Dear Prof. Manmohan Sarin and reviewers,

We appreciate all your detailed and valuable suggestions on our manuscript (egusphere-2025-3951). We have carefully considered the comments and revised the manuscript accordingly. Please see the point-by-point response below and changes are marked blue in the revised manuscript.

Thanks very much!

Most sincerely,

Yujue Wang and Wei Xu on behalf of all co-authors

Point-by-point response to review comments

Note: *Review comments are in italicized font.* Our responses are indented and in normal font. Revised text in the manuscript or supplementary is in blue color.

Referee #1

This study presents a very detailed analysis of the organic aerosol fraction in the marine atmosphere. The author made efforts to try to find a better way of parameterizing the primary and secondary OC in marine aerosols using common measurements of seawater (chl-a and Na+). However, it is not clear about the main conclusion of applying the proposed new method and its performance. Key gaps in interpreting the fluorescent components are missing. I expect further elaborations on these issues together with other major comments. Please see the following comments for details.

> **Response**: Thank you very much for your overall positive comments on our manuscript. We appreciate your detailed comments regarding the performance of the proposed parameterization and the interpretation of the fluorescent components, which we have carefully addressed and revised the manuscript according to the following comments. For the different contributions of MPOC identified by the mass concentrations and the fluorescent properties, we have elaborated in the *major comment 10* and revised the manuscript accordingly.

Major comments:

The authors define marine primary organic carbon MPOC as those emitted from marine bubble bursting. How about those emitted by other processes like wave breaking? The definition of MSOC is even unclear. In the Abstract, it is defined as "secondary organic carbon (MSOC) formed via gas-to-particle conversion remains poorly quantified". Are these gases those only emitted from the ocean?
How about the primary OC and gas precursors transported from the terrestrial environments?
In summary, it is unclear what does "marine" mean here. Does it only refer to marine atmosphere?

> **Response**: Thanks for pointing out the unclear definition. In this study, marine primary organic carbon (MPOC) is the organic carbon in the sea spray aerosols, which includes those emitted from bubble bursting and wave breaking. Marine secondary organic carbon (MSOC) includes the organic aerosols formed via gas-to-particle conversion of gaseous precursors and via the oxidation/aging processes of primary organic aerosols. The gaseous precursors and aged organic aerosols could be from both ocean emissions and long-range transported from

terrestrial environments. We have defined MPOC and MSOC in the abstract and the main text (lines 19−21, 236−238, 252−253).

We added the back trajectory analysis of air masses (Fig. S1) to analyze the influence of terrestrial outflows. The air masses during the cruises were mainly transported from open oceanic regions, and thus the impacts of terrestrial outflows were limited. Related descriptions on the potential impacts of transported terrestrial air masses are added in the revised version (lines 164−166, 315−324).

Yes, "marine" here refers to marine atmospheres. Atmospheric aerosols over the oceans could be from diverse sources, including primary and secondary marine emissions, and transported pollutants (Brooks and Thornton, 2018). We cannot exclude the potential impacts of transported terrestrial compounds based on the cruise observations. The secondarily-formed marine organic aerosols in this work could be sourced both from marine emissions and transported from terrestrial environments.

Lines 19−21 in the abstract:

However, the abundance of marine primary organic carbon (MPOC) generated by sea spray and secondary organic carbon (MSOC) formed via gas-to-particle conversion or atmospheric oxidation/aging processes remains poorly quantified, which hinders our understanding on the climate effects of marine aerosols.

Lines 236−238:

Based on the correlation analysis of the observed parameters, we proposed a parameterization scheme to separate the marine primarily-emitted OC (MPOC) in the SSA generated through wave breaking or bubble bursting processes and the secondarily formed organic carbon (MSOC) in the marine aerosols over WPO.

Lines 252−253:

The MSOC here includes the organic aerosols formed via gas-to-particle conversion of gaseous precursors and oxidation/aging processes of primary OC.

Lines 164−166:

The air masses were mainly transported from open oceanic regions, and thus the impacts of terrestrial outflows were limited during the cruises (Fig. S1).

Lines 315−324:

It is noted that, based on the shipboard in-situ observation, we cannot exclude the potential impacts of gaseous precursors or aged organic aerosols long-range transported from terrestrial environments, which were mostly in the MSOC fraction. The organic aerosols transported from terrestrial environments were secondary or aged organic aerosols, and tend to be water-soluble organic compounds (Boreddy et al., 2018; De Jonge et al., 2024; Miyazaki et al., 2010). Based on the air mass back trajectories (Fig.

S1) and the weak correlations between OC and EC stated in section 3.1, the impacts of transported continental outflows were limited during the cruises.

Newly added Figure S1:

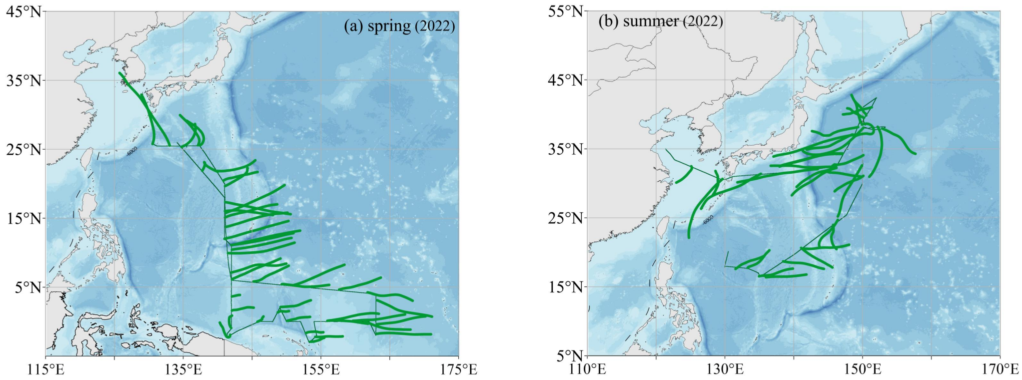

Figure S1 The 24-hr back trajectories of air masses during the cruises in (a) spring and (b) summer.

1. *L115 "Organic matters were the dominant components in the fine particles, which respectively contributed 18%−75% (40% on average) and 13%−74% (48% on average) of the PM2.5 mass in spring and summer." The authors measured the mass concentrations of organic matters and inorganic ions. In addition to these measured composition in PM2.5, how about the contribution of other composition, e.g., other metal elements, ions, elemental carbon, in the mass of PM2.5?*

Response: In this study, we measured organic carbon (organic matter), water-soluble ions (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^- and SO_4^{2-}), and elemental carbon (EC), which were summed as the total mass of $PM_{2.5}$. The average EC concentrations were 0.066 and 0.055 μgC m^{-3} during the spring and the summer campaigns, of which the contributions were much lower than OM. Previous studies have suggested that organics and water-soluble ions are the major contributors to the marine aerosol mass. Other metal elements (e.g., Al, Fe, Ti, Sr, Ba, Mn, etc.) accounted for <3.5% of the marine aerosol mass based on the measurements over the East China Sea (Hsu et al., 2010). Related descriptions have been added in lines 130−132, 141−142.

We agree with the reviewer that metal elements (e.g., Fe, Al, etc.) are also important in marine aerosols. We did not analyze metal elements in this work due to the limited mass loading of the collected marine aerosol samples. We will try to measure the metal elements in our future studies.

Lines 130−132:

The EC concentrations were 0.066 ± 0.056 μgC m^{-3} and 0.055 ± 0.052 μgC m^{-3} during the spring and the summer observations, much lower than those observed over coastal areas typically influenced by continental outflows (Sahu et al., 2009; Zhang et al., 2025).

Lines 140−143:

The mass concentrations of $PM_{2.5}$ were calculated by summing the measured OM, EC, and water-soluble ions. Metal elements were not measured in this study, which contributed <3.5% of the marine aerosol mass concentration over the East China Sea (Hsu et al., 2010). Without considering the metal elements, we may overestimate the organic proportion in marine aerosols.

2. *L118-122: how are the sampling and measurement uncertainties of OC in PM2.5 and TSP?*

What could be the reasons that "organic fractions were dominant in the submicron marine aerosols"? Could these reasons be consistent with the main sources of MPOC of MSOC?

Response: The sampling and measurement uncertainties of OC in the aerosol samples are described in lines 143−147.

Sea spray aerosols (SSA) or primary marine aerosols could be formed by film drops and jet drops during wave breaking and bubble-bursting processes. Submicron SSA are mainly formed by film drops produced from bursting bubble-cap film, which is enriched with hydrophobic organic matters contained within the sea surface microlayer (Burrows et al., 2014; Wang et al., 2017). In contrast, jet drops formed from the base of bursting bubbles are mainly produce larger supermicron particles from bulk seawater, which comprises sea salts and smaller fraction of organics (Wang et al., 2017). Thus, the dominance of organics in submicron marine aerosols was due to the enriched organic compounds in the air−water interface, which could be efficiently transferred into the submicron sea spray aerosols through film drops during wave breaking and bubble-bursting processes. This is now added in lines 138−140, 232−234.

The reason is consistent with the main source of MPOC related to marine biological activity, indicated by seawater *Chl-a*. Marine phytoplankton could produce gel-like aggregates and contribute to extracellular polymer particles, water-insoluble polysaccharide-containing transparent exopolymer, and protein-containing organics, etc. in seawater (Aller et al., 2017; Lawler et al., 2020). These hydrophobic organics could be enriched in the surface seawater and easily transferred into the submicron aerosols over the ocean by film drops. Related descriptions have been added in lines 182−185.

Lines 138−140:

Film drops could efficiently transfer hydrophobic organic compounds enriched in the air−water interface into the submicron aerosols, which explained the size-selective enrichment of organics in marine aerosols (Cochran et al., 2016; Prather et al., 2013; Quinn et al., 2015; Wang et al., 2017).

Lines 143−147:

During the sampling, positive artifacts of OC may exist due to the absorption of gaseous organic vapor on the filters, and negative artifacts may exist due to the evaporation of volatile organic compounds (Huebert and Charlson, 2000). The OC concentration was measured using thermal-optical analysis. Quantification uncertainty may be introduced due to the formation of pyrolyzed OC, which complicates the accurate determination of the OC/EC split point (Cao et al., 2025; Chow et al., 2004).

Lines 182−185:

Marine phytoplankton could produce gel-like aggregates and contribute to extracellular polymer particles, water-insoluble polysaccharide-containing transparent exopolymer, and protein-containing organics, etc. in seawater (Aller et al., 2017; Lawler et al., 2020). These organic substances could be enriched in the surface seawater and then transferred into the atmospheric aerosols within the marine boundary layer.

Lines 232−234:

Organic matters in marine aerosols are enriched in the submicron SSA, which is mainly formed by film drops from bursting bubble-cap films (Wang et al., 2017). In contrast, the majority of the sea salts exist in larger supermicron or coarse-mode particles generated by jet drops from the base of bursting bubbles (Wang et al., 2017).

3. *L177-179: is the representativeness of Na+ of overall SSA production also useful in coastal marine atmosphere considering the anthropogenic/terrestrial sources of Na+? The conclusion is derived based on $PM_{2.5}$ samples. Many marine studies collect TSP or PM_{10} samples. For these samples, can we interpret the Na+ measurements in this way as well?*

Response: Thanks for reminding the potential sources of Na^+ from terrestrial sources. We have conducted cruise observations of the aerosols in coastal marine atmospheres over the East Asian marginal seas. Among the marine aerosol sources resolved by PMF source apportionment, Na^+ mainly existed in sea spray aerosols and dust aerosols (Figure R1), and the Na^+ from anthropogenic sources are neglectable (Zhang et al., 2025). When using Na^+ as the indicator of SSA production, Na^+ transported by terrestrial dust storms should be excluded. Related description has been added in lines 227−230.

For the collected TSP samples, the OC concentrations did not display obvious correlation with the seawater *Chl-a* due to different production processes of OC and sea salts. We added related discussion in lines 230−234 and Fig. S4. The Na^+ in the PM_{10} or TSP samples could be used as an indicator of the bulk SSA production. However, it might not be a good input to estimate the organics from SSA.

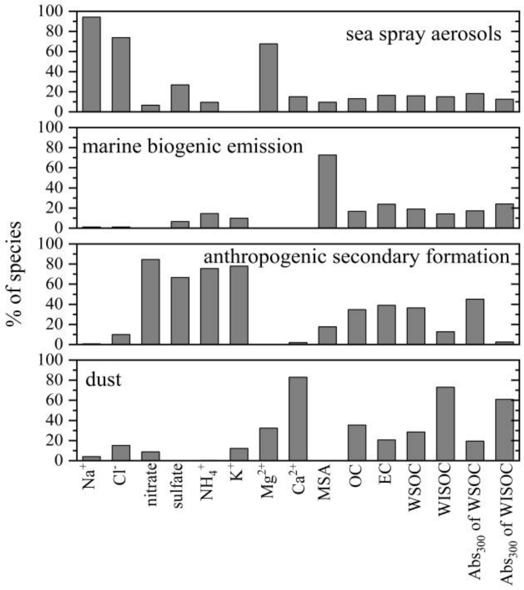

Figure R1 Explained variation of the marine aerosol source factors apportioned by PMF model over East Asian marginal seas (Figure S7 in Zhang et al., 2025)

Lines 227−230:

It should be noted that dust storms also transport Na^+ to marine atmospheres, especially over the marginal seas (Zhang et al., 2025). When using Na^+ in marine aerosols as the indicator of SSA production, the Na^+ contributed by transported dust storms should be excluded, especially during dust episodes.

Lines 230−234:

For the collected TSP samples, the OC concentrations did not display an obvious correlation with the seawater *Chl-a* (Fig. S4). This is because the dominant production processes of OC and sea salts are different. Organic matters in marine aerosols are enriched in the submicron SSA, which is mainly formed by film drops from bursting bubble-cap films (Wang et al., 2017). In contrast, the majority of the sea salt mass exist in larger supermicron or coarse-mode particles generated by jet drops from the base of bursting bubbles (Wang et al., 2017).

Newly added Figure S4:

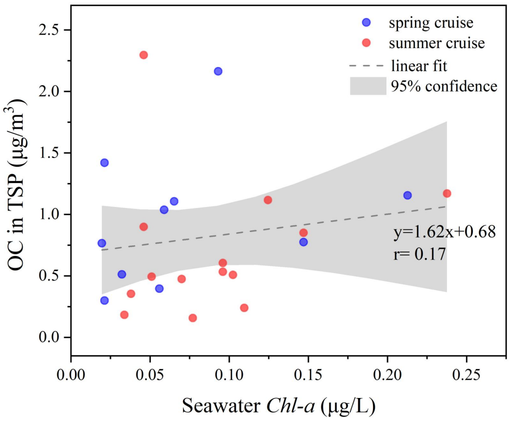

Figure S4 Scatter plot of the OC concentration in the collected TSP samples and seawater *Chl-a* during the cruises.

4. *L244-246 "The estimated MSOC matched better with the WSOC in the marine aerosols when using a combination of [Chl-a] and [Na+] as the input parameters and considering the variation of sea spray aerosols (Fig. 3c, 3g)". I don't see the evidence why the estimated MSOC matched better with the WSOC when using a combination of [Chl-a] and [Na+] and considering the variation of sea spray aerosols. The "r" value in Fig. 3c and 3g is the same (0.73). Please further explain.*

Response: The performance of the parameterization was evaluated based on the fitting line slope and correlation coefficient (r) of WSOC and estimated MSOC. It means that the estimated MPOC shows a similar variation trend to WIOC if with a r value closer to 1, and a good comparison with the WIOC mass concentrations if with a fitting line slope closer to 1. The fitting line slope of WSOC and estimated MSOC was closer to 1 when using equation 1 and 3, and more estimated MSOC concentrations fall within the WSOC/MSOC 3:1 and 1:3 lines compared with the results based on equations 1 and 2. We now have added related descriptions to be clear (lines 302−305, 315−318).

Lines 302−305:

Both the correlation coefficients (r) and the slopes of the fitting line between WIOC and estimated MPOC are used to evaluate the performance of different MPOC parameterization approaches. It means that the estimated MPOC shows a similar variation trend to WIOC if with a r value closer to 1, and a good comparison with the WIOC mass concentrations if with a fitting line slope closer to 1.

Lines 315−318:

Based on equations 1 and 3, the estimated MSOC concentrations in half of the samples fall within the WSOC/MSOC 3:1 and 1:3 lines, and the fitting line slope (0.50) was closer to 1 (Fig. 3h). Using equations 1 and 2, the fitting line slope of WSOC and estimated MSOC was 0.46, and 46% of the estimated MSOC concentrations fall within the WSOC/MSOC 3:1 and 1:3 lines (Fig. 3c).

5. *L248 and Fig. 3d & 3h: how about WSOC? Can you compare the estimated MSOC with those estimated by the parameters/methods of Gantt and Vignati?*

Response: The comparisons of WSOC and the estimated MSOC using the methods from Gantt et al. (2011) and Vignati et al. (2010) are now added in Figure 3 and the main text (lines 332−335).

Lines 332−335:

The estimated MSOC using the parameterizations from Gantt et al. (2011) or Vignati et al. (2010) showed similar variation trends to the WSOC in the collected aerosols samples. The comparison of the estimated MSOC and the WSOC concentrations using formulations in literatures (slopes in Fig. 3e, 3j), however, were not as good as those estimated in this study (slopes in Fig. 3h).

Revised Figure 3 (panels e and j are added):

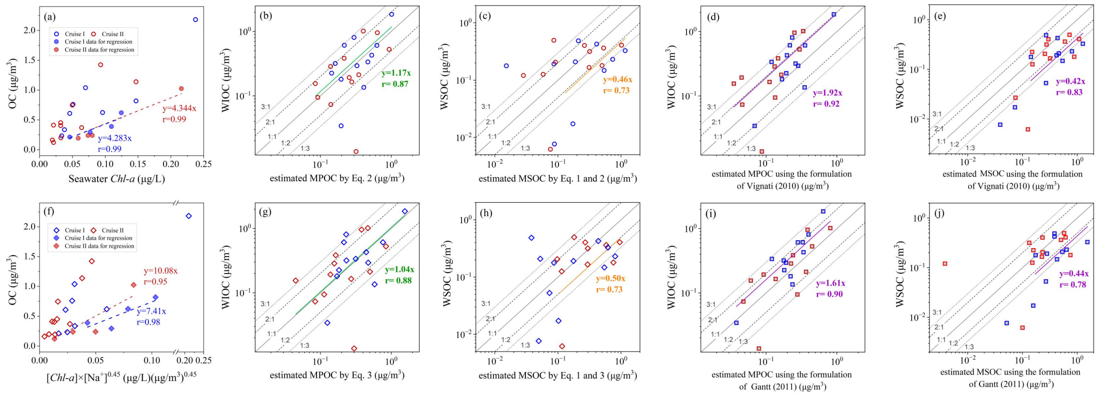

Figure 3 The scatter plots of OC in marine aerosols as a function of (a) seawater [*Chl-a*] and (f) ([*Chl-a*] × $[Na^+]^{0.45}$) during the two cruises; (b, g) Comparison of WIOC and the estimated MPOC based on the regression in panel (a) and panel (f); (c, h) Comparison of WSOC and the estimated MSOC; (d, i) Comparison of WIOC and the estimated MPOC, and (e, j) Comparison of WSOC and the estimated MSOC using the formulation of Vignati (2010) and Gantt (2011). The dashed lines in panels (a, f) are the regression line of [OC] and [*Chl-a*] or ([*Chl-a*] × $[Na^+]^{0.45}$) with 0–30% percentile ratios, indicated by solid markers, during Cruise I (blue) and Cruise II (red). The regressions line in panels (b−e, g−j) represent the correlation between WIOC and the estimated MPOC or between WSOC and the estimated MSOC in each panel during the two cruises.

6. *L253-254 "These parameterizations perform well to trace the variation trends of MPOC. However, they might lead to an underestimation of the primary MOA over the Northwest Pacific Ocean." What could be reasons? Is the difference between MPOC and WIOC can (partly) explain the different performance? Can location/terrestrial aerosols play a role in the difference? Please further explain/discuss.*

Response: Thanks for the suggestions. We agree with the review that locations could be an important reason for the difference, and have added the description in lines 338−340. The seawater compositions, marine environment or atmospheric meteorological conditions in the North Atlantic and Northwest Pacific Oceans are different. These different environmental conditions result in different quantitative relations between seawater *Chl-a* and MPOC in these oceanic regions.

Based on the weak correlations between MPOC and EC in marine aerosols (lines 162−164) and the air mass back trajectories (added in Figure S1 and lines 164−166), the influence of transported terrestrial aerosols was limited during the cruises. What's more, the organic aerosols transported from terrestrial environments were secondary or aged organic aerosols, and tend to be water-soluble organic compounds. Thus, the potential terrestrial aerosols contributed the WSOC and MSOC fractions in the marine aerosols.

The seawater *Chl-a* in the parametrization was determined using the spatial average of the satellite-derived *Chl-a* concentrations in Gantt et al. (2011), which was in-situ measured using the collected surface seawater in this work. This could be an additional reason for different parameterizations (added in lines 340−343). Previous studies (Gantt et al., 2011; Vignati et al., 2010) and this study compared the MPOC with WIOC, which is widely regarded from marine primary emissions. Thus, the difference between MPOC and WIOC might not be the main reason for different parameterizations.

Lines 338−340:

This is mainly due to different seawater compositions, marine environment or atmospheric meteorological conditions in the North Atlantic and the West Pacific Oceans, which result in different quantitative relations between seawater *Chl-a* and MPOC in these oceanic regions.

Lines 340−343:

What's more, the seawater *Chl-a* was determined using the spatial average of the satellite-derived *Chl-a* concentrations in Gantt et al. (2011). This could be an additional reason for the different parameterizations between *Chl-a* and MPOC compared with the results based on the in-suit measured *Chl-a* in this work.

Lines 162−164:

The correlation coefficients between OC and EC were lower (Cruise I: r=0.48; Cruise II: r = 0.17) than those between OC and seawater *Chl-a*, suggesting that the potential impacts of transported anthropogenic pollutants were limited during the cruises.

Lines 164−166:

The air masses were mainly transported from open oceanic regions, and thus the impacts of terrestrial outflows were limited during the cruises (Fig. S1).

Newly added Figure S1:

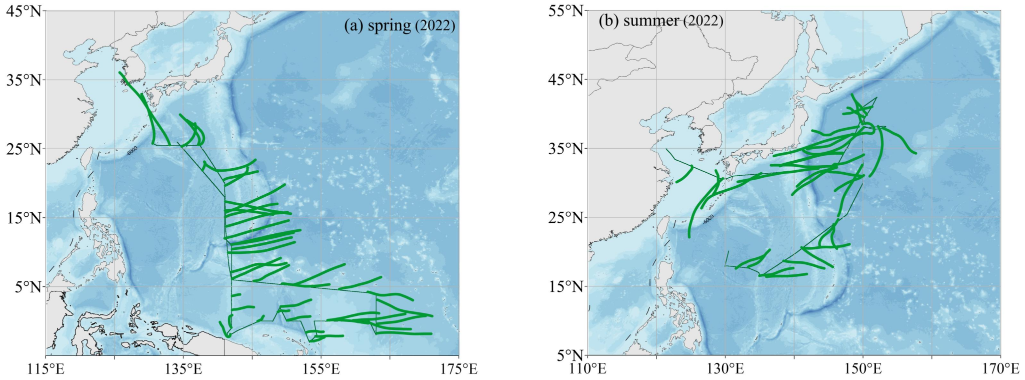

Figure S1 The 24-hr back trajectories of air masses during the cruises in (a) spring and (b) summer.

7. *For 3.3, overall, I don't see clear conclusion between the two methods, [chl-a] vs [chl-a] + [Na+]. From the "4 Summary", it seems that the authors prefer the second method. However, based on the comparison of Fig. 3b vs 3f and 3c vs 3g, it is not clear to me that the second method performs better. The "r" value is quite the same (0.87 vs 0.88 and 0.73 vs 0.73). Even, the first method performs better by comparing the fraction of points falling between the 1:2 and 2:1 lines in 3b and 3f, which are 69% (L225) and 58% (L243), respectively.*

Response: We used both the correlation coefficients (r) and the slopes of the fitting line between WIOC and estimated MPOC to evaluate the performance of different MPOC parameterization approaches. It means that the estimated MPOC shows a similar variation trend to WIOC if with a r value closer to 1, and a good comparison with the WIOC concentrations if with a slope closer to 1. The estimated MPOC using the two methods showed good correlations with WIOC, with the correlation coefficients of 0.87 and 0.88. However, the MPOC concentration was underestimated using [*Chl-a*], with a fitting line slop of 1.17 compared with 1.036 using [*Chl-a*] $\times[Na^+]^{0.45}$ as the input. Related descriptions have been added in lines 302−305, 309−311.

Lines 302−305:

Both the correlation coefficients (r) and the slopes of the fitting line between WIOC and estimated MPOC are used to evaluate the performance of different MPOC parameterization approaches. It means that the estimated MPOC shows a similar variation trend to WIOC if with a r value closer to 1, and a good comparison with the WIOC mass concentrations if with a fitting line slope closer to 1.

Lines 309−311:

When using p=0.45, both the fitting line slope (1.036) and r value (0.88) suggested an overall better performance than using other p values (Fig. S5, 3g). Without the [Na^+] as an input parameter, the fitting line slope and r of WIOC and MPOC were respectively 1.17 and 0.87 (Fig. 3b), suggesting an underestimation of MPOC.

8. *4 and the associated discussion of spatial distribution. How does the season difference affect the spatial distribution?*

Response: Aerosol samples were collected among the oceanic region within 15°N−30°N during the two cruises, which were compared to elaborate the seasonal difference. Related analysis has been added in lines 389−396 and Figures S7, S8.

Lines 386−396:

Aerosol samples were collected among 15°N−30°N during both the spring and the summer cruises, which were compared to elaborate the seasonal difference. The variations of the estimated MPOC and MSOC along the latitude are shown in Fig. S7, S8. Among the observation region within 15°N−30°N, the average MPOC was comparable in spring (0.16 μgC m^{-3}) and summer (0.18 μgC m^{-3}), with the average *Chl-a* concentration 0.042 and 0.044 μg L^{-1}, respectively. Among the oceanic regions with similar concentrations of seawater *Chl-a*, the MPOC abundance in marine aerosols was comparable without seasonal difference. Among 15°N−30°N, the elevation of MPOC concentrations was consistent with the elevated seawater *Chl-a* concentration without seasonal difference (Fig. S7a). This is consistent with the finding that marine biogenic activities drive the MPOC production. The average MSOC concentration was 0.24 μgC m^{-3} within 15°N−30°N in summer, higher than that in spring (0.19 μgC m^{-3}). The elevated MSOC was driven by the increase of seawater TOC concentrations (Fig. S8b). What's more, the stronger solar radiation in summer (Fig. S6) favored the photochemical VOC production in SML, their further photo-oxidation reactions, and the MSOC formation in the atmosphere.

Newly added Figures S7, S8:

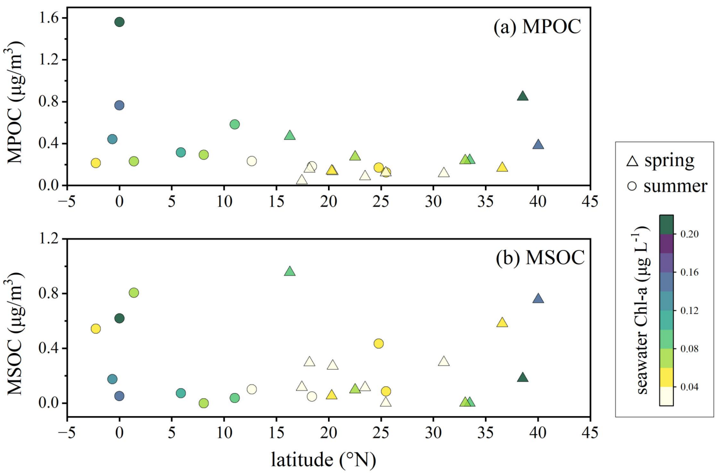

Figure S7 Spatial distribution of the estimated MPOC and MSOC concentrations. The data is colored by the corresponding seawater *Chl-a* concentrations.

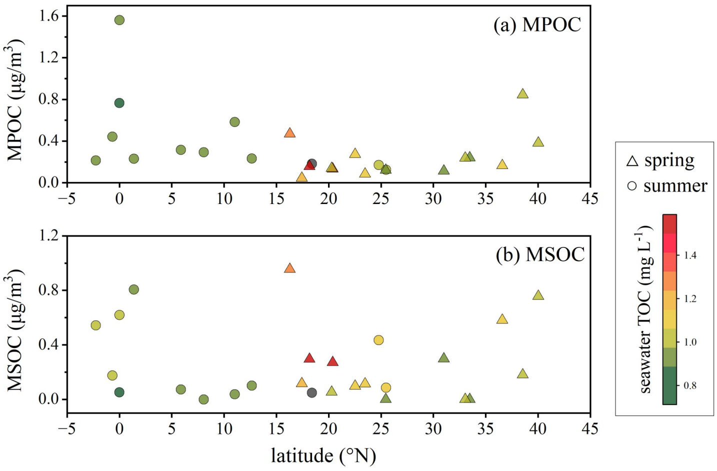

Figure S8 Spatial distribution of the estimated MPOC and MSOC concentrations. The data is colored by the corresponding seawater TOC concentrations.

9. L281-284: the link between TOC in the seawater and "abiotic VOC precursors from the photochemical production in the sea surface microlayer" is missing. Please further elaborate.

Response: The abiotic VOCs (e.g., isoprene) could be formed via photochemical degradation of soluble organic substances in the surface seawater. We have added their link in lines 376−380.

Lines 376−380:

Previous studies suggested that interfacial photochemical degradation of dissolved organic matters in seawater could be an important source of marine VOCs (e.g., isoprene) on a global scale (Bruggemann et al., 2018; Cui et al., 2023; Wang et al., 2023a; Yu and Li, 2021). For remote oceanic regions with high solar radiation but low biological activities, interfacial photochemistry of surface organics could be a major source of abiotic VOCs in the marine boundary layer (Bruggemann et al., 2018; Cui et al., 2023).

10. L310 "This was consistent to the higher mass contribution by MPOC than MSOC in the marine aerosol samples," There are many samples especially in summer (> half) that have quite low MPOC/OC fractions (< 40%) while the C1 fractions (~ 70% by average) are still very high as the other samples with high MPOC/OC fractions (~ 80%). Therefore, I couldn't buy it with your statement at the current stage. Please further elaborate.

And by the way, Fig. S4 doesn't show any measurements of Chl-a (L316)

Response: The difference in MPOC/OC fractions and PRLIS (C1) fractions was due to the different florescent efficiency of organic molecules from different sources. We now have deleted this sentence and added related statements in lines 420−432.

The seawater *Chl-a* data is added in Figure S9.

Lines 420−432:

It is noted that the proportion of different fluorescent compounds did not represent their mass contributions, as the florescent efficiency of organic compounds was related to their chemical structures. Organic molecules with substantial conjugation of π-bonds or double bound structures are known to be especially efficient at emitting fluorescence, particularly when N atoms are present (Chen et al., 2016a; Pöhlker et al., 2012). Amino acids, vitamins, and humic-like substances have been identified as efficient fluorophores (Graber and Rudich, 2006; Laskin et al., 2015; Pöhlker et al., 2012). The sea-to-air transfer of phytoplankton-produced protein-containing organics leads to a significant enhancement of fluorescent compounds in SSA (Aller et al., 2017; Lawler et al., 2020; Miyazaki et al., 2018a). The PRLIS, or named protein-like organic matter (PLOM), has been identified as a common component in the oceanic organic matter, and enriched in marine aerosols (Chen et al., 2016b). However, the biogenic SOA (e.g., isoprene oxidation products abundant in marine atmospheres) molecules, without conjugated double bounds, are weakly fluorescent or do not display fluorescent properties (Carlton et al., 2009; Laskin et al., 2015). Thus, the WSOC contributed by biogenic SOA was not included in the detected fluorescent components, and the observed proportions of PRLIS emitted by sea sprays was higher than those of the WIOC mass contribution in the marine aerosols.

Revised Figure S9:

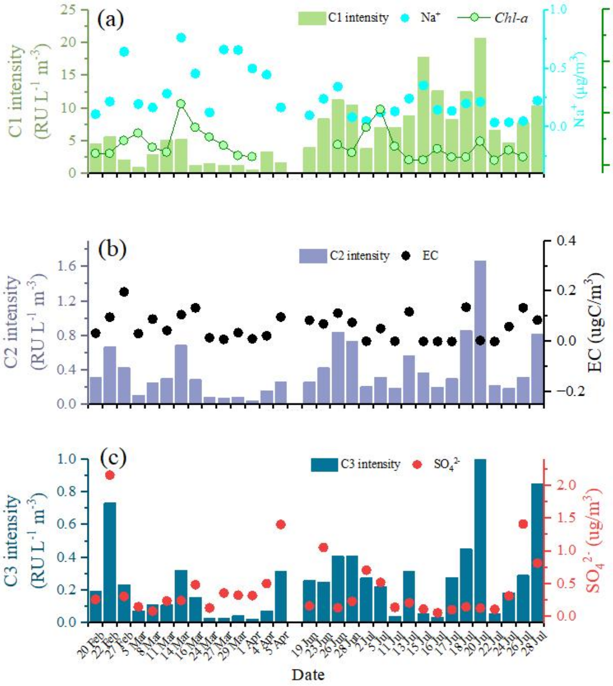

Figure S9 Variations of fluorescence component intensity identified by three-component solutions based on PARAFAC model analysis and related aerosol components: (a) C1, Na^+, and *Chl-a*, (b) C2 and EC, (c) C3 and SO_4^{2-}.

Minor comments:

1. *L49 "Ocean surface is one of the largest active reservoirs of organic carbon on Earth, resulting from phytoplankton, algal as well as the related senescence and lysis (Hedges, 1992; Quinn and Bates, 2011)." How large is this reservoir compared to other reservoirs?*

   Response: Ocean surface reserve about 18% of the active organic carbon. Other reservoirs include soil humus, land plant tissue and surface marine sediments. We have added the proportion in line 51.

   Lines 50−52:

   Ocean surface is one of the largest active reservoirs of organic carbon on Earth (~18%), resulting from phytoplankton, algal as well as the related senescence and lysis (Hedges, 1992; Quinn and Bates, 2011).

2. *L86: please add the details of the source for the satellite-derived chl-a data.*

   Response: The details of the satellite-derived *Chl-a* data are now added in the revised version (lines 89−91).

   Lines 89−91:

   The satellite-derived *Chl-a* data were provided by Copernicus Marine Environmental Monitoring Service (CMEMS) with a spatial resolution of 4 km and a monthly temporal resolution (https://marine.copernicus.eu/). Here, we utilized the satellite-derived *Chl-a* data during March and June 2022 to support our conclusion.

3. *L97: "organic aerosol concentration". Please be specific. I guess it is "organic aerosol mass concentration".*

   Response: Thanks for the reminding. Yes, it is organic aerosol mass concentration. We have revised to be specific (lines 105−106).

   Lines 105−106:

   The mass concentration of organic aerosols was calculated by multiplying OC by a conversion factor 1.6 (Wang et al., 2023b).

4. *L116-117: please show their values (at least a selection of the values) to give the audience a direct sense of "comparable".*

   Response: Revised accordingly. We now have added the values in lines 132−135.

   Lines 132−135:

   The observed OC concentrations during our cruises were comparable to previous studies over the North Pacific Ocean (0.5−0.7 μgC m^{-3}), and lower than those observed at an island in the West Pacific Ocean (1.7±1.0 μgC m^{-3}) (Hoque et al., 2015; Hoque et al., 2017; Kunwar and Kawamura, 2014).

5. *L138-139: have you tested the difference of the Chl-a concentration between spring and summer? Are they statistically different?*

Response: Thanks for the reminding. We test the difference of the *Chl-a* concentrations in spring and summer, and added in lines 168−169.

Lines 168−169:

However, the difference was not significant, with a *P* value of 0.33.

6. *In section 3.2, please further explain why we would expect good or poor correlations between chl-a concentration and organic fraction?*

Response: As suggested, we have added related explanations in section 3.2 (lines 201−208).

Lines 201−208:

Seawater *Chl-a* concentration is one of the most important factors driving the variation of organic fraction in the SSA, and they display good correlations when the wind speed does not vary a lot. However, wind speed should be combined with surface *Chl-a* to predict the organic fraction in SSA if the wind speed varies obviously during the observation or simulation periods (Gantt et al., 2011; Grythe et al., 2014). This is due to the influence of wind on the coverage of sea surface microlayer (SML) in the sea surface, which is enriched in organic compounds. For a given chemical composition of seawater, the largest coverage of sea surface by SML and a higher organic fraction in SSA are expected during calm winds. However, the SML would be destructed by mixing into the underlying seawater and the organic fraction in SSA decreased when surface wind exceeded 8 m s^{-1} (Gantt et al., 2011).

7. *L185 "Under certain marine environment conditions (e.g., Chl-a, wind speed, SST etc.), the abundance of MPOC should be constant." Please further explain what these conditions are with respect to Chl-a, wind speed, SST, etc.*

Response: Related descriptions are added in lines 240−245.

Lines 240−245:

Seawater *Chl-a* concentration is the most important factors driving the variation of organic fraction in the SSA, and has been widely used to estimate the organic fraction in SSA (Gantt et al., 2011; Vignati et al., 2010). For given chemical composition of seawater, the largest organic fraction in SSA is expected during calm winds. An increase in wind speed above 3–4 m s^{-1} will cause a rapid decrease of organic fraction due to the destructing of the SML coverage, and the lowest organic fraction is expected for wind exceeded 8 m s^{-1} (Gantt et al., 2011). Seawater temperature is related to the production efficiency and the number concentrations of SSA (Christiansen et al., 2019).

8. *Equation L2 and L3: I highly suggest explicitly showing the equation parts like [OC], [Chl-a], etc. in the equations, rather than using those dots. This will make it much easier for readers.*

Response: Revised accordingly.

9. Fig 3 caption and the texts referring to Fig 3: Please check the reference carefully. For example:

1. the "panels (b, d)" in L208 should be "panels (b, f)".
2. The "panels (c, f)" in L209 should be "panels (c, g)".
3. L231, 233: Fig. 3d should be "3e"
4. L244: 3d should be "3f"

Response: Thanks very much for the reminding. We have checked carefully and corrected the reference in Figure 3 caption and related discussion in the main text.

10. L242: why "p=0.45"? Why not other values within 0.35-0.65?

Response: We selected p=0.45 based on both the fitting line slope and correlation coefficient of WIOC and estimated MPOC. When using *p*=0.45, both the fitting line slope (1.036) and r value (0.88) suggested an overall better performance than using other *p* values (Fig. S5). We added the descriptions in lines 309−310 and revised Figure S5 to be clear.

Lines 309−310:

When using p=0.45, both the fitting line slope (1.036) and r value (0.88) suggested an overall better performance than using other *p* values (Fig. S5, 3g).

Revised Figure S5:

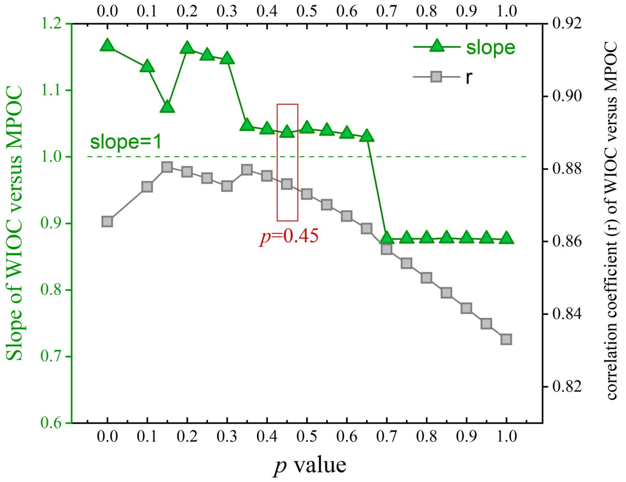

Figure S5 The variations of the fitting line slopes and correlation coefficients (r) of WIOC and estimated MPOC, using Eq. 3 with the *p* value changing from 0−1.

11. S2 caption: please also add the meaning of the colors.

Response: Revised accordingly.

Revised Figure S3 caption:

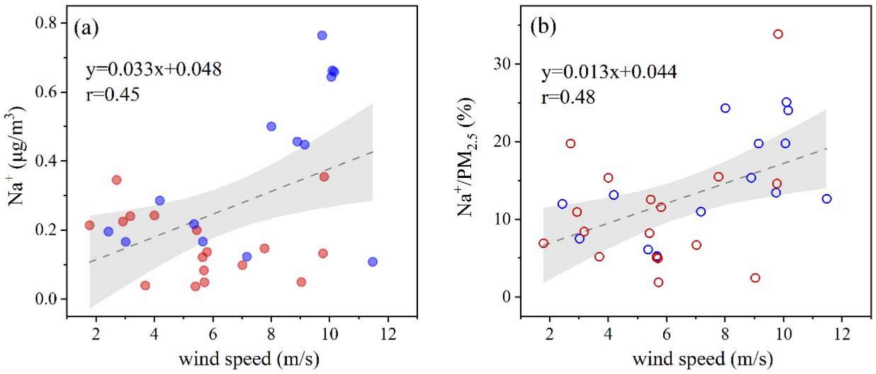

Figure S3 The variation of Na^+ concentration and $Na^+/PM_{2.5}$ as a function of the wind speed during the cruises. The data obtained during the spring Cruise I is in blue, and the data during the summer Cruise II is in red.

References:

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Lawler, M. J., Lewis, S. L., Russell, L. M., Quinn, P. K., Bates, T. S., Coffman, D. J., Upchurch, L. M., and Saltzman, E. S.: North Atlantic marine organic aerosol characterized by novel offline thermal desorption mass spectrometry: polysaccharides, recalcitrant material, and secondary organics, Atmospheric Chemistry and Physics, 20, 16007-16022, 10.5194/acp-20-16007-2020, 2020.

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Zhang, Y., Wang, Y., Li, S., Yi, Y., Guo, Y., Yu, C., Jiang, Y., Ni, Y., Hu, W., Zhu, J., Qi, J., Shi, J., Yao, X., and Gao, H.: Sources and Optical Properties of Marine Organic Aerosols Under the Influence of Marine Emissions, Asian Dust, and Anthropogenic Pollutants, J. Geophys. Res., [Atmos.], 130, 10.1029/2025jd043472, 2025.

Referee #2

General Comment

This paper is based on the analysis of PM2.5 samples collected over the Northwest Pacific Ocean during two campaigns in the spring season during 19 Feb.−9 April, 2022, and in the summer during 19 June−30 July, 2022. The chemical composition data have been investigated to the spatial/regional variability but mainly of Marine organic aerosol (MOA) fractions (WSOC and WIOC, primary and secondary), and Chl-a data as a supporting parameter. While the paper presents a quality data set, the discussion of the results is not up to mark. The basis or tracer data is not enough to quantify the contributions of WSOC and WIOC, primary and secondary to MOA.

Another concern is the ignoring/undermining of the impact of terrestrial biogenic outflow on the OC levels and their composition. Though the impact of long-range transport is mentioned, evidence (like back trajectory analysis) is not provided.

The data have been further utilised to parameterise the marine sources. But it is not well presented or summarized how seasonality in sea surface parameters and prevailing environmental conditions could impact the seasonal MOA composition. Further, following specific comments provide the issues to be addressed.

Response: Thanks very much for the suggestions and specific comments. We now have carefully addressed the comments and revised the manuscript accordingly.

(1) Quantifying the contributions of WSOC and WIOC, primary and secondary OC to MOA

Response: In this work, the contribution of WSOC and WIOC was quantified by the measurement result. The organic carbon (OC) and WSOC in the MOA were quantified by analyzing the collected marine aerosol samples using a Sunset Laboratory thermal/optical carbon analyzer and a TOC analyzer (TOC-L, Shimadzu, Japan). The WIOC in MOA was calculated by the difference between OC and WSOC concentrations in each sample. The contribution of WSOC and WIOC in MOA could then be quantified by the measured concentrations.

We quantified the concentrations and contribution of primary and secondary OC in MOA using the tracers of seawater *Chl-a* and sea salt (Na^+). Seawater *Chl-a* or a combination of *Chl-a* and wind is a widely used oceanic tracer to predict the organic fraction in primary marine aerosols (sea spray aerosols) (O'dowd et al., 2004; O'dowd et al., 2008; Rinaldi et al., 2013; Spracklen et al., 2008). Here, we checked the variations of OC or WIOC in MOA as a function of *Chl-a*, wind speed, and sea salt (Na^+) in section 3.2, and then proposed a parameterization scheme to estimate the primary and secondary OC in MOA in section 3.3. The performance of MPOC parameterization was then evaluated by comparing the estimated MPOC concentrations with WIOC, which is generally considered as a proxy for MPOC in previous studies.

The estimated primary OC and secondary OC matched well with the WIOC and WSOC, and the performance of the parameterization was better than the widely used functions proposed based on the results in the North Atlantic Ocean (Gantt et al., 2011; Vignati et al., 2010). Thus, we concluded that seawater *Chl-a* and aerosol components (OC, Na^+) could be used to separate the primary and secondary OC in MOA. This is an easy approach not dependent on other organic tracers, usually obtained through complex analysis procedures, and this approach is not limited to the time resolution of sample collection.

(2) The potential impact of terrestrial biogenic outflow on the OC levels and composition

Response: We agree with the reviewer that we cannot exclude the potential impacts of terrestrial biogenic outflows on the OC levels or compositions in the MOA. The OC contributed by terrestrial outflows is aged/secondary organic aerosols, which are mostly included in the WSOC and the oxygenated humic-like substances (HULIS, identified C3 in the fluorescence components) in MOA. Previous studies have suggested that aged or secondarily formed organic aerosols are mostly water-soluble (Boreddy et al., 2018; De Jonge et al., 2024; Miyazaki et al., 2010), as described in lines 187−189. The impacts of terrestrial biogenic outflows on OC compositions were included in and could be partially reflected by the variation of oxygenated HULIS (C3) in the collected aerosol samples. C3 showed a similar variation to sulfate, which indicated their sources related to secondary formation or aged processes. Oxygenated HULIS included the secondarily-formed and aged organic aerosols from both marine and terrestrial sources. Related analysis is now added in lines 318−322, 411−412.

As suggested, the back trajectory analysis of air masses during the cruises is now added in the revised version (Figure S1). Based on the back trajectories of air masses and the weak correlation between OC and EC (a tracer of terrestrial combustion emissions), we proposed that the impact of terrestrial outflows might be limited during the cruises. The air masses were mainly transported from open ocean based on the back trajectory analysis. What's more, the correlation coefficients between OC and EC (Cruise I: $r=0.48$; Cruise II: $r = 0.17$) were lower than those between OC and seawater *Chl-a* (Cruise I: $r = 0.81$, $p < 0.01$; Cruise II: $r = 0.67$, $p < 0.01$), as stated in lines 162−164. This suggested that the potential impacts of transported terrestrial outflows were limited compared to the marine biological emissions during the cruises. Related analysis is now added in lines 95−96, 322−324, 369−370.

Lines 318−322:

It is noted that, based on the shipboard in-situ observation, we cannot exclude the potential impacts of gaseous precursors or aged organic aerosols long-range transported from terrestrial environments, which were mostly in the MSOC fraction. The organic aerosols transported from terrestrial environments were secondary or aged organic aerosols, and tend to be water-soluble organic compounds (Boreddy et al., 2018; De Jonge et al., 2024; Miyazaki et al., 2010).

Lines 411−412:

Oxygenated HULIS included the secondarily-formed and aged organic aerosols from both terrestrial and marine sources.

Lines 95−96:

The 24-hr backward trajectories of air masses (Fig. S1) originating at 500 m above the ground level were calculated along the observation cruises every 24 hr using the HYSPLIT model (Version 5.2.1, NOAA).

Lines 322−324:

Based on the air mass back trajectories (Fig. S1) and the weak correlations between OC and EC stated in section 3.1, the impacts of transported continental outflows were limited during the cruises.

Lines 369−370:

Based on the air mass back trajectories (Fig. S1), the impacts of transported terrestrial outflows were limited among the observation regions.

Newly added Figure S1:

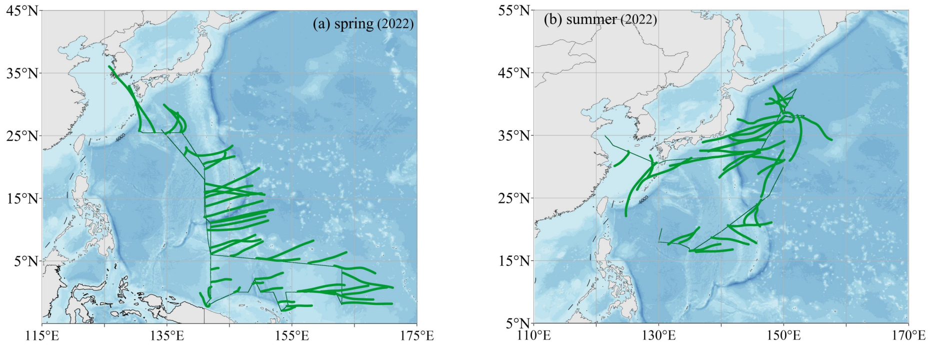


Figure S1 The 24-hr back trajectories of air masses during the cruises in (a) spring and (b) summer.

(3) Seasonal variations of sea surface parameters and MOA compositions

Response: Aerosol samples were collected among the oceanic region within 15°N−30°N during the two cruises. The sea surface parameters (*Chl-a*, TOC) and MOA compositions within 15°N−30°N were compared to elaborate the seasonal difference. Related analysis has been added in lines 386−396 and Figures S7, S8.

Lines 386−396:

Aerosol samples were collected among 15°N−30°N during both the spring and the summer cruises, which were compared to elaborate the seasonal difference. The variations of the estimated MPOC and MSOC along the latitude are shown in Fig. S7, S8. Among the observation region within 15°N−30°N, the average MPOC was comparable in spring (0.16 μgC m^{-3}) and summer (0.18 μgC m^{-3}), with the average *Chl-a* concentration 0.042 and 0.044 μg L^{-1}, respectively. Among the oceanic regions with similar concentrations of seawater *Chl-a*, the MPOC abundance in marine aerosols was comparable without seasonal difference. Among 15°N−30°N, the elevation of MPOC concentrations was consistent with the elevated seawater *Chl-a* concentration without seasonal difference (Fig. S7a). This is consistent with the finding that marine biogenic activities drive the MPOC production. The average MSOC concentration was 0.24 μgC m^{-3} within 15°N−30°N in summer, higher than that in spring (0.19 μgC m^{-3}). The elevated MSOC was driven by the increase of seawater TOC concentrations (Fig. S8b). What's more, the stronger solar radiation in summer (Fig. S6) favored the photochemical VOC production in SML, their further photo-oxidation reactions, and the MSOC formation in the atmosphere.

Newly added Figures S7, S8:

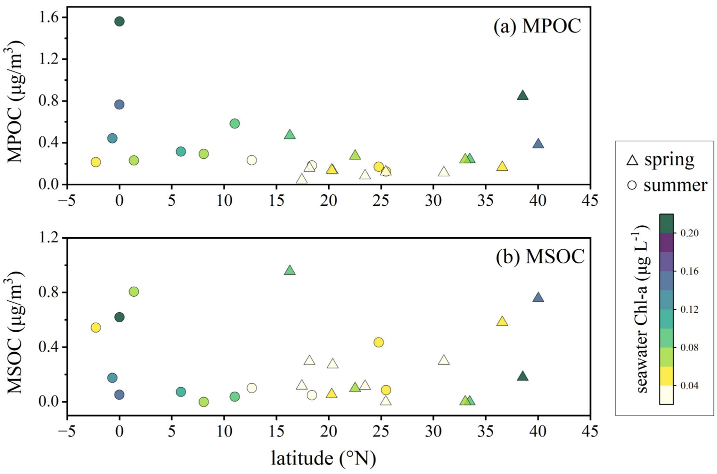


Figure S7 Spatial distribution of the estimated MPOC and MSOC concentrations. The data is colored by the corresponding seawater *Chl-a* concentrations.

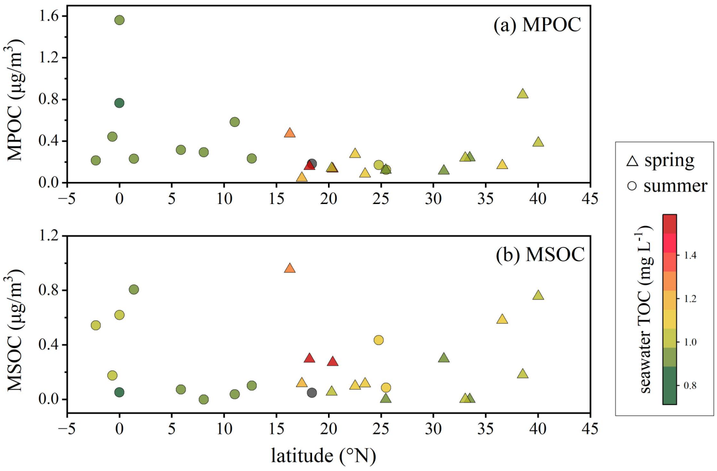


Figure S8 Spatial distribution of the estimated MPOC and MSOC concentrations. The data is colored by the corresponding seawater TOC concentrations.

General Comment

Line 57-58: "regions with high emission rates of MOA are largely related to the spatial" Why high emissions rates only? You may revise "regional emission rates of MOA are largely related to the spatial"

Response: Revised accordingly (line 58).

Lines 58−59:

A recent modeling study suggested that regional emission rates of MOA are largely related to the spatial distribution of ocean biological productivity (Zhao et al., 2021).

Line 60: "Due to the relationship between sea surface phytoplankton and MOA" I feel this phrase is not required.

Response: Deleted as suggested.

Line 64: "Observation-based parameterization..." Provide a reference to support this sentence.

Response: Related references (Brooks and Thornton, 2018b; Quinn et al., 2015a) are now cited.

Line 77: As shown in Figure 1(a) the track covers between 02S-34 N, so it would be appropriate to mention the region as "Northwest Pacific Ocean"? covering southern latitudes

Response: Thanks for the reminding. We change "the Northwest Pacific Ocean" to "the West Pacific Ocean (WPO)" throughout the main text in the revised version.

Line 80-81: "suspended particles (TSP) samples and PM2.5" can be revised as "suspended particles (TSP) and PM2.5"

Response: Revised accordingly.

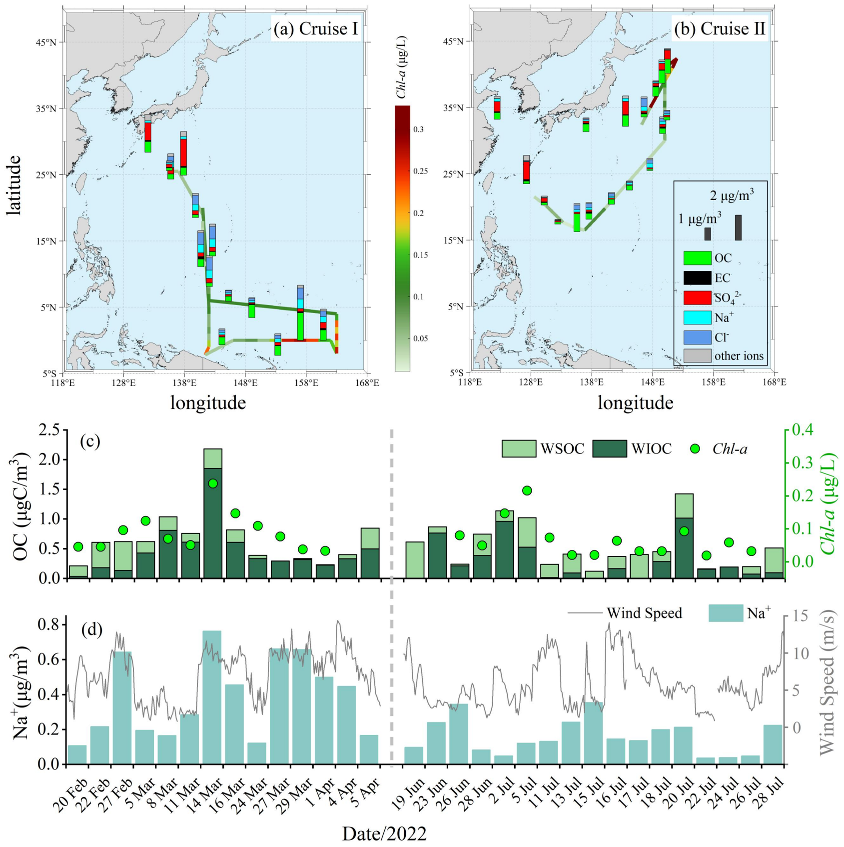
Line 114: "observation, and" should be "observation and"

Response: Revised accordingly.

Figure 1(b). I suggest to mention the year of the campaigns. You may provide a common x-axis title as (Date/Year).

Response: Thanks for the suggestion. The x-axis title of Figure 1 has been revised accordingly, and the year of the campaigns is mentioned.

Revised Figure 1:



Line 131: "$r = 0.67$, $p < 0.01$, Fig. 1)," $r = 0.67$ may not represent a strong correlation. Also I dont find correlation plots in Figure 1, perhaps referring to Fig 2?

Response: Thanks for the reminding. This sentence is revised accordingly (lines 158−159).

Lines 158−159:

The abundance of MOA displayed similar spatial distribution (Fig. 1) and strong or medium correlations with the sea surface *Chl-a* concentration…

Line 136: But how about the impact/outflow of terrestrial biogenic emissions. ?

Response: As suggested in the general comment, we now have added the back trajectory analysis of air masses to address the potential impacts of terrestrial biogenic emissions during the cruises. The air masses were mainly transported from open oceanic regions based on the back trajectory analysis. The potential impacts of transported terrestrial outflows could be limited during the cruises. Related analysis is added in lines 95−96, 164−166 and Figure S1.

We agree with the reviewer that we cannot exclude the potential impacts of terrestrial biogenic outflows on OC in the marine aerosol samples. Based on the data in this work, we cannot distinguish the contribution of terrestrial biogenic emissions from the WSOC or the oxygenated HULIS. We will try to evaluate this impact combining other organic tracers in our future studies.

Lines 95−96:

The 24-hr backward trajectories of air masses (Fig. S1) originating at 500 m above the ground level were calculated along the observation cruises every 24 hr using the HYSPLIT model (Version 5.2.1, NOAA).

Lines 164−166:

The air masses were mainly transported from open oceanic regions, and thus the impacts of terrestrial outflows were limited during the cruises (Fig. S1).

Newly added Figure S1:

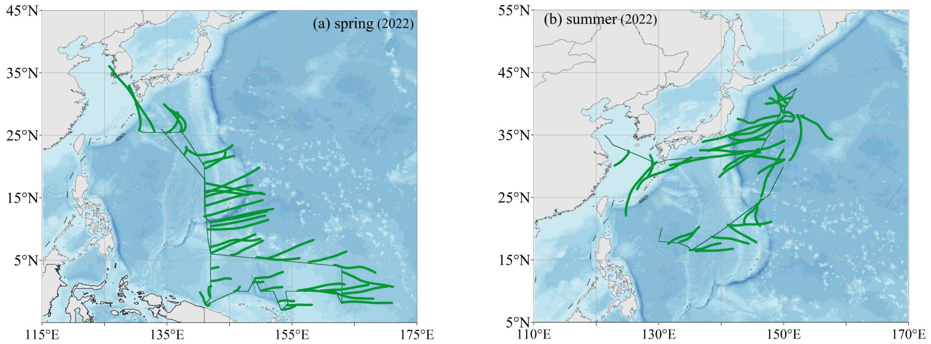


Figure S1 The 24-hr back trajectories of air masses during the cruises in (a) spring and (b) summer.

Line 138-139: "spring 138 (0.09 ± 0.06 μg L-1) than in summer (0.07 ± 0.05 μg L-1)." I think the values are not very different. So the explanation for the differences in OC levels is not robust. I feel the transport from regions of Papua New Guinea and Indonesia needs a consideration. Following works may provide more insights.
https://doi.org/10.1038/s41612-022-00311-0
https://doi.org/10.5194/essd-15-5403-2023

Response: We checked the significant level of the difference, and stated that the difference is not significant (P=0.33).

Thanks for your reminding of the related literatures. Based on the back trajectories of air masses, the influence of transported terrestrial emissions was limited. The potential influence of transported MOA or biogenic VOC precursors from the coastal oceanic regions of Papua New Guinea and Indonesia is now described in lines 170−173.

Lines 170−173:

For the samples collected near the equator, the MOA or biogenic VOC precursors could also be transported from coastal oceanic regions of Papua New Guinea and Indonesia with higher marine biological activity and higher isoprene emission fluxes (Cui et al., 2023; Zhang and Gu, 2022). This could be an additional reason for the higher OC level during the spring cruise and the highest OC concentration observed on 14 March.

Line 141-142 and other places: "mass ratios of 70% ± 27% in spring and 48% ± 35% in" should be "mass ratios of 70 ± 27% in spring and 48 ± 35% in"

Response: Revised accordingly.

Line 145: "WSPC in marine aerosols." should be "WSOC in marine aerosols."

Response: It is corrected.

Line 147-148: "and transferred to atmospheric aerosols over the ocean." Rewrite with a better phrase.

Response: This sentence is rewritten to be clear (lines 184−185).

Lines 184−185:

These organic substances could be enriched in the surface seawater and then transferred into the atmospheric aerosols within the marine boundary layer.

Line 152: "marine reactive organic gases (Boreddy et al., 2018; De Jonge et al., 2024; Miyazaki et al., 2010)." Here I would suggest citing studies which provide measurements of reactive trace gases implying the roles in SOA in marine air. Following works may be cited:
Sources and distribution of light NMHCs in the marine boundary layer of the northern Indian Ocean during winter: Implications to aerosol formation. Journal of Geophysical Research: Atmospheres, 129, e2023JD039433. https://doi.org/10.1029/2023JD039433
Elevated levels of biogenic nonmethane hydrocarbons in the marine boundary layer of the Arabian Sea during the intermonsoon. Journal of Geophysical Research: Atmospheres, 125, e2020JD032869. https://doi.org/10.1029/2020JD032869..

Response: Thanks for your kind reminding of the related papers. We now have cited these works in the revised manuscript (lines 189−191).

Lines 189−191:

Reactive gaseous precursors of organic aerosols are widely observed over different oceanic regions (Tripathi et al., 2024; Tripathi et al., 2020; Wang et al., 2023a), which contribute to the SOA formation in the marine boundary layer.

Further, in this section, I suggest comparing the composition of WSOC, WISOC, and other species presented in typically continentally influenced air over this region. The *following work may be useful in this regard (It would be good if you can find some other similar works and highlight those)*

Anthropogenic aerosols observed in Asian continental outflow at Jeju Island, Korea, in spring 2005, J. Geophys. Res., 114, D03301, doi:10.1029/2008JD010306.

Response: We have cited related works and compared the concentrations or contributions of WSOC, WIOC and EC in this work and those observed in the atmospheres typically influenced by continental outflows (lines 130−132, 174−179).

Lines 130−132:

The EC concentrations were 0.066 ± 0.056 μgC m^{-3} and 0.055 ± 0.052 μgC m^{-3} during the spring and the summer observations, much lower than those observed over coastal areas typically influenced by continental outflows (Sahu et al., 2009; Zhang et al., 2025).

Lines 174−179:

The observed concentrations of WSOC and WIOC over the open Pacific Ocean were lower than those observed in the atmosphere under severe influence of continental outflows (Sahu et al., 2009; Zhang et al., 2025). Marine organic aerosols were dominated by the water-insoluble fractions, with the WIOC/OC mass ratios of 70 ± 27% in spring and 48 ± 35% in summer (Fig. 1). The proportion of water-soluble organics in MOA over the WPO was lower than that observed over the East Asian marginal seas in autumn (75%), during which severe impacts of continental anthropogenic pollutants were observed (Zhang et al., 2025).

Line 155: "..through the ocean bubble bursting." why through bubble bursting only as other air-sea exchange processes can also contribute?. Please explain and revise accordingly.

Response: The seawater organic substances could be transferred into sea spray aerosols through bubble bursting and wave breaking processes. We now have revised this sentence accordingly (lines 193−194).

Lines 193−194:

The similar variation trends and good correlations between WIOC in marine aerosols and seawater *Chl-a* (Fig. 1, 2) suggested the origins of MOA from seawater through ocean bubble bursting or wave breaking.

Line 158-160: "in the present .." I think similar discussion is presented in previous section (line 143-144). I suggest to concise the discussion related to correlation in one section only.

Response: This sentence is simplified accordingly (lines 197−198). We try to address the difference between WIOC and WSOC in section 3.1 (lines 179−181 in the revised version), and the difference between organic concentration and organic fractions in section 3.2 (lines 197−198 in the revised version).

Lines 197−198:

Over the West Pacific Ocean, we observed better correlations between OC or WIOC concentrations and *Chl-a* than those between organic or water-insoluble organic mass fractions and *Chl-a* (Fig. 2a−2d).

Line 170: "concentrations of OC or WIOC in PM2.5 did not show obvious correlations with" I disagree here. The levels OC and WIOC tend to decrease rapidly with wind speed (may not be linear). The discussion should be based on dependencies or trend (with Chl-a and wind speed) but not strictly in terms of correlation/ anti-correlation (as data points are scattered)

Response: Thanks for the reminding. This sentence is revised accordingly (lines 216−217).

Lines 216−217:

During our cruises over the WPO, the concentrations of OC or WIOC in $PM_{2.5}$ showed a decreasing trend with the increase of wind speed (Fig. 2e, 2f).

Line 172: "which was due to the elevated proportions of inorganic sea salts in the marine aerosols under the high-wind" This justification is not convincing. That can change only the magnitude of the slope but not the direction (positive/negative), check.

Response: The reason is revised as (lines 218−219): The organic-enriched SML in the sea surface would be destructed under high wind speed conditions, which results in a decrease of organic substances transported into the SSA (Gantt et al., 2011).

Line 185: "Under certain marine environment conditions (e.g., Chl-a, wind speed, SST etc.)," What is certain here , as values of these parameters are not given. And why MPOC should be constant anyway?

Response: Surface seawater *Chl-a* and wind speed are the most important marine environmental factors determining the abundance of primary marine organic aerosols (organics in sea spray aerosols). Thus, for a given *Chl-a* and wind speed condition, the abundance of MPOC primarily generated from surface seawater should be constant. We now have added related descriptions to be clear (lines 238−245).

Lines 238−245:

For a given marine environment condition (a given *Chl-a*, wind speed, sea surface temperature (SST), etc.), the abundance of MPOC should be constant (Gantt et al., 2011). Seawater *Chl-a* concentration is the most important factors driving the variation of organic fraction in the SSA, and has been widely used to estimate the organic fraction in SSA (Gantt et al., 2011; Vignati et al., 2010). For given chemical composition of seawater, the largest organic fraction in SSA is expected during calm winds. An increase in wind speed above 3–4 m s^{-1} will cause a rapid decrease of organic fraction due to the destructing of the SML coverage, and the lowest organic fraction is expected for wind exceeded 8 m s^{-1} (Gantt et al., 2011). Seawater temperature is related to the production efficiency and the number concentrations of SSA (Christiansen et al., 2019).

Line 188: "Based on the shipboard observations in present study," Should be "In the present study,"

Response: Revised accordingly.

Line 191: "contribution of SOA (e.g., methanesulfonic acid from DMS oxidation," Check if DMS oxidation provides SOA, I know it gives sulfate. But then do you consider sulfate as SOA ? Check following and other papers on DMS
Processes controlling DMS variability in marine boundary layer of the Arabian Sea during post-monsoon season of 2021. Journal of Geophysical Research: Atmospheres, 130, e2024JD042547. https://doi.org/10.1029/2024JD042547

Response: The oxidation of DMS could form both sulfate and methanesulfonic acid (MSA) (Barnes et al., 2006). MSA is an important SOA compound in the marine boundary layer. Related papers are cited in the main text (lines 251−252).

Line213-214: "Based on the correlation analysis above," should be "Based on the correlation analysis"

Response: Revised accordingly.

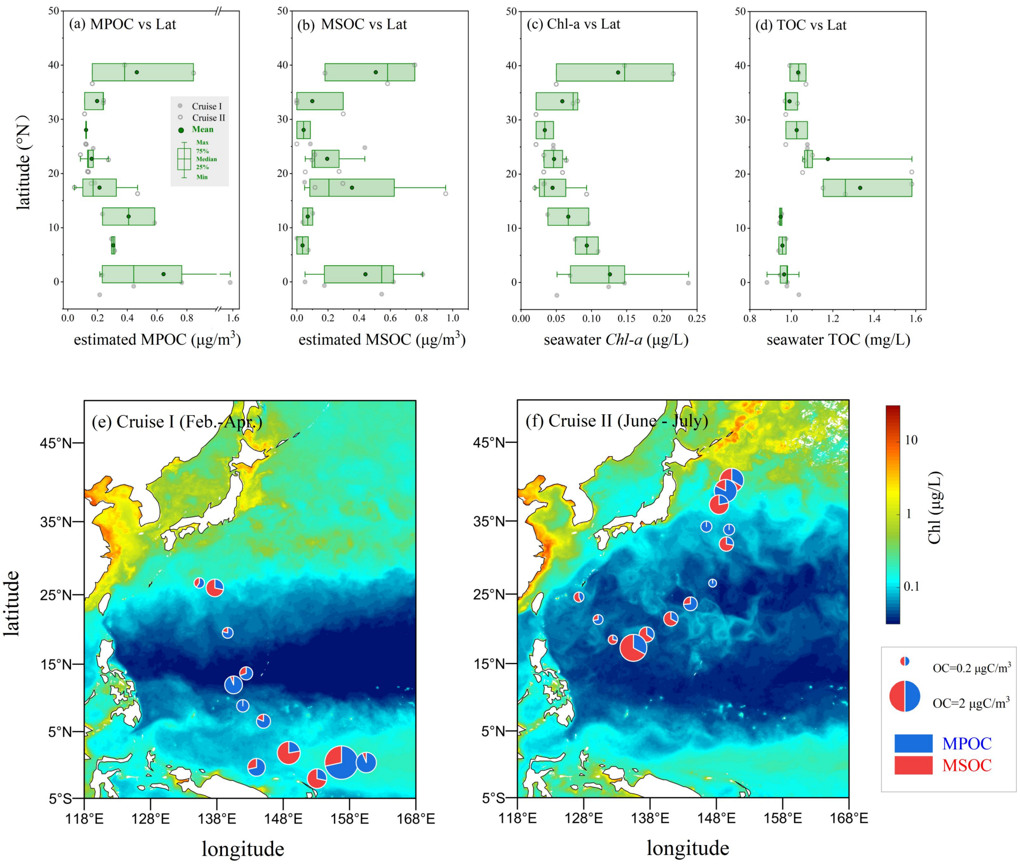
Line 215-219: "In the classic OC/EC ratio method...Yu et al., 2021)." These sentences appear out of context , may be removed.

Response: We have deleted these sentences here as suggested.

Figure 4 (a-d): Y-axis should be Latitude . Also check Fig 4 (e-f) if axes titles are correct (latitude /longitude or vice versa) .

Response: Thanks for the reminding. We now have corrected the axes titles of Figure 4.

Revised Figure 4:



Line 272:"high concentrations over the oceanic regions among 5°S–5°N and 35°N–40°N," Here for these two region, why the contributions from the land vegetation should also be discussed (due to vicinity to land/island regions)

Response: The potential contributions of organic aerosols from land or coastal regions are analyzed in lines 369−373.

Lines 369−373:

Based on the air mass back trajectories (Fig. S1), the impacts of transported terrestrial outflows were limited among the observation regions. Marine organic aerosols or biogenic VOC precursors could also be transported from coastal oceanic regions with higher *Chl-a* levels and higher isoprene emission fluxes (Cui et al., 2023; Zhang and Gu, 2022), which could be an additional reason for the higher MOA concentrations within 5°S–5°N and 35°N–40°N.

LIne 279-280" "from surface seawater as well as the production of biologic VOCs (e.g., isoprene, DMS, etc.) from phytoplankton" should be revised as "from seawater as well as the production of VOCs from phytoplankton" but again terrestrial contributions should be discussed.

Response: Revised accordingly.

Line 284-286: "The strong.." This sentence seems speculative also Chl-a map does *not support this localized feature.*

Response: Surface net solar radiation (SSR) data is now added to address the stronger solar radiation within 15°N–20°N during the summer cruise than during spring (lines 93−95, 382−383, Figure S6).

Lines 93−95:

Surface net solar radiation (SSR) data were obtained from the hourly data of the ECMWF Reanalysis v5 (ERA5) product (Hersbach et al., 2020), with a spatial resolution of 0.25°.

Lines 382−383:

The strong solar radiation during the summertime (19 June- 30 July) Cruise II, as shown in Fig. S6, favored the photochemical VOC production and the SOA formation in marine atmospheres.

Newly added Figure S6:

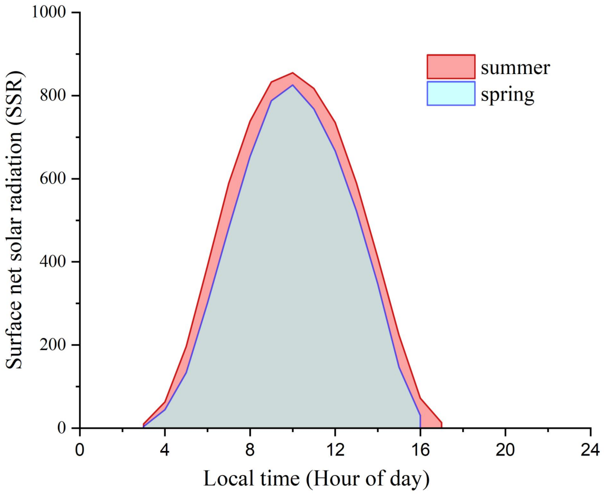


Figure S6 Diurnal variation of the surface net solar radiation (SSR) within the 15°N–20°N during the spring and the summer cruises.

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Referee #3

This manuscript presented an investigation on the marine organic aerosol over Northwest Pacific Ocean. It is important to gain better understanding regarding the contribution of primary and secondary organics over the marine environments. This study provided valuable data sets of organic aerosol compositions in this understudied ocean. This manuscript proposed a parameterization approach to estimate this contribution and compared to other methods. This manuscript is overall well written and fits to the scope of this journal. There are a few issues I suggest to be addressed in the revision.

Response: Thanks very much for the overall positive comments and detailed suggestions on our manuscript. We have carefully addressed the comments below.

1. The abstract should present or summarize more on the quantitative results of the study.

Response: Thanks for the suggestion. The estimated concentrations of MPOC and MSOC, and their spatial variations have been added in the abstract to present the quantitative results.

Revised abstract (lines 26−31):

The parameterization approach of MPOC using [*Chl-a*]×$[Na^+]^{0.45}$ was validated through comparing with the water-insoluble organic carbon in the aerosol samples. The estimated MPOC (0.33 ± 0.32 μgC m^{-3}) averagely accounted for 56%−66% of the total organic carbon in the collected samples, which was mainly attributed to the protein-like substances transferred into the sea spray aerosols from seawater. Over the West Pacific Ocean, the MPOC and MSOC displayed peak concentrations over the regions 5°S–5°N (0.64 ± 0.56 and 0.44 ± 0.32 μgC m^{-3}) and 35°N–40°N (0.46 ± 0.35 and 0.51 ± 0.30 μgC m^{-3}). The variation and spatial distribution of MPOC and MSOC along the latitude were driven by the marine biological activities. High MSOC concentrations were also observed over the region of 15°N–20°N (0.35 ± 0.41 μgC m^{-3}), …

2. L96-98, please elaborate the use of 1.6 factor to covert OC to organic aerosol concentration.

Response: We added the description on the selection of OM/OC conversion factor (1.6) in lines 106−112.

Lines 106−112:

The OM/OC conversion factor (1.6) was selected based on previous observation results of marine organic aerosols. Over the North Atlantic, an OM/OC mass ratio of 1.8 was adopted for WSOC based on the speciation of WSOC performed on the samples, and a conversion factor of 1.2 was applied for WIOC (Cavalli, 2004a). An average OM/OC ratio of 1.75 was observed in the submicron organic aerosol samples over the Atlantic

Ocean (Huang et al., 2018). A higher proportion of water-soluble secondary organic aerosols (SOA), with higher OM/OC ratios than primary MOA, was observed in Huang et al. (2018) than in this study. Here, the proportions of WIOC are comparable to (summer cruise) or higher than (spring cruise) those of WSOC, and thus an OM/OC ratio of 1.6 was selected here.

3. L177-178, do you mean indicator of the overall organic production? If not, this is misleading and please provide evidences or references to support and justify this statement.

Response: Yes, we want to state that Na^+ in fine particles could be used as a better indicator of the overall organic production levels than the wind speed in marine atmospheres based on the correlation analysis. We revised this sentence to be clear (lines 222−224).

Lines 222−224:

We proposed that, for the filter-based observation or the samplings with a similar time resolution, Na^+ in fine particles could be used as a better indicator of the overall organic production levels than the wind speed in marine atmospheres.

4. L187-188, L252, It would be helpful if the parameterizations from Gantt et al (2011) and Vignati et al. (2010) can be shown somewhere in the main text or supplementals, so readers can have a direct comparison.

Response: Thanks for the suggestion. The parameterizations from Gantt et al (2011) and Vignati et al. (2010) have been added in lines 327−330.

Lines 327−330:

Vignati et al (2010) estimated the organic mass fraction in sea spray aerosol (OM_{SSA}) using seawater [*Chl-a*]: $\%OM_{SSA} = 43.5 \times [Chl\text{-}a](\mathrm{mg\ m^{-3}}) + 13.805$. Gantt et al (2011) predicted the OM_{SSA} using a combination of [*Chl-a*] and 10 m wind speed (U_{10}): $OM_{SSA}(\mathrm{Chl-a}, U_{10}) = \frac{OM_{SSA}^{max}}{1+\exp(-2.63[Chl-a]+0.18U_{10})}$, where OM_{SSA}^{max} is the maximum OM_{SSA} observed during the cruises.

5. L207, L220, It is not clear why the 0-30 percentiles of ratios are chosen. Do the results change if different percentiles are selected?

Response: The reason for choosing data with 0-30% percentiles of ratios and related descriptions now have been added in lines 279−285.

Lines 279−285:

This is similar to the classic OC/EC ratio method (Turpin and Huntzicker, 1995). The OC/EC ratios in POC is usually calculated based on the dataset with the lowest 10%−20% percentile OC/EC ratios observed during the campaign, which is then used to separate the POC and SOC in each aerosol sample (Lim and Turpin, 2002; Yu et al., 2021). We used the data with the lowest 30% percentile of [OC]/[*Chl-a*] ratios, considering the number of data points to calculate the fitting curve of MPOC versus [*Chl*-*a*]. With more data points, the data with the lowest 10%‒20% percentile [OC]/[*Chl*-*a*] ratios could be used to estimate the [MPOC]/[*Chl*-*a*] ratios, and the estimated MPOC abundance may be a little higher than the results using the lowest 30% percentile data.

Biogenically driven marine organic aerosol production over the West Pacific Ocean

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Abstract. Marine organic aerosols play crucial roles in cloud formation and climate regulation within the marine boundary layer. However, the abundance of marine primary organic carbon (MPOC) generated by sea spray and secondary organic carbon (MSOC) formed via gas-to-particle conversion or atmospheric oxidation/aging processes remains poorly quantified, which hinders our understanding on the climate effects of marine aerosols. In this work, two shipboard cruises were conducted over the West Pacific Ocean to estimate abundance and compositions of marine organic aerosols. We propose an observation-based approach to quantify the MPOC and MSOC using a combined parameterization of the observed Na^+ in fine aerosol particles and the surface chlorophyll-*a* (*Chl-a*), an indicator of marine biological activity. The parameterization approach of MPOC using $[Chl\text{-}a] \times [Na^+]^{0.45}$ was validated through comparing with the water-insoluble organic carbon in the aerosol samples. The estimated MPOC (0.33 ± 0.32 μgC m^{-3}) averagely accounted for 56%−66% of the total organic carbon in the collected samples, which was mainly attributed to the protein-like substances transferred into the sea spray aerosols from seawater. Over the West Pacific Ocean, the MPOC and MSOC displayed peak concentrations over the regions 5°S–5°N (0.64 ± 0.56 and 0.44 ± 0.32 μgC m^{-3}) and 35°N–40°N (0.46 ± 0.35 and 0.51 ± 0.30 μgC m^{-3}). The variation and spatial distribution of MPOC and MSOC along the latitude were driven by the marine biological activities. High MSOC concentrations were also observed over the region of 15°N–20°N (0.35 ± 0.41 μgC m^{-3}), which was due to an additional contribution by the oxidation of volatile organic precursors from the photochemical production of seawater organics. This study proposes a parameterization approach to quantify the MPOC and MSOC over the Pacific Ocean or other oceanic areas.

Our results highlight the marine biogenically driven formation of marine organic aerosols, and different quantitative relations of MPOC with seawater *Chl-a* and other parameters are needed based on in-situ observations across oceanic regions.

1 Introduction

Marine aerosols are one of the most important natural aerosols on a global scale (De Leeuw et al., 2011; Quinn et al., 2015b). Observation and modeling studies have proved that marine aerosols are an important source of cloud condensation nuclei (CCN) and ice-nucleating particles (INPs) over remote oceanic areas, and play a vital role in Earth's radiation balance (Demott et al., 2016; Quinn et al., 2017; Sinclair et al., 2020; Vergara-Temprado et al., 2017; Wolf et al., 2019; Xu et al., 2022). Sea salt, sulfate, and organic matters (OM) make up the major components of marine aerosols, and the chemical nature determines the hygroscopicity, ice nucleation, and climate impacts of marine aerosols (Huang et al., 2022; Zhao et al., 2021). Marine organic aerosols (MOA) have attracted attention due to their effects on CCN formation over the remote ocean (Zhao et al., 2021). Limited understanding on the formation, flux and composition of MOA results in the estimation uncertainty of climate regulation by marine aerosols (Brooks and Thornton, 2018a; Quinn and Bates, 2011; Quinn et al., 2015b).

Organics are a major fraction in marine aerosols, contributing 3%−90% of submicron aerosol mass (Huang et al., 2018; O'dowd et al., 2004; O'dowd et al., 2008; Shank et al., 2012). MOA could be primarily released from the ocean surface or secondarily formed via the oxidation and gas-to-particle conversion of volatile organic compounds (VOCs), including dimethyl sulfide (DMS), isoprene, etc., in the marine boundary layer (Fu et al., 2011; Trueblood et al., 2019). Ocean surface is one of the largest active reservoirs of organic carbon on Earth (~18%), resulting from phytoplankton, algal as well as the related senescence and lysis (Hedges, 1992; Quinn and Bates, 2011). Wave breaking and bubble bursting at the ocean surface would inject quantities of organic-enriched sea spray aerosols (SSA) into marine atmospheres (Hu et al., 2024; Quinn et al., 2014). Organic matters are predominant in the fine or submicron SSA, which are usually dominated by water-insoluble organic carbon (WIOC) (Cavalli, 2004b; Cravigan et al., 2020; Miyazaki et al., 2020). However, the majority of the water-soluble organic carbon (WSOC) in MOA is contributed by secondary processes via the VOC oxidation or aged organic aerosols (Schmitt-Kopplin et al., 2012; Trueblood et al., 2019).

A recent modeling study suggested that regional emission rates of MOA are largely related to the spatial distribution of ocean biological productivity (Zhao et al., 2021). During phytoplankton blooms, the organic content elevated to as high as 63% of submicron aerosols, compared to a proportion of 15% during the low biological activity periods (O'dowd et al., 2004). Seawater chlorophyll-a (*Chl-a*) or its combination with wind speed and aerosol size has been used to parameterize the organic fraction in SSA (Gantt et al., 2012; Gantt et al., 2011). However, the abundance of various organics in SSA remains highly uncertain and is a current challenge to understand their role in cloud formation (Albert et al., 2012; Brooks and Thornton, 2018a).

Observation-based parameterization of primary and secondary MOA is urgently needed to constrain the modeling results (Brooks and Thornton, 2018b; Quinn et al., 2015a). In this work, two shipboard observations of atmospheric aerosols were conducted from the temperate to the tropical regions over the West Pacific Ocean (WPO) during spring and summer. Chemical compositions of marine aerosols, including organic carbon and inorganic ions, and seawater parameters were simultaneously obtained during the cruises. We derived a parameterization to estimate the primarily emitted organic aerosols in the SSA from wave breaking and bubble bursting, and separated the primary and secondary MOA based on the observation results. The derived formulation of primary MOA was validated by the measured water-insoluble organics and protein-like organic matter in marine aerosols, which are primarily generated by sea spray. We further investigated the spatial distribution, fluorescence characteristics of MOA, and the driving factors of MOA formation over the WPO. Our results provide an easy observation-based approach to divide the primary and secondary MOA based on the aerosol components and seawater *Chl-a*, as well as an observation-based parameterization of the primary MOA for further improving the parameterization of sea spray organic aerosols in large-scale models.

2 Materials and Methods

2.1 Cruises and sample collection

Two shipboard cruise observations were conducted over the West Pacific Ocean (Fig. 1). Cruise I was conducted in spring during 19 Feb.−9 April, 2022 on the R/V *KeXue* research vessel, and Cruise II was conducted in summer during 19 June−30 July, 2022 onboard of the R/V *Dongfanghong 3* research vessel. High-volume particle samplers (Qingdao Genstar Electronic Technology, China) were placed on the upper deck of the ship to collect the total suspended particles (TSP) and $PM_{2.5}$ (particles with a diameter of <2.5 μm) samples in marine atmospheres. To avoid the contamination of ship exhausts, the aerosol samplers were placed upwind on the foredeck of the ship. The quartz fiber filters were pre-baked at 500°C for 6 h before sample collection. The field blank aerosol sample was collected during each cruise.

Surface seawater samples were collected by a CTD (conductivity-temperature-depth) assembly (Seabird911). The concentration of the in-situ seawater *Chl-a* was measured using a fluorescence spectrophotometer (F-4700, Hitachi, Japan) (Wang et al., 2023a). Surface *Chl-a* concentrations were also obtained based on the satellite-derived data (Siemer et al., 2021; Tuchen et al., 2023). The satellite-derived *Chl-a* data were provided by Copernicus Marine Environmental Monitoring Service (CMEMS) with a spatial resolution of 4 km and a monthly temporal resolution (https://marine.copernicus.eu/). Here, we utilized the satellite-derived *Chl-a* data during March and June 2022 to support our conclusion. The concentration of soluble organic carbon in the seawater was measured by a total organic carbon (TOC) analyzer (TOC-L, Shimadzu, Japan). Air temperature and wind speed were monitored by the shipborne meteorological station. Surface net solar radiation (SSR) data were obtained from the hourly data of the ECMWF Reanalysis v5 (ERA5) product (Hersbach et al., 2020), with a spatial resolution of 0.25°. The 24-hr backward trajectories of air masses (Fig. S1) originating at 500 m above the ground level were calculated along the observation cruises every 24 hr using the HYSPLIT model (Version 5.2.1, NOAA).

2.2 Aerosol chemical composition analysis

An aliquot of the filter sample was extracted by Milli-Q water (>18.2 MΩ·cm) in ultrasonication, and filtered through 0.22 μm PTFE filters. The extracted solutions were analyzed by ion chromatograph systems (ICS-Aquion and ICS-2100 DIONEX) to obtain the concentrations of water-soluble inorganic ions (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^- and SO_4^{2-}) and methanesulfonic acid (MSA). The WSOC in the aerosol samples was measured by the TOC analyzer (TOC-L, Shimadzu, Japan). Organic carbon (OC) and elemental carbon (EC) were analyzed using a Sunset Laboratory thermal/optical carbon analyzer. Concentration of the water-insoluble organic carbon (WIOC) was calculated by the difference between OC and WSOC concentrations in each sample. The mass concentration of organic aerosols was calculated by multiplying OC by a conversion factor 1.6 (Wang et al., 2023b). The OM/OC conversion factor (1.6) was selected based on previous observation results of marine organic aerosols. Over the North Atlantic, an OM/OC mass ratio of 1.8 was adopted for WSOC based on the speciation of WSOC performed on the samples, and a conversion factor of 1.2 was applied for WIOC (Cavalli, 2004a). An average OM/OC ratio of 1.75 was observed in the submicron organic aerosol samples over the Atlantic Ocean (Huang et al., 2018). A higher proportion of water-soluble secondary organic aerosols (SOA), with higher OM/OC ratios than primary MOA, was observed in Huang et al. (2018) than in this study. Here, the proportions of WIOC are comparable to (summer cruise) or higher than (spring cruise) those of WSOC, and thus an OM/OC ratio of 1.6 was selected here. The mass concentrations of $PM_{2.5}$ or TSP were obtained by summing the measured OM, EC, and water-soluble ions in each aerosol sample. The aerosol samples with EC> 0.2 μgC m^{-3} might be influenced by the ship exhausts (Lawler et al., 2020), which thus were excluded in our discussion. A total of 14 sets of aerosol samples during Cruise I and 17 sets of samples during Cruise II would be used for further discussion in this work.

2.3 Fluorescence spectra analysis

Filter aerosol samples were extracted by methanol and filtered through a 0.22 μm PTFE syringe filter. The methanol-extracted solutions were measured by a fluorescence spectrometer (F98, Lengguang Technology, China) to obtain the excitation (Ex) and emission (Em) spectra of MOA. Excitation−emission spectra were scanned within 200−600 nm using a 1 cm optical path length. Pre-processing of the fluorescence spectra data included instrument correction, inner filter correction, Raman and scattering removal, and blank subtraction, which was conducted according to Stedmon and Bro (2008) and Murphy et al. (2013). Fluorescent components in MOA were identified by excitation-emission matrix-parallel factor (EEM-PARAFAC) analysis (Murphy et al., 2013; Stedmon and Bro, 2008). The fluorescence intensity was reported using the unit of RU L^{-1} m^{-3} after considering the extracted solution volume and air volume of each sample (Fu et al., 2015).

3 Results and Discussion

3.1 Overview of marine organic aerosols during the cruises

The concentrations of the water-soluble ions and carbonaceous aerosols in the fine particles ($PM_{2.5}$) along the cruises are presented in Fig. 1. The average OC concentration in $PM_{2.5}$ was 0.67 μgC m^{-3} (0.21−2.18 μgC m^{-3}) during the spring observation and 0.54 μgC m^{-3} (0.12−1.42 μgC m^{-3}) during the summer observation. The EC concentrations were 0.066 ± 0.056 μgC m^{-3} and 0.055 ± 0.052 μgC m^{-3} during the spring and the summer observations, much lower than those observed over coastal areas typically influenced by continental outflows (Sahu et al., 2009; Zhang et al., 2025). The observed OC concentrations during our cruises were comparable to previous studies over the North Pacific Ocean (0.5−0.7 μgC m^{-3}), and lower than those observed at an island in the West Pacific Ocean (1.7±1.0 μgC m^{-3}) (Hoque et al., 2015; Hoque et al., 2017; Kunwar and Kawamura, 2014). Organic matters were the dominant components in the fine particles, which respectively contributed 18%−75% (40% on average) and 13%−74% (48% on average) of the $PM_{2.5}$ mass in spring and summer. This is consistent with previous findings that the organic fractions were dominant in the submicron marine aerosols (Facchini et al., 2008; O'dowd et al., 2004). Film drops could efficiently transfer hydrophobic organic compounds enriched in the air−water interface into the submicron aerosols, which explained the size-selective enrichment of organics in marine aerosols (Cochran et al., 2016b; Prather et al., 2013; Quinn et al., 2015a; Wang et al., 2017). The mass concentrations of $PM_{2.5}$ were calculated by summing the measured OM, EC, and water-soluble ions. Metal elements were not measured in this study, which contributed <3.5% of the marine aerosol mass concentration over the East China Sea (Hsu et al., 2010). Without considering the metal elements, we may overestimate the organic proportion in marine aerosols. During the sampling, positive artifacts of OC may exist due to the absorption of gaseous organic vapor on the filters, and negative artifacts may exist due to the evaporation of volatile organic compounds (Huebert and Charlson, 2000). The OC concentration was measured using thermal-optical analysis. Quantification uncertainty may be introduced due to the formation of pyrolyzed OC, which complicates the accurate determination of the OC/EC split point (Cao et al., 2025; Chow et al., 2004).

Taking the spring observation as an example, the OC mass in most $PM_{2.5}$ samples was roughly equal to that in the corresponding TSP samples (Fig. S2), which were simultaneously collected using two aerosol samplers during the cruise. The campaign-averaged OC concentrations were comparable in the $PM_{2.5}$ (0.67 μgC m^{-3}) and the TSP (0.69 μgC m^{-3}) samples. Thus, our further discussion on marine organic aerosols would focus on the results obtained from the $PM_{2.5}$ samples.

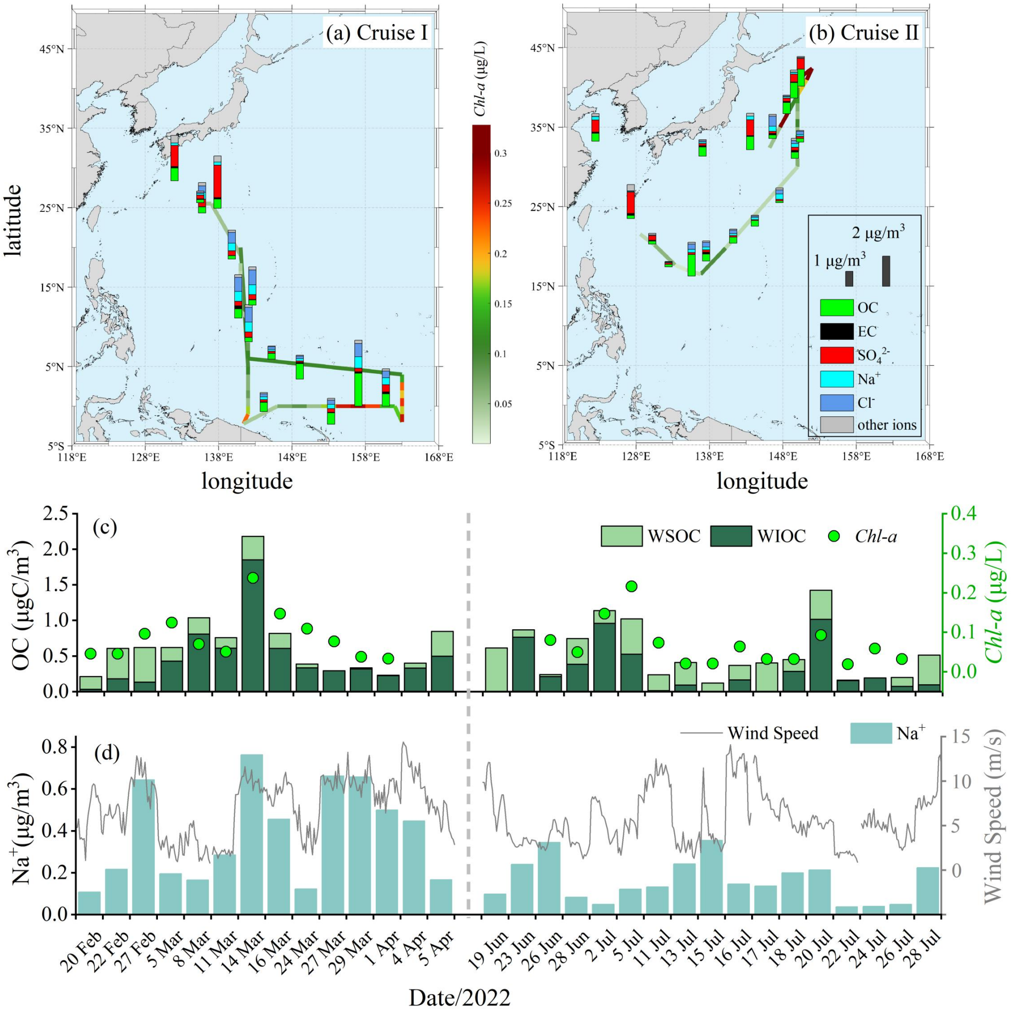

Figure 1 Spatial distributions of water-soluble ions and carbonaceous aerosols (organic carbon, OC, and elemental carbon, EC) in the $PM_{2.5}$ samples during (a) Cruise I conducted during spring, and (b) Cruise II conducted during summer. Time series of (c) the water-soluble OC (WSOC), water-insoluble OC (WIOC), and *Chl-a*, and (d) Na^+ concentration in aerosol samples and the wind speed during the cruises. In panels (a) and (b), the ship route is colored by the concentration of seawater *Chl-a*. Other ions include NO_3^-, NH_4^+, K^+, Mg^{2+} and Ca^{2+}.

The abundance of MOA displayed similar spatial distribution (Fig. 1) and strong or medium correlations with the sea surface *Chl-a* concentration (Cruise I: $r = 0.81$, $p < 0.01$; Cruise II: $r = 0.67$, $p < 0.01$), an indicator of the marine biological activity (Brooks and Thornton, 2018a; Miyazaki et al., 2020). During the biologically active periods, the sea surface layer was enriched in organics, which would be readily transferred into sea spray aerosols through wave breaking and bubble-bursting processes (Cochran et al., 2016a; Cochran et al., 2017; Crocker et al., 2022; Wang et al., 2015). The correlation coefficients between OC and EC were lower (Cruise I: $r=0.48$; Cruise II: $r = 0.17$) than those between OC and seawater *Chl-a*, suggesting that the potential impacts of transported anthropogenic pollutants were limited during the cruises. The air masses were mainly transported from open oceanic regions, and thus the impacts of terrestrial outflows were limited during the cruises (Fig. S1). The OC concentration levels in marine aerosols were higher during the spring cruise than during the summer cruise (Fig. 1c). This was due to the relatively higher phytoplankton activities along the cruise in spring, indicated by the higher seawater *Chl-a* in spring (0.09 ± 0.06 μg L^{-1}) than in summer (0.07 ± 0.05 μg L^{-1}). However, the difference was not significant, with a P value of 0.33. The highest OC concentration occurred on 14 March during the spring cruise, when the highest seawater *Chl-a* (0.24 μg L^{-1}) was observed (Fig. 1). For the samples collected near the equator, the MOA or biogenic VOC precursors could also be transported from coastal oceanic regions of Papua New Guinea and Indonesia with higher marine biological activity and higher isoprene emission fluxes (Cui et al., 2023; Zhang and Gu, 2022). This could be an additional reason for the higher OC level during the spring cruise and the highest OC concentration observed on 14 March.

The observed concentrations of WSOC and WIOC over the open Pacific Ocean were lower than those observed in the atmosphere under severe influence of continental outflows (Sahu et al., 2009; Zhang et al., 2025). Marine organic aerosols were dominated by the water-insoluble fractions, with the WIOC/OC mass ratios of 70 ± 27% in spring and 48 ± 35% in summer (Fig. 1). The proportion of water-soluble organics in MOA over the WPO was lower than that observed over the East Asian marginal seas in autumn (75%), during which severe impacts of continental anthropogenic pollutants were observed (Zhang et al., 2025). The observed WIOC concentrations showed stronger correlations with the seawater *Chl-a* ($r = 0.79$, $p < 0.01$ in spring and $r = 0.63$, $p < 0.05$ in summer) than the correlations between WSOC and *Chl-a* ($r = 0.32$ in spring and $r = 0.42$ in summer). This indicated the closer linkage of marine biological-related organics with the WIOC than with the WSOC in marine aerosols. Marine phytoplankton could produce gel-like aggregates and contribute to extracellular polymer particles, water-insoluble polysaccharide-containing transparent exopolymer, and protein-containing organics, etc. in seawater (Aller et al., 2017; Lawler et al., 2020). These organic substances could be enriched in the surface seawater and then transferred into the atmospheric aerosols within the marine boundary layer. Previous studies suggested that seawater organics injected into aerosol particles through wave breaking or bubble bursting tend to be more hydrophobic and water insoluble (Cavalli, 2004b; Facchini et al., 2008; Miyazaki et al., 2010; O'dowd et al., 2004). Water-soluble organics in marine aerosols are usually related to the aged organic aerosols through long-range transportation or the SOA formed via the oxidation of marine reactive organic gases (Boreddy et al., 2018; De Jonge et al., 2024; Miyazaki et al., 2010). Reactive gaseous precursors of organic aerosols are widely observed over different oceanic regions (Tripathi et al., 2024; Tripathi et al., 2020; Wang et al., 2023a), which contribute to the SOA formation in the marine boundary layer.

3.2 Correlations of MOA with other parameters

The similar variation trends and good correlations between WIOC in marine aerosols and seawater *Chl-a* (Fig. 1, 2) suggested the origins of MOA from seawater through ocean bubble bursting or wave breaking. Seawater *Chl-a* is a widely used oceanic parameter to indicate the marine biological activity or the enrichment of organics in marine aerosols (O'dowd et al., 2004; O'dowd et al., 2008; Rinaldi et al., 2013; Spracklen et al., 2008), which has been employed to predict the organic fraction in marine aerosols. Over the West Pacific Ocean, we observed better correlations between OC or WIOC concentrations and *Chl-a* than those between organic or water-insoluble organic mass fractions and *Chl-a* (Fig. 2a−2d). Some studies reported poor correlations between seawater *Chl-a* and the organic fraction in SSA, and proposed that the organic enrichment is also controlled by physical processes, especially the wind-driven sea spray production processes (De Leeuw et al., 2011; Lewis and Schwartz, 2004; Salter et al., 2014). Seawater *Chl-a* concentration is one of the most important factors driving the variation of organic fraction in the SSA, and they display good correlations when the wind speed does not vary a lot. However, wind speed should be combined with surface *Chl-a* to predict the organic fraction in SSA if the wind speed varies obviously during the observation or simulation periods (Gantt et al., 2011; Grythe et al., 2014). This is due to the influence of wind on the coverage of sea surface microlayer (SML) in the sea surface, which is enriched in organic compounds. For a given chemical composition of seawater, the largest coverage of sea surface by SML and a higher organic fraction in SSA are expected during calm winds. However, the SML would be destructed by mixing into the underlying seawater and the organic fraction in SSA decreased when surface wind exceeded 8 m s^{-1} (Gantt et al., 2011). Thus, researchers usually combine wind speed with surface *Chl-a* to predict the organic fraction in SSA (Gantt et al., 2011; Grythe et al., 2014).

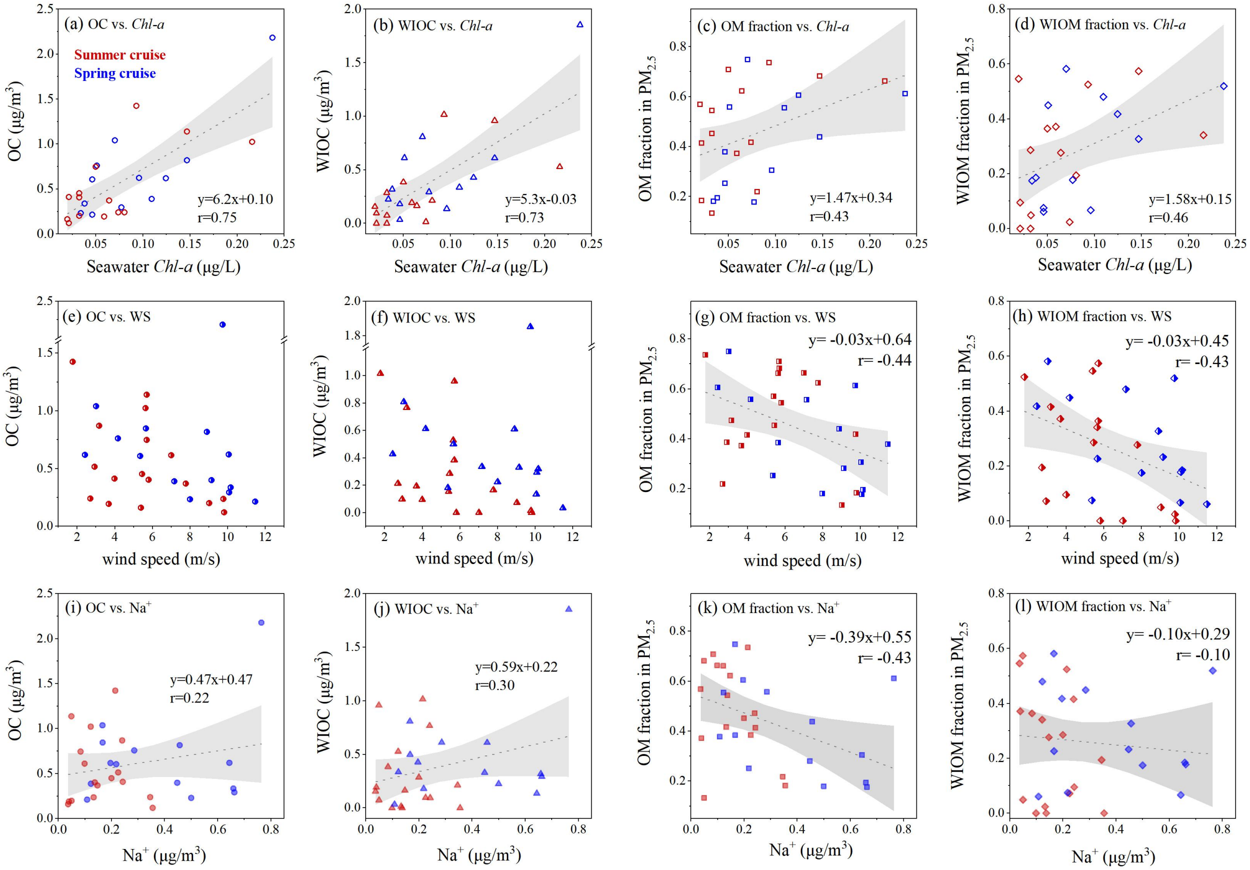

Figure 2 The scatter plots of OC, WIOC concentrations or fractions in marine aerosols as a function of (a-d) surface seawater *Chl-a*, (e-h) wind speed (WS) and (i-l) [Na^+] in $PM_{2.5}$ samples during the two cruises. The data points during the springtime Cruise I and the summertime Cruise II are in blue and red, respectively. The regression line in each panel represents the correlation between the two parameters during the two cruises with a 95% confidence band.

During our cruises over the WPO, the concentrations of OC or WIOC in $PM_{2.5}$ showed a decreasing trend with the increase of wind speed (Fig. 2e, 2f). The organic fraction in marine aerosols displayed a negative correlation with the wind speed (Fig. 2g, 2h). The organic-enriched SML in the sea surface would be destructed under high wind speed conditions, which results in a decrease of organic substances transported into the SSA (Gantt et al., 2011). The concentration or proportion of Na^+ in the marine aerosols showed positive correlations with the wind speed during the observations (Fig. S3). Atmospheric SSA are primarily released as a mixture of inorganic sea salt and organic matters from the ocean surface. We observed weak positive correlations between OC or WIOC and Na^+ concentrations (Fig. 2i, 2j). We proposed that, for the filter-based observation or the samplings with a similar time resolution, Na^+ in fine particles could be used as a better indicator of the overall organic production levels than the wind speed in marine atmospheres. The [Na^+] represents the bulk sea salt abundance generated by wave breaking and bubble bursting, and reflects the overall effects of wind speeds and other meteorological conditions on SSA production during the period of filter sample collection. Russell et al. (2010) found strong correlations between ocean-derived submicron organic aerosols and Na^+ concentrations (Russell et al., 2010). It should be noted that dust storms also transport Na^+ to marine atmospheres, especially over the marginal seas (Zhang et al., 2025). When using Na^+ in marine aerosols as the indicator of SSA production, the Na^+ contributed by transported dust storms should be excluded, especially during dust episodes. For the collected TSP samples, the OC concentrations did not display an obvious correlation with the seawater *Chl-a* (Fig. S4). This is because the dominant production processes of OC and sea salts are different. Organic matters in marine aerosols are enriched in the submicron SSA, which is mainly formed by film drops from bursting bubble-cap films (Wang et al., 2017). In contrast, the majority of the sea salt mass exist in larger supermicron or coarse-mode particles generated by jet drops from the base of bursting bubbles (Wang et al., 2017).

3.3 Estimation of primary and secondary MOA

Based on the correlation analysis of the observed parameters, we proposed a parameterization scheme to separate the marine primarily-emitted OC (MPOC) in the SSA generated through wave breaking or bubble bursting processes and the secondarily formed organic carbon (MSOC) in the marine aerosols over WPO. For a given marine environment condition (a given *Chl-a*, wind speed, sea surface temperature (SST), etc.), the abundance of MPOC should be constant (Gantt et al., 2011). Seawater *Chl-a* concentration is the most important factors driving the variation of organic fraction in the SSA, and has been widely used to estimate the organic fraction in SSA (Gantt et al., 2011; Vignati et al., 2010). For given chemical composition of seawater, the largest organic fraction in SSA is expected during calm winds. An increase in wind speed above 3–4 m s^{-1} will cause a rapid decrease of organic fraction due to the destructing of the SML coverage, and the lowest organic fraction is expected for wind exceeded 8 m s^{-1} (Gantt et al., 2011). Seawater temperature is related to the production efficiency and the number concentrations of SSA (Christiansen et al., 2019). In other words, it is a consistent relation between the MPOC and the sea surface *Chl-a* when the marine environment conditions remain stable. Sea surface *Chl-a* and wind speed have been utilized to parameterize the MPOC in global models (Gantt et al., 2012). Based on the shipboard observations in the present study, the mass ratio of the bulk OC (unit: μgC m^{-3}) versus seawater *Chl-a* (unit: μg L^{-1}) ranged from 3.0×10^{-3} to 1.9×10^{-2}. The increased ratios or fitting line slope of OC versus *Chl-a* relevant to the lowest ones were attributed to the favorable marine conditions for the SSA generation, and the elevated contribution of SOA (e.g., methanesulfonic acid from DMS oxidation, isoprene SOA contributed by phytoplankton emission) (Barnes et al., 2006; De Jonge et al., 2024; Gupta et al., 2025; Ma et al., 2024; Wang et al., 2023b). The MSOC here includes the organic aerosols formed via gas-to-particle conversion of gaseous precursors and oxidation/aging processes of primary OC. The idea is conceptually similar to the classic OC/EC ratio method (Lim and Turpin, 2002; Turpin and Huntzicker, 1995), which uses EC as the tracer and has been widely used to estimate the primary and secondary OC in the continental atmospheres. Here we explored a formulation to estimate the MPOC and MSOC based on the observed OC and Na^+ in marine aerosols and the seawater *Chl-a*:

$$[\mathrm{OC}] = [\mathrm{MPOC}] + [\mathrm{MSOC}] \qquad \text{Eq. 1}$$

$$[\mathrm{MPOC}] = [Chl-a] \times \left(\frac{[\mathrm{OC}]}{[Chl-a]}\right)_{SSA} \qquad \text{Eq. 2}$$

$$[\mathrm{MPOC}] = ([Chl-a] \times [\mathrm{Na}^{+}]^{p}) \times \left(\frac{[\mathrm{OC}]}{[Chl-a] \times [Na^{+}]^{p}}\right)_{SSA} \qquad \text{Eq. 3}$$

where the [OC] is the total OC concentration in the marine aerosols, and [*Chl-a*] is the concentration of surface seawater *Chl-a*. The $\left(\frac{[\mathrm{OC}]}{[Chl-a]}\right)_{SSA}$ in equation (2) represents the ratio of [OC] versus [*Chl-a*] in SSA, and $\left(\frac{[\mathrm{OC}]}{[Chl-a] \times [\mathrm{Na}^{+}]^{p}}\right)_{SSA}$ in equation (3) is [OC] versus ($[Chl\text{-}a] \times [\mathrm{Na}^+]^p$) in the primary SSA.

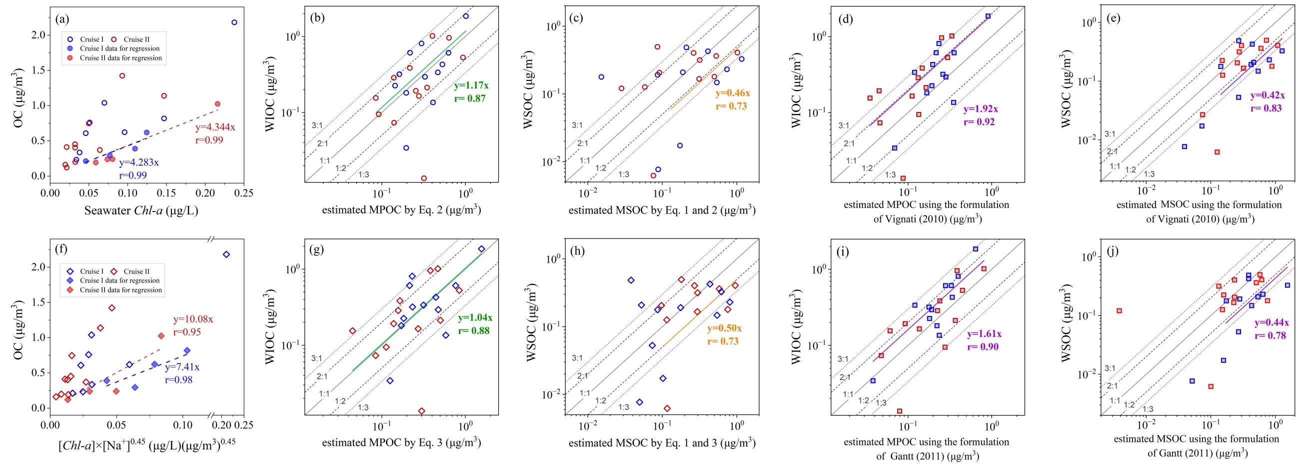

Figure 3 The scatter plots of OC in marine aerosols as a function of (a) seawater [*Chl-a*] and (f) ($[Chl\text{-}a] \times [\mathrm{Na}^+]^{0.45}$) during the two cruises; (b, g) Comparison of WIOC and the estimated MPOC based on the regression in panel (a) and panel (f); (c,

h) Comparison of WSOC and the estimated MSOC; (d, i) Comparison of WIOC and the estimated MPOC, and (e, j) Comparison of WSOC and the estimated MSOC using the formulation of Vignati (2010) and Gantt (2011). The dashed lines in panels (a, f) are the regression line of [OC] and [*Chl-a*] or ([*Chl-a*] × [Na^+]$^{0.45}$) with 0–30% percentile ratios, indicated by solid markers, during Cruise I (blue) and Cruise II (red). The regressions line in panels (b−e, g−j) represent the correlation between WIOC and the estimated MPOC or between WSOC and the estimated MSOC in each panel during the two cruises.

Here, Eq. 2 or Eq. 3 is used to estimate the concentrations of MPOC based on the seawater [*Chl-a*] without or with considering the simultaneously measured Na^+ concentrations as the input parameters. When p=0, Eq. 3 is the same formulation as Eq. 2 without the Na^+ as an input parameter. Based on the correlation analysis, the MOA abundance was mainly driven by the *Chl-a* abundance. We used *Chl-a* as the parameter to predict the concentration of MPOC in Eq. 2. For the samples with the lowest 30% percentile of [OC]/[*Chl-a*] ratios, we proposed that the generation of organic aerosols was dominated by the primary sea spray. The dataset with 0–30% percentile of [OC]/[*Chl-a*] ratios, indicated by the solid markers in Fig. 3a, was used to calculate the fitting line of MPOC versus [*Chl-a*]. This is similar to the classic OC/EC ratio method (Turpin and Huntzicker, 1995). The OC/EC ratios in POC is usually calculated based on the dataset with the lowest 10%−20% percentile OC/EC ratios observed during the campaign, which is then used to separate the POC and SOC in each aerosol sample (Lim and Turpin, 2002; Yu et al., 2021). We used the data with the lowest 30% percentile of [OC]/[*Chl-a*] ratios, considering the number of data points to calculate the fitting curve of MPOC versus [*Chl-a*]. With more data points, the data with the lowest 10%−20% percentile [OC]/[*Chl-a*] ratios could be used to estimate the [MPOC]/[*Chl-a*] ratios, and the estimated MPOC abundance may be a little higher than the results using the lowest 30% percentile data. The ratios of [MPOC]/[*Chl-a*] were 4.28 during cruise I and 4.34 during cruise II (slopes of the fitting lines in Fig. 3a), which were then used to estimate the MPOC during each cruise. The performance of MPOC parameterization was evaluated by comparing the estimated concentrations of MPOC with the WIOC concentrations, which is generally considered as a proxy for MPOC (Fig. 3b). The average mass ratio of WIOC versus MPOC was 1.17 (r=0.87), and 69% of the data points fall within the 1:2 and 2:1 line (Fig. 3b). The shipboard observations suggested that the OC concentrations primarily generated by sea spray over the WPO could be approximately estimated by 4.3×[*Chl-a*] when other related parameters were absent.

A combined parameterization scheme of multiplying seawater [*Chl-a*] by [Na^+]^p was also used to predict the concentration of MPOC (Eq. 3). A weak correlation between OC and Na^+ was observed here (Fig. 2i, 2j), and we thus combined [Na^+] as the input parameter to reflect the variation of the bulk sea spray aerosol abundance. In the scatter plot of OC and ([*Chl-a*] × [Na^+]^p), taking p=0.45 as an example in Fig. 3f, we proposed that the generation of organic matters were dominated by the primary sea spray in the samples with the lowest 30% percentile of [OC]/([*Chl-a*] × [Na^+]^p) ratios. The dataset with 0–30% percentile of [OC]/([*Chl-a*] ×[Na^+]$^{0.45}$) ratios, indicated by the solid markers in Fig. 3f, was used to calculate the fitting line of MPOC versus [OC]/([*Chl-a*] ×[Na^+]$^{0.45}$). The fitting line was then employed to estimate the MPOC in other marine aerosol samples based on the seawater *Chl-a* and the aerosol Na^+ concentrations. In each sample, the increased OC concentration relevant to the MPOC fitting line is attributed to the additional contribution by MSOC.

We compared the estimated MPOC and the measured WIOC to evaluate the performance of the MPOC parameterization and determine the p value in equation 3. Both the correlation coefficients (r) and the slopes of the fitting line between WIOC and estimated MPOC are used to evaluate the performance of different MPOC parameterization approaches. It means that the estimated MPOC shows a similar variation trend to WIOC if with a r value closer to 1, and a good comparison with the WIOC mass concentrations if with a fitting line slope closer to 1. We tested the performance of the MPOC formulation when changing the *p* value from 0−1, with an interval of 0.05. The variations of the fitting line slopes and correlation coefficients of WIOC and MPOC are shown in Fig. S3. When using a *p* value of 0.35–0.65, the estimated MPOC matched well with WIOC concentrations, with the fitting line slopes of 1.03–1.05 and the r values of 0.86–0.88. When using p=0.45, both the fitting line slope (1.036) and r value (0.88) suggested an overall better performance than using other *p* values (Fig. S5, 3g). Without the [Na^+] as an input parameter, the fitting line slope and r of WIOC and MPOC were respectively 1.17 and 0.87 (Fig. 3b), suggesting an underestimation of MPOC. In further analysis, we employed Eq. 3 with *p*=0.45 to estimate the MPOC. A total of 58% of the estimated data points fell within the WIOC/MPOC 2:1 and 1:2 lines (Fig. 3g), and 73% fell within the 3:1 and 1:3 lines, during the two cruises. The estimated MSOC matched better with the WSOC in the marine aerosols when using a combination of [*Chl-a*] and [Na^+] (equations 1 and 3) as the input parameters and considering the variation of sea spray aerosols (Fig. 3c, 3h). Based on equations 1 and 3, the estimated MSOC concentrations in half of the samples fall within the WSOC/MSOC 3:1 and 1:3 lines, and the fitting line slope (0.50) was closer to 1 (Fig. 3h). Using equations 1 and 2, the fitting line slope of WSOC and estimated MSOC was 0.46, and 46% of the estimated MSOC concentrations fall within the WSOC/MSOC 3:1 and 1:3 lines (Fig. 3c). It is noted that, based on the shipboard in-situ observation, we cannot exclude the potential impacts of gaseous precursors or aged organic aerosols long-range transported from terrestrial environments, which were mostly in the MSOC fraction. The organic aerosols transported from terrestrial environments were secondary or aged organic aerosols, and tend to be water-soluble organic compounds (Boreddy et al., 2018; De Jonge et al., 2024; Miyazaki et al., 2010). Based on the air mass back trajectories (Fig. S1) and the weak correlations between OC and EC stated in section 3.1, the impacts of transported continental outflows were limited during the cruises.

The MPOC was also estimated using the formulations in literatures (Gantt et al., 2011; Vignati et al., 2010) based on the observed seawater *Chl-a*, OC and Na^+ in aerosols as well as the wind speed observed during the cruises over the WPO (Fig. 3d, 3i). Vignati et al (2010) estimated the organic mass fraction in sea spray aerosol (OM_{SSA}) using seawater [*Chl-a*]: $\%OM_{SSA} = 43.5 \times [Chl\text{-}a](\mathrm{mg\ m^{-3}}) + 13.805$. Gantt et al (2011) predicted the OM_{SSA} using a combination of [*Chl-a*] and 10 m wind speed (U_{10}): $OM_{SSA}(\mathrm{Chl-a}, U_{10}) = \frac{OM_{SSA}^{max}}{1+\exp\left(-2.63[Chl-a]+0.18U_{10}\right)}$, where OM_{SSA}^{max} is the maximum OM_{SSA} observed during the cruises. The estimated MPOC displayed good correlations with the observed WIOC. However, the abundance of MPOC was underestimated approximately by 38%−48% through comparing with the WIOC concentrations (Fig. 3d, 3i). The estimated MSOC using the parameterizations from Gantt et al. (2011) or Vignati et al. (2010) showed similar variation trends to the WSOC in the collected aerosols samples. The comparison of the estimated MSOC and the WSOC concentrations using formulations in literatures (slopes in Fig. 3e, 3j), however, were not as good as those estimated in this study (slopes in Fig. 3h). The MPOC source functions in Gantt et al. (2011) and Vignati et al. (2010) were proposed based on the observation over the North Atlantic, which has been widely employed in large-scale models. These parameterizations perform well to trace the variation trends of MPOC. However, they might lead to an underestimation of the primary MOA over the West Pacific Ocean. This is mainly due to different seawater compositions, marine environment or atmospheric meteorological conditions in the North Atlantic and the West Pacific Oceans, which result in different quantitative relations between seawater *Chl-a* and MPOC in these oceanic regions. What's more, the seawater *Chl-a* was determined using the spatial average of the satellite-derived *Chl-a* concentrations in Gantt et al. (2011). This could be an additional reason for the different parameterizations between *Chl-a* and MPOC compared with the results based on the in-suit measured *Chl-a* in this work. The results highlight different quantitative relations of MPOC with seawater *Chl-a* and other parameters in different areas, which are needed to be provided through in-situ observations across different oceanic regions and to constrain in global models.

3.4 Spatial distribution and driving factors of primary and secondary MOA

Based on the validated formulation, concentrations of MPOC and MSOC in the marine aerosols over the WPO are estimated. Here we employed Eq. 3, with p=0.45, for the estimation of MPOC. The concentrations and relative contributions of MPOC and MSOC along the latitude are shown in Fig. 4. The estimated MPOC was respectively 0.43 ± 0.40 and 0.24 ± 0.21 μgC m^{-3}, averagely accounting for 66% ± 27% and 56% ± 30% of the total OC in marine aerosols, during the springtime Cruise I and the summertime Cruise II. The dominant contribution of MOA by the marine fresh carbon pool was also observed during the Arctic cruises, during which the MPOC contributed 80% of the carbonaceous fraction based on the stable carbon isotopic signature (Gu et al., 2023). The estimated MSOC concentrations were comparable in spring (0.25 ± 0.28 μgC m^{-3}) and in summer (0.27 ± 0.30 μgC m^{-3}) over the WPO. The SOA fraction among the total organic aerosols was higher during the summer cruise (44% on average) than during the spring (34% on average).

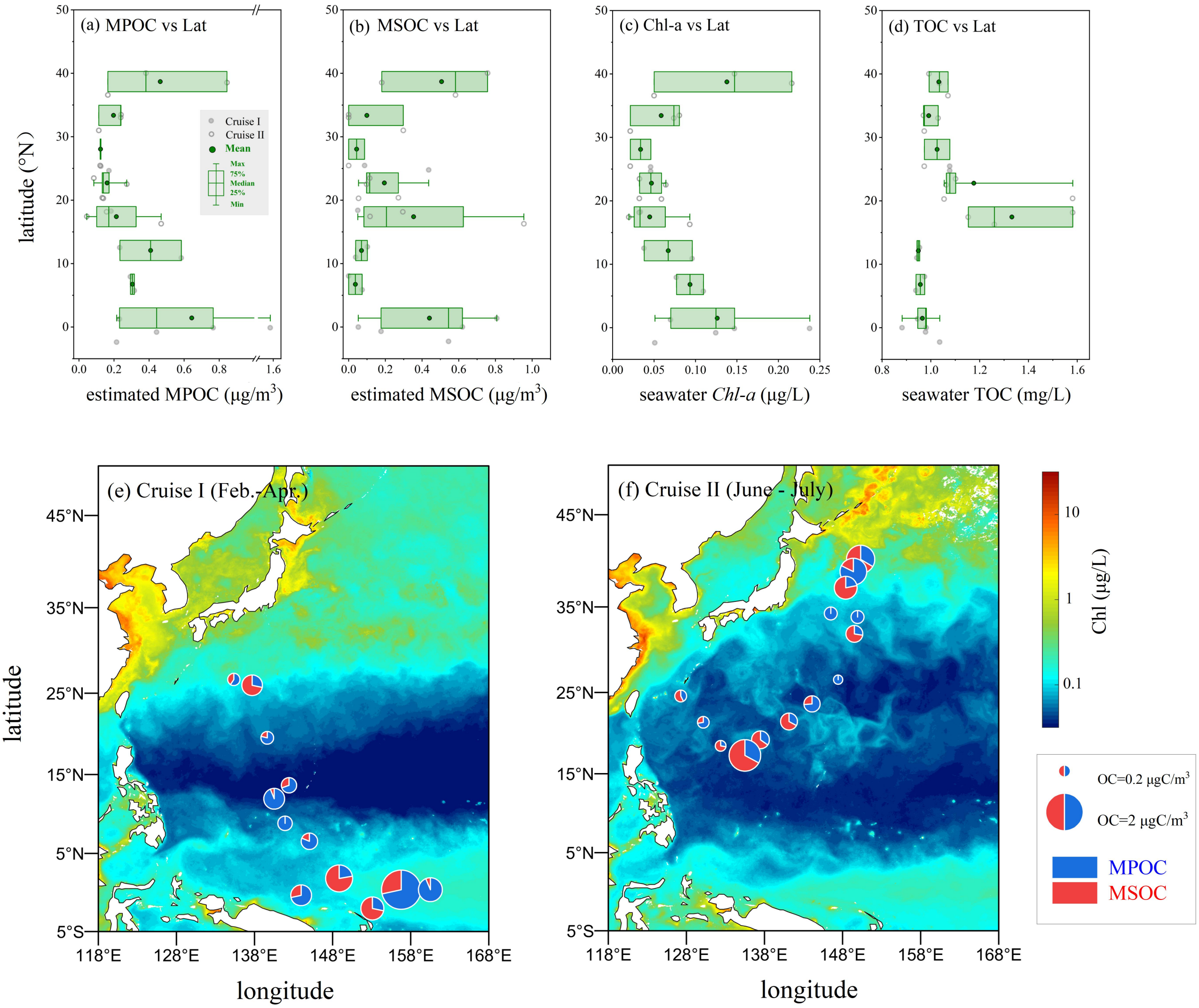

Figure 4 The variations of (a) estimated MPOC, (b) MSOC, (c) seawater *Chl-a* and (d) TOC along the latitude during the two cruises over the WPO. Spatial distributions of the estimated MPOC and MSOC during (e) the springtime Cruise I, and (f) the summertime Cruise II. Ocean is coloured by the sea surface *Chl-a* concentrations in March (panel e) and July (panel f), 2022. The marker size in panels (e, f) represents the observed OC concentration in each sample.

Both MPOC and MSOC displayed high concentrations over the oceanic regions among 5°S–5°N (0.64 ± 0.56 and 0.44 ± 0.32 μgC m^{-3}) and 35°N–40°N (0.46 ± 0.35 and 0.51 ± 0.30 μgC m^{-3}), which were consistent to the spatial distribution of the sea surface *Chl-a* (Fig. 4). In contrast to the findings over the North Atlantic that plankton had little impact on the chemical compositions of SSA (e.g., organic mass fraction) (Bates et al., 2020), we observed a positive correlation between MOA and seawater *Chl-a* and the driving effects of surface *Chl-a* on the abundance of primary MOA over the WPO (Fig. 2, 4). In addition, the observation areas within 35°N–40°N were the Kuroshio Oyashio Extension (KOE) region, where the nutrient enrichment driven by upwelling favored the phytoplankton growth and resulted in elevated seawater *Chl-a* levels (Wang et al., 2023a). The extreme physical disturbance in the KOE further promoted the sea spray-generated organics from seawater as well as the production of VOCs from phytoplankton. Based on the air mass back trajectories (Fig. S1), the impacts of transported terrestrial outflows were limited among the observation regions. Marine organic aerosols or biogenic VOC precursors could also be transported from coastal oceanic regions with higher *Chl-a* levels and higher isoprene emission fluxes (Cui et al., 2023; Zhang and Gu, 2022), which could be an additional reason for the higher MOA concentrations within 5°S–5°N and 35°N–40°N.

The MSOC also displayed a peak over the areas among 15°N–20°N (0.35 ± 0.41 μgC m^{-3}, Fig. 4b), which could be attributed to the additional contribution by abiotic VOC precursors from the photochemical production in the sea surface microlayer and their further oxidation in marine boundary layer (Bruggemann et al., 2018). Previous studies suggested that interfacial photochemical degradation of dissolved organic matters in seawater could be an important source of marine VOCs (e.g., isoprene) on a global scale (Bruggemann et al., 2018; Cui et al., 2023; Wang et al., 2023a; Yu and Li, 2021). For remote oceanic regions with high solar radiation but low biological activities, interfacial photochemistry of surface organics could be a major source of abiotic VOCs in the marine boundary layer (Bruggemann et al., 2018; Cui et al., 2023). Higher concentration levels of the surface seawater TOC concentrations were observed along the summer cruise within 15°N–20°N (Fig. 4d). The strong solar radiation during the summertime (19 June- 30 July) Cruise II, as shown in Fig. S6, favored the photochemical VOC production and the SOA formation in marine atmospheres. During the summer cruise, the estimated MSOC/OC ratios over the oceanic regions of 15°N–20°N were 65%−72%, and the SOA formation drove the elevation of MOA concentrations over this area during summer (Fig. 4f).

Aerosol samples were collected among 15°N−30°N during both the spring and the summer cruises, which were compared to elaborate the seasonal difference. The variations of the estimated MPOC and MSOC along the latitude are shown in Fig. S7, S8. Among the observation region within 15°N−30°N, the average MPOC was comparable in spring (0.16 μgC m^{-3}) and summer (0.18 μgC m^{-3}), with the average *Chl-a* concentration 0.042 and 0.044 μg L^{-1}, respectively. Among the oceanic regions with similar concentrations of seawater *Chl-a*, the MPOC abundance in marine aerosols was comparable without seasonal difference. Among 15°N−30°N, the elevation of MPOC concentrations was consistent with the elevated seawater *Chl-a* concentration without seasonal difference (Fig. S7a). This is consistent with the finding that marine biogenic activities drive the MPOC production. The average MSOC concentration was 0.24 μgC m^{-3} within 15°N−30°N in summer, higher than that in spring (0.19 μgC m^{-3}). The elevated MSOC was driven by the increase of seawater TOC concentrations (Fig. S8b). What's more, the stronger solar radiation in summer (Fig. S6) favored the photochemical VOC production in SML, their further photo-oxidation reactions, and the MSOC formation in the atmosphere.

3.5 Fluorescence characteristics of MOA

The fluorescence spectrum of MOA was analyzed to gain a further understanding on the composition characteristics of MOA over the WPO. Based on the EEM PARAFAC analysis, three fluorescent components were identified in marine organic aerosols during each cruise observation (Fig. 5, S4). Similar fluorescence components were resolved during the Cruise I (Fig. 5a-5c) and Cruise II (Fig. 5d-5f). Each component was named based on the fluorescence characteristics and the temporal variation of the fluorescent intensity. Component 1 (C1) shows a peak (Ex/Em = 285/307 nm) identical to the protein-like substances (PRLIS) (Chen et al., 2016b). The PRLIS are enriched in the surface seawater and could be injected into SSA via bubble bursting (Miyazaki et al., 2018a; Santander et al., 2021). The similar variations of C1 intensity and Na^+ in the marine aerosols, especially during the summer observation (Fig. S9), suggested the origins of PRLIS from marine biological materials (Fu et al., 2015; Santander et al., 2022). Thus, C1 was designated as marine PRLIS. Component 2 (C2) has a peak Ex/Em = 320-335/389 nm (Fig. 5), which is related to terrestrial humic-like substances (HULIS) (Chen et al., 2016b). Component 3 (C3) displayed the fluorescence characteristics of oxygenated HULIS, with a peak Ex/Em = 365-370/450-455 nm (Fig. 5). The intensities of C2 and C3 showed similar variations to the concentrations of EC and sulfate in marine aerosols (Fig. S9), which indicated their sources related to combustion emission and secondary formation (Tang et al., 2024). Oxygenated HULIS included the secondarily-formed and aged organic aerosols from both terrestrial and marine sources.

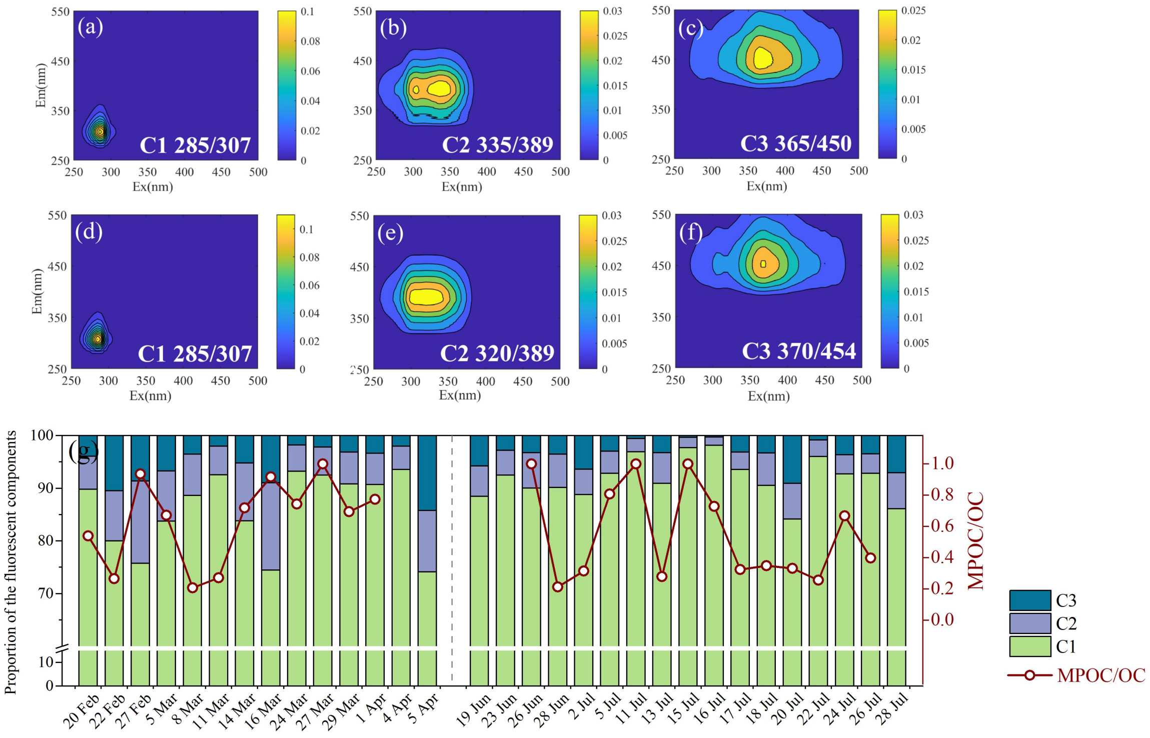

Figure 5 (a-f) The excitation (Ex) and emission (Em) spectra of the identified fluorescent components: C1: marine protein-like substances (PRLIS), C2: terrestrial humic-like substances (HULIS), C3: oxygenated HULIS. (g) Time series of the fluorescent component relative abundances and the MPOC/OC mass ratios in the marine aerosols over the West Pacific Ocean.

The fluorescent components of the MOA were dominated by the PRLIS primarily emitted by the sea spray (C1 shown in Fig. 5a, 5d), which contributed 74%−94% (86% on average) and 84%−98% (92% on average) of the MOA fluorescent intensity during Cruise I and Cruise II, respectively (Fig. 5g). It is noted that the proportion of different fluorescent compounds did not represent their mass contributions, as the florescent efficiency of organic compounds was related to their chemical structures. Organic molecules with substantial conjugation of π-bonds or double bound structures are known to be especially efficient at emitting fluorescence, particularly when N atoms are present (Chen et al., 2016a; Pöhlker et al., 2012). Amino acids, vitamins, and humic-like substances have been identified as efficient fluorophores (Graber and Rudich, 2006; Laskin et al., 2015; Pöhlker et al., 2012). The sea-to-air transfer of phytoplankton-produced protein-containing organics leads to a significant enhancement of fluorescent compounds in SSA (Aller et al., 2017; Lawler et al., 2020; Miyazaki et al., 2018b). The PRLIS, or named protein-like organic matter (PLOM), has been identified as a common component in the oceanic organic matter, and enriched in marine aerosols (Chen et al., 2016b). However, the biogenic SOA (e.g., isoprene oxidation products abundant in marine atmospheres) molecules, without conjugated double bounds, are weakly fluorescent or do not display fluorescent properties (Carlton et al., 2009; Laskin et al., 2015). Thus, the WSOC contributed by biogenic SOA was not included in the detected fluorescent components, and the observed proportions of PRLIS emitted by sea sprays was higher than those of the WIOC mass contribution in the marine aerosols.

During the summertime Cruise II, the fluorescent intensity and the relative contribution of marine-emitted PRLIS (C1) were higher than those during Cruise I. During the summer cruise, the contribution of the marine PRLIS among the total fluorescent organic aerosols displayed a similar variation trend to the mass fraction of the estimated MPOC (Fig. 5g). The variation of the marine PRLIS (C1) intensity was consistent to the seawater *Chl-a* concentration in summer (Fig. S9). This further indicated the dominant contribution of primary MOA in marine organic aerosols, which could be attributed to the marine biological materials and injected into the atmosphere through bubble bursting. The marine biological PRLIS could be related to tryptophan-like or tyrosine-like components as well as the non-nitrogen-containing organic compounds in atmospheric aerosols (Chen et al., 2016b).

4 Summary

In-situ shipboard observations were conducted to investigate the abundance and composition of MOA over the open Pacific Ocean. We proposed a formulation to separate and estimate the primary and secondary MOA based on the seawater *Chl-a* or its combination with Na^+ in marine aerosols. Based on the validated formulation, the estimated MPOC accounted for 56%−66% of the total OC in the marine aerosol samples, which were mostly related to the protein-like substances from seawater biological materials. Both the MPOC and the MSOC displayed peak concentrations among 5°S–5°N and 35°N–

40°N over the West Pacific Ocean. The spatial distribution of MOA along the latitude was driven by the marine biological activities, indicated by the seawater *Chl-a*. For the secondary MOA, high concentrations were also observed over the region of 15°N–20°N, which was attributed to an additional contribution by the secondary oxidation of VOCs generated from the photochemical production of seawater organics.

This study provides a parameterization to estimate the primary and secondary MOA based on the shipboard observation evidence, and highlights the marine biogenically driven MOA formation over the North Pacific Ocean. For the observation studies, our results provide an easy approach to separate the primary and secondary MOA with different chemical natures, based on the seawater *Chl-a* and aerosol components (OC, Na^+). The approach is not dependent on the organic tracers, usually obtained through complex analysis procedures, or limited to the time resolution of sample collection. In previous studies, fractions of organics in marine aerosols have been estimated based on an empirical relationship of satellite-derived oceanic *Chl-a*, or a combination with wind speed and aerosol size distribution (Gantt et al., 2012; Li et al., 2024; Wang et al., 2024). Here, we gain the quantitative relations of primarily-generated marine organic aerosols with sea salts and *Chl-a* based on measurement results of the marine aerosols and seawater. For the modelling studies, the sea salt flux has been better estimated than that of marine organic aerosols in global models (Gantt and Meskhidze, 2013). The MPOC formulation here would help to improve the parameterization of MOA in models and better understand the climate effects of marine aerosols on a global scale.

Data availability

The data is available via https://zenodo.org/records/16831992 (Wang, 2025)

Author contributions

Y.W. designed the research. Y.Y., Y.Z., S.L., H.Z., and S.Y. conducted the measurements. Y.Y. and Y.W analyzed the data. Y. W., Y.Y. and W.X. wrote the manuscript with contributions from all authors.

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Competing interests

The authors declare no conflict of interests.

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Supplementary Information

Biogenically driven marine organic aerosol production over the West Pacific Ocean

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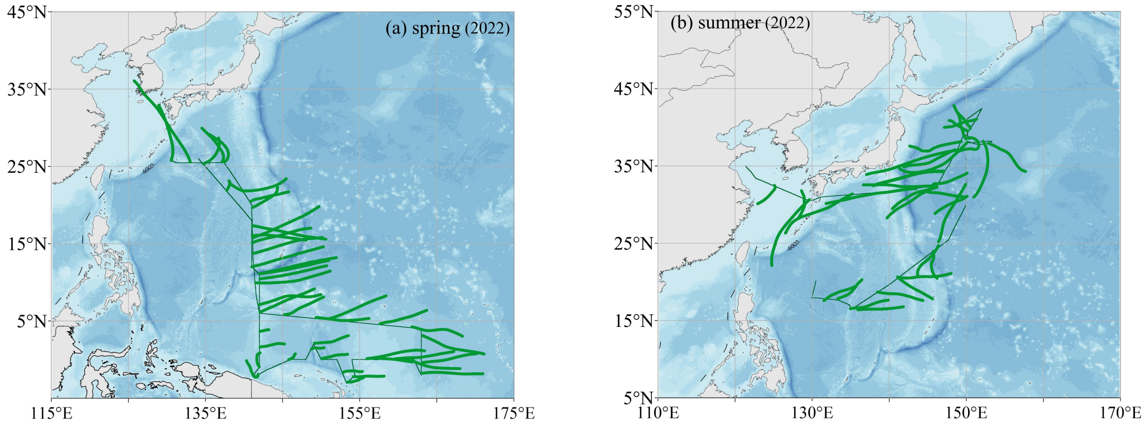

Figure S1 The 24-hr back trajectories of air masses during the cruises in (a) spring and (b) summer.

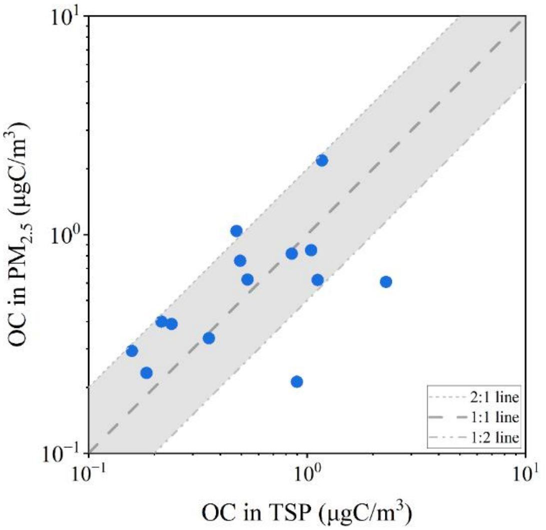

Figure S2 Comparison of the OC concentrations in the $PM_{2.5}$ and the TSP samples during the spring observation (Cruise I)

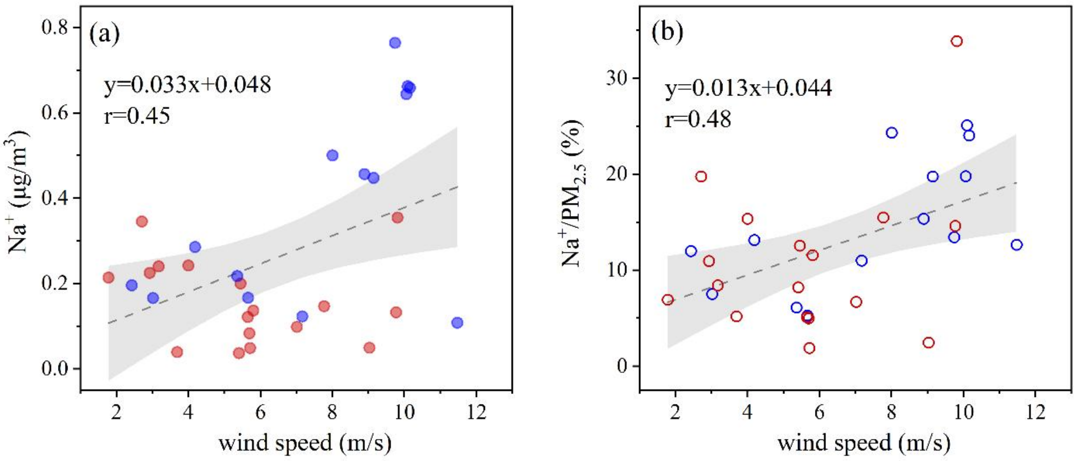

Figure S3 The variation of Na$^+$ concentration and Na$^+$/PM$_{2.5}$ as a function of the wind speed during the cruises. The data obtained during the spring Cruise I is in blue, and the data during the summer Cruise II is in red.

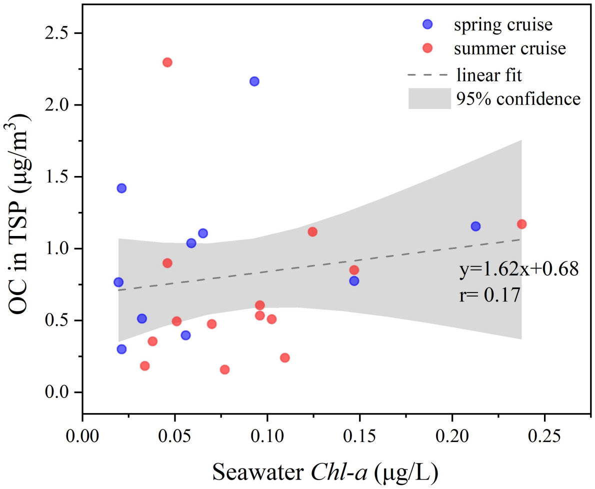

Figure S4 Scatter plot of the OC concentration in the collected TSP samples and seawater *Chl-a* during the cruises.

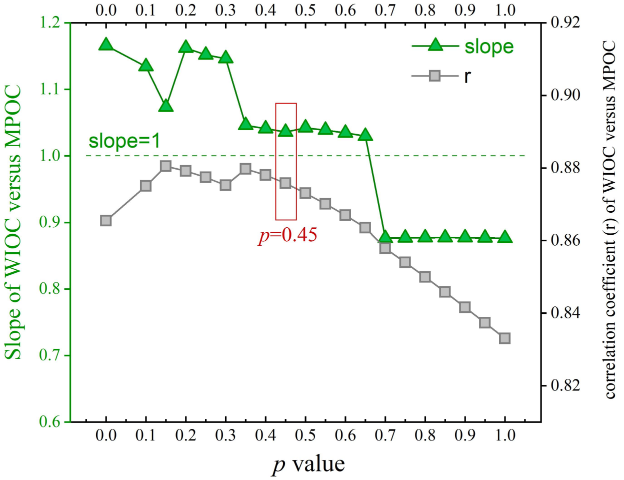

Figure S5 The variations of the fitting line slopes and correlation coefficients (r) of WIOC and estimated MPOC, using Eq. 3 with the p value changing from 0−1.

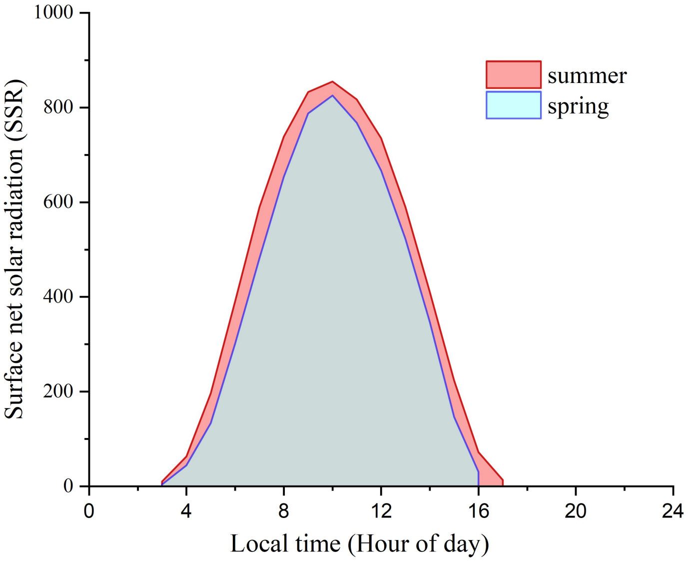

Figure S6 Diurnal variation of the surface net solar radiation (SSR) within the 15°N–20°N during the spring and the summer cruises.

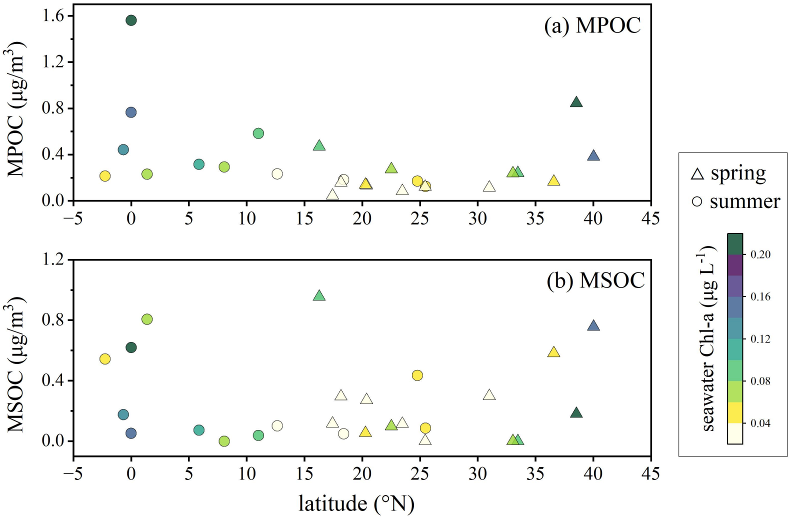

Figure S7 Spatial distribution of the estimated MPOC and MSOC concentrations. The data is colored by the corresponding seawater *Chl-a* concentrations.

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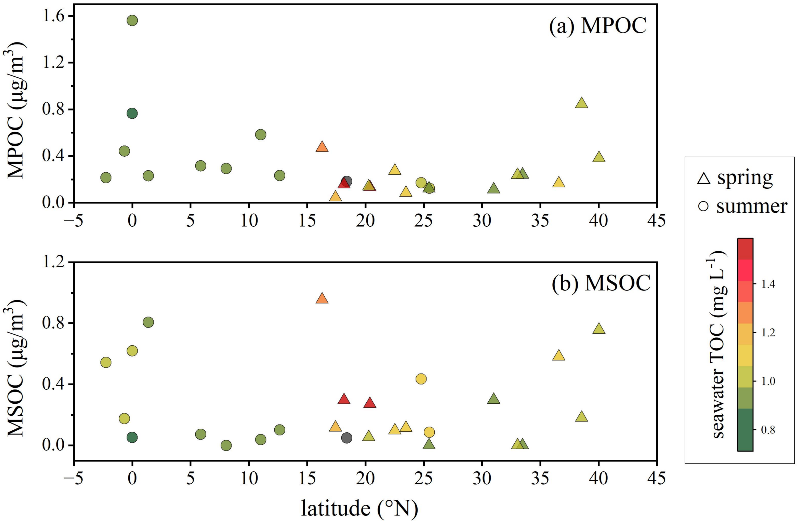

Figure S8 Spatial distribution of the estimated MPOC and MSOC concentrations. The data is colored by the corresponding seawater TOC concentrations.

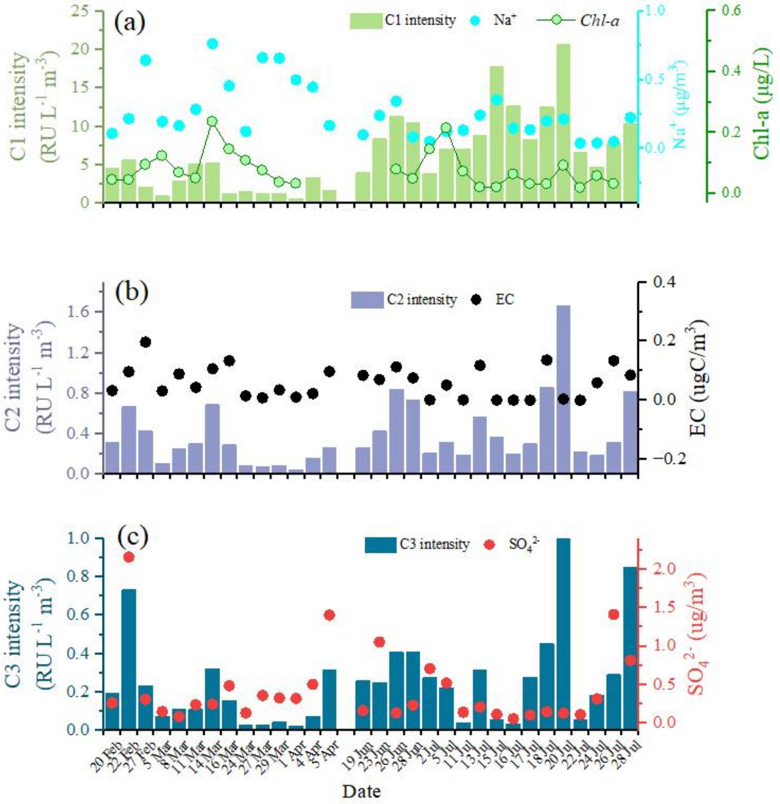

Figure S9 Variations of fluorescence component intensity identified by three-component solutions based on PARAFAC model analysis and related aerosol components: (a) C1, Na^+, and *Chl-a*, (b) C2 and EC, (c) C3 and SO_4^{2-}.