

We thank the Reviewer for the insightful comments. We have revised our manuscript according to the suggestions of the Reviewer's comments and our responses to the comments are as follows: Reviewer's comments are in black, authors' responses are in blue, and changes to the manuscript are in red color text. Figures prepared for reply are named as Figure R-.

Reviewer #2:

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.

Author reply:

We thank the Reviewer for the thoughtful comments and valuable suggestions that will contribute without doubt to improve our original manuscript. Based on these major and minor points below, we have revised our manuscript.

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.

Author reply:

For the aromatic acids we studied, benzoic and hydroxybenzoic acids as well as vanillic and syringic acids in seawater have both natural and anthropogenic sources, and benzene dicarboxylic acids are mainly derived from anthropogenic sources. We have rewritten some sentences and summarized in a table as follows.

Line 41-45:

In various observations, aromatic acids have been detected in both natural and anthropogenic sources (Zhao et al., 2019; Zangrando et al., 2019; Dekiff et al., 2014). Among them, natural sources of aromatic acids produced by algal releases account for most of marine aromatic acids, especially benzoic acid, and most of hydroxybenzoic acids (Mostafa et al., 2017; Fotso Fondja Yao et al., 2010a; Castillo et al., 2023; Abdel-Hamid A. Hamdy, 2020).

Line 50-53:

This is consistent with previous researches that phthalic acid is primarily derived from anthropogenic sources (Ren et al., 2023a), whereas hydroxybenzoic acid has both anthropogenic and natural sources (Zhao et al., 2019; Castillo et al., 2023).

Line 89-90:

Further details are provided in Table S1, which lists sources and concentrations of these aromatic acid identified in seawater and atmospheric samples over the ocean.

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., 2010b; Liu et al., 2022b)● sedimentary organic matter (10–65 $\mu\text{g g}^{-1}$) (Deshmukh et al., 2016)● bacteria isolated from sea bass viscera (314 ppb) (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 (34–491 ng L^{-1}) (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., 2023b; 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., 2023b) ● the end product of photochemical oxidation of SOA (3.6 ng m⁻³) (Ding et al., 2021) ● 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., 2023b; 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 (76.8 mg L⁻¹) (Castillo et al., 2023; Mostafa et al., 2017; Klejdus et al., 2017) 	<ul style="list-style-type: none"> ● pharmaceuticals and drugs of abuse (0.4–53.3 ng L⁻¹) (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 (57.7 mg L⁻¹) (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 (4.58–49.9 ng L⁻¹) (Zhao et al., 2019; Lu et al., 2021; Alygizakis et al., 2016)
vanillic acid	<ul style="list-style-type: none"> ● sea algae (3–47 ng L⁻¹) (Zangrando et al., 2019; Klejdus et al., 2017) ● lignin decomposition (Wang et al., 2015; Hu et al., 2022; Xu et al., 2017) 	<ul style="list-style-type: none"> ● combustion of both softwood and hardwood (Simoneit, 2022)
syringic acid	<ul style="list-style-type: none"> ● sea algae (0.3–0.6 ng L⁻¹) (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)

- 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.

Author reply:

We have modified the original text as follows.

Line 59-67:

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). SSAs can act as carrier agents for the vertical transport of much more than just sea salt and often include organic surfactants in the ocean, as already shown by field and laboratory studies (Cochran et al., 2016; Franklin et al., 2022; Rastelli et al., 2017). Recent data indicated that the surface activity and octanol-water partitioning coefficients (K_{ow}) of organic compounds may affect their transport efficiency from the water phase to the atmosphere (Olson et al., 2020). Moreover, molecular structure may induce changes of organic acids properties (i.e., surface tension, toxicity), which further affect their transport potential and global emission flux (Lee et al., 2017; Rastelli et al., 2017; Frossard et al., 2019; Van Acker et al., 2021).

- 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.

Author reply:

We have added more contents to make the manuscript more complete.

Lines 99-100:

A jet-based laboratory SSA simulation chamber, made of 316L stainless steel material, was used to mimic the SSA generation (Fig.S2).

Lines 104-106:

When used, plunging jets were cycled in seawater by the pump (Shenchen V6-6, China) at a flow rate of 1 L min⁻¹ through a stainless-steel nozzle (inner diameter 4.3 mm).

Line 103:

The detailed dimensions of the SSA simulation chamber are provided in Table S2.

Table S2. Dimensions and operating conditions of the SSA simulation chamber.

Characteristic	Value
Nozzle diameter (mm)	4.3
Seawater depth (cm)	15
Seawater volume (L)	9
Headspace depth (cm)	22
Headspace volume (L)	15
Zero sweep air (L min ⁻¹)	3
Headspace residence time (min)	5
Plunging jet flow rate (L min ⁻¹)	1

The reasons for the selection of the relevant parameters are as follows:

1) A stainless steel nozzle with an inner diameter of 4.3 mm is used to generate plunging jets. The nozzle can be changed easily. The size of the nozzle is consistent with that in previous studies (Salter et al., 2014; Sha et al., 2021).

2) We have studied the cases with headspace heights of 14 cm, 19 cm, and 22 cm in our previous work (Liu et al., 2022a) and found that the SSA production was most efficient at a headspace height of 22 cm. Hence, we chose it for SSA generation.

3) We have tried pump flow rates of 0.50 L min⁻¹, 0.75 L min⁻¹, 1.00 L min⁻¹, 1.25 L min⁻¹, and 1.50 L min⁻¹ in our previous study (Liu et al., 2022a) and found that when the pump flow rate was 1 L min⁻¹ or higher, the performance of the SSA generator was good (see Table R1). Therefore, we used a pump flow rate of 1 L min⁻¹ to maintain a high SSA production rate and a relatively long working lifetime of the corresponding accessories.

Table R1. Effect of pump flow rate on SSA production.

Pump flow rate (L min ⁻¹)	SSA production (particles s ⁻¹)	Mass concentration (μg m ⁻³)
0.50	5.78×10^6	198.96
0.75	9.90×10^6	317.31
1.00	1.37×10^7	417.86
1.25	1.58×10^7	469.59
1.50	1.63×10^7	495.42

4) We have previously studied SSA production at purge air flow rates of 2 L min⁻¹, 3 L min⁻¹, 4 L min⁻¹, 5 L min⁻¹, 6 L min⁻¹ to simulate the sea breeze (Liu et al., 2022a). We found that the SSA production increased with the purge air flow rate when the flow rate was less than 3 L min⁻¹, and decreased with the increase of the purge air when the flow rate was more than 3 L min⁻¹.

5) Based on a previous study of the effect of relative humidity on the growth of sea salt particles, the inlet humidity would affect the morphology of SSA (Tang et al., 1997). Hence, we chose to keep the relative humidity at about 40% in our study. Particles will deliquesce at higher humidity (Bryan et al., 2022). In addition, it has been demonstrated that acid has no effect on NaCl deliquesce (Ming and Russell, 2001). Combining the above parameters, we obtained a particle size distribution similar to that from field observations (Quinn et al., 2017; Xu et al., 2022).

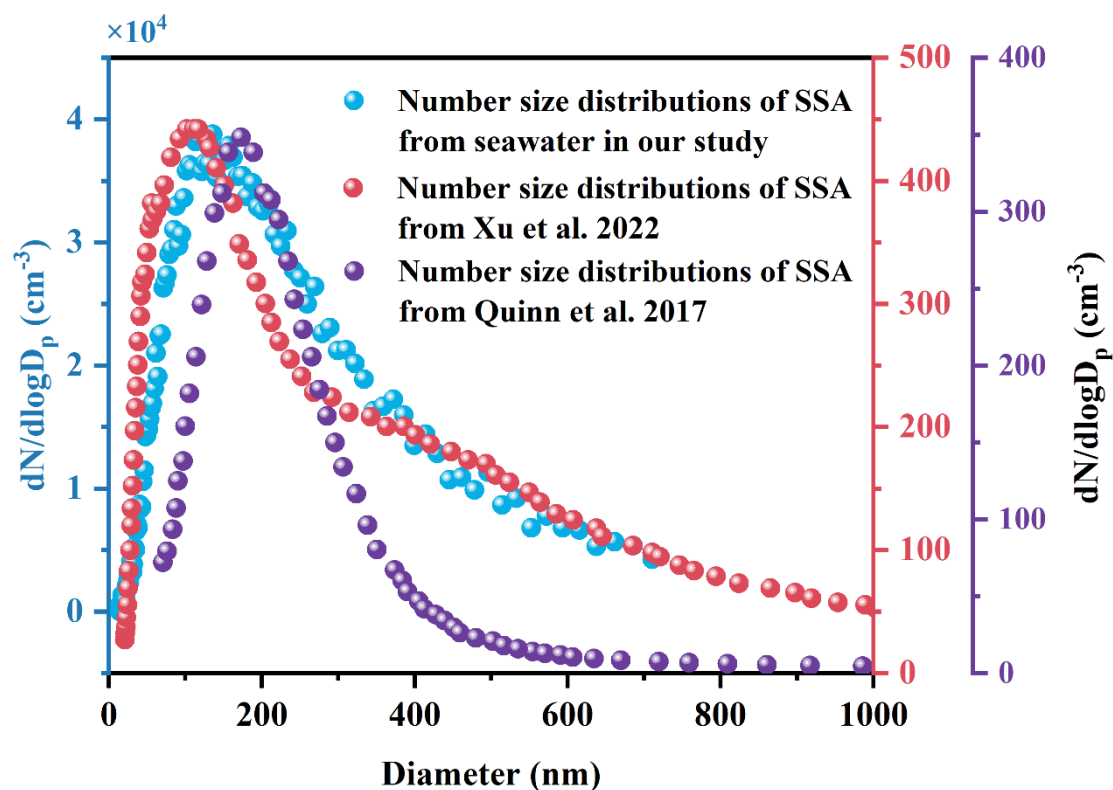


Fig. S2. Number size distribution of SSA generated with the SSA simulation chamber in this study compared with field studies.

- 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?

Author reply:

1) Seawater temperature:

We controlled the laboratory temperature around 22–25°C during the experiments and examined the seawater temperature in the SSA simulation chamber before and after the experiments, and the corresponding results are supplemented in Table S3.

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.

2) SSA simulation chamber cleaning:

Between experiments, the SSA simulation chamber was cleaned with ethanol first, then rinsed with ultrapure water, blown with zero air to reduce the influence of organic acids residue. The above steps run the pump to allow for thorough cleaning of the system.

S1. Quality assurance/quality control.

Seawater was collected from the coastal area of Shazikou on March 27, 2023, with a volume of 500 L (Fig. S1). Considering the storage inconvenience caused by huge consumption of seawater, all our seawater was pre-filtered through a polyethersulfone filter (47 mm diameter, 0.2 μm pore size, Supor®-200, Pall Life Sciences, USA) and stored in the dark at 18 °C for less than one month. Quinn et al. (2015) have shown that the fraction that passes through the filter is regarded as dissolved organic carbon and includes colloidal and truly dissolved materials. For each experiment, we measured particle number concentrations generated by filtered seawater and cations concentrations in seawater, and we found good agreement between each set of experiments (see Fig. S2).

In order to avoid the influence of organic matter in quartz fiber filters and access the accuracy of the experiment, pre-baked quartz fiber filters were used in sampling. Before each set of experiments, experimental blanks were conducted using filtered seawater. Experimental blanks were conducted with the same procedure of SSA samples. Seawater and filter samples were stored at -20°C until analyzed. In order to reduce the influence of organic acids residue after each experiment, the SSA simulation chamber was cleaned with ethanol first, then the system was cleaned with ultra-pure water for several times. The above steps also run the pump to allow for thorough cleaning of the system. Thereafter, the system was blown with zero air and sealed for preservation. The Dekati DLPI was also ultrasonicated with methanol and water (V:V=1:1) and dried after the experiment.

Filtered seawater (without added aromatic acid) was used as the experimental blank, and the same experimental and analytical methods were used as those for the experimental samples. As a result, no target aromatic acid was found in both seawater and filters. This may be due to the fact that we did not perform any concentration operation during the seawater sample processing. The standard curves for each aromatic acid are linear, as shown in Fig. S5.

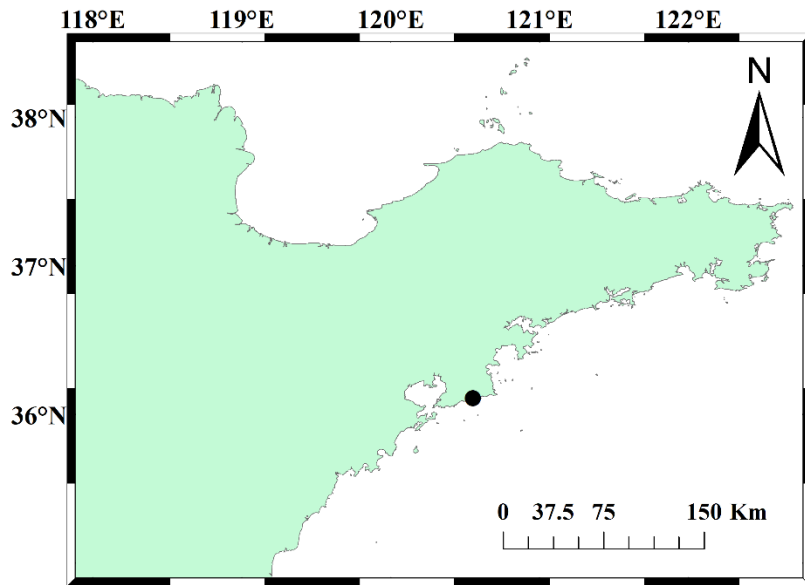


Fig. S1. Sampling site at Shazikou along the Yellow Sea coast, Qingdao, China.

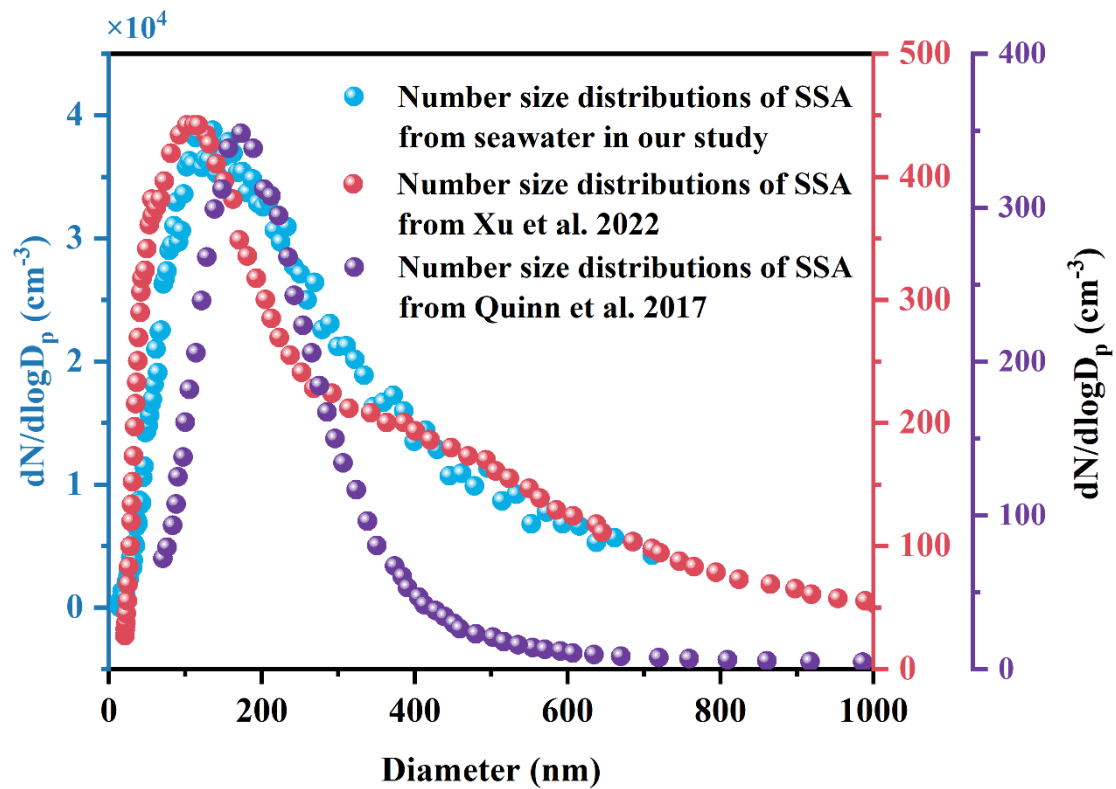


Fig. S2. Number size distribution of SSA generated with the SSA simulation chamber in this study compared with field studies.

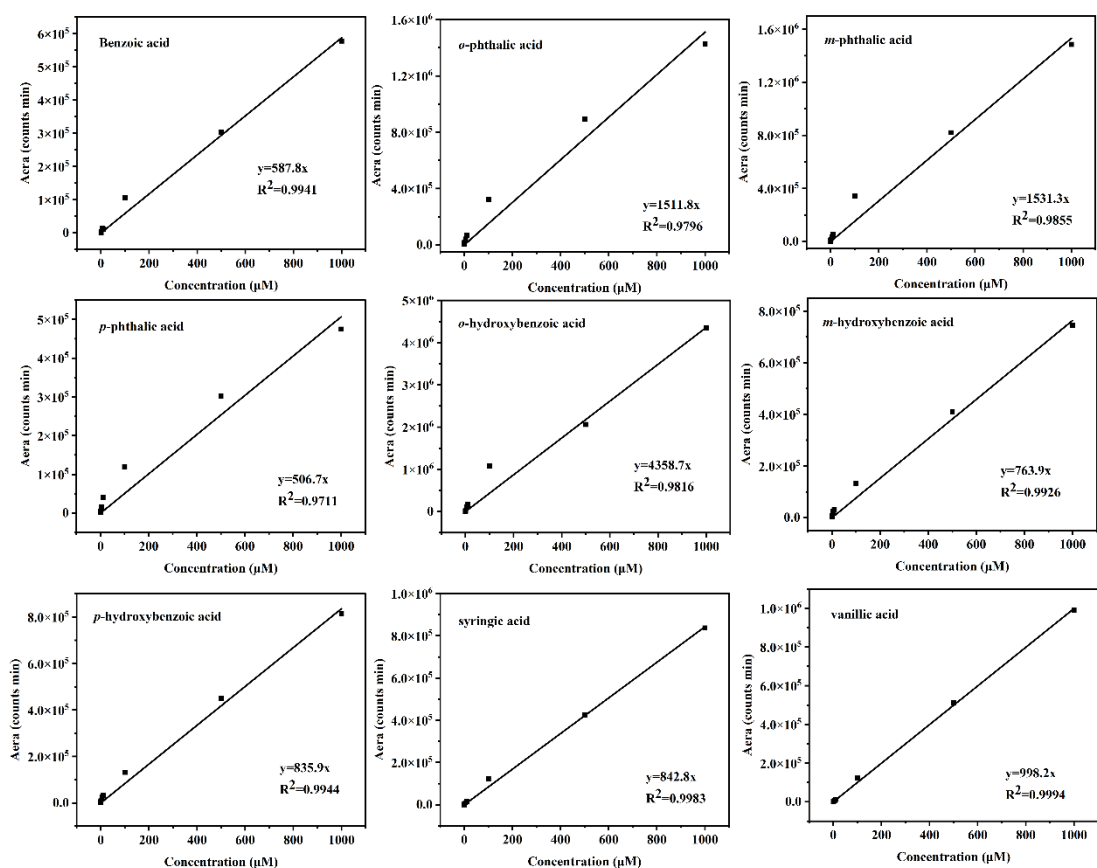


Fig. S5. The standard curves for aromatic acids were constructed within a concentration range of 0.01-1000 μM , with more than seven data points.

3) Experimental setup:

We used a three-way valve to allow excess gas to escape, ensuring that the chamber operates at atmospheric pressure. The DeKati DLPI+ was connected to the same outlet as the SMPS system. The stabilization of the particles total number concentration suggests that there is little SSA particle adsorption in the chamber. The decrease in aromatic acid concentration may then be due to the transfer of SSA. Therefore, the sample collection for the SMPS system and the DeKati DLPI+ were conducted separately.

We inserted these sentences and figures in the revised manuscript and supplement to make it clearer.

Lines 111-112:

The outlet was fitted with a three-way valve to transfer the sample airflow and the excess gas.

Lines 128-129:

After the size distribution stabilized, a total of 40 mL of seawater is collected from a tap located 1.5 cm above the bottom of the chamber.

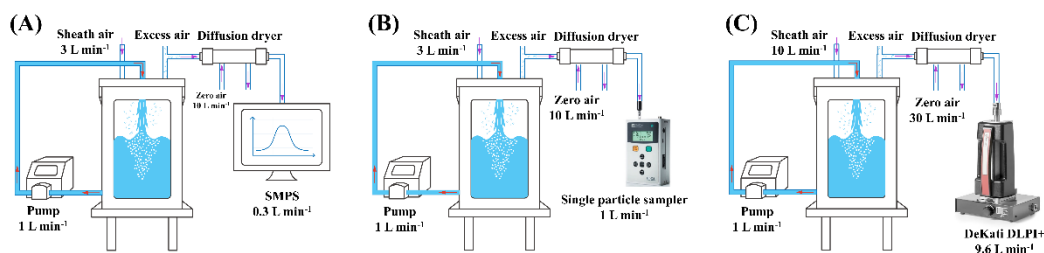


Fig. S3. Schematic picture of the plunging jet-sea spray aerosol generator: SMPS sampling (A), single particle sampling (B), and DeKati DLPI+ sampling (C). The red arrows represent the flow direction of seawater, and the purple arrows represent the flow of gases and aerosol particles.

- 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.

Author reply:

- 1) The bubble plume reached the bottom of the SSA simulation chamber.
- 2) The physical diagram of the experiment and the top view of the bubble bursting are shown below. Furthermore, due to the similar enrichment levels of 1 μ M and 1 mM aromatic acids in SSA, we speculate that the main factor influencing the enrichment factor is aerosolization rather than the "wall effect".

The following text was added in the revised manuscript.

Line 107-108:

During the experiment, it was assumed that the interaction of the bubble plume generated by the plunging jet with the wall was negligible (Fig. S4).

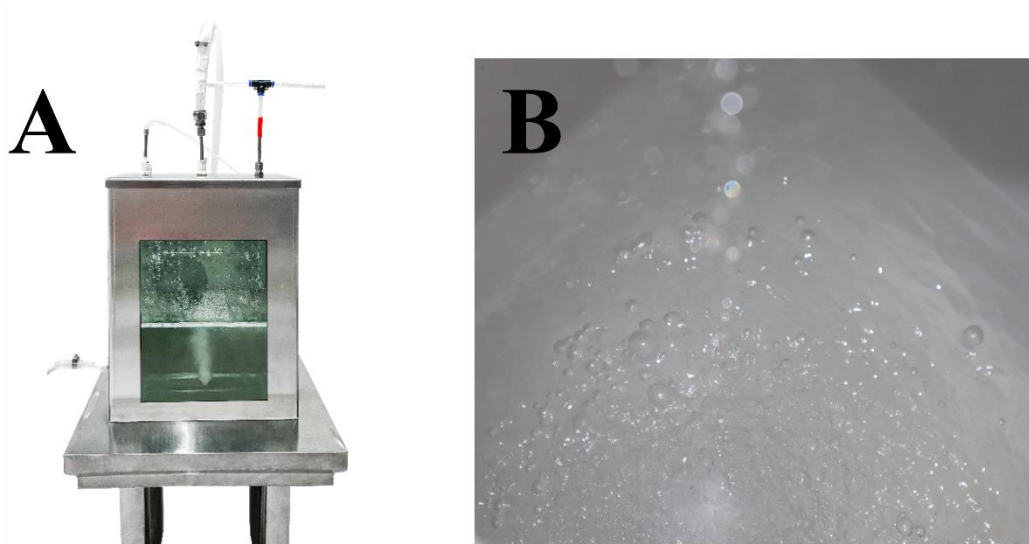


Fig. S4. Physical diagram of the SSA simulation chamber (A) and the top view of the bubble generation in the chamber (B).

- 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 analyzed in the same way as the samples.

Author reply:

During the experiment, we used filtered seawater (no aromatic acids added) as the experimental blank and then analyzed in the same way as the samples. We measured the concentration of aromatic acid in filtered seawater and filters all below the detection limit. The TEM images of the filtered seawater (no aromatic acids added) have been shown in Fig. 4A.

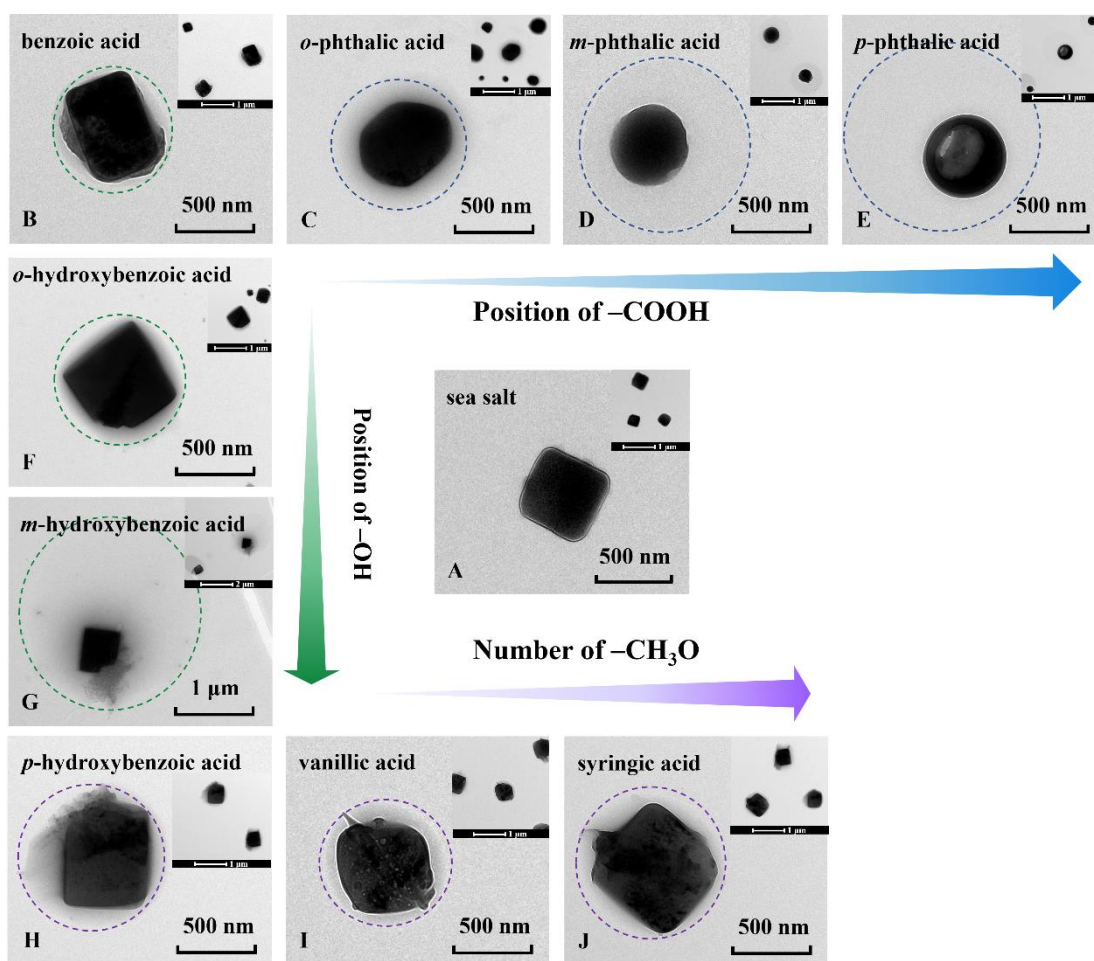


Fig. 4. Particle morphology observed using TEM of sea salt (A) and mixed particles composed of aromatic acids-coated sea salt particles (B–K).

- 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.

Author reply:

1) The experiment consisted of a total of 25 sets with target compound concentrations of 10^{-3} and 1 mM (Table S3). Each aromatic acid was added individually to seawater for the experiment. The SMPS recorded particle number concentrations, and sampling was performed once the size distributions stabilized. Seawater samples, totaling 40 mL, were collected from a tap located 1.5 cm above the bottom of the chamber. After collection, quartz fiber filter (QFF) containing aromatic acids was subjected to a 40-minute ultrasonic extraction in 4 mL of Milli-Q water to dissolve submicron SSA particles. The solution was then filtered through a 0.45 μm syringe filter to eliminate filter fragments, for subsequent concentration calculations and EF analysis. After each experiment, the SSA chamber underwent thorough cleaning as outlined in QA/QC (see S1).

We have inserted the following text in the revised manuscript to make the experimental process clearer.

Lines 176-179:

The experiment consisted of a total of 25 sets with target compound concentrations of 10^{-3} and 1 mM (Table S3). Each aromatic acid was added individually to seawater for the experiment. Surface tension of seawater was examined using a surface tensiometer (Sigma 700, Biolin Scientific, Sweden) equipped with a Wilhelmy plate, which was calibrated at 25 °C with 30 mL ultrapure water.

Lines 128-129:

After the size distribution stabilized, a total of 40 milliliters of seawater was collected from a tap located 1.5 cm above the bottom of the chamber.

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
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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.

2) During the experimental process, filtered seawater (without the addition of aromatic acids) was employed as a procedural blank. Subsequently, analysis was conducted using the same methods as applied to the samples. However, the targeted aromatic acids were not detected in the seawater, possibly due to their concentrations being below the detection limit. We have added relevant information in the Supplement.

Filtered seawater (without added aromatic acid) was used as the experimental blank, and the same experimental and analytical methods were used as those for the experimental samples. As a result, no aromatic acid was found in both seawater and filtered samples. This may be due to the fact that we did not perform any concentration operation during the water sample processing.

3) ASW were selected as the bulk water to characterize the influence of organic matter in our previous study (Song et al., 2022). We concluded that considering ASW as bulk waters will result in a deviation of SSA production from that produced by seawater. And the particles size distribution obtained by the lognormal-mode-fitting approach in our previous work (Fig. S2) is very similar to the distributions observed in previous studies (Xu et al., 2022; Quinn et al., 2017).

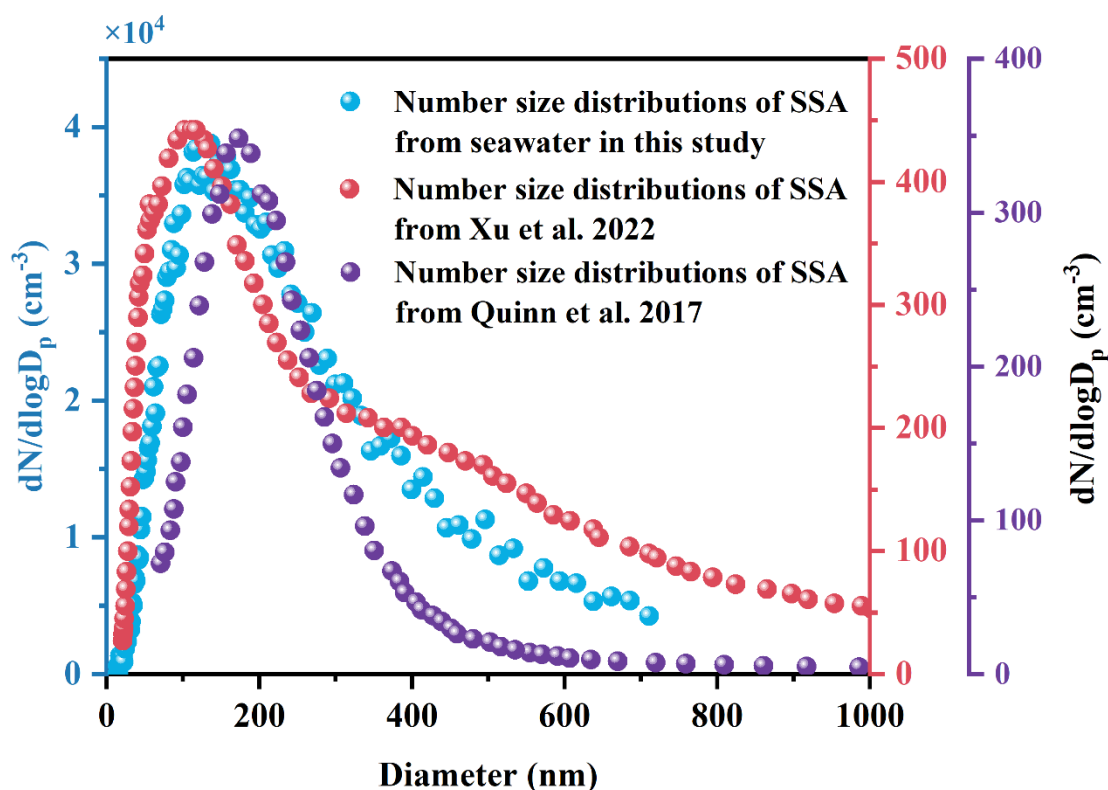


Fig. S2. Number size distribution of SSA generated with the SSA simulation chamber in this study compared with field studies.

Additionally, to enhance the rigor of our results, we have “spiked” the target analytes into artificial seawater, and the following text has been added in the revised manuscript. Lines 177-179:

Each aromatic acid was added individually to seawater and artificial seawater (ASW) for the experiment. Surface tension of seawater was examined using a surface tensiometer (Sigma 700, Biolin Scientific, Sweden) equipped with a Wihelmy plate, which was calibrated at 25 °C with 30 mL ultrapure water.

Lines 181-183:

However, the average surface tension value of ASW was 74.5 mN m⁻¹ (Fig. S6), indicating that the presence of surfactants in seawater enhances its surface activity.

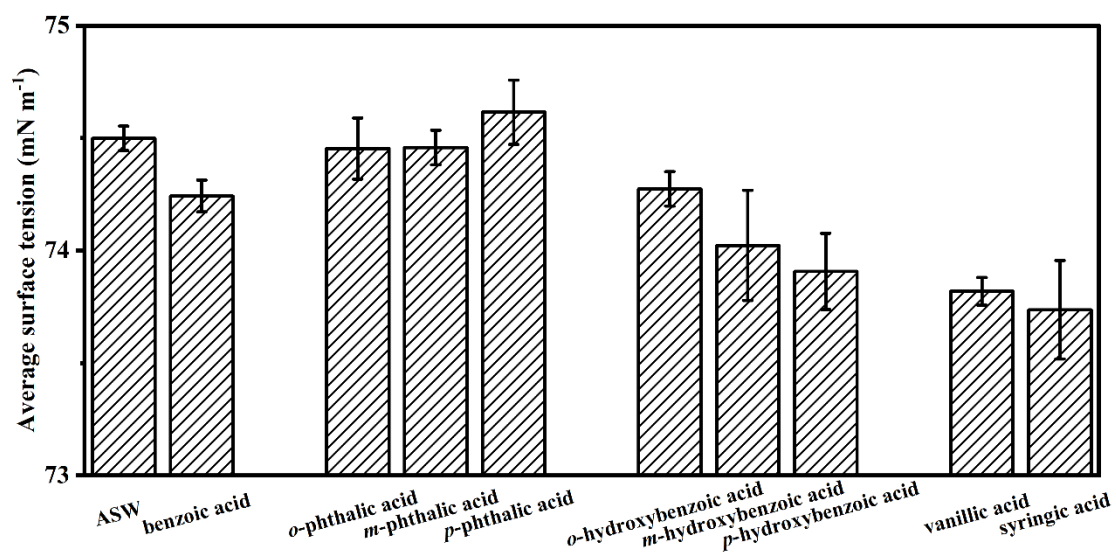


Fig. S6. Measured surface tension values of artificial seawater (ASW) and aromatic acid-containing ASW.

From the results, we can see that the effect trend of aromatic acids on seawater surface tension is generally similar, not only in seawater but also in artificial seawater.

Furthermore, we also measured the SSA particle number concentration with aromatic acids and the EF of aromatic acids, the results are as follows.

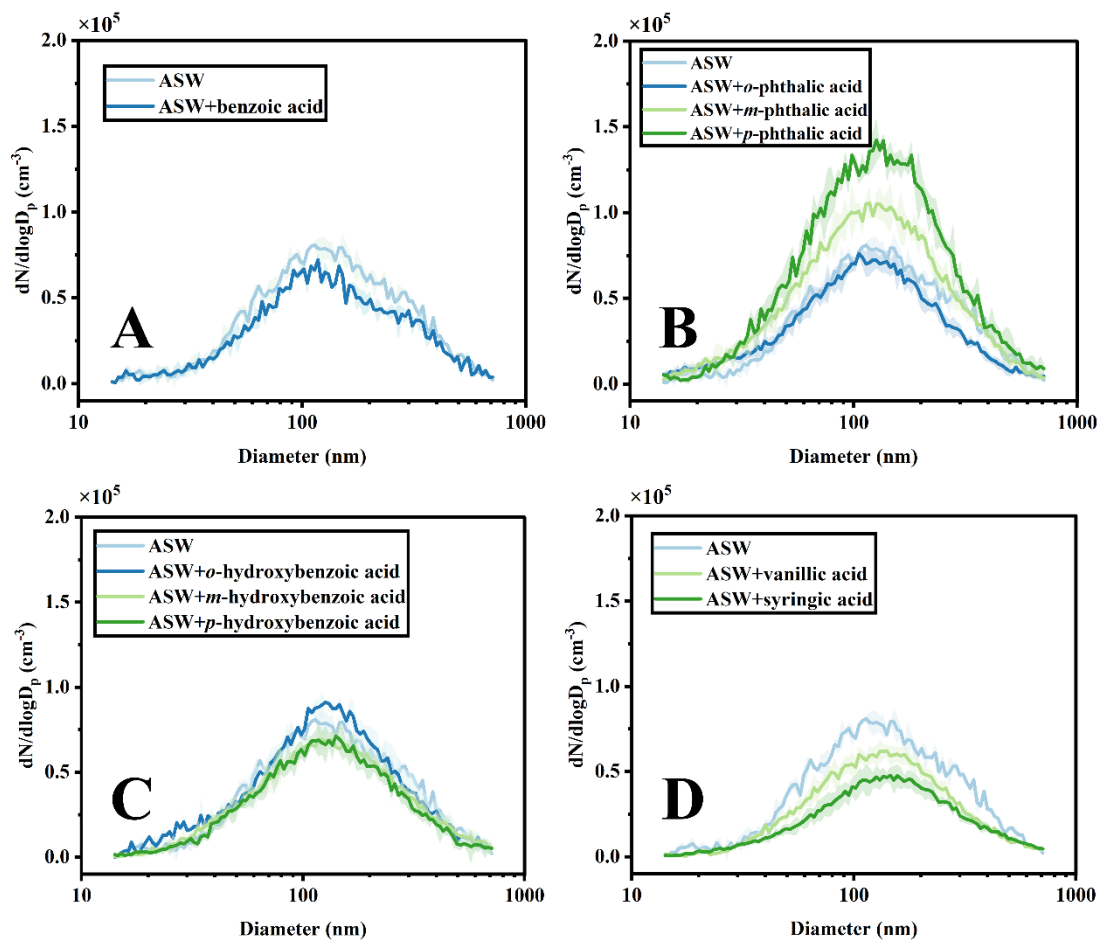


Fig. S7. Number concentration distribution of sea salt particles and SSA particles containing benzoic acids (A), benzene dicarboxylic acids (B), hydroxybenzoic acids (C), vanillic acid and syringic acid (D).

From the results, we can see that the effect trend of aromatic acids on SSA production is generally similar, not only in seawater but also in artificial seawater.

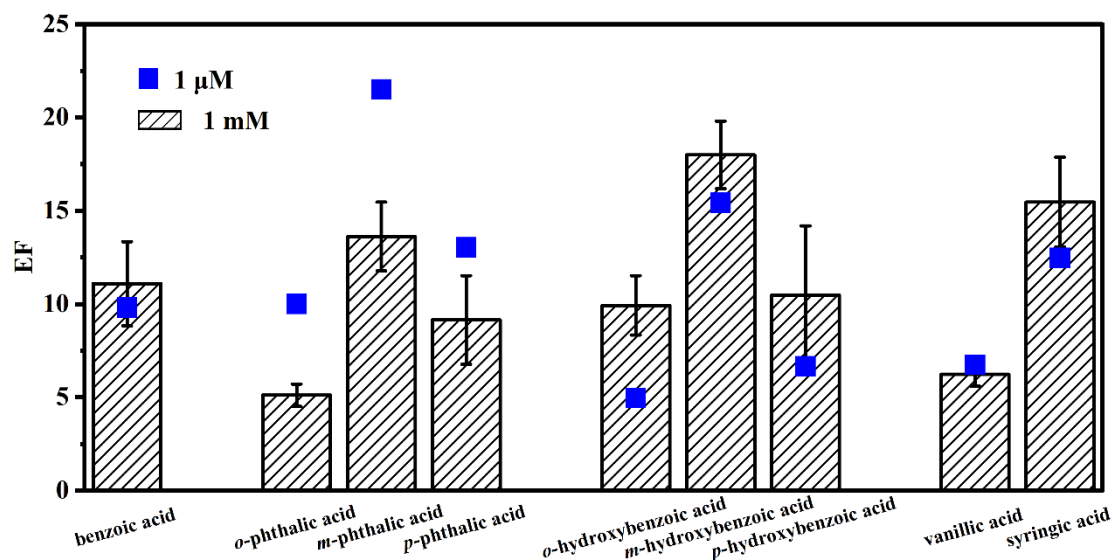


Fig. S8. Enrichment factors of aromatic acids at different concentrations from artificial seawater to the atmosphere.

Comparing the EF of aromatic acids in SW and ASW, it was observed that the EF trends of benzene dicarboxylic acids in seawater follows the pattern: *o*-phthalic acid < *m*-phthalic acid < *p*-phthalic acid. However, in ASW, the EF of *p*-phthalic acid was lower than that of *m*-phthalic acid. Based on the findings of Li et al. (2023), we hypothesize that in ASW, *p*-phthalic acid acts as an $\cdot\text{OH}$ scavenger to produce TAOH. Hence, the EF of *p*-phthalic acid is lower than that of *m*-phthalic acid. Meanwhile, organic compounds in SW preferentially react with $\cdot\text{OH}$ (Anastasio and Newberg, 2007), thus the EF of *p*-phthalic acid is the highest among the benzene dicarboxylic acids. Furthermore, differences in aromatic acid concentration did not change the enrichment pattern of organic acids.

- 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.

Author reply:

1) We added 1 mM of individual aromatic acids to natural seawater. In artificial seawater, 10^{-3} and 1mM aromatic acids were added, and the specific experimental groups can be found in Table S3.

Lines 176-177:

The experiment consisted of a total of 25 sets with target compound concentrations of 10^{-3} and 1 mM (Table S3).

2) We used the same seawater for all experiments. Seawater was collected from the coastal area of Shazikou on March 27, 2023, with a volume of 500 L (Fig. S1). Considering the storage inconvenience caused by huge consumption of seawater, all our seawater was pre-filtered through a polyethersulfone filter (47 mm diameter, 0.2 μm pore size, Supor®-200, Pall Life Sciences, USA) and stored in the dark at 18 °C for less than one month. For each experiment, we measured particle number concentrations generated by filtered seawater and cations concentrations in seawater, and we found good agreement between each set of experiments (see Fig. R1). Furthermore, for comparing the properties of SSA particles containing aromatic acids, we normalized the particles size distribution of seawater before adding aromatic acids.

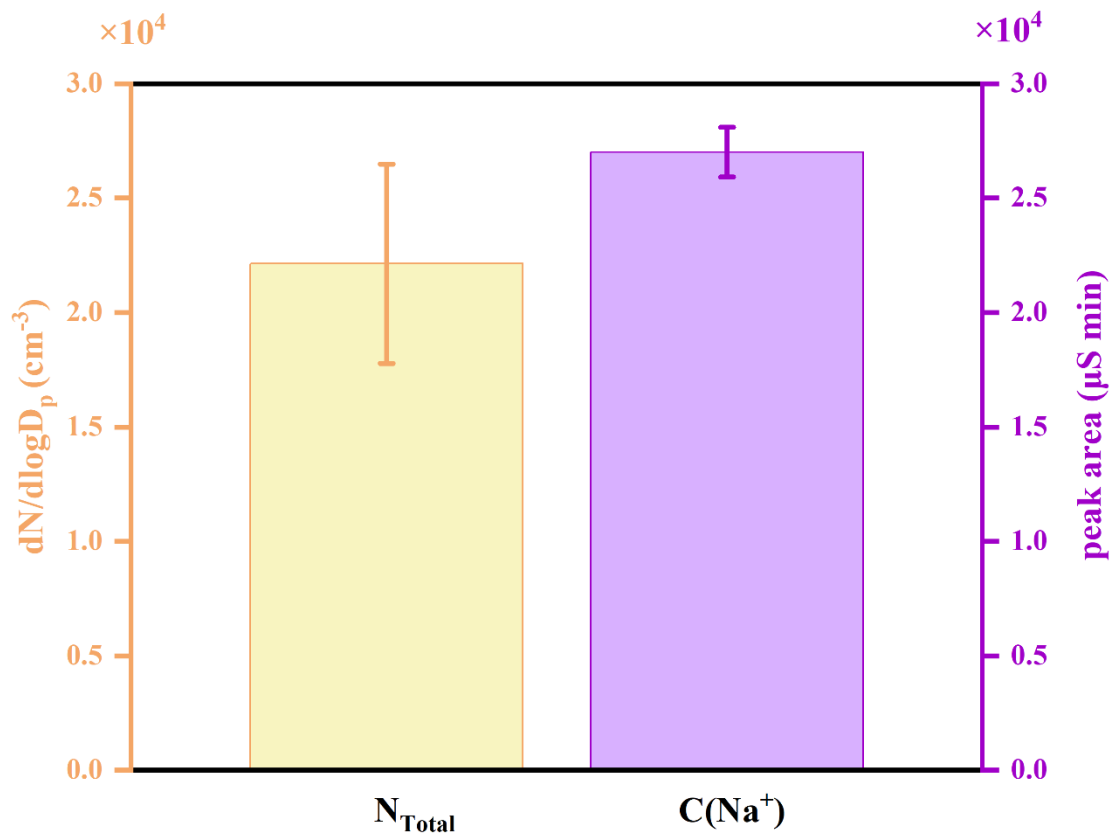


Fig. R1. Mean value of total particle number concentration of SSA (N_{Total}) and concentration of Na^+ ($C(\text{Na}^+)$) for each experiment.

3) We measured the standard curve for aromatic acids and it was linear over the measurement range. Therefore, we used the peak area directly for the EF calculation.

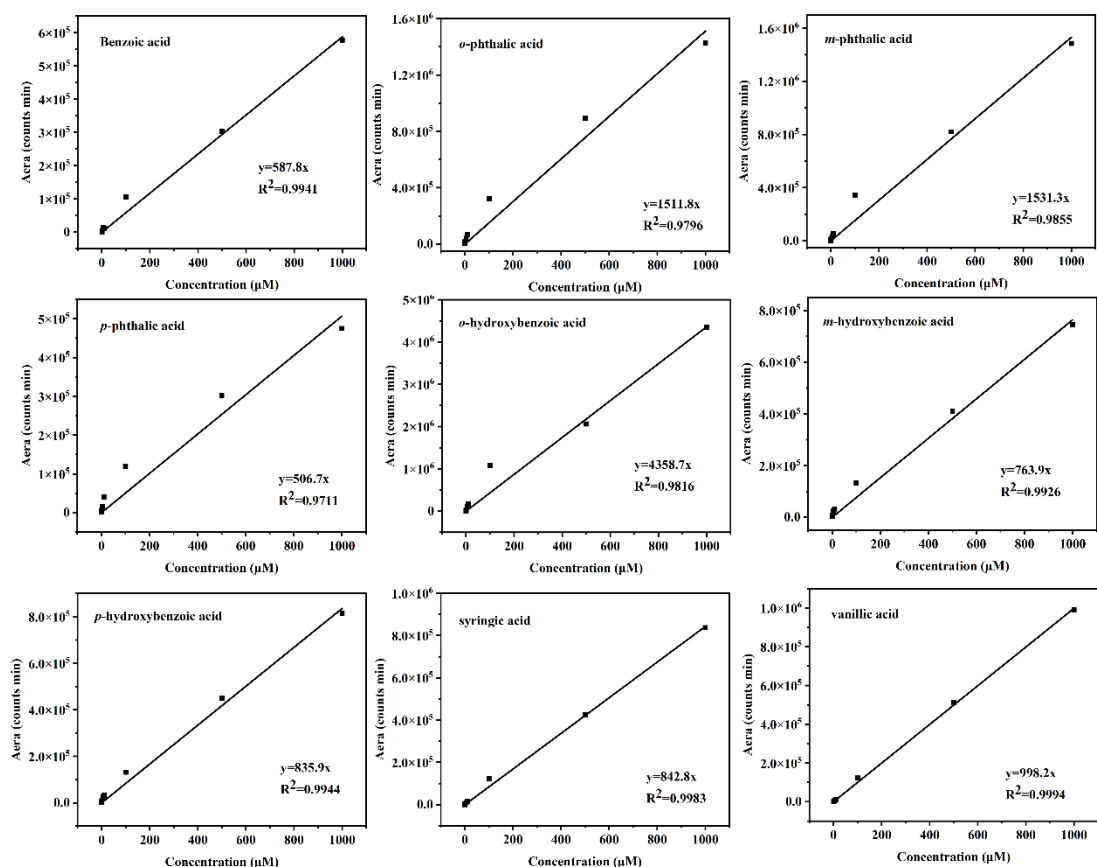


Fig. S5. Standard curves for aromatic acids were constructed within a concentration range of 0.01-1000 μM , with more than seven data points.

Table R2. Peak areas of aromatic acids in seawater and SSA filter samples.

	Group 1		Group 2		Group 3		Group 4	
	SW	SSA	SW	SSA	SW	SSA	SW	SSA
benzoic acid	159632	2732.77	196860	5206.99	185135	1540.1	167892	2508.47
<i>o</i> -phthalic acid	123785	1226.6	168974	1802.2	156324	1888.7		
<i>m</i> -phthalic acid	466987	6606.85	458769	5432.25	491623	4880.15		
<i>p</i> -phthalic acid	454593	10626.92	496102	21471.43	445985	15614.82	471582	20110.87
<i>o</i> -hydroxybenzoic acid	257548	2482	232946	3844.08	238358	5167.06		
<i>m</i> -hydroxybenzoic acid	170482	8532.59	160309	7072.09	190181	4732.93		
<i>p</i> -hydroxybenzoic acid	169753	3635.99	153257	4467.54	131186	2297.87	171062	4329.58
vanillic acid	357000	4948.01	287470	4696.83	279462	12750.11		
syringic acid	486640	23833.92	467129	29328.75	491816	11993.31		

• 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.

Author reply:

1) Impactor sampling:

The present results are averaged over all stages of the impactor with particle size less than 1 μm .

Lines 144-145:

Submicron SSA particles were impacted onto 25-mm diameter quartz fiber filters (QFF, 1851–025, Waterman, UK), which were baked in muffle furnace at 450 °C for 3 h.

2) Quantification of concentration:

We extracted the substrates in ultrapure water and then subsampled these extracts for analyses. We further measured the standard curve for the aromatic acids and it was linear over the measurement range. Therefore, we used the peak area directly for the subsequent EF calculation.

Table R2. Peak areas of aromatic acids in seawater and SSA filter samples.

	Group 1		Group 2		Group 3		Group 4	
	SW	SSA	SW	SSA	SW	SSA	SW	SSA
benzoic acid	159632	2732.77	196860	5206.99	185135	1540.1	167892	2508.47
<i>o</i> -phthalic acid	123785	1226.6	168974	1802.2	156324	1888.7		
<i>m</i> -phthalic acid	466987	6606.85	458769	5432.25	491623	4880.15		
<i>p</i> -phthalic acid	454593	10626.92	496102	21471.43	445985	15614.82	471582	20110.87
<i>o</i> -hydroxybenzoic acid	257548	2482	232946	3844.08	238358	5167.06		
<i>m</i> -hydroxybenzoic acid	170482	8532.59	160309	7072.09	190181	4732.93		
<i>p</i> -hydroxybenzoic acid	169753	3635.99	153257	4467.54	131186	2297.87	171062	4329.58
vanillic acid	357000	4948.01	287470	4696.83	279462	12750.11		
syringic acid	486640	23833.92	467129	29328.75	491816	11993.31		

3) Sampling time:

We obtained seawater samples after the SMPS-monitored total number concentrations had stabilized, and prior to DLPI+ sampling.

Lines 128-129:

After the size distribution stabilized, a total of 40 milliliters of seawater is collected from a tap located 1.5 cm above the bottom of the chamber.

- 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.

Author reply:

Based on the concentration values summarized from Table S1 in our study, we calculated the annual emission flux of SSA within the concentration range reported in the literature. For concentration selection, we chose the actual detected concentrations of aromatic acids in seawater rather than the concentrations found in sea algal extracts. The concentrations we used are shown in Table S5. We calculated the range of aromatic acid emissions within the concentration range reported in the literature (Zangrando et al., 2019; Zhao et al., 2019). The following reference has been cited in the revised manuscript.

Lines 351-353:

For example, the seawater concentrations of syringic acid, *p*-hydroxybenzoic acid, benzoic acid are 0.3, 4.58, 34 ng L⁻¹, respectively (Zhao et al., 2019).

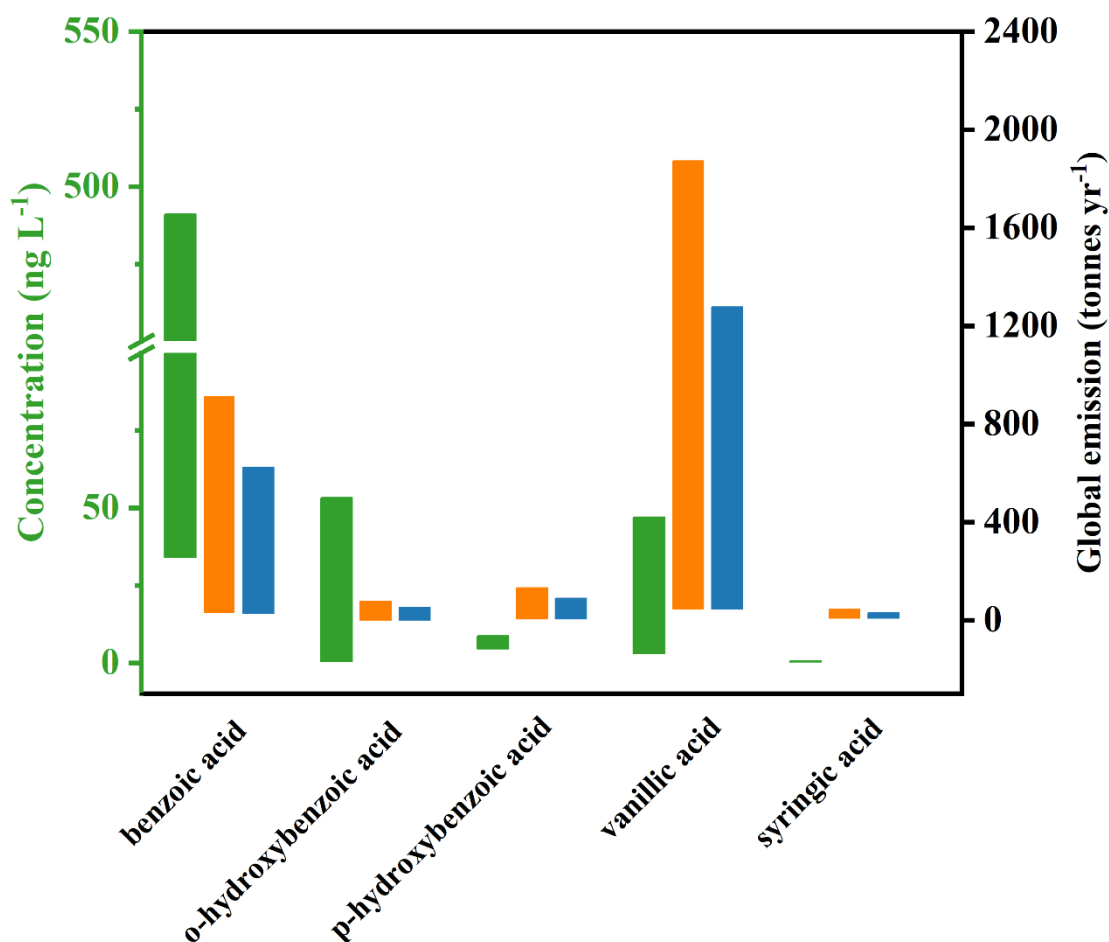


Fig. 7. Estimated annual global aromatic acids emission (tons yr⁻¹) via SSA. Yellow and blue stacked columns represent emissions based on Textor et al. (2006) and Jonas et al. (2021), respectively.

Minor points

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

Author reply:

We have replaced "captured" by "taken up" in the revised manuscript.

Lines 30-32:

On the one hand, aromatic acids are readily to be taken up by marine organisms, leading to their enrichment within these organisms or transportation to remote areas (Fu et al., 2011; Shariati et al., 2021; Wang and Kawamura, 2006).

- Line 31: Would read better as: "...leading to their enrichment within these organisms or transportation to remote areas (Fu et al.,...)".

Author reply:

We have modified corresponding sentences.

- 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)."

Author reply:

We have modified corresponding sentences. Please, refer to our reply to the first minor comment for the detailed changes.

Lines 32-33:

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.

Author reply:

We have modified corresponding sentences in the revised manuscript.

Lines 35-38:

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.

Author reply:

We thank the Reviewer for pointing this out. A new sentence has been shown in the revised manuscript.

Lines 38-39:

SSA may contain organic acids that play a major role in Earth's climate (Moore et al., 2011; Zhu et al., 2019).

- 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."

Author reply:

We have modified corresponding sentences in the revised manuscript.

Lines 49-50:

Importantly, in addition to biodegradation, the identification of phthalic acid in organisms raises the possibility of its origin from the ingestion of marine plastics (Almulhim et al., 2022).

- Line 53: Would read better as "This phenomenon has prompted discussions about the concept of" missing aromatic acids."

Author reply:

We have modified corresponding sentences in the revised manuscript.

Lines 53-55:

Recent laboratory studies have shown that personal-care products, especially sunscreen (e.g., *o*-hydroxybenzoic acid), are reduced in levels during algal blooms (Franklin et al., 2022). 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."

Author reply:

We have modified corresponding sentences in the revised manuscript.

Lines 55-56:

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).”

Author reply:

We have modified corresponding sentences in the revised manuscript.

Lines 57-59:

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.”

Author reply:

We have modified corresponding sentences in the revised manuscript.

Lines 67-69:

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.

Author reply:

This reference has been removed.

- 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)."

Author reply:

We have modified corresponding sentences in the revised manuscript.

Lines 72-74:

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 behavior 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.

Author reply:

We have deleted this sentence in the Introduction of the revised manuscript.

- Line 78: There should be a new paragraph here: "In this study..."

Author reply:

We have modified it in the revised manuscript.

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