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Continental pollutants modulate organic nitrogen and light absorption of marine organic aerosols over East Asian marginal seas

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Abstract: Organic nitrogen (ON) in marine aerosols is not only an important fraction of atmospheric nitrogen deposition but also a vital contributor to light-absorbing organic aerosols. However, ON abundance, sources, or its influence on organic aerosol absorption remain unclear in marine atmosphere. Here, shipboard observations were conducted in spring, summer, and autumn over the Yellow Sea and Bohai Sea (YBS) to understand the spatiotemporal distributions and sources of aerosol ON over East Asian marginal seas. Aerosol ON was 0.35 ± 0.25 μgN/m³, accounting for 4%–60% of total nitrogen in marine aerosols. Concentrations of ON were the highest in autumn due to severe impacts of anthropogenic pollutants, followed by those in spring and summer. Anthropogenic secondary pollutants (aged biomass burning and secondary nitrate formation) were the most important sources of aerosol ON, contributing 36%–76% of ON, 46%–83% of water-soluble ON, and 39%–89% of water-insoluble ON. In spring, 55% of ON, 45% of water-soluble ON, and 54% of water-insoluble ON were attributed to dust, and its contribution increased to >80% during a dust episode. In summer, marine sources associated with biological activity were important for aerosol ON formation. Nitrogen-containing organic compounds played vital roles in regulating light absorption by organic aerosols over the YBS. Elevated organic aerosol absorption was not only attributed to

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higher ON concentrations, but also related to increased absorption capability at higher ON/OC ratios. Our results highlight transported continental ON drove the light absorption by marine organic aerosols over marginal seas.

1 Introduction

Atmospheric organic nitrogen (ON) is a vital fraction among the total nitrogen (ON, NO_3^-–N, and NH_4^+–N) or the organic matter in ambient aerosols (Altieri et al., 2021). Deposition of particulate ON was estimated 23 Tg N yr^{-1}, and dominated the global atmospheric ON deposition (Li et al., 2023). Terrestrial aerosol nitrogen deposition can account for one-third of the external nitrogen supply to the open ocean per year, thereby influencing the marine nitrogen cycle and ecosystems (Duce et al., 2008; Jickells et al., 2017). Anthropogenic nitrogen deposition is especially important for maintaining primary productivity in the upper oligotrophic ocean (Dai et al., 2023; Tang et al., 2021). Aerosol ON also plays vital roles in the light absorption of organic aerosols (Xu et al., 2024). A recent modeling study suggests that absorptive nitrogenous components in organic aerosols contribute 61% of their global absorptive optical depth (Li et al., 2025b). However, the abundance, distribution, sources, or environmental effects of ON are much less understood compared to the inorganic nitrogen (IN, NO_3^-–N and NH_4^+–N) in marine aerosols.

Concentrations of ON in marine aerosols range from <0.01 to 3.6 μg N/m^3 (Li et al., 2019; Li et al., 2023; Luo et al., 2016; Shi et al., 2010), and ON contributes 5%−84% of total aerosol nitrogen in various atmospheric environments (Luo et al., 2016; Violaki et al., 2015; Xiao et al., 2018). Previous studies usually used water-soluble ON to represent the ON abundance in marine aerosols, resulting in an underestimation of ON in the marine atmosphere. A recently developed aerosol nitrogen analyzer system enables sensitive quantification of aerosol ON from remote open ocean to polluted urban environments (Sun et al., 2026; Yu et al., 2021b; Yu et al., 2024; Yu et al., 2023a). Cruise observations found that the average concentration of aerosol ON in the Northern Hemisphere (83 ng N/m^3) was much higher than in the Southern Hemisphere (15 ng N/m^3) (Sun et al., 2026). During cruises in different oceanic regions, the highest aerosol ON levels were recorded over coastal East Asia as a result of strong influence of anthropogenic pollutants (Sun et al., 2026). High aerosol ON abundance was usually observed in areas influenced by biomass burning or with high anthropogenic pollutants. Downwind of biomass-burning regions, aerosol ON can account for as much as 40%-80% of the total nitrogen deposition (Li et al., 2023).

Aerosol ON can originate from various sources, including biomass burning emissions (Wang et al., 2017), anthropogenic pollutants (Luo et al., 2018; Wang et al., 2019b), dust (Nie et al., 2014), atmospheric oxidation/aging processes (Laskin et al., 2010; Lin et al., 2015), marine emissions (Triesch et al., 2021), etc. Biomass burning activities can emitted abundant heterocyclic N-bases and nitroaromatic compounds into ambient atmosphere (Lin et al., 2017). Particulate ON in biomass burning plumes can be long-range transported and influence aerosol light absorption and marine ecosystems in downwind areas (Tang et al., 2021; Wang et al., 2019a). Aerosol ON can also be formed via oxidation of organic precursors or aging of organic aerosols in the presence of NO_x or NH_3 (Laskin et al., 2015; Li et al., 2020a; Shi et al., 2023). Some organic

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nitrogen compounds (e.g., imines, amino acids, protein-like organic matters) can be emitted from the ocean through sea spray or sea-to-air exchange (Chen et al., 2016; Triesch et al., 2021; Zhang et al., 2025).

The East Asian marginal seas are a typical region subject to the combined effects of continental air masses (e.g., anthropogenic pollutants, Asian dust storms) and marine emissions. Our recent observations suggested that the formation and light absorption of marine organic aerosols over the East Asian marginal seas were obviously influenced by transported anthropogenic pollutants, Asian dust, and marine emissions, and highlighted distinct seasonal differences (Zhang et al., 2025). Nitrogenous organic components have been recognized as vital chromophores in light-absorptive organic aerosols (Li et al., 2025b). To elaborate ON distribution, sources, and their effects on the optical properties of marine aerosols over marginal seas, shipboard cruise observations were conducted in spring, summer, and autumn over the Bohai Sea and the Yellow Sea (YBS). Aerosol ON was quantified, and its spatiotemporal variations in different seasons were analyzed. The sources of ON were quantified using source apportionment model. We further investigate the effects of organic nitrogen compounds on the light absorption and absorption capability of marine organic aerosols under the joint influence of terrestrial air mass outflows and marine emissions. Our results highlight that transported continental pollutants regulated ON formation and light absorption by marine organic aerosols over East Asian marginal seas.

2 Materials and Methods

2.1 Cruise observations and sample collection

Shipboard cruise observations were conducted over the Bohai Sea and the Yellow Sea (YBS) during autumn (22 Oct.–2 Nov.) in 2022 and during spring (13–28 April) and summer (13 July–13 Aug.) in 2023. During the campaigns, total suspended particles (TSP) and fine particles ($PM_{2.5}$) were simultaneously collected on prebaked quartz fiber filters using a high-volume aerosol sampler (1.05 m^3 min^{-1}). The sampler was placed at the front of the upper deck to avoid potential contamination from ship exhaust. Each sample was collected for 15–20 hrs. A total of 9, 22, and 20 sets of aerosol samples were obtained during the autumn, spring, and summer cruises, respectively. A field blank sample was collected during each cruise.
Air temperature and relative humidity (RH) were monitored using an onboard meteorological station during the observations. The 72-hr backward trajectories of air masses at an altitude of 500 m were calculated using the HYSPLIT model (Fig. S1) (Zhang et al., 2025). Satellite-derived chlorophyll-a (Chl-a) in surface seawater was obtained from NASA Ocean Color (https://oceancolor.gsfc.nasa.gov/).

2.2 Measurements of organic and inorganic nitrogen

Total ON in aerosol samples was determined using an aerosol IN&ON analyzer system, which integrates an online aerosol carbon analyzer and a NO_x analyzer (Yu et al., 2023b). Programmed thermal evolution facilitates the separation of aerosol

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ON from IN, and quantification of ON is achieved through multivariate curve resolution treatment of carbon and nitrogen thermal fractions (Yu et al., 2021a). The IN&ON analyzer has been widely used to quantify aerosol ON from various atmospheric environments. Water-soluble organic carbon (WSOC) and water-soluble total nitrogen (WSTN) are analyzed by the TOC/TN analyzer (TOC-L, Shimadzu, Japan). Inorganic nitrogen (NO_3^--N and NH_4^+-N), other inorganic ions (SO_4^{2-}, Cl^-, Na^+, K^+, Ca^{2+}, Mg^{2+}), methanesulfonic acid (MSA), and oxalic acid were quantified using ion chromatograph systems (ICS-Aquion, ICS-2100 DIONEX). Concentrations of WSON and WION were calculated as follows:

$$\mathrm{WSON} = \mathrm{WSTN} - [\mathrm{NH_4^+} - \mathrm{N}] - [\mathrm{NO_3^-} - \mathrm{N}]$$

$$\mathrm{WION} = \mathrm{ON} - \mathrm{WSON}$$

Organic carbon (OC) and elemental carbon (EC) in atmospheric aerosols were measured using a carbon analyzer (Sunset Laboratory) based on thermal-optical method. Water-insoluble organic carbon (WIOC) was calculated by the difference between OC and WSOC. Concentration of organic matter (OM) in the aerosol samples was calculated by multiplying OC by 1.6.

2.3 UV−Visible absorption spectra

Light absorption properties of extracted organic aerosols were measured using a UV-visible spectrometer (UV-8000, Jingmi, China). Methanol and water were used to extract the total organic matter and the water-soluble organic matter (WSOM) in aerosols. Absorption spectra of the extracted solutions were detected using a UV-visible spectrometer within the wavelength range of 200-700 nm. Absorption coefficients (Abs_λ, Mm^{-1}) and mass absorption efficiency (MAE_λ) at a wavelength λ were calculated using the following equations:

$$Abs_\lambda = \frac{(A_\lambda - A_{700}) \times V_l \times \ln(10)}{l \times V_a}$$

$$MAE_\lambda = \frac{Abs_\lambda}{C}$$

where A_λ is the measured absorbance of extracted solutions at wavelength λ. A_{700} is used to correct the baseline shift of the UV-visible spectrometer during analysis. The L is the optical path length (10 mm). V_l and V_a are volumes of the extraction solution and the air passing through the extracted filters. The light absorption of methanol-soluble components was used to represent the absorption by total organic aerosols. The difference between methanol-extracted solutions and WSOM was the absorption by water-insoluble organic matter (WIOM).

2.4 Source apportionment of aerosol ON

To identify the aerosol ON from different sources, we used the positive matrix factorization (PMF) receptor model (PMF version 5.0, EPA) to apportion the sources of aerosol ON, WION, and WSON over the YBS. The input parameters include:

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OC, EC, WSOC, MSA, oxalic acid, ON, WION, WSON, and Abs_{300} by WIOM and WSOM, water-soluble cations (NH_4^+, K^+, Na^+, Ca^{2+}, Mg^{2+}), and water-soluble anions (SO_4^{2-}, NO_3^-, Cl^-). Uncertainties of the parameters were calculated according to Manousakas et al. (2017) and Li et al. (2020b). Fig. S2 shows the variations of Q/Q_{exp} with different solution numbers. Base Model Displacement and bootstrap combined displacement methods were used to evaluate robustness of the PMF results, and 85% of the runs were acceptable.

3 Results and Discussion

3.1 Spatiotemporal variations of ON in marine aerosols over the YBS

Spatial variation and time series of ON in the collected $PM_{2.5}$ samples are shown in Fig. 1. The average concentration of ON in marine aerosols was 0.35 ± 0.25 μgN/m³ during the cruise observations over the YBS. The campaign-averaged concentrations of NO_3^--N and NH_4^+-N were 0.63±1.06 and 1.59±2.14 μgN/m³, respectively. Organic nitrogen averagely accounted for 22% (range: 4%–60%) of the total nitrogen (ON+NH_4^+-N+NO_3^--N) and 6% (range: 2%–13%) of organic matters in marine aerosols over the YBS. Under the joint influence of terrestrial transport and marine emissions, ON abundance in aerosols over the YBS was higher than those reported over open ocean, such as the Southern Ocean, North Atlantic, and Pacific Ocean (Cornell et al., 2003; Miyazaki et al., 2016; Miyazaki et al., 2011; Saliba et al., 2020). The observed aerosol ON over marginal seas was lower than those in polluted continental environments (e.g., urban Beijing, coastal Qingdao sites) (Shi et al., 2010; Wu et al., 2021; Xu et al., 2017).

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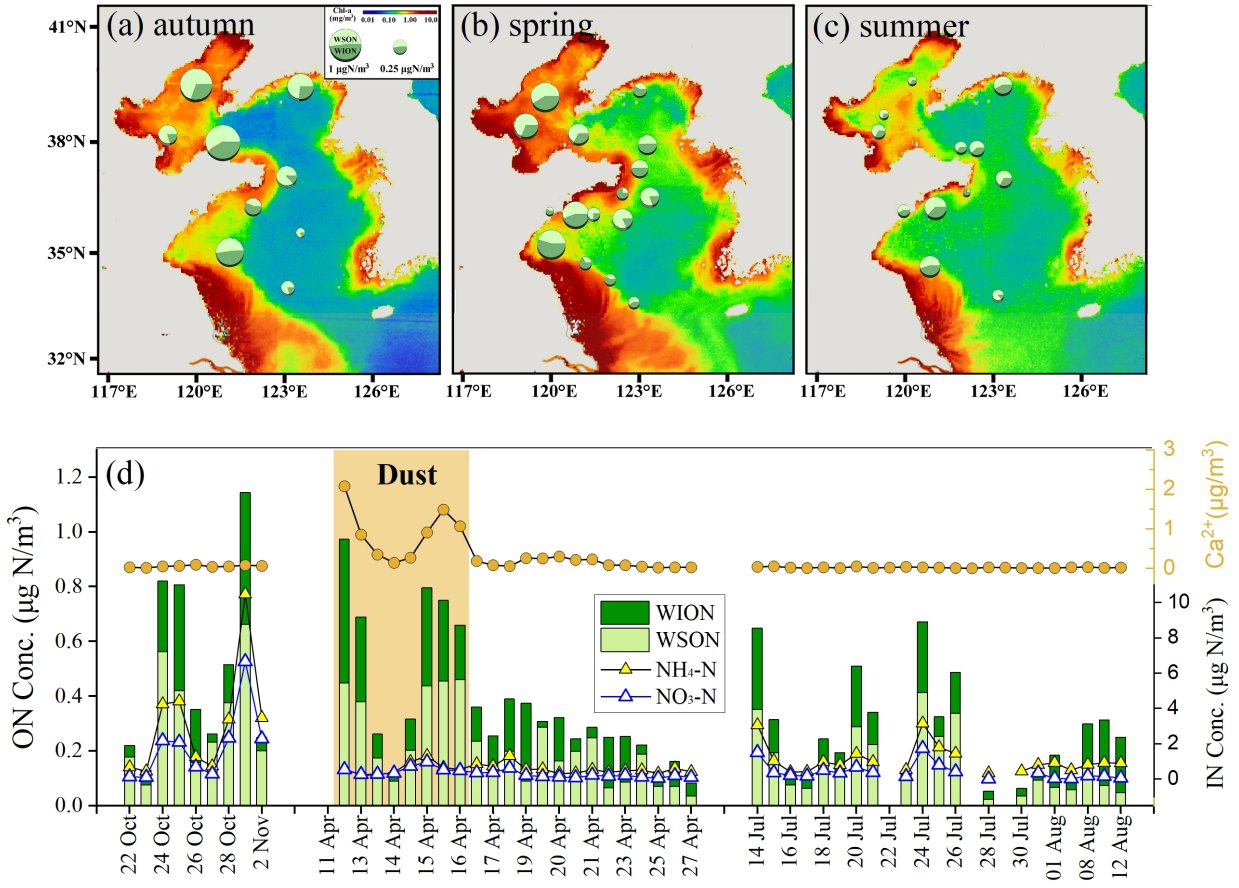

Figure 1. (a, b, c) Spatial distribution of organic nitrogen (ON) in $PM_{2.5}$ samples during cruises over the YBS. (d) Time series of ON, inorganic nitrogen (IN, including NH_4^+-N and NO_3^--N), and Ca^{2+} in marine aerosols during the observations. A dust episode was marked by orange shading in panel (d).

Aerosol ON over the YBS exhibited obvious seasonal variation trends (Fig. 2), following the abundance order of autumn (0.50 ± 0.32 μgN/m³)> spring (0.37 ± 0.24 μgN/m³)> summer (0.27 ± 0.18 μgN/m³). The observation regions were dominated by continental air masses in autumn and spring and by marine air masses in summer (Fig. S1). Under the severe impacts of continental air mass outflows, aerosol ON in autumn or spring was higher than that in summer (Fig. 1, 2). Concentrations of K^+ and NO_3^- in the collected samples were the highest during the autumn cruise (0.26 and 8.20 μg/m³) than during the other two seasons (0.066 and 1.37 μg/m³ in spring, 0.076 and 1.82 μg/m³ in summer), indicating the severer impacts of biomass burning and anthropogenic pollutants in autumn. In addition, a dust episode was observed during 12-16 April during the spring cruise, when Ca^{2+} concentrations in TSP increased obviously (Fig. 1d). During the dust episode, ON abundance increased to 0.57 ± 0.30 μgN/m³, higher than the ON concentrations observed on non-dust days during spring cruise.

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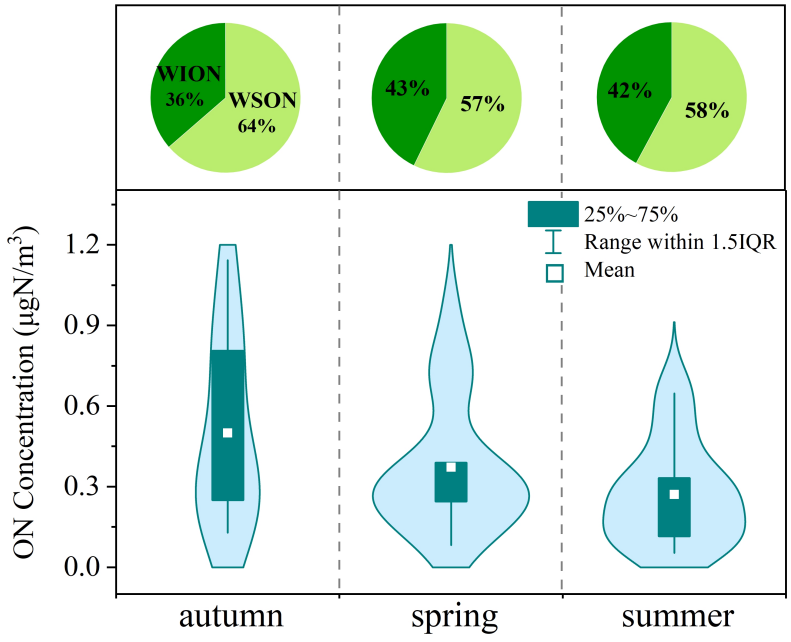

Figure 2. Seasonal variation of ON concentrations and relative contributions of water-soluble ON (WSON) and water-insoluble ON (WION). The values in the pie charts are the average contribution of WSON versus WION in each season.

During the observations over the YBS, WSON (0.21 ± 0.15 μgN/m³) in marine aerosols was higher than WION (0.15 ± 0.12 μgN/m³). Previous studies suggested that aged or secondarily formed organic aerosols tend to be more soluble than primarily emitted organic aerosols (Fang et al., 2023; Zhang et al., 2025). For example, abundant WSON can be formed via oxidation of volatile organic compounds (VOCs) in the presence of anthropogenic NO_x or in NH_3-aged organic aerosols (Laskin et al., 2015). East Asian marginal seas are obviously influenced by continental air pollutants, which undergo oxidation and aging processes during transport, thereby contributing water-soluble ON in marine aerosols over the YBS. The water-insoluble fractions of aerosol ON were higher in spring (43%) or summer (42%) than in autumn (36%). Our recent observation suggested that long-range transport of Asian dust is a major source of water-insoluble organic aerosols in spring over the East Asian marginal seas (Zhang et al., 2025). The proportion of water-insoluble ON among the total aerosol ON increased under the influence of spring dust storms. During summer, dominated by marine air masses, the relative contribution of marine-related sources increased. This resulted in a higher proportion of water-insoluble ON in summer than in autumn (Fig. 2). Marine biological activities emit nitrogen-containing organic components (e.g., protein-like macromolecules), which can be primarily emitted to and enriched in sea spray aerosols (Chen et al., 2016; Facchini et al., 2008).

3.2 Sources and influence factors of aerosol ON

Inter-species correlation and PMF source apportionment were analyzed to investigate the sources of aerosol ON over the YBS. As shown in Fig. S3, a five-factor solution was identified as reasonable and stable results. The resolved source factors

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include: (1) aged biomass burning, characterized by high loadings of K^+, EC, WSOC, and secondary inorganic ions; (2) dust, dominated by mineral dust tracers (Ca^{2+} and Mg^{2+}); (3) secondary nitrate formation, with high loadings of inorganic nitrate aerosols; (4) Marine biogenic emission, characterized by MSA, a typical atmospheric oxidation product of dimethyl sulfide emitted by phytoplankton (Kurosaki et al., 2022; Stefels et al., 2007); (5) Sea spray aerosol, dominated by Na^+, Cl^-, and MSA. The PMF-resolved ON, WSON, and WION from different sources matched well with the measured concentrations in marine aerosols (Fig. S4).

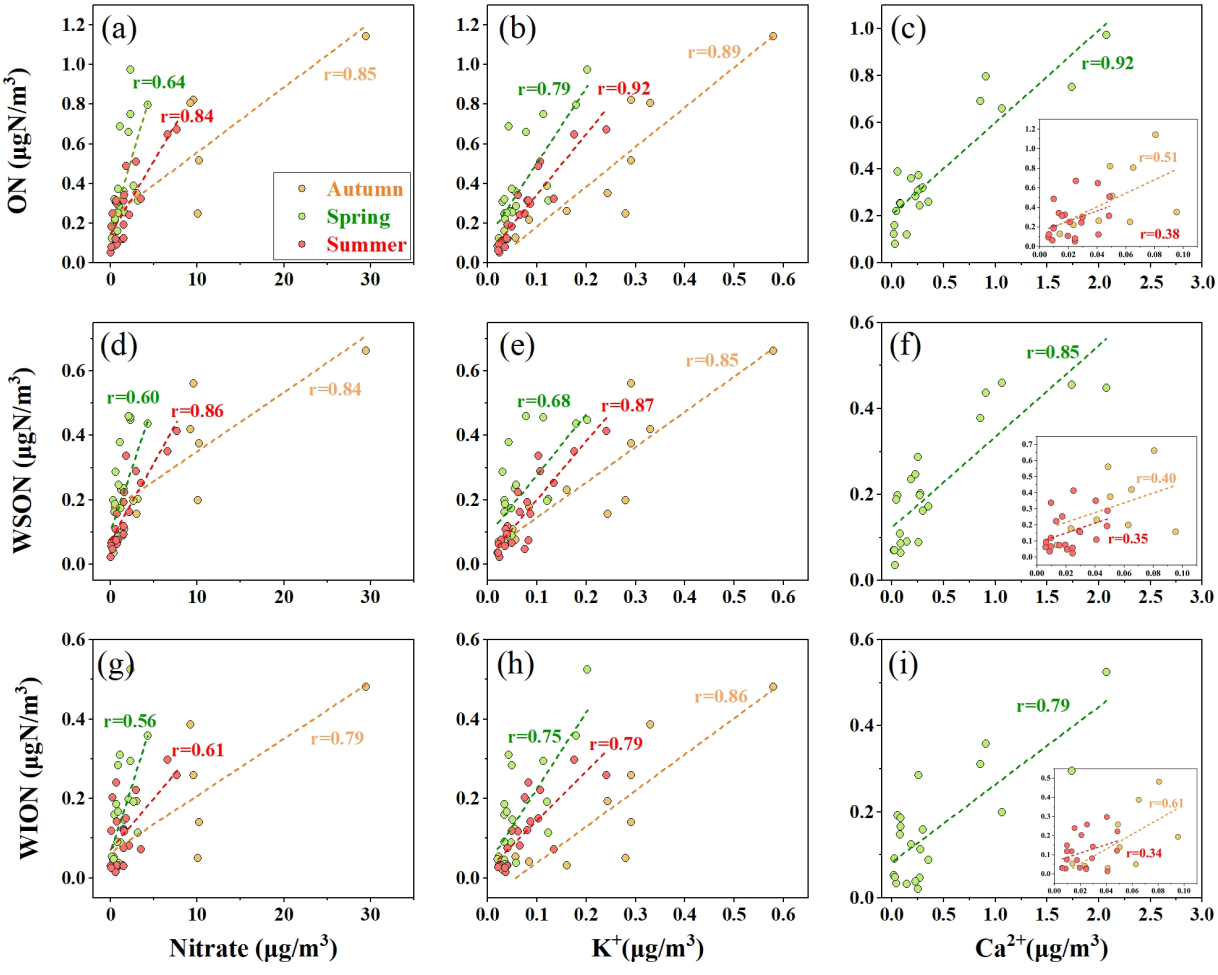

Figure 3. Correlation between (a-c) ON, (d-f) WSON, or (g-i) WION and secondary nitrate aerosols, K^+, and Ca^{2+} in $PM_{2.5}$ over the YBS. Data for autumn, spring, and summer are represented in orange, green, and red, respectively. The dashed lines and r-values are the fitted curves and correlation coefficients of the data in each season.

Anthropogenic secondary pollutants, including aged biomass burning and secondary nitrate formation, were important sources of organic nitrogen in atmospheric aerosols over the YBS. The concentrations of ON, WSON, and WION showed moderate or strong correlations with K^+ and nitrate in marine aerosols (Fig. 3). The source apportionment result suggested

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that aged biomass burning and secondary nitrate formation totally contributed 36%–76% of ON, 46%–83% of WSON, and 39%–89% of WION during the observations (Fig. 4). Driven by continental air masses, the concentrations of K^+ and nitrate in the collected aerosol samples were much higher in autumn (0.26±0.15 and 8.2±9.1 μg/m³) than in the other two seasons (0.066±0.050 and 1.4±1.1 μg/m³ in spring, 0.076±0.056 and 1.8±2.1 μg/m³ in summer). Aerosol ON contributed by anthropogenic sources substantially exceed that by dust or natural sources (marine biogenic emission and sea spray aerosol) during the autumn cruise (Fig. 4). Previous studies have suggested that biomass burning and VOC oxidation with NO_x involved in would produce atmospheric ON in terrestrial environments (Liu et al., 2019; Wang et al., 2019b), which can be long-rang transported to marine atmosphere. Secondary formation of aerosol ON was usually related to aqueous reaction processes (Chen et al., 2026). The contribution of aged biomass burning to WSON was higher than that to WION in aerosols (Fig. 4). Biomass burning is an important contributor to water-soluble organic components in the atmosphere, and aged organic aerosols tend to be more water-soluble than primarily emitted organic aerosols. A recent study suggested that wildfires contributed 37% of global atmospheric ON deposition and that 40%-80% of total N deposition was attributable to aerosol ON downwind of biomass burning areas (Li et al., 2023).

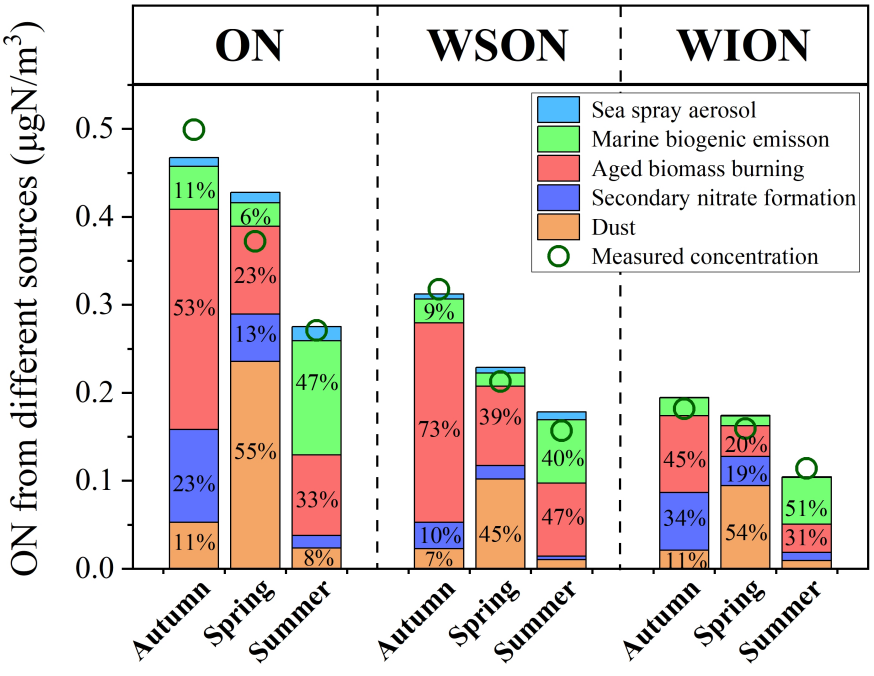

Figure 4. Aerosol ON, WSON, and WION from different sources resolved by PMF during cruise observations over the YBS. The relative contribution of each source to the total ON, WSON, or WION is listed in the corresponding stacked column.

We noted that ON abundance from secondary nitrate formation increased with the increase in RH in autumn or spring (Fig. S5). High-humidity conditions favor the secondary formation of aerosol ON via heterogeneous or aqueous-phase reactions. For example, high RH promotes the aqueous formation of nitrated aromatic compounds (e.g., methyl-nitrocatechols) (Shi et

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al., 2023; Vidovic et al., 2018; Wang et al., 2019b). Aqueous reactions between dicarbonyls and ammonium, amine, or amino acids produce N-heterocycle compounds (De Haan et al., 2009; Marrero-Ortiz et al., 2019). During the summertime cruise, however, we did not observe the influence of ambient RH on aerosol ON from secondary nitrate formation (Fig. S5c). This is because humidity was not a limiting factor for aqueous formation of secondary orgnaic aerosols (SOA) in summer (Wang et al., 2023), when the ambient RH was higher than in other seasons.

During the spring cruise, ON, WSON, and WION showed strong positive correlations with Ca^{2+} in the aerosol samples (Fig. 3), indicating dust storms as an important source of aerosol ON over the YBS. However, the correlations between ON and Ca^{2+} during the other two seasons were weaker than in spring (Fig. 3). Based on the PMF result, contribution of dust to ON, WSON, and WION in marine aerosols elevated to 55%, 45%, and 54% in spring, compared to 6%-11% in autumn and summer (Fig. 4). As shown in Fig. 1, a dust storm originating in Mongolia was recorded in spring (Wang et al., 2025; Zhang et al., 2025). The highest Ca^{2+} concentration was observed on 12 April. Based on the source apportionment result, 95% of ON, 83% of WSON, and 88% of WION were attributed to dust on 12 April during the dust episode (Fig. S4). On one hand, dust surface favors the gas-to-particle partitioning or the uptake of gaseous organic precursors (Li et al., 2025c). On the other hand, some mineral components provided favorable conditions for nitration reactions and further ON formation in aerosols (Nie et al., 2014). For the spring samples, ON from dust displayed a positive correlation with nitrate aerosol concentration (Fig. S6). Aged dust particles could facilitate the formation of SOA via aqueous-phase reactions, especially on nitrate-coating particles (Li et al., 2025a). Dust storms, mixing with anthropogenic nitrate pollutants during transport through East Asia, are a vital contributor to the inorganic and organic nitrogen deposition of marginal seas.

Marine sources, including marine biogenic emissions and sea spray aerosol, became a dominant contributor to ON in summer, accounting for 53% of ON, 45% of WSON, and 52% of WION in aerosols over the YBS (Fig. 4). Marine-related ON was mainly associated with biogenic emissions. Amino acids, protein-like organic matter, or other organic nitrogen compounds can be emitted from the ocean and enriched in sea spray aerosols (Triesch et al., 2021; Wang et al., 2016). Marine biogenic emissions contributed 6%-11% of ON, 7%-9% of WSON, and 6%-10% of WION during the autumn and spring cruises, higher than the contribution of sea spray aerosols (<3%). Previous studies have suggested that marine-generated ON is usually related to biological activity or nitrogen-fixing microorganisms in seawater (Aller et al., 2017; Dobashi et al., 2023).

3.3 Influence of ON on light absorption by marine organic aerosols

A recent study suggests that brown nitrogen, the absorptive nitrogenous components of organic aerosols, dominates their light absorption (Li et al., 2025b). Brown nitrogen contributes 76% of surface light absorption by organic aerosols over the US, and 61% of their global absorptive optical depth (Li et al., 2025b). To understand the role of ON in regulating the absorption of marine organic aerosols over the East Asian marginal seas, we analyzed the correlations between aerosol ON and light absorption (Abs_{300}), absorption capability (MAE_{300}) of organic aerosols (Fig. 5). For the water-soluble organics,

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Abs$_{300}$ of WSOM displayed strong correlations with WSON in the marine aerosols across seasons over the YBS (Fig. 5a). This revealed the importance of nitrogen-containing organic components for organic aerosol absorption. A strong dependence of WSOM absorption coefficient at 365 nm on WSON concentrations was also observed in atmospheric aerosols at inland city sites (Chen et al., 2026). However, for the water-insoluble fraction, we only observed a strong correlation between Abs$_{300}$ of WIOM and WION in the spring samples (Fig. 5b). For the autumn or the summer cruises, we did not observe an obvious variation trend between WIOM Abs$_{300}$ and WION in the collected aerosol samples. This is consistent with our recent findings that absorption of brown carbon (BrC) over the YBS was generally dominated by water-soluble organics, and the contribution of water-insoluble organic aerosols increased during dust episodes (Zhang et al., 2025). Water-soluble organic nitrogen drove the light absorption by organic aerosols over the YBS. During the spring dust period, both WSON and WION played important roles in the light absorption by marine organic aerosols over the East Asian marginal seas.

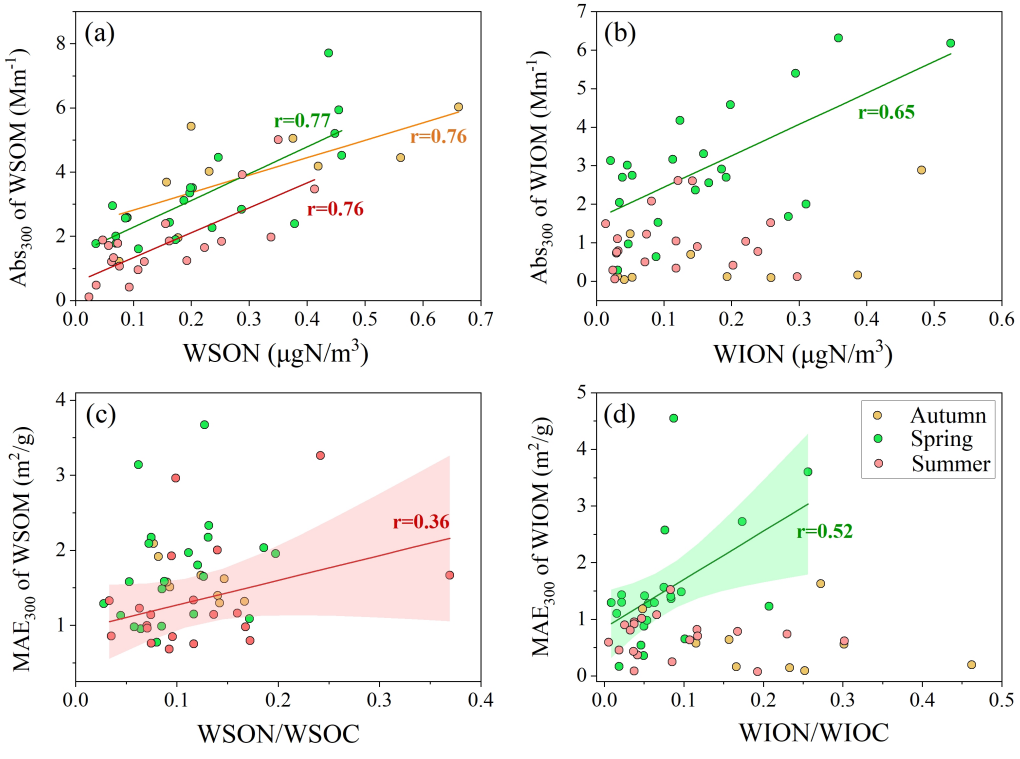

Figure 5. (a, b) Variations of Abs$_{300}$ by WSOM, Abs$_{300}$ by WIOM as a function of WSON, WION. (c, d) Variations of MAE$_{300}$ of WSOM, MAE$_{300}$ of WIOM as a function of WSON/WSOC, WION/WSOC mass ratios. The fitting line for the summer, spring, and autumn data is represented in red, green, and yellow, respectively.

The elevated light absorption by organic aerosols was not only attributed to the higher ON concentrations but also associated with an increase in the absorption capability of organic aerosols. We plotted the variations of MAE$_{300}$ as a function of ON/OC ratios in Fig. 5 (c, d). For the summer samples, MAE$_{300}$ of WSOM showed an increasing trend as WSON/WSOC

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ratios increased (Fig. 5c). During the spring cruise, MAE_{300} of WIOM elevated as the increasing of WION/WIOC ratios (Fig. 5d). Previous studies have concluded that organic molecules contributing to BrC absorption usually have a large degree of unsaturation and contain one or more nitrogen atoms (Laskin et al., 2015). These nitrogen-containing organic molecules are more prone to forming conjugated chromophores, thereby enhancing absorption by organic aerosols within the near-UV wavelength region (Laskin et al., 2015; Lin et al., 2016). For the water-insoluble organic aerosols in autumn or summer, there was no obvious variation trend for absorption capability with an increase in the WION/WIOC mass ratios (Fig. 5d).

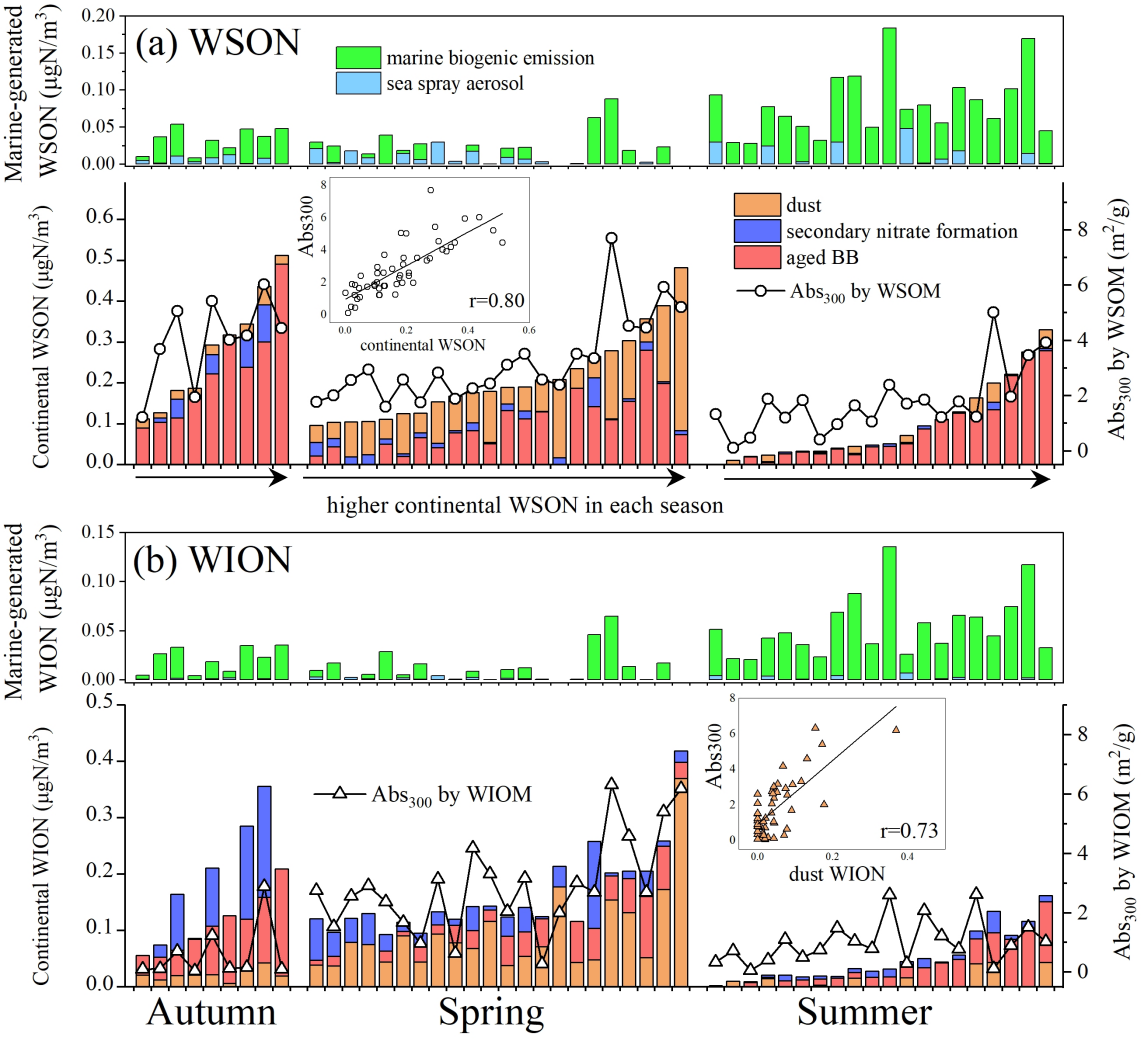

Figure 6. (a) Variations of marine-generated WSON (WSON from marine biogenic emission and sea spray aerosol), continental WSON (WSON from dust, secondary nitrate formation, and aged biomass burning), and Abs_{300} by WSOM. (b) Variations of marine-generated WION, continental WION, and Abs_{300} by WIOM. The inserted chart in panel (a) is the correlation between continental WSON and Abs_{300} by WSOM. The inserted chart in panel (b) is the correlation between WION from dust and Abs_{300} by WIOM. The samples during each cruise are ranked by the concentrations of continental

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WSON in aerosol samples.

Variations of aerosol ON from continental and marine sources, as well as BrC absorption coefficients, are shown in Fig. 6 to understand the roles of ON from different sources in regulating the light absorption by marine organic aerosols. Continental WSON or WION includes those from dust, secondary nitrate formation, and aged biomass burning. Marine-generated WSON or WION includes those from marine biogenic emissions and sea spray aerosols. In Fig. 6, samples during each cruise are listed in ascending order of continental WSON concentrations shown in panel (a). The Abs_{300} by WSOM or WIOM followed similar variation trends to the abundance of continental WSON or WION, and was different from those of marine-generated organic nitrogen (Fig. 6). Light absorption of WSOM showed a strong positive correlation (r=0.80) with continental WSON (inserted chart in Fig. 6a). The Abs_{300} by WIOM displayed a consistent variation trend to continental WION, especially to the dust WION. As shown in the inserted chart in Fig. 6(b), a strong positive correlation (r=0.73) was observed between Abs_{300} by WIOM and dust WION. This clearly suggested the vital roles of transported continental aerosol ON in regulating the light absorption by organic aerosols over marginal seas. It is noted that organic matter in dust aerosols among the source regions usually has weak light absorption capability. The light absorption of dust-related organics would increase via interactions with anthropogenic pollutants (e.g., nitrate aerosols) during long-range transport through East Asia.

4 Conclusion

Organic nitrogen is an important fraction of the total nitrogen in atmospheric aerosols over the East Asian marginal seas. Aerosol ON concentration was 0.35 ± 0.25 μgN/m³, and accounted for 4%–60% of the total nitrogen in marine aerosols during the cruises over the YBS. Aerosol ON abundance was the highest in autumn (0.50 ± 0.32 μgN/m³), followed by those in spring and summer. The obvious seasonal variation trend was attributed to the severe impacts of anthropogenic pollutants in autumn and dust storms in spring. The concentration of WSON (0.21 ± 0.15 μgN/m³) was higher than WION (0.15 ± 0.12 μgN/m³) in marine aerosols over the YBS. During the dust episode in spring, ON increased to 0.57 ± 0.30 μgN/m³, and the proportion of water-insoluble ON increased.

Sources of aerosol ON in marine atmosphere were apportioned using the PMF model. Anthropogenic secondary pollutants, including aged biomass burning and secondary nitrate formation, contributed 36%–76% of ON, 46%–83% of WSON, and 39%–89% of WION during the observations. Anthropogenic secondary pollutants dominated the formation of aerosol ON in autumn. An Asian dust storm originating from Mongolia was recorded during the spring cruise. A total of 55% of ON, 45% of WSON, and 54% of WION in marine aerosols was attributed to dust in spring, much higher than its contribution in autumn or summer (6%-11%). The contribution of dust can increase to >80% during the dust episode. Dust particles mixed with anthropogenic pollutants during long-range transport through East Asia, and were a vital contributor of organic nitrogen over marginal seas. During the summer cruise dominated by marine air masses, marine sources were important for aerosol ON formation, particularly those associated with marine biological activity.

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Organic nitrogen played a vital role in regulating the light absorption of organic aerosols over the YBS. Water-soluble ON drove light absorption by marine organic aerosols. Under the impacts of spring dust storms, both WSON and WION played important roles in organic aerosol absorption over the East Asian marginal seas. Elevated light absorption by organic aerosols was not only attributed to the higher ON concentrations, but also related to the increasing of absorption capability with higher ON/OC ratios (WSON/WSOC for summer, and WION/WIOC for spring). Transported continental aerosol ON, including those from dust, secondary nitrate formation, and aged biomass burning, modulated the light absorption by organic aerosols over marginal seas. Marine-generated ON, however, played a minor role in the light absorption by organic aerosols in marine atmosphere.

Author contributions

Chao Yu: Investigation, Visualization, Writing – original draft; Lin Zheng: Data curation, Investigation, Validation, Writing – review and editing; Yujue Wang: Conceptualization, Funding acquisition, Supervision, Visualization, Writing – original draft, Writing – review and editing; Meijing Guo, Xu Yu, Yuqi Guo, and Sisi Song: Data curation, Methodology, Investigation, Writing – review and editing; Kyoung-Soon Jang: Writing – review and editing; Jian Zhen Yu: Resources, Writing – review and editing; Xiaohong Yao and Huiwang Gao: Supervision, Resources, Writing – review and editing

Declaration of competing interest

The authors declare there are no conflicts of interest for this manuscript.

Data availability

The dataset is available upon request from the corresponding author.

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