https://doi.org/10.5194/egusphere-2025-3951
Preprint. Discussion started: 28 August 2025

Biogenically driven marine organic aerosol production over the Northwest Pacific Ocean

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https://doi.org/10.5194/egusphere-2025-3951
Preprint. Discussion started: 28 August 2025

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) emitted from marine bubble bursting or secondary organic carbon (MSOC) formed via gas-to-particle conversion remains poorly quantified, which hinders our understanding on the climate effects of marine aerosols. In this work, two shipboard cruises were conducted over the Northwest Pacific Ocean to estimate abundance and compositions of marine organic aerosols. We propose an observation-based approach to quantify the MPOC and MSOC using combined parameterization of the observed Na^+ in fine aerosol particles and the surface chlorophyll-*a*, an indicator of marine biological activity. The parameterization approach was validated through comparing with the water-insoluble organic carbon in the aerosol samples. The estimated MPOC averagely accounted for 56%−66% of the total organic carbon in the collected marine aerosol samples, which was mainly attributed to the protein-like substances transferred into the sea spray aerosols from seawater. Over the Northwest Pacific Ocean, the MPOC and MSOC displayed peak concentrations over the regions 5°S–5°N and 35°N–40°N. 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, 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 chlorophyll-*a* and other parameters are needed based on the 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., 2015). 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 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 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 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, 2018; Quinn and Bates, 2011; Quinn et al., 2015).

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, 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, 2004; 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 regions with high 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). Due to the relationship between sea surface phytoplankton and MOA, 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 of understanding their role in cloud formation (Albert et al., 2012; Brooks and Thornton, 2018).

Observation-based parameterization of primary and secondary MOA is urgently needed to constrain the modeling-results. In this work, two shipboard observations of atmospheric aerosols were conducted from the temperate to tropical regions over the Northwest Pacific Ocean (NPO) 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 from bubble bursting, and separate 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 NPO. 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 Northwest 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) samples 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 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.

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 organic aerosol concentration was calculated by multiplying OC by a conversion factor 1.6 (Wang et al., 2023b). 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. 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. The observed OC concentrations during our cruises were comparable to previous studies over the North Pacific Ocean (Hoque et al., 2015; Hoque et al., 2017; Kunwar and Kawamura, 2014). 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. S1), 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. 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). Thus, our further discussion on marine organic aerosols would focus on the results obtained from the $PM_{2.5}$ samples.

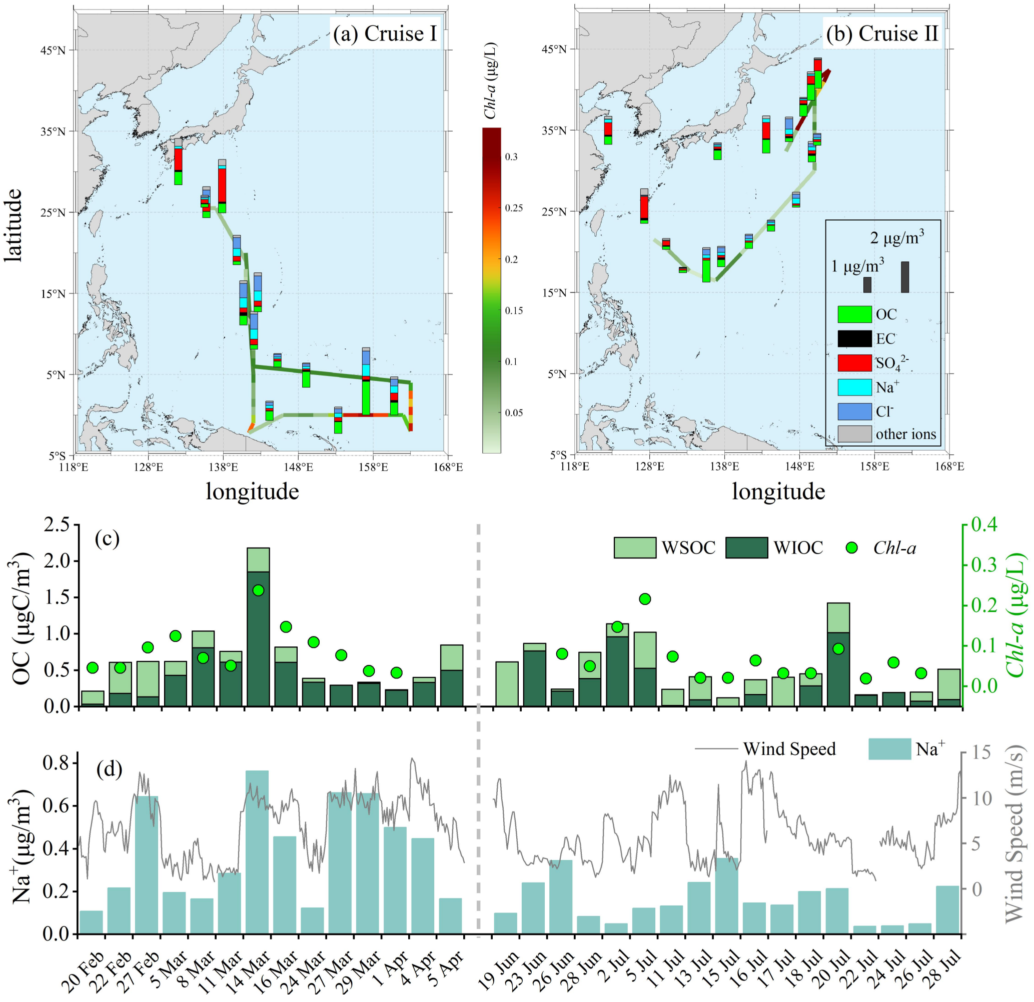

Figure 1 Spatial distributions of water-soluble ions and carbonaceous aerosols (organic carbon, OC, and elemental carbon, EC) in the $PM_{2.5}$ samples during (a) Cruise I conducted during spring, and (b) Cruise II conducted during summer. Time series of (c) the water-soluble OC (WSOC), water-insoluble 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 and strong correlations with the sea surface *Chl-a* concentration (Cruise I: r = 0.81, p < 0.01; Cruise II: r = 0.67, p < 0.01, Fig. 1), an indicator of the marine biological activity (Brooks and Thornton, 2018; 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 bubble-bursting processes (Cochran et al., 2016; 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 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}). 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).

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 WIOC 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 WSPC 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), which could be enriched in the surface seawater and transferred to atmospheric aerosols over the ocean. 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, 2004; 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 secondary organic aerosols (SOA) formed via the oxidation of marine reactive organic gases (Boreddy et al., 2018; De Jonge et al., 2024; Miyazaki et al., 2010).

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 organics through the ocean bubble bursting. Seawater *Chl-a* is a widely used oceanic parameter to indicate the marine biological activity or the enrichment of organic carbon 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. In the present work over the NPO, we observed better correlations between OC or WIOC concentrations and *Chl-a* ($r=0.75$, 0.73, $p<0.01$) than those between organic or water-insoluble organic mass fractions and *Chl-a* ($r=0.43$, 0.46), as shown in 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 the physical processes, especially the wind-driven sea spray production processes (De Leeuw et al., 2011; Lewis and Schwartz, 2004; Salter et al., 2014). 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).

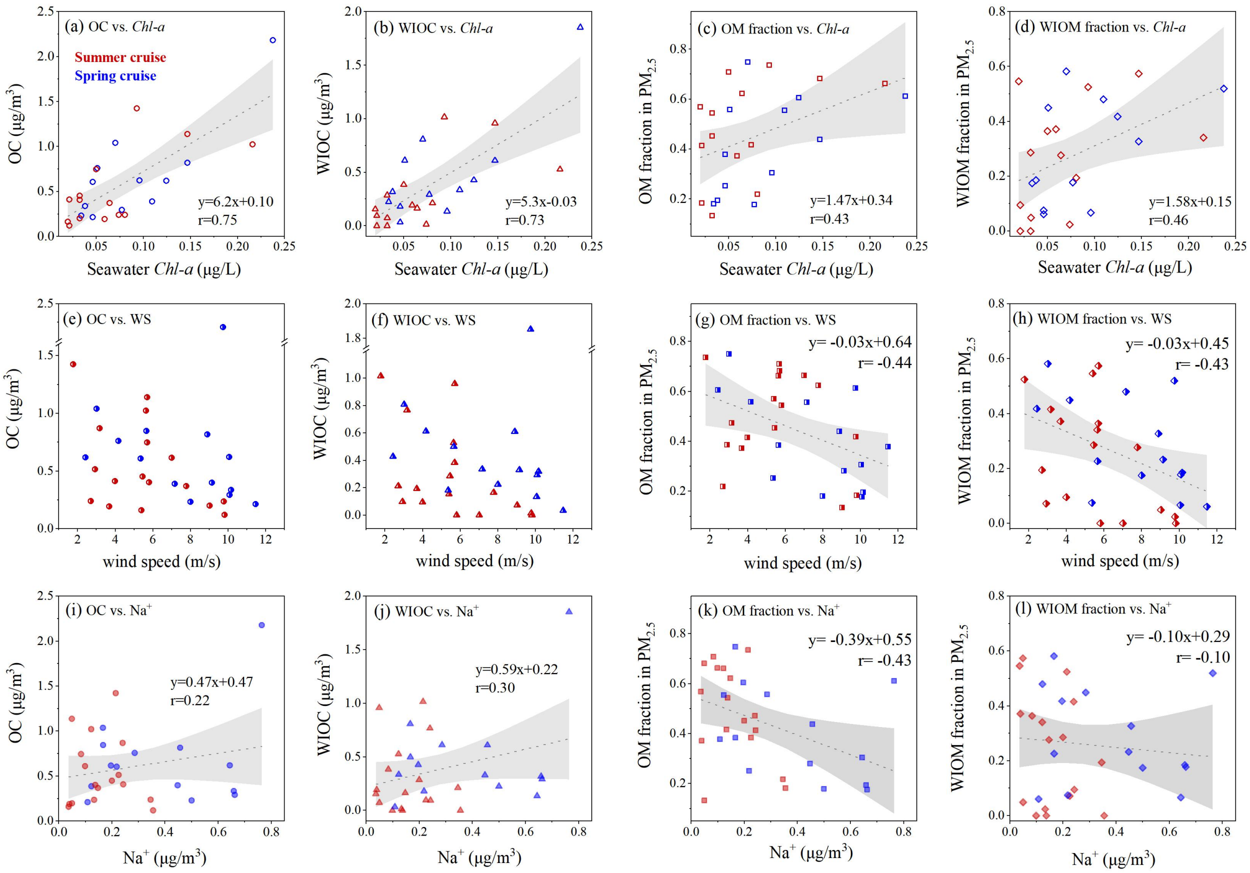

Figure 2 The scatter plots of OC, WIOC concentrations or fractions in marine aerosols as a function of (a-d) surface seawater *Chl-a*, (e-h) wind speed (WS) and (i-l) [Na^+] in $PM_{2.5}$ samples during the two cruises. The data points during the springtime Cruise I and the summertime Cruise II are in 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 NPO, the concentrations of OC or WIOC in $PM_{2.5}$ did not show obvious correlations with the wind speed (Fig. 2e, 2f). The organic fraction in marine aerosols displayed a negative correlation with the wind speed (Fig. 2g, 2h), which was due to the elevated proportions of inorganic sea salts in the marine aerosols under the high-wind speed conditions. The concentration or proportion of Na^+ in the marine aerosols showed positive correlations with the wind speed during the observations (Fig. S2). Higher wind speed would promote sea salts production, and its influence on the MOA generation was not obvious. Atmospheric SSA are primarily released as a mixture of inorganic sea salt and organic matters from 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 a better indicator of the overall SSA production levels than the wind speed in marine atmospheres. The [Na^+] represents the bulk sea salt abundance generated by bubble bursting, and reflects the overall effects of wind speeds and other

https://doi.org/10.5194/egusphere-2025-3951
Preprint. Discussion started: 28 August 2025

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).

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) by bubble bursting and the secondarily formed organic carbon (MSOC) in the marine aerosols over NPO. Under certain marine environment conditions (e.g., *Chl-a*, wind speed, SST etc.), the abundance of MPOC should be constant. 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 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) (De Jonge et al., 2024; Ma et al., 2024; Wang et al., 2023b). 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*:

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

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

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

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

https://doi.org/10.5194/egusphere-2025-3951
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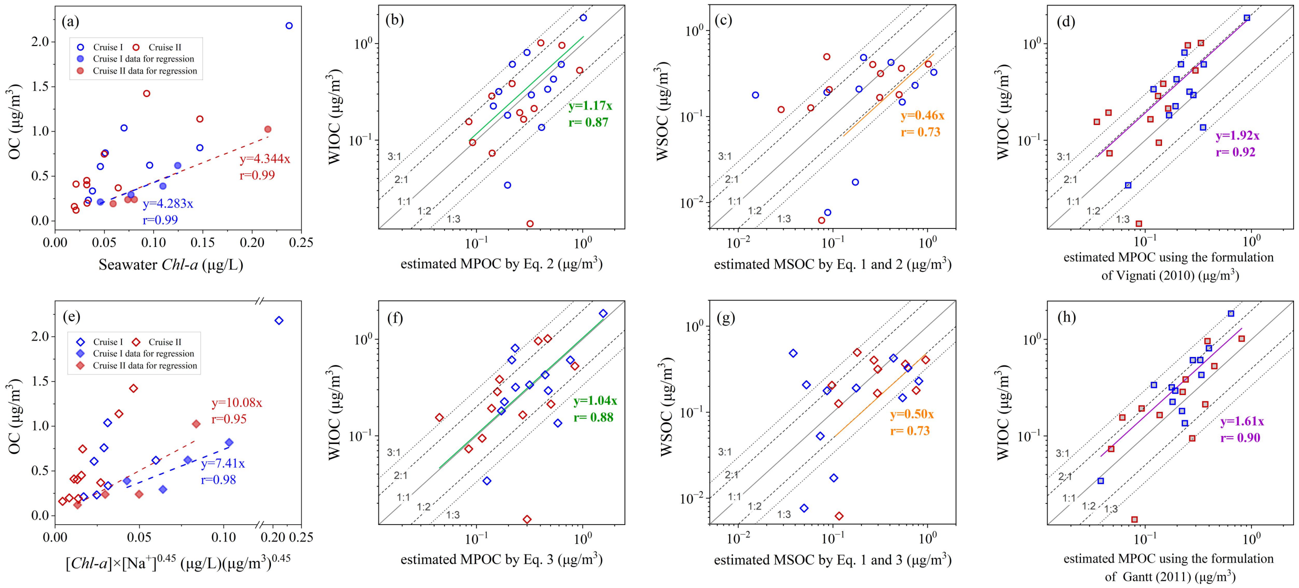


Figure 3 The scatter plots of OC in marine aerosols as a function of (a) seawater [*Chl-a*] and (e) ([*Chl-a*] × [Na^+]$^{0.45}$) during the two cruises. (b, f) Comparison of WIOC and the estimated MPOC based on the regression in panel (a) and panel (e). (c, g) Comparison of WSOC and the estimated MSOC. (d, h) Comparison of WIOC and the estimated MPOC using the formulation of Vignati (2010) and Gantt (2011). The dashed lines in panels (a, e) 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 regression line (green) in panels (b, d) represents the correlation between WIOC and the estimated MPOC during the two cruises. The regression line (orange) in panels (c, f) represents the correlation between WSOC and the estimated MSOC during the two cruises.

Here Eq. 2 or Eq. 3 is used to estimate the concentrations of MPOC generated by bubble bursting, based on the seawater *Chl-a* concentrations without or with 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 above, 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. In the classic OC/EC ratio method, EC is used as the tracer of primary combustion emissions and the OC/EC ratio from the primary emissions is assumed to be constant during the observation (Turpin and Huntzicker, 1995). The OC/EC ratios in POC is usually calculated based on the dataset with the lowest 10%−20% percentile observed OC/EC ratios, which is then used to estimate the POC and SOC in each aerosol sample (Lim and Turpin, 2002; Yu et al., 2021). Here, 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*]. 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 then 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 NPO could be approximately estimated by 4.3×[*Chl-a*] when other related parameters were absent.

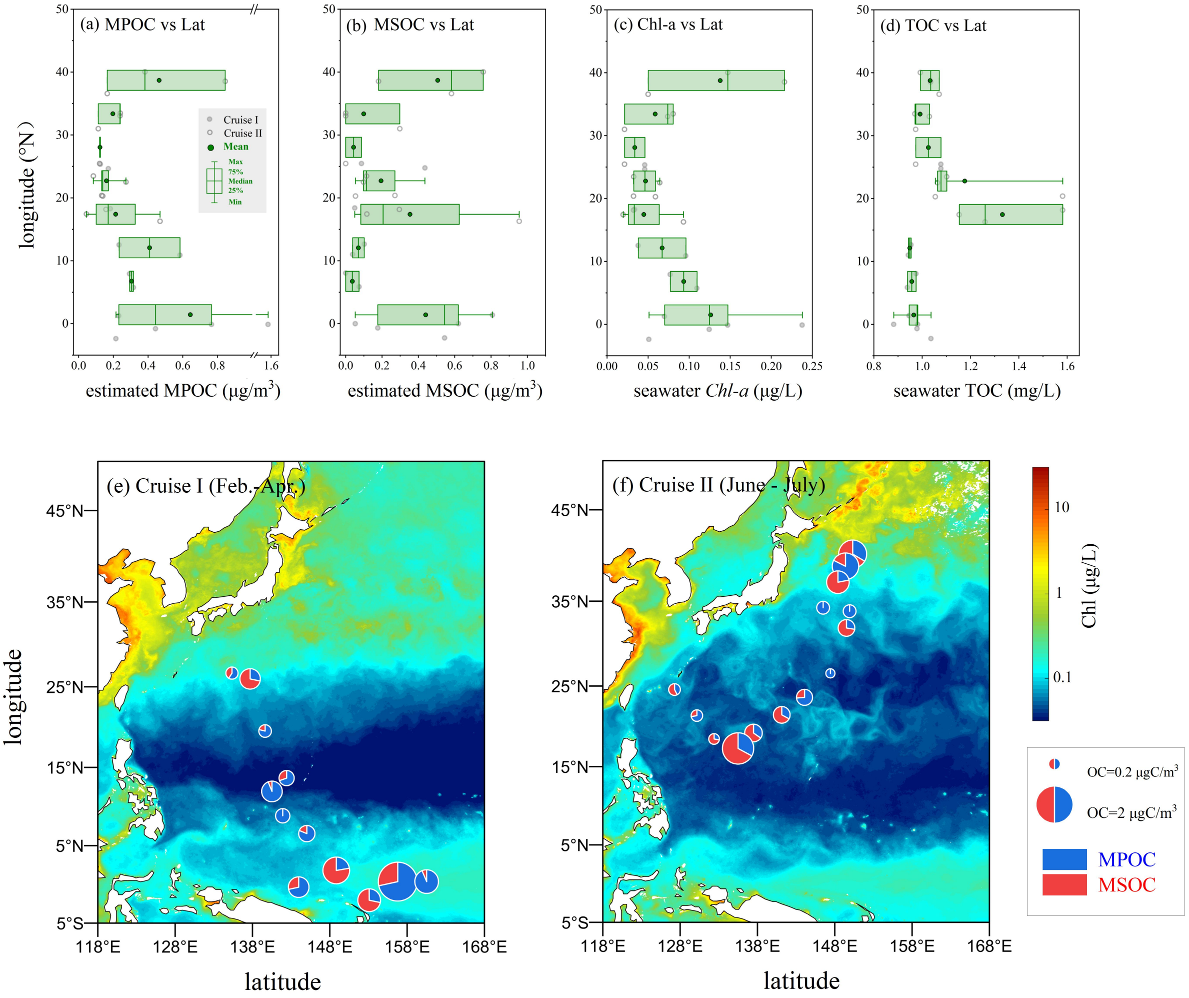
A combined parameterization scheme of multiplying seawater [*Chl-a*] by $[\mathrm{Na}^+]^p$ were also used to predict the concentration of MPOC (Eq. 3). A weak correlation between OC and Na^+ was observed here (Fig. 2i, 2j), and we thus combined $[\mathrm{Na}^+]$ as the input parameter to reflect the variation of the bulk sea spray aerosol abundance. In the scatter plot of OC and ([*Chl-a*] × $[\mathrm{Na}^+]^p$), taking p=0.45 as an example in Fig. 3d, 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*] × $[\mathrm{Na}^+]^p$) ratios. The dataset with 0–30% percentile of [OC]/([*Chl-a*] ×$[\mathrm{Na}^+]^{0.45}$) ratios, indicated by the solid markers in Fig. 3d, was used to calculate the fitting line of MPOC versus [OC]/([*Chl-a*] ×$[\mathrm{Na}^+]^{0.45}$). 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. 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 (r) 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. In further analysis, we employed Eq. 3 with p=0.45 to estimate the MPOC, which performs well based on the fitting line slope (1.04) and r value (0.88) between WIOC and MPOC. A total of 58% of the estimated data points fell within the WIOC/MPOC 2:1 and 1:2 lines (Fig. 3d), 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 $[\mathrm{Na}^+]$ as the input parameters and considering the variation of sea spray aerosols (Fig. 3c, 3g). 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 (Fig. 3g).

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 NPO (Fig. 3d, 3h). The estimated MPOC displayed good correlations with the observed WIOC. However, the abundance of MPOC was underestimated by ~ 50% through comparing with the WIOC concentrations (Fig. 3d, 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 Northwest Pacific Ocean. 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 NPO 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 NPO. The SOA fraction among the total organic aerosols was higher during the summer cruise (44% on average) than during the spring (34% on average).



https://doi.org/10.5194/egusphere-2025-3951
Preprint. Discussion started: 28 August 2025

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 NPO. 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 and 35°N–40°N, 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 NPO (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 surface seawater as well as the production of biologic VOCs (e.g., isoprene, DMS, etc.) from phytoplankton.

The MSOC also displayed a peak over the areas among 15°N–20°N (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). 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 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).

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 NPO. 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., 2016). The PRLIS are enriched in the surface seawater and could be injected into SSA via bubble bursting (Miyazaki et al., 2018; Santander et al., 2021). The similar variations of C1 intensity and Na^+ in the marine aerosols, especially during the summer observation (Fig. S4), 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., 2016). 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. S4), which indicated their sources related to combustion emission and secondary formation (Tang et al., 2024).

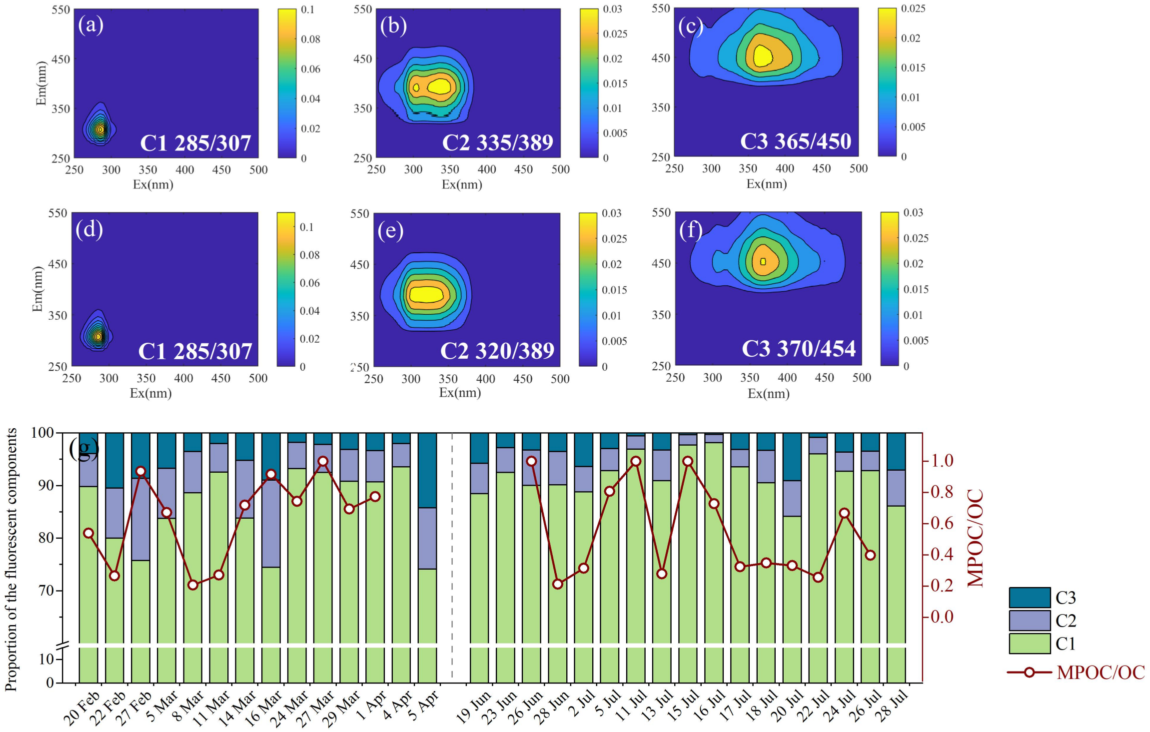


Figure 5 (a-f) The excitation (Ex) and emission (Em) spectra of the identified fluorescent components: C1: marine protein-like substances (PRLIS), C2: terrestrial humic-like substances (HULIS), C3: oxygenated HULIS. (g) Time series of the fluorescent component relative abundances and the MPOC/OC mass ratios in the marine aerosols over the Northwest 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). This was consistent to the higher mass contribution by MPOC than MSOC in the marine aerosol samples, as discussed in section 3.3. 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., 2016). 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. S4). 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., 2016).

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 Northwest Pacific Ocean. The spatial distribution of MOA along the latitude were 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 highlight 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.

Acknowledgments

This study was supported by the National Key Research and Development Program of China (2024YFC2815800, 2023YFC3705503), the National Natural Science Foundation of China (42205103; 42411540229), the Taishan Scholars of Shandong Province, China (tsqn202306101), the Fundamental Research Funds for the Central Universities (202441011), and the Shandong Provincial Natural Science Foundation (ZR2022QD105). The Fund of Key Laboratory of Marine Ecological Conservation and Restoration, Ministry of Natural Resources/ Fujian Provincial Key Laboratory of Marine Ecological Conservation and Restoration (EPR2025009); State Environmental Protection Key Laboratory of Formation and Prevention of Urban Air Pollution Complex (No. 2025080172).

Date and samples were collected onboard of R/V *Dongfanghong 3* and R/V *KeXue* implementing the open research cruise NORC2024-584 supported by NSFC Shiptime Sharing Project (42349584), the Laoshan Laboratory (LSKJ202201701, LSKJ202400202), and the Fundamental Research Funds for the Central Universities (202372001, 202472001)

Competing interests

The authors declare no conflict of interests.

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