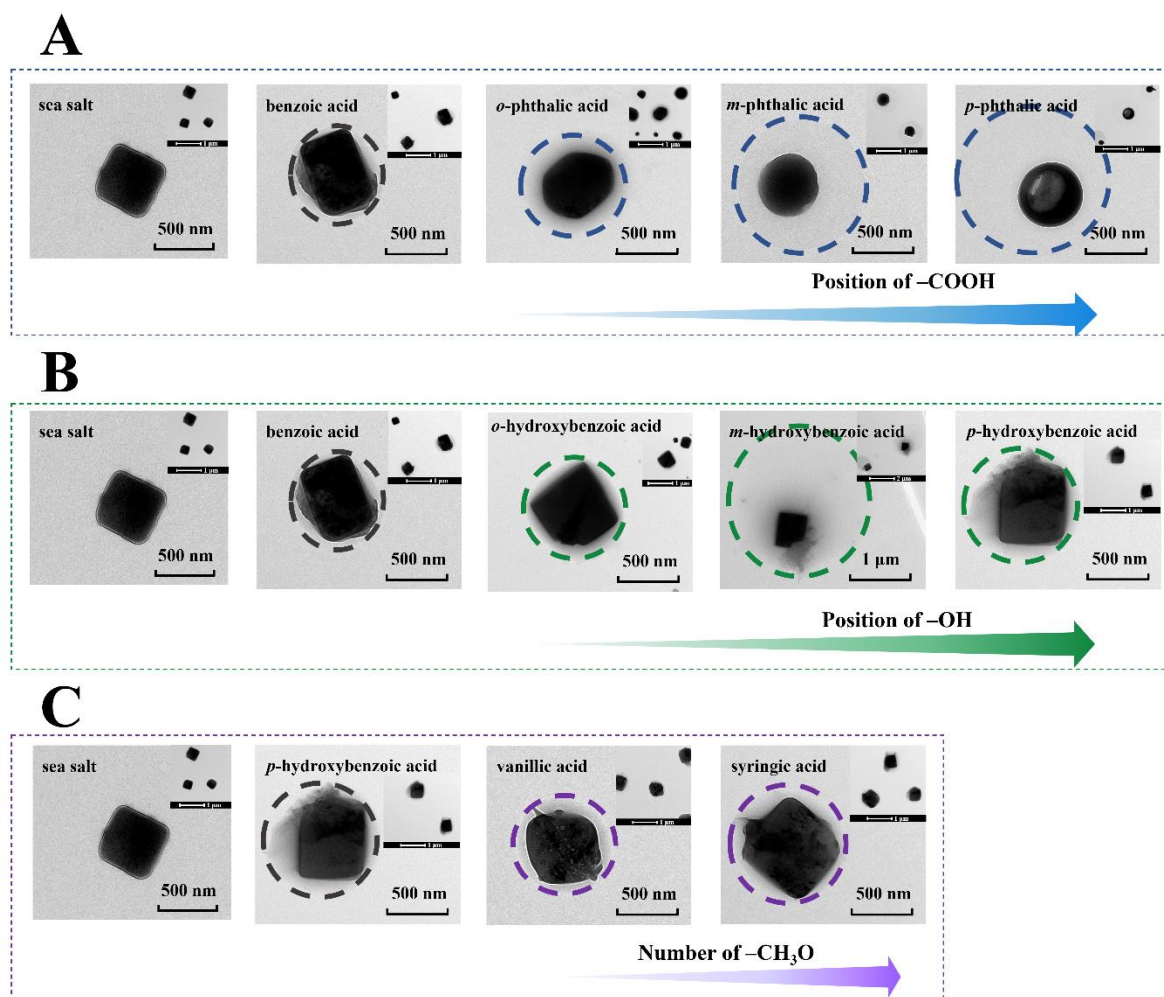


Fig. 4. Particle morphology observed using TEM of sea salt particles, and benzenedicarboxylic acids- (A), hydroxybenzoic acids- (B), *p*-hydroxybenzoic acid-, vanillic acid-, and syringic acid-coated sea salt particles (C).

Secondly, we revised the enrichment factor figure for aromatic acids by incorporating chemical structural formulas into the figures, aiming to illustrate more clearly the impact of aromatic acid structures on their enrichment at the sea-air interface.

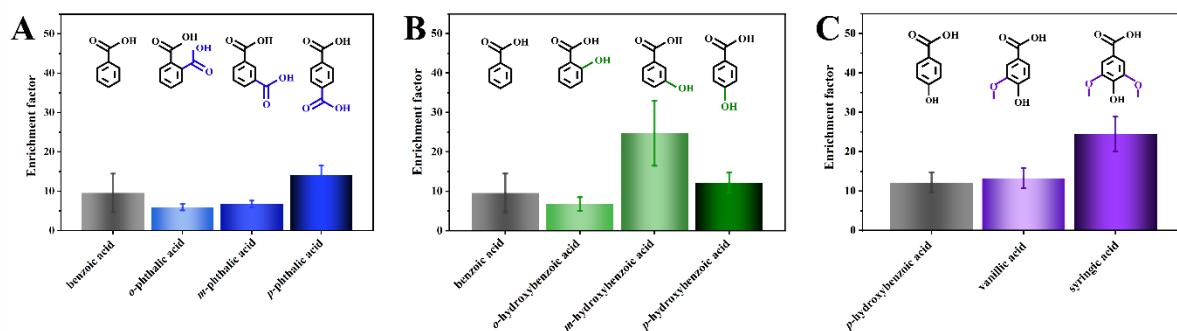


Fig. 5. Enrichment factors of benzenedicarboxylic acids (A), hydroxybenzoic acids (B), *p*-hydroxybenzoic acid, vanillic acid, and syringic acid (C) from seawater to the atmosphere.

Then, we transformed the enrichment factor figure of cations into bar graphs to better illustrate the trend of each ion's variation.

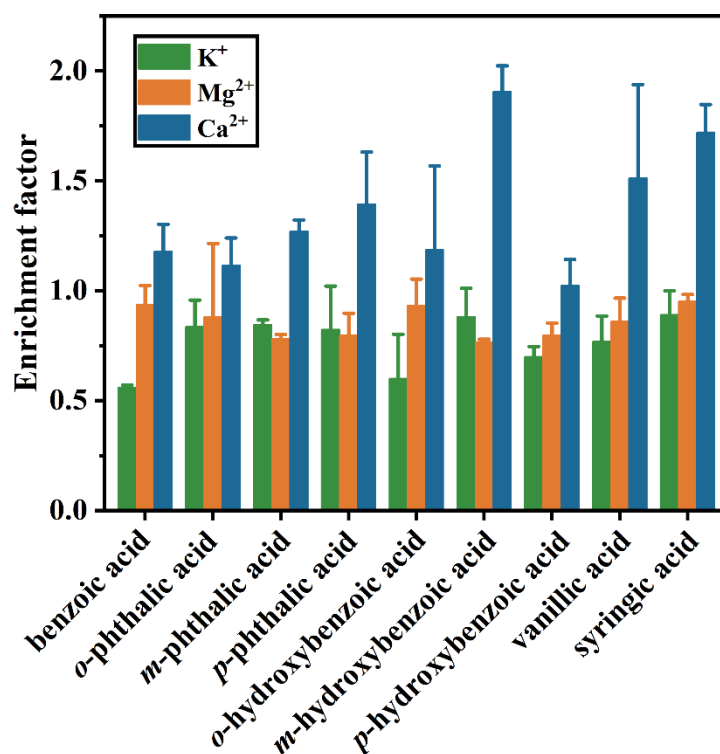


Fig. 6. Enrichment factors of K^+ , Mg^{2+} , and Ca^{2+} in submicron SSA during the experiment.

Finally, we revised Fig. 7 by separating the concentration range figure of aromatic acids from the emission flux estimates, employing colors judiciously to present the content more intuitively to the readers.

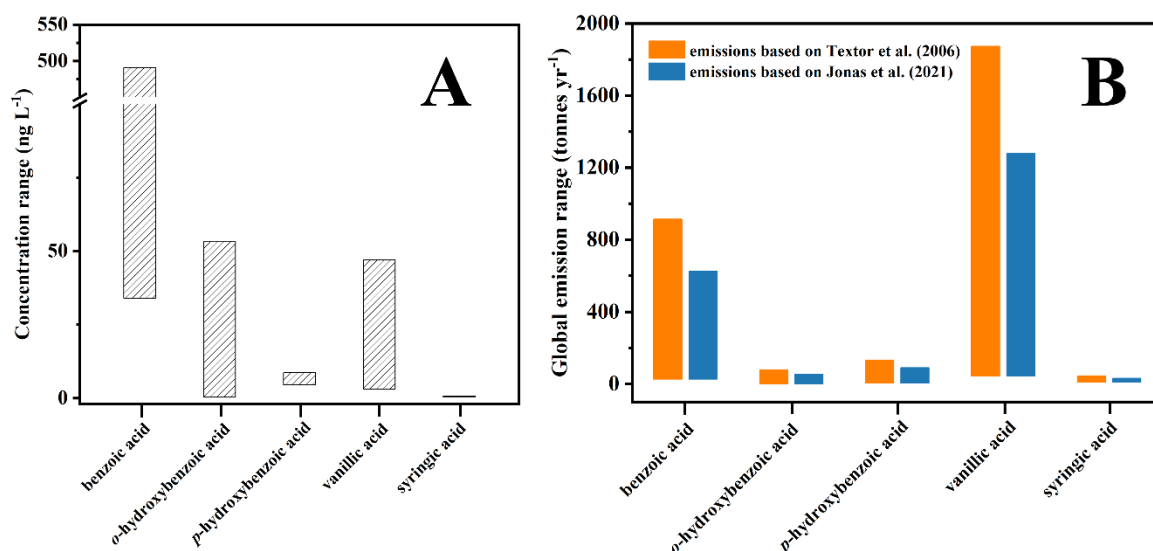


Fig. 7. Concentration range of aromatic acids in seawater (A) and the estimated range of annual global aromatic acids emission (tons yr⁻¹) via SSA (B). Yellow and blue stacked columns represent emissions based on Textor et al. (2006) and Jonas et al. (2021), respectively.

In light of these issues, I regret to inform you that I cannot recommend the publication of this manuscript at this time. Addressing the highlighted concerns, particularly improving writing quality, enhancing structural organisation, and refining figure presentation, is crucial to meet what I view as the required standards for acceptance in ACP.

Author reply:

We thank the Reviewer for the constructive comments. We have comprehensively adjusted the writing quality, organizational structure, and graphical representation of the manuscript according to the reviewer's suggestions. We believe that the quality of the manuscript has significantly improved.

Reviewer #3:

I have the feeling that the authors answered many points from the critical review (2) quite good. The sea spray generation system and quality control are much better explained and the relevance of the aromatic amino acids is much clearer now. However, I have two big issues, that were also raised in the critical review:

1. I am surprized that no amino acids were originally detected in the seawater. I believe this is due to the insensitive analytical approach (LC-MS, without enrichment or sample preparation steps). Hence the water was spiked with a very high concentration of amino acids (1 mM) that is unrealistically high regarding seawater (typical concentrations of amino acids are in the low ng/L range). The fact that the experiments were done with very high spiked concentration is clearly a limitation of the study and should be pointed out more precisely.

Author reply:

We thank the Reviewer for the thoughtful comment. The absence of detection of aromatic acids in seawater is indeed attributed to the lack of enrichment of the seawater samples. The concentration of aromatic acids in seawater ranges from 1 pM to 0.5 mM (Table S1). To investigate the influence of concentration, we introduced a set of experiments using artificial seawater with a concentration of 1 μ M for aromatic acids, and the specific experimental groups can be found in Table S3. We found a consistent enrichment trend of aromatic acids at both concentrations (Fig. S8).

And we have now pointed out this limitation in our manuscript.

Lines 103–106:

For simplified consideration, aromatic acids were added separately to artificial seawater to achieve concentrations of 1 μ M and 1 mM, in order to verify the effects of background systems and concentrations on the EF of aromatic acids, which may not accurately reflect realistic conditions but provide an approximated trend of EFs instead.

Table S1. Sources and concentrations of aromatic acids identified in seawater and atmospheric samples over the ocean.

Aromatic acids	Natural sources	Anthropogenic sources
benzoic acid	<ul style="list-style-type: none">● sea algae (Abdel-Hamid A. Hamdy, 2020; Al-Zereini et al., 2010; Fotso Fondja Yao et al., 2010; Liu et al., 2022b)● sedimentary organic matter (10–65 $\mu\text{g g}^{-1}$) (Deshmukh et al., 2016)● bacteria isolated from sea bass viscera (0.3 μM) (Martí-Quijal et al., 2020)● snow pit samples (2.11 ng g^{-1}) (Mochizuki et al., 2016)	<ul style="list-style-type: none">● emerging endocrine disrupting compounds (0.3–4.0 nM) (Zhao et al., 2019)● fuel combustion (Boreddy et al., 2017)● industrial wastewater, automobile exhaust and tobacco smoke (Cuadros-Orellana et al., 2006)
<i>o</i> -phthalic acid		<ul style="list-style-type: none">● plasticizer (16.7–657 ng g^{-1} d.w.) (Ren et al., 2023; Sanjuan et al., 2023);● plastic waste burning (8.3–84.9 ng m^{-3}) (Zhu et al., 2022)● the end product of photochemical oxidation of SOA (15.5 ng m^{-3}) (Ding et al., 2021)● biomass burning and fossil fuel combustion sources (0.4–7.9 ng m^{-3}) (Shumilina et al., 2023; Yang et al., 2020; Boreddy et al., 2022)
<i>m</i> -phthalic acid		<ul style="list-style-type: none">● plasticizer (Ren et al., 2023)● the end product of photochemical oxidation of SOA (3.6 ng m^{-3}) (Ding et al., 2021)

		<ul style="list-style-type: none"> ● biomass burning and fossil fuel combustion sources (0.01–2.3 ng m⁻³) (Yang et al., 2020; Boreddy et al., 2022; Kawamura, 2014)
<i>p</i> -phthalic acid		<ul style="list-style-type: none"> ● plasticizer (0.51–6.8 mg kg⁻¹) (Ren et al., 2023; Di Giacinto et al., 2023; Di Renzo et al., 2021) ● plastic waste burning (10.8–80.7 ng m⁻³) (Zhu et al., 2022); the end product of photochemical oxidation of SOA (4.3 ng m⁻³) (Ding et al., 2021) ● biomass burning and fossil fuel combustion sources (0.05–2.5 ng m⁻³) (Yang et al., 2020; Boreddy et al., 2022; Kawamura, 2014)
<i>o</i> -hydroxybenzoic acid	<ul style="list-style-type: none"> ● sea algae (0.5 mM) (Castillo et al., 2023; Mostafa et al., 2017; Klejdus et al., 2017) 	<ul style="list-style-type: none"> ● pharmaceuticals and drugs of abuse (2.8–385.9 pM) (Alygizakis et al., 2016)
<i>m</i> -hydroxybenzoic acid	<ul style="list-style-type: none"> ● sea algae (Al-Zereini et al., 2010; Castillo et al., 2023) 	
<i>p</i> -hydroxybenzoic acid	<ul style="list-style-type: none"> ● sea algae (0.4 mM) (Castillo et al., 2023; Klejdus et al., 2017; Tian et al., 2012; Hawas and Abou El-Kassem, 2017) ● sea fungus (Rukachaisirikul et al., 2010; Shao et al., 2007) ● sponge Mycale species (Xuefeng Zhou, 2013); metabolite (Jingchuan Xue, 2015; Liao and Kannan, 2018) ● sediment samples (6.85–437 ng g⁻¹ dw) (Liao et al., 2019) 	<ul style="list-style-type: none"> ● Pharmaceuticals and personal care products (Lu et al., 2023) ● emerging endocrine disrupting compounds (0.03–0.4 nM) (Zhao et al., 2019; Lu et al., 2021; Alygizakis et al., 2016)
vanillic acid	<ul style="list-style-type: none"> ● sea algae (0.02–0.3 nM) (Zangrando et al., 2019; Klejdus 	<ul style="list-style-type: none"> ● combustion of both softwood and hardwood

	<p>et al., 2017)</p> <ul style="list-style-type: none"> ● lignin decomposition (Wang et al., 2015; Hu et al., 2022; Xu et al., 2017) 	(Simoneit, 2022)
syringic acid	<ul style="list-style-type: none"> ● sea algae (1.5–3 pM) (Poznyakovsky et al., 2021; Zangrando et al., 2019; Klejdus et al., 2017) ● lignin decomposition (Hu et al., 2022; Xu et al., 2017) 	<ul style="list-style-type: none"> ● pharmaceuticals (Fisch et al., 2017) ● hardwood burning (Simoneit, 2022)

Table S3. Summary of experimental conditions.

Exp. No.	Experiment type	Concentration (mM)	pH	Salinity (psu)	Sampling time (h)	RH (%)	Temperature difference (°C) ^a
1	SW	0	7.92	34.2	5	35	2.0
2	SW+benzoic acid	1	7.72	34.3	5	34	1.5
3	SW+ <i>o</i> -hydroxybenzoic acid	1	7.60	34.5	5	36	1.0
4	SW+ <i>m</i> -hydroxybenzoic acid	1	7.68	34.1	5	40	2.0
5	SW+ <i>p</i> -hydroxybenzoic acid	1	7.84	34.3	5	38	1.5
6	SW+ <i>o</i> -phthalic acid	1	7.58	34.2	5	36	2.0
7	SW+ <i>m</i> -phthalic acid	1	7.80	34.5	5	37	2.5
8	SW+ <i>p</i> -phthalic acid	1	7.85	34.4	5	42	2.0
9	SW+vanillic acid	1	7.81	34.2	5	43	3.0
10	SW+syringic acid	1	7.84	34.3	5	39	2.0
11	ASW	0	7.96	35.1	5	33	1.5
12	ASW+benzoic acid	1	7.68	34.6	5	35	1.0
13	ASW+ <i>o</i> -hydroxybenzoic acid	1	7.76	34.9	5	34	0.5
14	ASW+ <i>m</i> -hydroxybenzoic acid	1	7.99	35.3	5	36	1.5
15	ASW+ <i>p</i> -hydroxybenzoic acid	1	7.85	34.7	5	38	2.0
16	ASW+ <i>o</i> -phthalic acid	1	7.93	34.5	5	35	1.0
17	ASW+ <i>m</i> -phthalic acid	1	7.88	34.9	5	36	1.0

18	ASW+ <i>p</i> -phthalic acid	1	7.97	34.6	5	34	1.5
19	ASW+vanillic acid	1	7.89	35.2	5	35	1.0
20	ASW+syringic acid	1	7.99	34.8	5	39	1.0
21	ASW+benzoic acid+ <i>o</i> -hydroxybenzoic acid+ <i>o</i> -phthalic acid+vanillic acid+syringic acid	10 ⁻³	7.95	35.1	20	41	3.5
22	ASW+benzoic acid+ <i>m</i> -hydroxybenzoic acid+ <i>m</i> -phthalic acid+vanillic acid+syringic acid	10 ⁻³	7.98	34.6	20	38	1.5
23	ASW+benzoic acid+ <i>p</i> -hydroxybenzoic acid+ <i>p</i> -phthalic acid+vanillic acid+syringic acid	10 ⁻³	7.88	34.9	20	40	2.0
24	NaCl	0	7.68	35.3	5	38	1.0
25	NaCl+ <i>m</i> -hydroxybenzoic acid	1	7.54	34.7	5	36	1.5

^a The temperature difference in the SSA simulation chamber before and after the experiment.

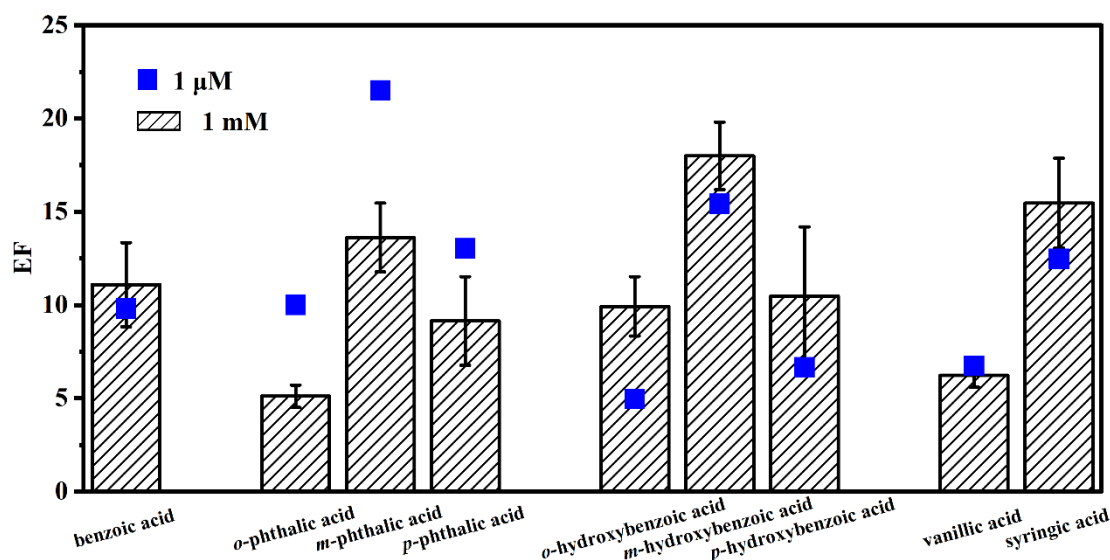


Fig. S8. Enrichment factors of aromatic acids at different concentrations from artificial seawater to the atmosphere.

2. As mentioned by the reviewer, the extrapolation to annual global amino acid emissions is very crude and also in the revised form, the effect of e.g. seawater temperature is not considered. This part should be overworked again.

Author reply:

We thank the Reviewer for the valuable comment and we agree that temperature has an impact on SSA emission fluxes. In this study, we have roughly estimated the annual emissions of aromatic acids at experimental temperature. Firstly, we greatly appreciate your input, however, due to the current study focuses on the enrichment behavior of structurally different aromatic acids under room temperature conditions and the influencing factors of their enrichment trend, I'm very sorry that we did not add additional experimental content at this time. Secondly, the enrichment process in field studies is much more complicated than in laboratory experiments. In field measurements for instance, seawater temperature affects SSA emission, while sea surface wind speed may affect SSA particle release and size, etc. Finally, the concentration of target compounds in seawater is a more significant contributor to the estimates of k_{SSA} (Sha et al., 2021). Therefore, the effect of temperature is not discussed in detail in the current manuscript. Nevertheless, we will carefully consider your suggestions and explore them to the best of our ability in future work. And we point out in the manuscript the limitations of temperature in this manuscript.

Lines 369–373:

Furthermore, the enrichment process in the field is much more complicated than in controlled laboratory experiments. For example, seawater temperature affects SSA release, while wind speed at the sea surface may influence the amount and size of SSA particles emitted, etc. As such, further research on the environmental enrichment mechanism of aromatic acids in SSA is required to reduce the uncertainty in the estimation of aromatic acid emissions.

Finally, some more recent studies on amino acids in the marine environment could be included in this work.

Author reply:

We added some recent studies on aromatic acids in the marine and atmosphere environment in our manuscript.

Lines 40–41:

In addition, organic acids, such as carboxylic acids, aromatic acids also contribute significantly to ocean acidification (Kumari et al., 2022).

Lines 51–55:

This is consistent with previous studies that phthalic acid is primarily derived from anthropogenic sources, such as plasticizer, biomass burning and fossil fuel combustion (Boreddy et al., 2022, Ding et al., 2021, Ren et al., 2023, Sanjuan et al., 2023, Shumilina et al., 2023, Yang et al., 2020, Zhu et al., 2022), whereas hydroxybenzoic acid has both anthropogenic and natural sources (Castillo et al., 2023, Liao et al., 2019, Lu et al., 2021, Lu et al., 2023, Zhao et al., 2019).

Lines 61–62:

Although recent studies outline the critical sources of aromatic acids, much less is known about its transportation via SSA (Castillo et al., 2023, Hu et al., 2022).

Altogether I think that the manuscript could be published if these points are addressed (once more). I hope this helps, due to time constraints I am currently not able to provide a more detailed review.

Author reply:

Thank you for the very helpful comments from the reviewer. We have made corresponding revisions in the manuscript.

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