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 #1:

General comments

I find this manuscript well-written / well-referenced, scientifically interesting for the SSA community, and the experimental quality is good. I find it great that the authors try connect SSA experiments with functional group level chemistry.

I have some minor concerns, that should be easily to address. I would recommend publication with only minor revisions.

Author reply:

We thank the Reviewer for the positive assessment of our manuscript and the constructive comments. We have revised our manuscript according to the suggestions of the Reviewer's comments and our responses to the comments are as follows.

Specific comments

1. Real seawater composition. In line 94 you write that you sample and transport seawater to the SSA laboratory. Is it possible to get more details? Conditions at sampling site (is it a productive area?), temperature, duration of storage, volume, was it filtered? Was it sampled on the same or different days? What time of year? You can add this information to the SI or just expand Table S1.

   Author reply:

   Based on the global net primary productivity estimated by Dai et al. (2023), the sampling site is a high-productivity area. We have indicated the storage conditions and time in the revised SI and expanded Table S3 to make the experimental process clearer.

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.

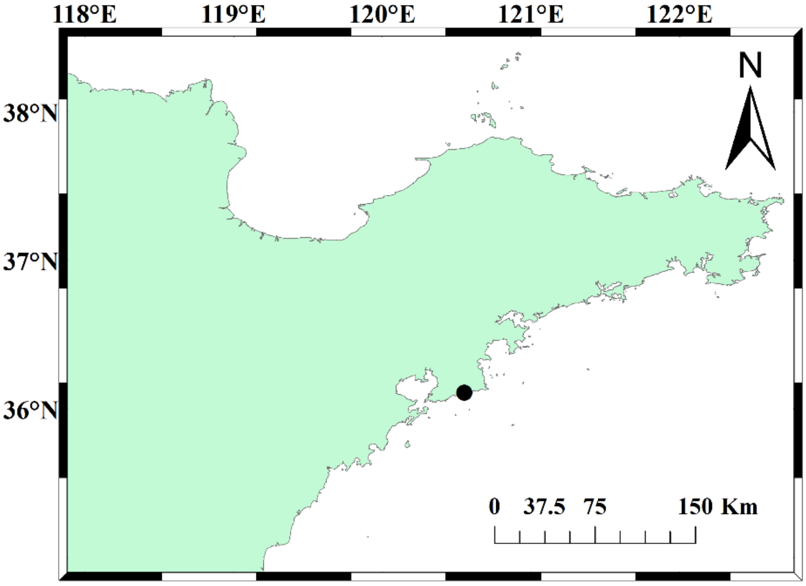

Fig. S1. Sampling site at Shazikou along the Yellow Sea coast, Qingdao, China.

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+*o*-hydroxybenzoic acid	1	7.60	34.5	5	36	1.0
4	SW+*m*-hydroxybenzoic acid	1	7.68	34.1	5	40	2.0
5	SW+*p*-hydroxybenzoic acid	1	7.84	34.3	5	38	1.5
6	SW+*o*-phthalic acid	1	7.58	34.2	5	36	2.0
7	SW+*m*-phthalic acid	1	7.80	34.5	5	37	2.5
8	SW+*p*-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+*o*-hydroxybenzoic acid	1	7.76	34.9	5	34	0.5
14	ASW+*m*-hydroxybenzoic acid	1	7.99	35.3	5	36	1.5
15	ASW+*p*-hydroxybenzoic acid	1	7.85	34.7	5	38	2.0
16	ASW+*o*-phthalic acid	1	7.93	34.5	5	35	1.0

17	ASW+*m*-phthalic acid	1	7.88	34.9	5	36	1.0
18	ASW+*p*-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+*o*-hydroxybenzoic acid+*o*-phthalic acid+vanillic acid+syringic acid	10^{-3}	7.95	35.1	20	41	3.5
22	ASW+benzoic acid+*m*-hydroxybenzoic acid+*m*-phthalic acid+vanillic acid+syringic acid	10^{-3}	7.98	34.6	20	38	1.5
23	ASW+benzoic acid+*p*-hydroxybenzoic acid+*p*-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+*m*-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.

- Also I have had challenges when I sampled fresh real seawater, that the SSA properties (size and number) changed as a function of time in the SSA chamber (due to microbial activity, degassing) – I therefore sometimes prepared artificial seawater from just inorganic sea salts. Could the authors elaborate on how reproducible the experiments are? And would the authors expect the results being similar using artificial inorganic mixture?

Author reply:

1) 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 to minimize microbiological effects.

2) We measured the total particle number concentration and concentration of Na^+ of seawater before each experiment, and all the experiments showed good repeatability (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. Therefore, perhaps it is likely that the same trend would exist in artificial seawater.

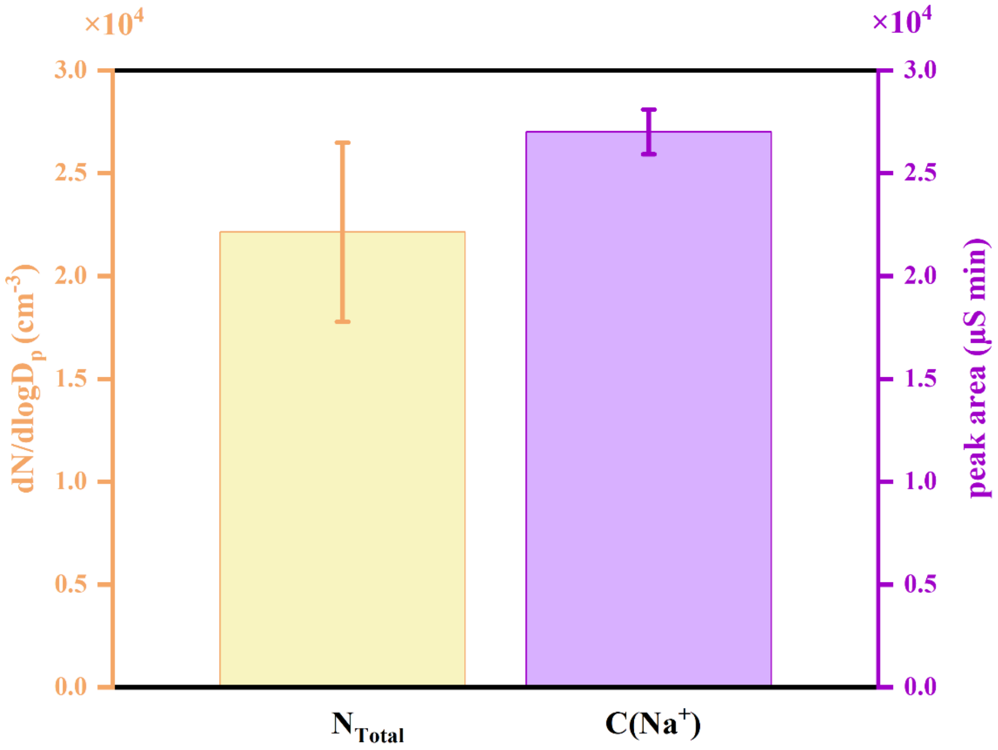

Fig. R1. Mean value of total particle number concentration of SSA (N_{Total}) and concentration of Na^+ ($C(Na^+)$) for each experiment.

3) To demonstrate this conclusion more rigorously, we added aromatic acid to the artificial seawater and observed its effect on the particle size distribution of SSA particles. The following text has been added in the revised manuscript.

Lines 241-242:

Moreover, the results showed that the effect trends of aromatic acids on SSA production in ASW were consistent with those observed in seawater (Fig. S7), eliminating the influence of organic matter.

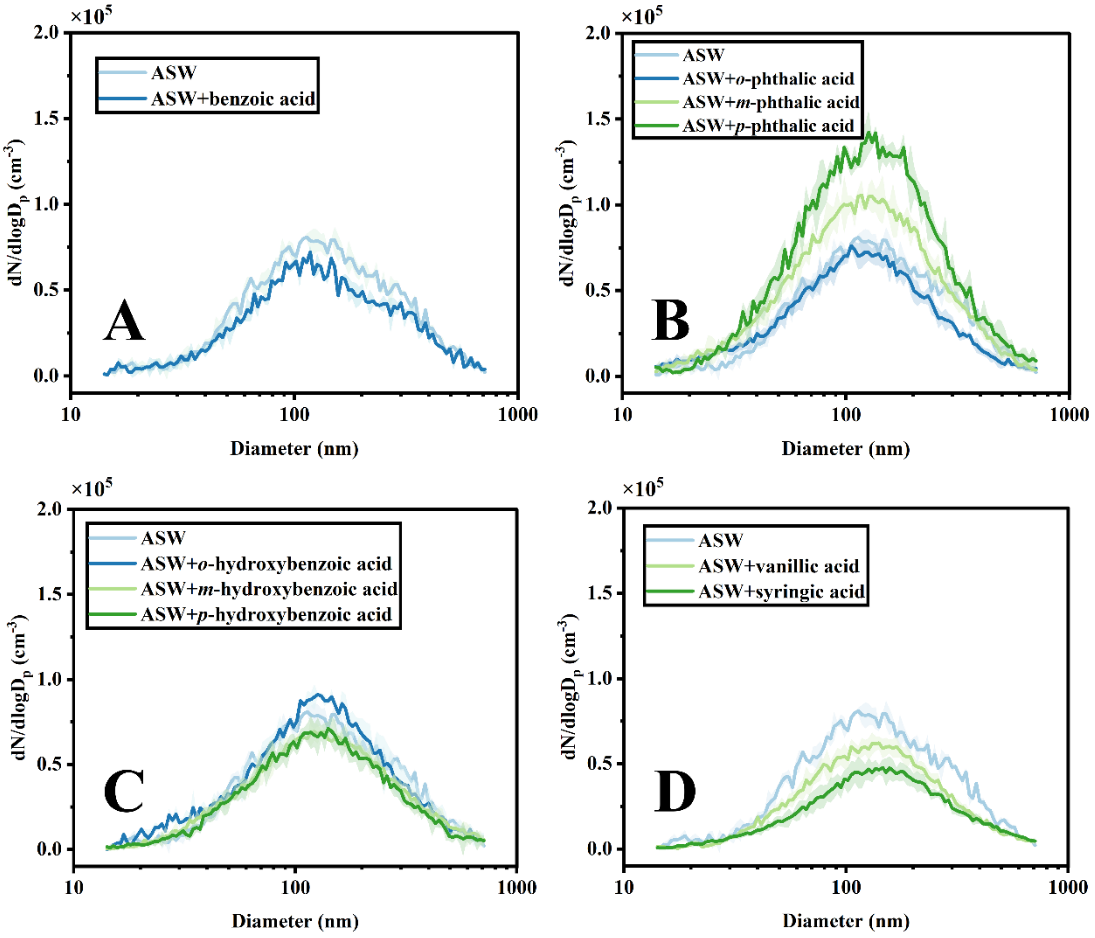

Fig. S7. Number concentration distribution of sea salt particles and SSA particles containing benzoic acids (A), benzene dicarboxylic acids (B), hydroxybenzoic acids (C), vanillic acid and syringic acid (D).

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.

Furthermore, we also measured the artificial seawater surface tension with aromatic acids and the EF of aromatic acids, and the results are as follows.

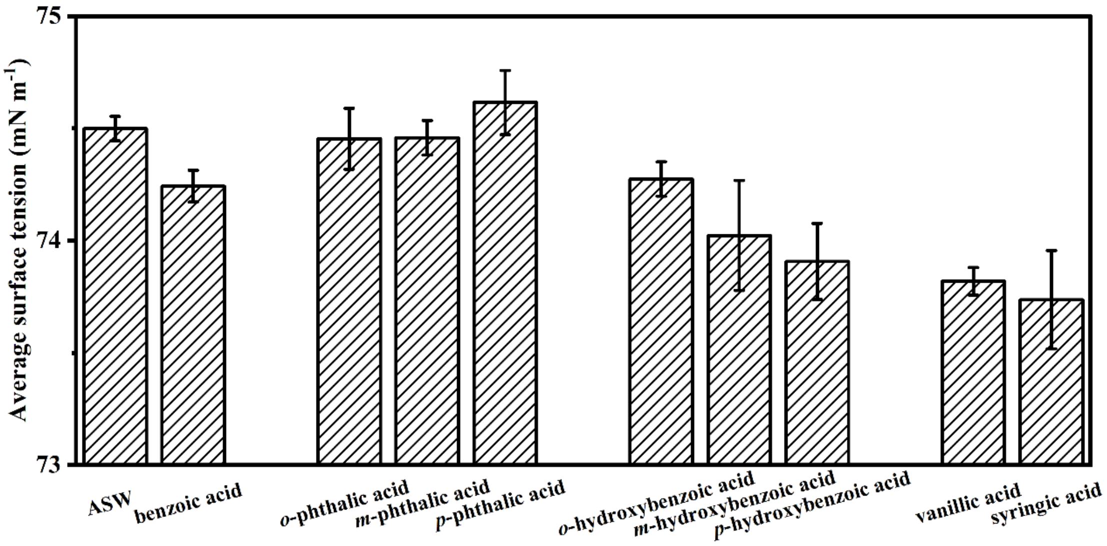

Fig. S6. Measured surface tension values of artificial seawater (ASW) and aromatic acid-containing ASW.

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.

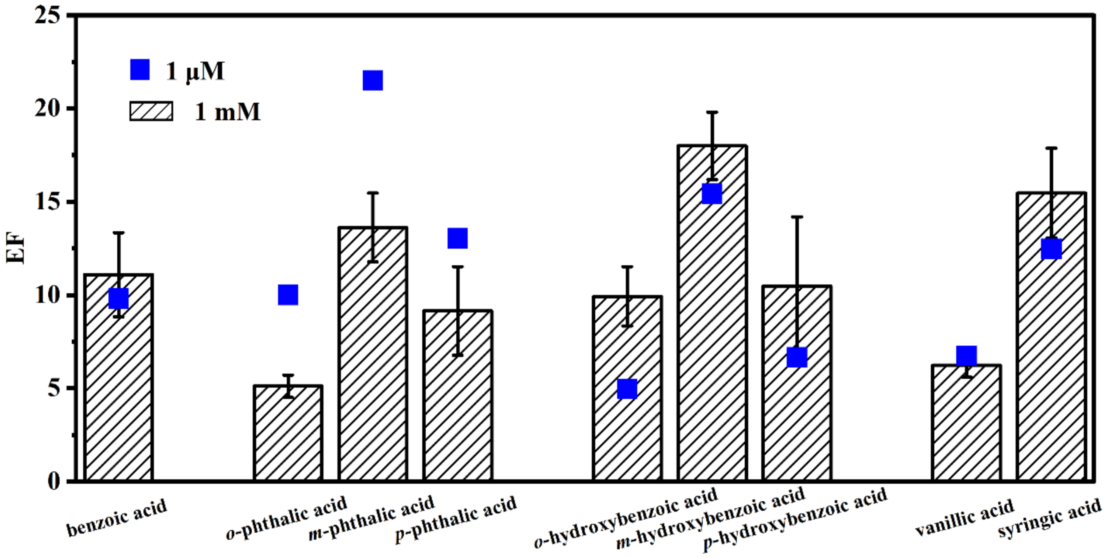

Fig. S8. Enrichment factors of aromatic acids at different concentrations from artificial seawater to the atmosphere.

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 ·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 ·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.

2. Experimental Setup. Would be helpful for the reader if Figure S1 was updated to include schematics of the entire setup, e.g. add where the DLPI+ was connected (before or after dryer?), single particle sampler (TEM). The air flow rate into SSA chamber (Line 109). Why does the range span from 3 all the way to 50 L min^{-1}? Is it because you have different setups at different times during a single experiment? Could you elaborate more on this.

Author reply:

The Dekati DLPI+ and single particle sampler were connected after dryer and sampling was carried out separately. We updated figure S3 to include schematics of the entire setup in the supplement to make it clearer. For the air flow rate, we would like to express that the zero air can be adjusted in the range of 3–50 L min^{-1}, e.g. zero air flow rate into the SSA simulation chamber was set at 3 L min^{-1}, while the zero air flow rate of the Nafion dryer tube was set at three times the outlet air flow rate when we measured the size distribution of SSA particles. When the total particle concentration has stabilized, we connected the single particle sampler to sample the SSA particles under the same air flow rate. The inlet flow rate for Dekai DLPI+ sampler sampling was set to 10 L min^{-1} to supply its pumping flow. We also updated Figure S3 to include air flow rate in the supplement to make it clearer.

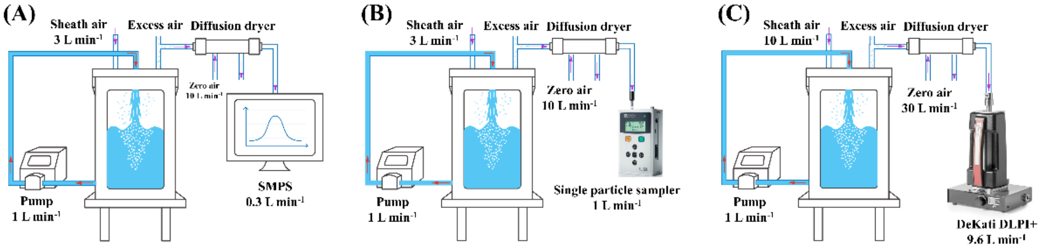

Fig. S3. Schematic picture of the plunging jet-sea spray aerosol generator: SMPS sampling (A), single particle sampling (B), and DeKati DLPI+ sampling (C). The red arrows represent the flow direction of seawater, and the purple arrows represent the flow of gases and aerosol particles.

3. TEM details. I am missing details about TEM instrument and conditions. When you do TEM, wouldn't the organic coating of SSA just vaporize in the vacuum? Also, wouldn't the SSA and aromatic acids be more internally mixed, when use a plunging jet / real bubble bursting?

Author reply:

We have added details about TEM instrument and conditions in the manuscript. The copper grid film was placed in a liquid nitrogen-frozen vacuum environment through a TEM holder, thereby inhibiting the evaporation of the organic coating of SSA. According to the classification of aerosol particles by Li et al. (2016), a particle consisting of two or more aerosol components can be defined as an internally mixed particle. Otherwise, it will be regarded as an externally mixed particle. For the mixing state of SSA particles, the OM coating and core-shell mixing structure were considered as internally mixed in previous studies (Li et al., 2016; Li et al., 2021).

Lines 138-141:

> TEM was performed using a TEM cryo-mount (Gatan 626) to load the samples, where the TEM grid was immersed in liquid nitrogen and then mounted on the holder by means of a cryo-transfer workstation. TEM with a high-angle annular-dark-field detector was used and then TEM images were obtained at an accelerating voltage of 200 kV.

Technical comments

Line 58: add mass, so it reads main mass component

Author reply:

We simplified the original sentence as follows.

Lines 59-61:

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).

Line 61: "... disturbing ecological systems... ", sound funny. Maybe change to "further impacting" or "further interaction with"

Author reply:

We have rephrased the original text as:

Lines 61-63:

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).

Line 163: The **k** look wierd, should be k_{SSA} as in Sha et al, right? Also chemical symbols should be upright, not italic.

Author reply:

Yes. We have replaced "**k**" by "k_{SSA}" in the revised manuscript.

Line 202: Unit, Part s^{-1}, would prefer just s^{-1} or particles s^{-1}.

Author reply:

We have modified corresponding sentences and figures in the revised manuscript. All the SSA production units previously noted as "part s^{-1}" of seawater are now expressed as "particles s^{-1}".

Line 207: what does increase in bubble bursting refer to? Is it foam stability or lifetime? Or smaller and more bubbles?

Author reply:

The increase in bubble bursting here refers to smaller and more bubbles.

Line 260: change ball to sphere

Author reply:

We have modified corresponding sentences in the manuscript.

Lines 275-276:

Notably, it can be seen that the core morphology of salt particles had changed significantly, where the cubic structure has changed into a sphere structure.

Line 323: “lousy” is informal slang, change to “very poor” or just “poor” or “around and just below 1”.

Author reply:

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

Lines 342-343:

In SSA, Ca^{2+} always exhibited high enrichment (EF > 1), while the EFs for K^+ and Mg^{2+} were around and just below 1.

Line 351: … plays a very important role… Tone down, add “might play”

Author reply:

We have modified corresponding sentences in the manuscript.

Lines 373-374:

This demonstrates that the EF might play a very important role in the global emission fluxes of organic matter, in addition to concentration.

Figure 1: Maybe same range on y-axis? Easier to compare across subplots. I am colorblind, do you need the colors? If you perfer using colors, then should be the same as in Figure 2.

Author reply:

We have modified corresponding figures in the manuscript. We used the same range on y-axis and removed unnecessary colors.

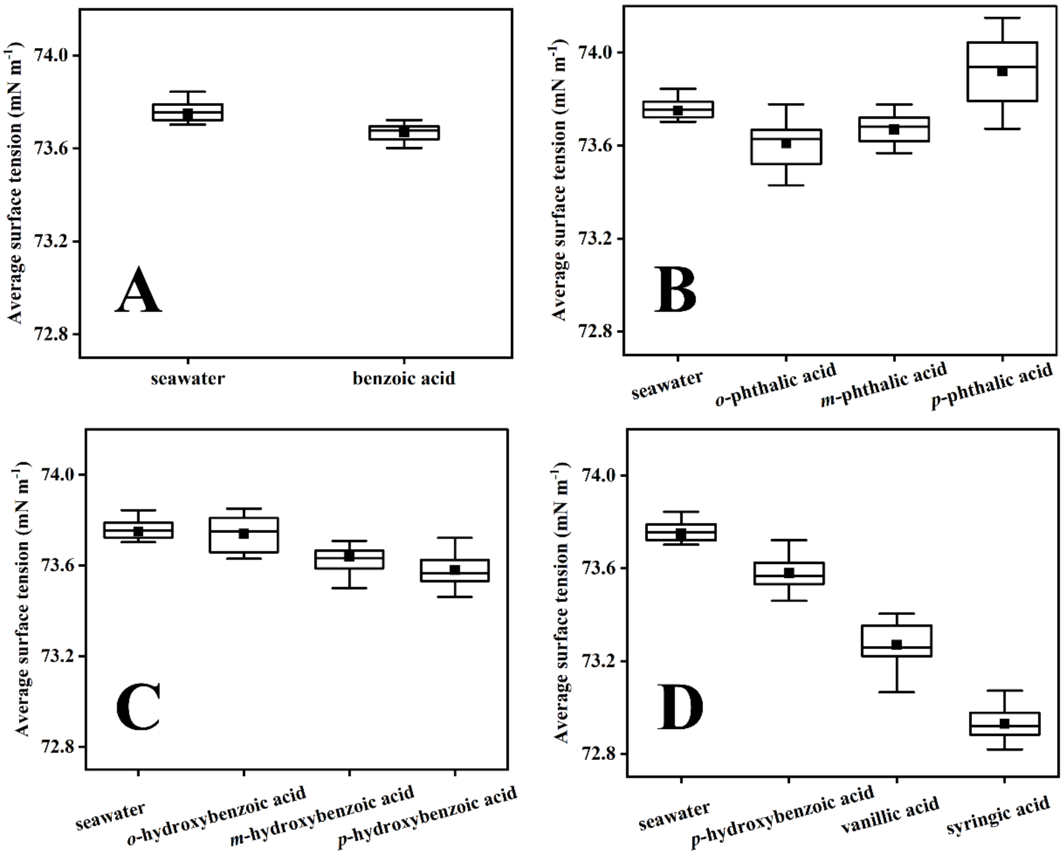

Fig. 1. Measured surface tension values of natural seawater and aromatic acid-containing seawater: benzoic acids (A), benzene dicarboxylic acids (B), hydroxybenzoic acids (C), *p*-hydroxybenzoic acid, vanillic acid, and syringic acid (D). The dark spots represent Mean values of at least three measurements and the boxes represent the ranges of 25th−50th−75th percentiles.

Figure 2. I would change the colors. Yellow is difficult to see. Look at this website for inspiration: https://colorbrewer2.org/#type=diverging&scheme=BrBG&n=4

Author reply:

We thank the Reviewer for providing us with this useful website. We have changed the colors of Figure 2 in the revised manuscript.

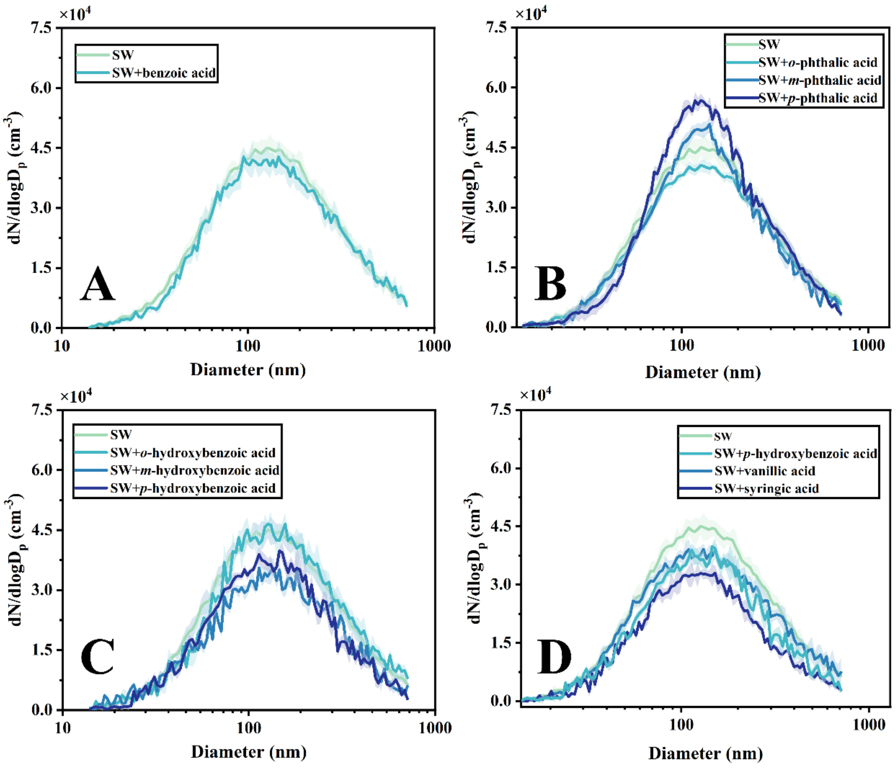

Fig. 2. Number concentration distribution of sea salt particles and SSA particles containing benzoic acids (A), benzene dicarboxylic acids (B), hydroxybenzoic acids (C), *p*-hydroxybenzoic acid, vanillic acid, and syringic acid (D).

Figure 3. Seawater = Seawater only. And maybe add some errorbar estimation with respect to SSA production? Here color is okay, but that white shadow looks funny.

Author reply:

We have added error bar estimation with respect to SSA production of Figure 3 in the revised manuscript.

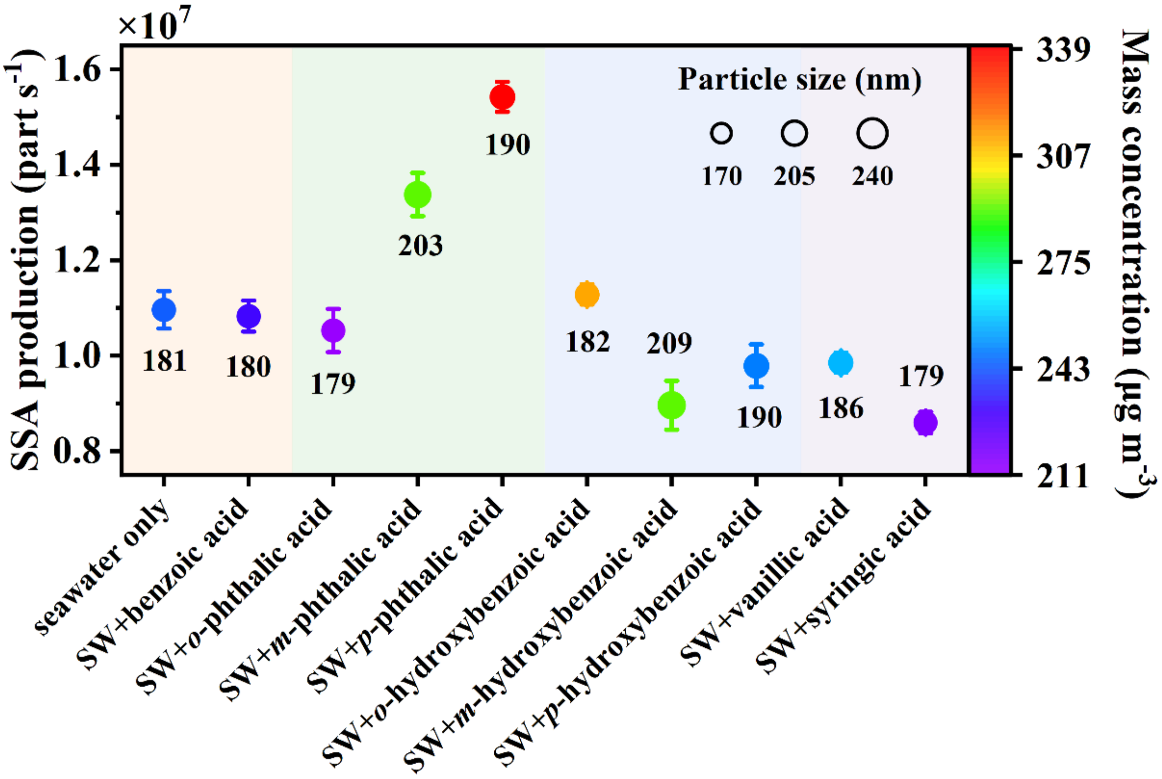

Fig. 3. SSA production, particle size, mass concentration distribution of aromatic acids. The symbol size represents the geometric mean diameter of SSA particles and is marked with numbers, and the symbol color indicates the particle mass concentration.

Figure 6: Maybe make different symbols and use one color? Feel free to ignore this comment.

Author reply:

We have tried to make different symbols in Figure 6 in the revised manuscript. Using the same color makes the symbols overlap. Hence, we used different colors.

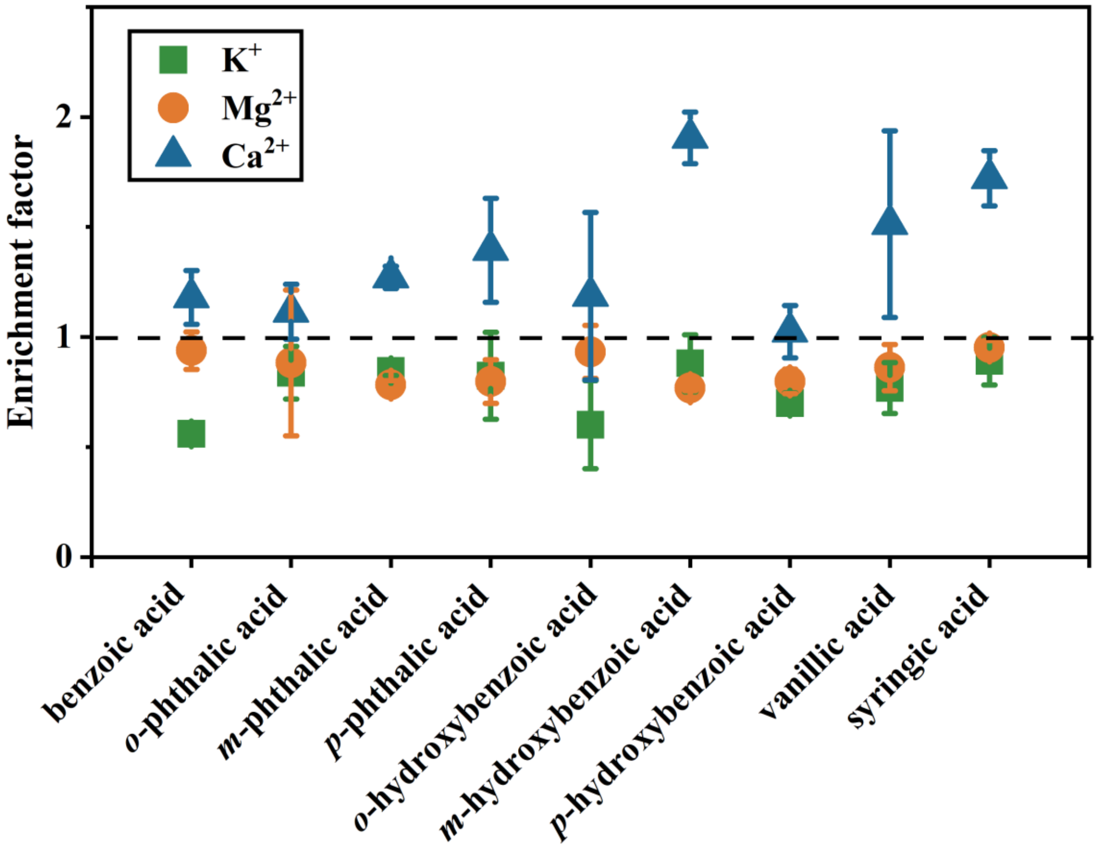

Fig. 6. Enrichment factors of K$^+$, Mg^{2+}, and Ca^{2+} in submicron SSA during the experiment.

Figure 7. The space between the bars are not the same. Also I would change colors (Feel free to ignore this comment)

Author reply:

We have made the space between the bars same and changed colors of Figure 7 in the revised manuscript.

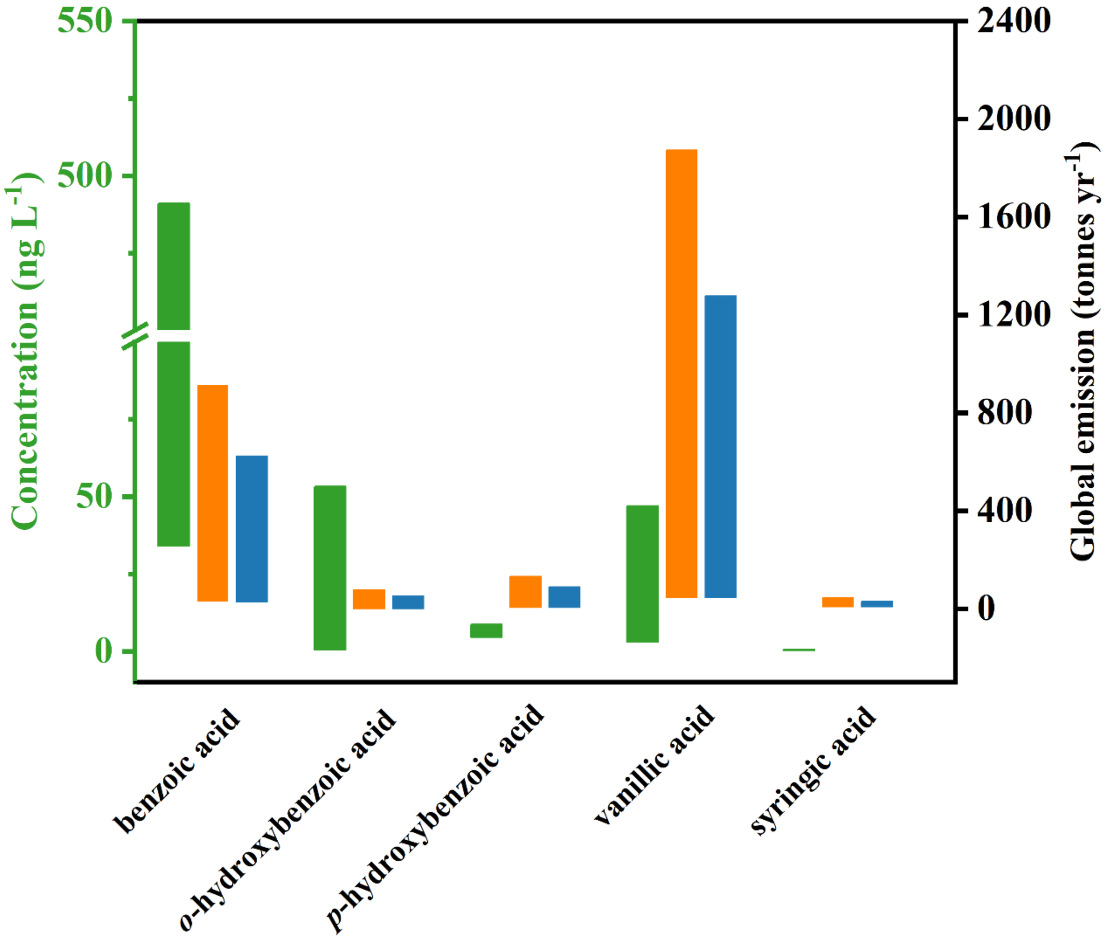

Fig. 7. Estimated annual global aromatic acids emission (tons yr^{-1}) via SSA. Yellow and blue stacked columns represent emissions based on Textor et al. (2006) and Jonas et al. (2021), respectively.

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