https://doi.org/10.5194/egusphere-2025-2154
Preprint. Discussion started: 28 May 2025

Atmospheric Organosulfate Formation Regulated by Continental Outflows and Marine Emissions over East Asian Marginal Seas

Shubin Li[1], Yujue Wang[1, 2, *], Yiwen Zhang[1], Yizhe Yi[1], Yuchen Wang[4], Yuqi Guo[1], Chao Yu[1], Yue Jiang[1], Jinhui Shi[1, 2], Chao Zhang[1, 2], Jialei Zhu[5], Wei Hu[5], Jianzhen Yu[6, 7], Xiaohong Yao[1, 2], Huiwang Gao[1, 2], Min Hu[3, *]

[1]Frontiers Science Center for Deep Ocean Multispheres and Earth System, Key Laboratory of Marine Environment and Ecology, Ministry of Education of China, Ocean University of China, Qingdao, China

[2]Laboratory for Marine Ecology and Environmental Science, Qingdao Marine Science and Technology Center, Qingdao, China

[3]State Key Joint Laboratory of Regional Environment and Sustainability, College of Environmental Sciences and Engineering, Peking University, Beijing 100871, China

[4]College of Environmental Science and Engineering, Hunan University, Changsha, Hunan, 410082, China

[5]Institute of Surface-Earth System Science, School of Earth System Science, Tianjin University, Tianjin, China

[6]Division of Environment & Sustainability, Hong Kong University of Science & Technology, Hong Kong, China

[7]Department of Chemistry, Hong Kong University of Science & Technology, Hong Kong, China

**Correspondence to*: Yujue Wang (wangyujue@ouc.edu.cn); Min Hu (minhu@pku.edu.cn)

Abstract. Organosulfates (OSs) represent an unrecognized fraction and a potentially important source of marine organic aerosols. Based on shipboard observations over East Asian marginal seas, we characterized OSs in marine aerosols during spring, summer, and autumn. The C_2–C_3 OSs and isoprene-/monoterpenes-derived OSs were quantified using synthesized standards. The total quantified OS concentrations ranged from 4.5 to 109.1 ng/m³, contributing 0.1%−3.2% of the mass concentration of marine organic aerosols. The highest OS concentrations, dominated by C_2–C_3 OSs and isoprene-OSs, were observed in summer, which surpassed the abundance of methane sulfonic acid, a key component in climate regulation by oceanic phytoplankton sulphur emissions. Abundant OS formation in summer was mainly attributed to the increased isoprene emissions from the ocean. During the spring and autumn cruises, transported continental pollutants resulted in the higher fraction of monoterpene-derived (nitrooxy-)OSs, as well as the elevated OS concentrations over regions surrounded by the continent. This work highlights the joint effects of marine emissions and continental outflows on the formation and distribution of atmospheric OSs over marginal seas.

1 Introduction

Marine atmospheric aerosols play a vital role in climate change through influencing cloud formation and solar radiative balance(Li et al., 2022). Marine phytoplankton could produce dimethylsulfide (DMS), which further forms methane

https://doi.org/10.5194/egusphere-2025-2154
Preprint. Discussion started: 28 May 2025

sulphonic acid (MSA) or sulfate aerosols and then regulates the cloud condensation nuclei (CCN) formation and climate in the marine boundary layer (Andreae and Rosenfeld, 2008; Kettle and Andreae, 2000; Kloster et al., 2006). This is named CLAW hypothesis, proposed by Charlson et al. (1987) and Ayers et al. (1991). While following observation evidence and modelling studies indicated that CCN formation in marine atmospheres is far more complex than was recognized by the CLAW hypothesis (Quinn and Bates, 2011). This is mainly attributed to the unknown organic fractions in marine aerosols, including those primarily emitted by sea spray and secondarily formed organic aerosols (SOA) via the oxidation of volatile organic compounds (VOCs).

Traditional SOA tracers, including those from the oxidation of isoprene and monoterpene, etc., could explain only <10% of marine organic aerosols (Fu et al., 2011; Guo et al., 2020). Majority of the marine SOA components remain unknown till now. Abundant isoprene could be emitted from the ocean, and isoprene SOA has been proved to be one of the most important fractions in marine organic aerosols (Hu et al., 2013). Another important SOA formation pathway from isoprene and monoterpene oxidation is facilitated by acidic sulfate particles under high humidity conditions, with organosulfates (OSs) as the products (Brüggemann et al., 2020). Sulfate aerosols could be readily formed via DMS oxidation and are generally abundant over various marine environments (Andreae, 1990; Li et al., 2018; Yan et al., 2024). Recent studies have indicated the existence and importance of organic sulfur compounds, including OSs, in marine aerosols (Bao et al., 2018; Ye et al., 2021).

Atmospheric OSs constitute a large portion of organic aerosols (OA) in environments with substantial interactions of biogenic and anthropogenic emissions (Hettiyadura et al., 2019; Meade et al., 2016; Surratt et al., 2008; Wang et al., 2018). A recent cruise observation over Asian marginal seas suggested that OSs derived from isoprene and monoterpenes could contribute about 7% of the OA mass concentration (Wang et al., 2023b). Wang et al (2023) also indicated that isoprene/monoterpene-derived OSs could surpass the traditionally identified SOA tracers generated from isoprene or monoterpene oxidation (e.g., methylglyceric acid, alkene triols, hydroxyglutaric acid, pinic acid etc.). While these OS compounds derived from isoprene or monoterpenes were rarely detected in marine aerosols collected at a remote island site located in the Southern Ocean or in the southern Indian Ocean (Claeys et al., 2010; Cui et al., 2019). These could be attributed to the low biogenic VOCs emission/flux or the degradation of OSs over long-term storage (Claeys et al., 2010; Cui et al., 2019).

The existence or abundance of atmospheric OSs in marine aerosols have not been well evaluated or quantified till now (Hawkins et al., 2010; Wang et al., 2023b; Ye et al., 2021), which limited the understanding on their formation processes or their roles in the sulfur cycle and aerosol climate effects in marine atmospheres. In this study, atmospheric OSs over East Asian marginal seas were quantified using synthesized OS standards. We characterized the particulate OSs derived from

https://doi.org/10.5194/egusphere-2025-2154
Preprint. Discussion started: 28 May 2025

isoprene and monoterpenes, and investigated their spatial distributions, seasonal variations, as well as the dominant environmental factors of OS formation. Our results suggested that, over marginal seas, the spatiotemporal distribution of OS abundance and composition was dependent on the relative importance of marine emissions and continental outflows. This work highlights the vital roles of OSs in altering the sulfur cycle in marine boundary layer, and further studies in open ocean are needed to understand the influence of OSs on the climate effects of marine aerosols.

2 Methods

2.1 Cruise observation and sample collection

Marine aerosol samples were collected during summer (16 July–26 July) and autumn (21 October–2 November) in 2021, and during spring (14 April–25 April) in 2022 over the Yellow Sea and Bohai Sea (YBS). The fine particle ($PM_{2.5}$) and total suspended particulate (TSP) samples were collected using high-volume aerosol samplers (KB-1000, Qingdao Genstar Electronic Technology, China). The quartz fiber filters were pre-baked at 500 °C for 4.5 hours and wrapped in pre-baked aluminum foil after sampling. Aerosol samplers were placed on the top deck of the vessel "Lanhai 101", approximately 8 m above the sea surface. Each aerosol sample was collected for 10−24 hrs, and a field blank sample was collected during each cruise.

During the observation, wind speed (WS), air temperature, and relative humidity (RH) were measured by a shipboard meteorological observatory. The *Chl-a* concentration in surface seawater was measured using a CE Turner Designs fluorometer. Concentrations of seawater isoprene were then estimated by empirical formulas based on previous studies (Ooki et al., 2015; Wang et al., 2023b). The 72 h backward trajectories of air masses from an altitude of 500 m above ground level were calculated using the HYSPLIT model (Version 5.2.1, NOAA), starting every 6 h (Fig. S1). Trajectories at the center site of the observation region were calculated to represent the air masses during each cruise over the YBS.

2.2 HPLC-MS analysis and OS quantification

An aliquot of each filter sample was extracted by methanol. The solutions were filtered using PTFE syringe filter (0.22 μm), and evaporated to dryness under a gentle stream of N_2 gas. The dried residues were redissolved in methanol containing 0.1% formic acid (100 μL). Organosulfate compounds were quantified using a QTRAP 4500 mass spectrometer (AB Sciex) coupled with an UHPLC system (Ultimate 3000, Thermo Scientific, DE) for the low-molecular-weight OSs, and an Exactive Plus-Orbitrap mass spectrometer (Thermo Scientific Inc.) with an UHPLC system (Ultimate 3000) for monoterpene-derived compounds. Mass spectrometry was operated using a negative-mode electrospray ionization. The monoterpene NOSs ($C_{10}H_{16}NO_7S^-$ and $C_9H_{14}NO_8S^-$) were identified in the extracted ion chromatogram mode, and other OS compounds were quantified in multiple-reaction monitoring (MRM) mode. In this work, C_2−C_3 OSs (HAS, GAS, and LAS), isoprene-OSs

https://doi.org/10.5194/egusphere-2025-2154
Preprint. Discussion started: 28 May 2025

(IEPOX-OS, MAE-OS, and $C_5H_7O_7S^-$), monoterpene-OSs, and nitrooxy-OSs (NOSs) were quantified using synthesized OS standards (Table S1) (Wang et al., 2018).

Chromatographic separation of the low-molecular-weight OSs, including C_2–C_3 OSs and isoprene-OSs/NOSs, was optimized using a BEH Amide column (2.1 mm×100 mm, 1.7 µm, Waters, USA) equipped with a pre-column. Hydrophilic interaction liquid chromatography separation is an accurate analytical method for quantifying the low-molecular-weight OSs (Hettiyadura et al., 2015). The injection volume was 2.0 µL. The column was maintained at 35°C. Mobile eluents were solvent A: ammonium acetate buffer (10 mM, pH 9) in ultrapure water and solvent B: 10 mM ammonium acetate buffer (10 mM, pH 9) in acetonitrile/water (95:5). The flow rate was 0.4 mL/min at 0–2.5 min, then decreased to 0.35 mL/min from 2.5 to 11.5 min, and increased back to 0.4 mL/min from 11.5 to 18 min. The gradient elution was set as follows: 100% B at 0–0.4 min; reduced to 88% B at 0.4–2.4 min and maintained until 11 min; increased to 100% B at 11–11.5 min, and maintained at 100% B until 18 min to re-equilibrate the column.

Monoterpene OSs/NOSs were analyzed using an Acquity UPLC HSS T3 column (2.1 mm×100 mm, 1.8 µm, Waters, USA) with a pre-column. The mobile eluents were solvent A (0.1% acetic acid in ultrapure water) and solvent B (0.1% acetic acid in methanol) at a flow rate of 0.3 mL/min. The gradient elution procedure was performed as follows: 5% B at 0−1.5 min; increased to 54% B over 13.7 min and held for 1.0 min; then increased to 90% B over 1.8 min and held for 5 min; decreased to 5% B over 0.5 min and held for 1.5 min to re-equilibrate the column for next injection. The column temperature was maintained at 45°C, and the injection volume was 5.0 µL.

2.3 Measurements of aerosol chemical composition

Elemental carbon (EC) and organic carbon (OC) in aerosol samples were quantified using a carbon analyzer (Model RT-3131, Sunset Laboratory, OR). The OA concentration was then calculated by multiplying OC by 1.6. Water-soluble cations (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}), anions (Cl^-, NO_3^-, SO_4^{2-}), and MSA were measured using ion chromatography systems (ICS-2100 and ICS-Aquion RFIC, Thermo Scientific). The concentrations of non-sea-salt potassium ion (nss-K^+) and non-sea-salt sulfate (nss-SO_4^{2-}) were calculated by $[K^+]-0.037\times[Na^+]$ and $[SO_4^{2-}]-0.2516\times[Na^+]$. The concentrations of $PM_{2.5}$ or TSP were reconstructed by summing the concentrations of inorganic ions, OA, and EC in each aerosol sample.

3 Results and discussion

3.1 Concentration and composition of marine atmospheric OSs

The total quantified OSs and nitrooxy-OSs ranged from 4.5 to 109.1 ng/m³ in marine aerosols during the shipboard observations over the YBS (Fig. 1, Table S1). The eleven quantified OS and NOS compounds contributed 0.1%−3.2% of the

https://doi.org/10.5194/egusphere-2025-2154
Preprint. Discussion started: 28 May 2025

OA mass concentrations over the YBS. The observed OS concentrations here were generally higher than the wintertime concentrations at inland sites, and lower than those in coastal regions (Kanellopoulos et al., 2022; Meade et al., 2016; Nguyen et al., 2014; Wang et al., 2020, 2021, 2022b) (Fig. 2). This was due to the active interactions between biogenic VOCs and sulfate aerosols under high RH conditions in coastal areas, which favored the aqueous-phase formation of OSs in the atmosphere. While the OS formation could be limited by the low biogenic VOC emissions or ambient RH in the wintertime inland environments (Wang et al., 2020). It is noted that, taking the autumn observation as an example, we compared the OS concentrations in the $PM_{2.5}$ and the TSP samples simultaneously collected during the cruise (Fig. S2). The majority of the data points fall along the 1:1 line (Fig. S2). The presence of OSs is dominant in fine particles, and thus our further discussion is focused on the results of the $PM_{2.5}$ samples.

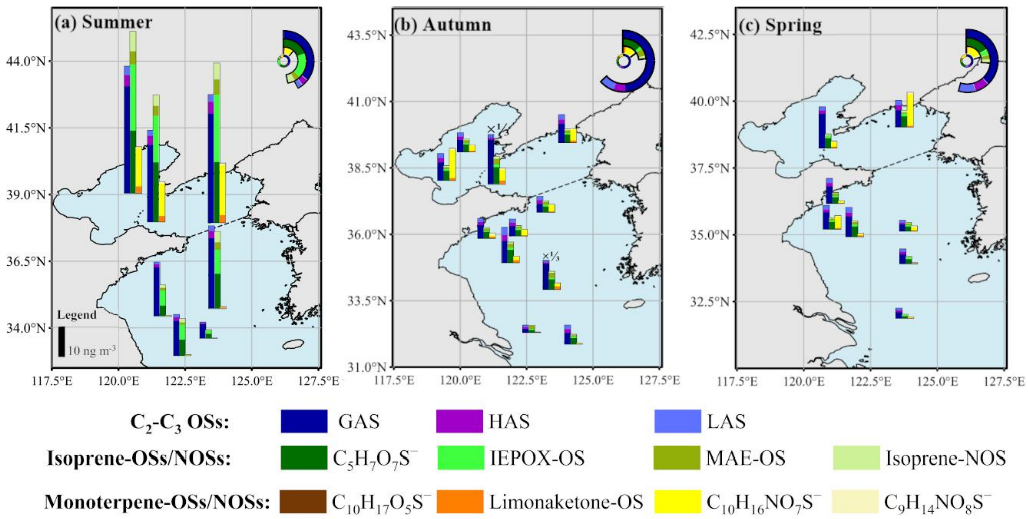


Figure 1. Spatial distributions of OSs in $PM_{2.5}$ over the YBS during (a) summer, (b) autumn in 2021, and (c) spring in 2022. The inserted charts in panels (a, b, c) are the contribution of different OS compounds. The dotted lines in the panels are the dividing line of the North Yellow Sea (nYS) and the South Yellow Sea (sYS).

The C_2–C_3 OSs, including glycolic acid sulfate (GAS), hydroxyacetone sulfate (HAS), and lactic acid sulfate (LAS), were the most abundant compound group across the observed seasons (Fig. 1, 2). The C_2–C_3 OSs concentrations were respectively 7.2±3.1, 24.2±12.4, and 12.8±14.4 ng/m³ in spring, summer, and autumn, comparable to the concentration levels at inland sites and lower than those in coastal areas (Fig. 2). In autumn and spring, the fraction of C_2–C_3 OSs, especially GAS, was much higher than other compound groups. The highest GAS concentration (47.8 ng/m³) over YBS was observed on 30 October during the autumn cruise. We noted that, in marine atmospheres, the contribution of C_2–C_3 OSs among the quantified OSs was much higher than those observed in various continental environments (Fig. 2). These low-molecular-

https://doi.org/10.5194/egusphere-2025-2154
Preprint. Discussion started: 28 May 2025

weight OSs could be formed via the oxidation of VOC precursors from both biogenic and anthropogenic origins (Wang et al., 2023a), and have been frequently observed as one of the most abundant OS groups in previous studies (Wang et al., 2018, 2020; Cai et al., 2020).

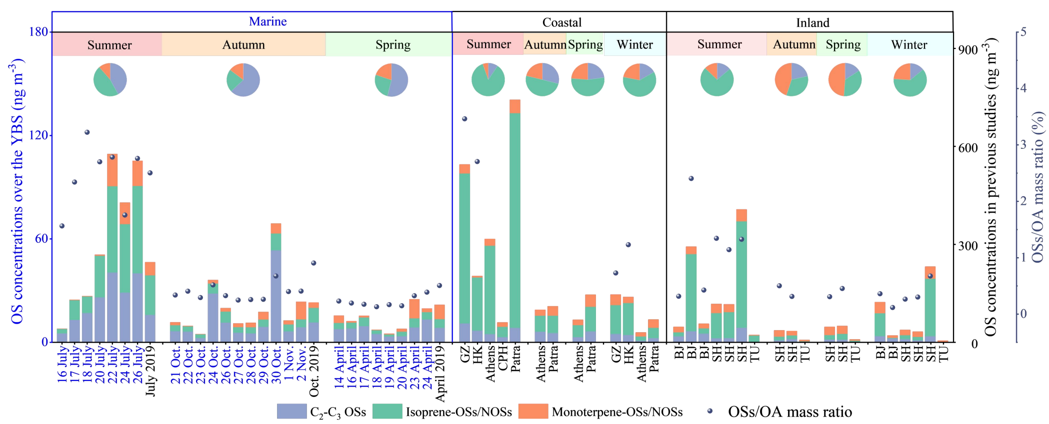


Figure 2. Atmospheric OS concentrations and mass ratios of (OSs+NOSs)/OA over the YBS in this study and in inland and coastal atmospheres reported in previous studies (Kanellopoulos et al., 2022; Meade et al., 2016; Nguyen et al., 2014; Wang et al., 2020, 2021, 2022b). The data labels in this work are denoted in blue, and those from previous studies are in black. The pie charts represent the average contribution of OS compound groups in each season. It is noted that the OS abundance over the YBS and at coastal or inland sites are represented by different y-axes concentration ranges.

The total concentration of quantified isoprene-OSs and NOSs ranged from 1.3 to 56.9 ng/m³, which were the most abundant group in summer over the YBS (Fig. 1). The predominance of isoprene OSs has been well documented at both coastal and inland sites (Fig. 2), which is attributed to the substantial biogenic isoprene emissions, especially during warmer seasons. Wu et al. (2021) reported abundant emission of isoprene from coastal and shelf seas (Wu et al., 2021), and isoprene OSs would then form via the interaction between sulfate aerosols and isoprene oxidation products (Surratt et al., 2010; Cooke et al., 2022). In the marine atmosphere over YBS, isoprene-derived OSs displayed a dominance by isoprene epoxydiol (IEPOX)-OS and $C_5H_7O_7S^-$ during summer, while by $C_5H_7O_7S^-$ during spring and autumn (Fig. 1). The $C_5H_7O_7S^-$ compound has been suggested as a further oxidized or aged form of IEPOX-OSs (Armstrong et al., 2022; Chen et al., 2020). The abundant presence of $C_5H_7O_7S^-$ in marine aerosols across seasons indicated the rapid oxidation and aging processes of isoprene SOA in marine atmospheres. The high contribution of $C_5H_7O_7S^-$ molecule among isoprene-derived OSs has been reported in marine aerosols, as well as in coastal and inland atmospheres (Hettiyadura et al., 2015; Kanellopoulos et al., 2022; Surratt et al., 2008; Wang et al., 2018, 2022b, 2023b).

The IEPOX-OS, a typical low-NO oxidation product of isoprene, was one of the dominant compounds during the summer

https://doi.org/10.5194/egusphere-2025-2154
Preprint. Discussion started: 28 May 2025

cruise (Fig. 1). For the summertime samples, the contribution of IEPOX-OS among isoprene-OSs here is higher than that in a previous study conducted in 2019 over the YBS (Wang et al., 2023b). This could be due to the reduction of NO_x emissions in the North China Plain(Li et al., 2024), resulting in a lower NO condition in 2022 than in 2019. It is also noted that a BEH Amide column was employed to separate the C_2-C_3 OSs and isoprene OSs in this work. The OS quantification here was more accurate than the study conducted in 2019, in which a reversed-phase column was used to separate the low-molecular-weight and highly polar OSs. This could be an additional reason for the different OS proportions between the two studies. The concentrations of methacrylic acid epoxide (MAE)-OS and isoprene-NOS, usually originated via NO/NO_2 pathway or under high-NO conditions(Worton et al., 2013), were much lower than those of IEPOX-OS and its aged product ($C_5H_7O_7S^-$) in the marine atmospheres (Fig. 1, Table S1).

The mass concentration and contribution of monoterpene-derived (nitrooxy-)OSs were lower than those of C_2−C_3 OSs and isoprene-derived OSs over the YBS (Fig. 1, 2). This compound group was dominated by monoterpene NOSs ($C_{10}H_{16}NO_7S^-$), which were formed via the oxidation of monoterpenes in the presence of anthropogenic NO_x (Surratt et al., 2008; Wang et al., 2018). The formation of monoterpene OSs/NOSs in marine atmospheres was driven by the transported continental pollutants. The concentration levels of monoterpene OSs/NOSs over the YBS were generally lower than those observed in continental atmospheres (Fig. 2) (He et al., 2014; Meade et al., 2016; Nguyen et al., 2014; Wang et al., 2020, 2021, 2022b).

3.2 Importance of OSs in marine atmospheres

The OS concentrations and mass contribution among marine OA were the highest in summer, followed by those in autumn and spring (Fig. 1, 2). The average OS concentration was 57.8±38.9, 20.4±19.7, and 13.3±8.3 ng/m³ in summer, autumn, and spring, respectively. During the summer cruise, OSs occupied 1.6%−3.2% (2.5% on average) of the marine OA mass concentrations, which were comparable to those observed in coastal regions and higher than those at the inland sites (Fig. 2). The elevated concentration levels and contributions of biogenic OSs, especially isoprene OSs and C_2−C_3 OSs, in summer were attributed to the increased biogenic VOC emissions from marine phytoplankton or photochemical reactions in surface microlayer (Conte et al., 2020). The higher seawater Chlorophyll-a (*Chl-a*) during summer indicated elevated isoprene production by phytoplankton, and higher temperature favored the sea-to-air transfer process of isoprene (Fig. S3). The vital importance of biogenic OSs to OA formation in summer has been highlighted in previous observations at both marine and continental sites (Hettiyadura et al., 2017; Kanellopoulos et al., 2022; Meade et al., 2016; Nguyen et al., 2014; Wang et al., 2020, 2021, 2022b). We cannot exclude the potential influence of terrestrial biogenic VOC emissions based on the observational evidence. The air masses were dominantly from the open ocean. During spring or autumn, the lower *Chl-a* and air temperature resulted in the decreasing of biogenic OS formation (Fig. 2). Though the seawater *Chl-a* was at similar

https://doi.org/10.5194/egusphere-2025-2154
Preprint. Discussion started: 28 May 2025

concentration levels in spring and autumn (Fig. S3), the OS abundance was lower in spring. The ambient temperature was lower in spring, and the oceanic phytoplankton had not revived from the low temperature conditions throughout winter. Thus, the biological activity and biogenic VOC production were likely at low levels during the spring cruise.

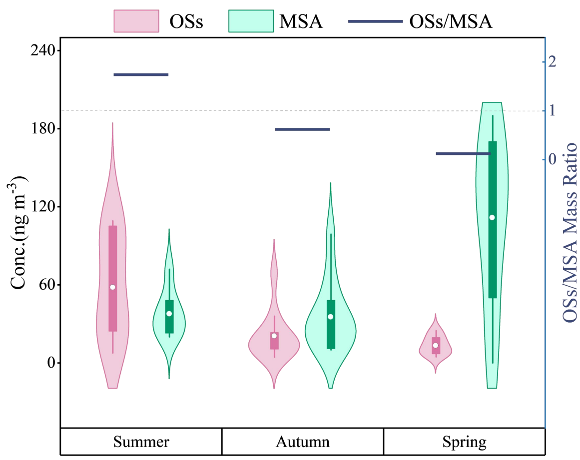


Figure 3. The OSs, MSA concentrations and OSs/MSA mass ratios in atmospheric aerosols over the YBS.

During summer, the active interactions between biogenic VOCs, especially isoprene, and acidic sulfate converted notable fractions of inorganic sulfate aerosols to OSs in marine atmospheres. The abundance of OSs was comparable to that of MSA in summer, and their mass ratios were higher than those in autumn and spring (Fig. 3). Atmospheric MSA contributes to the CCN formation in marine boundary layer, which is a vital species relevant to the CLAW hypothesis of oceanic phytoplankton-controlled climate regulation (Ayers et al., 1997; Charlson et al., 1987; Quinn and Bates, 2011). In addition, the OSs and MSA displayed strong correlations ($r = 0.86$, $p < 0.01$) in autumn (Fig. S4). This suggested that the atmospheric OSs and MSA formation was limited by the same environmental factors in autumn, which could be the lower biological activities indicated by the seawater *Chl-a* and temperatures (Fig. S3). The cruise observations indicated that organosulfate, besides MSA, should be taken into consideration when studying the sulfur cycle and its climate effects in marine atmospheres, especially over regions with high phytoplankton biomass and high temperature.

3.3 Seasonal variation of atmospheric OS composition

For the seasonal variations of OS composition, the chemical spaces of the autumn and spring samples are highly overlapped, which are different from that of the summer samples (Fig. 4a). The fraction of isoprene-derived (nitrooxy-)OSs was higher

https://doi.org/10.5194/egusphere-2025-2154
Preprint. Discussion started: 28 May 2025

during the summer cruise than those observed during the other two seasons. The autumn and spring samples generally showed a higher contribution by monoterpene-derived OS compounds. The seasonal variation was attributed to the relatively lower isoprene emissions, indicated by the lower seawater *Chl-a* (Fig. S3), and the more severe influence of anthropogenic pollutants transported from the continent in spring and autumn (Fig. S1). In addition to the air mass back trajectories, the more severe impacts of continental outflows in spring and autumn were also indicated by elevated elemental carbon (EC) concentrations (0.5 $\mu gC/m^3$ and 0.4 $\mu gC/m^3$ compared to 0.2 $\mu gC/m^3$ in summer). In marine atmospheres over the YBS, the relative contribution of monoterpene-derived (nitrooxy-)OSs was lower than that in continental atmospheres under more severe impacts of anthropogenic pollutants (Fig. 4a).

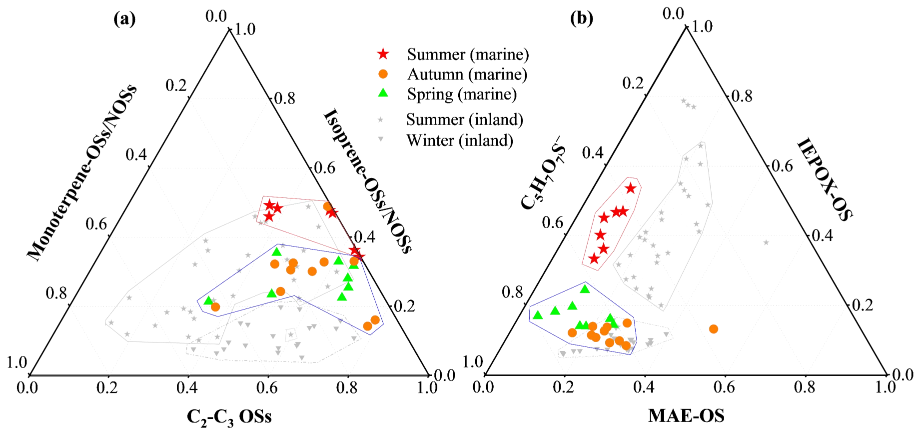


Figure 4. (a) Relative abundance of isoprene OSs, monoterpene OSs, and C_2−C_3 OSs, and (b) composition of isoprene OSs over the YBS in summer (red), autumn (orange) and spring (green). The results previously reported at the inland urban site (Wang et al., 2020) are colored gray.

In marine atmospheres over the YBS, different influences of marine emissions versus continental outflows across seasons resulted in the variation of C_2−C_3 OSs/isoprene-OSs mass ratios (Fig. S5). Strong correlations (r=0.79−0.97, p≤0.05) between isoprene-OSs and C_2−C_3 OSs suggested their consistent biogenic sources dominated by isoprene oxidation, which has been reported in previous studies(Schindelka et al., 2013; Surratt et al., 2008; Wang et al., 2020). In summer, the abundance of C_2−C_3 OSs was comparable to that of isoprene OSs. While, during autumn and spring, we observed higher mass ratios of C_2−C_3 OSs versus isoprene-OSs due to the additional sources of C_2−C_3 OSs contributed by anthropogenic sources (Fu, 2008; Huang et al., 2018; Liao et al., 2015).

The chemical space distributions of isoprene OSs also displayed obvious seasonal variations. The fraction of IEPOX-OSs was substantially higher, and that of MAE-OS was relatively lower in summer compared with those in spring and autumn

https://doi.org/10.5194/egusphere-2025-2154
Preprint. Discussion started: 28 May 2025

(Fig. 4b). The low-NO conditions in summer favored the IEPOX formation from isoprene oxidation via HO_2 pathway, while the formation of MAE via NO/NO_2 pathway would increase under the influence of continental pollutants in autumn and spring (Wang et al., 2020; Worton et al., 2013). During summer, the relative contribution of MAE-OS among isoprene-OSs in marine aerosols over the YBS was lower than those observed in continental atmospheres, indicated by the gray markers in Fig. 4b. This was due to the lower anthropogenic pollutants and NO conditions in marine atmospheres than in continental atmospheres. The proportions of $C_5H_7O_7S^-$, a further oxidation or aged forms of IEPOX-OSs(Armstrong et al., 2022; Chen et al., 2020), were also higher in autumn and spring than in summer. The dominant presence of $C_5H_7O_7S^-$ compared to IEPOX-OS indicated a highly oxidized state of marine SOA in spring and autumn.

3.4 Spatial distribution of OSs regulated by continental outflows

As shown in Fig. 5, atmospheric OSs concentrations over the Bohai Sea and the North Yellow Sea (nYBS, 51.3±37.4 ng/m^3) were notably higher than those over the South Yellow Sea (sYS, 16.1±11.9 ng/m^3). Surrounded by the continent, the nYBS region was under more severe impacts of transported anthropogenic pollutants compared with the relatively open sYS. This is also indicated by the variation of EC concentrations in atmospheric aerosols over nYBS and sYS areas (Fig. S6). Marine emissions dominated the biogenic OS formation over the YBS in summer, while we cannot exclude the potential influence of transported continental air masses, especially over the nYBS. This could be a reason for the higher OS concentrations over the nYBS than those over the sYS.

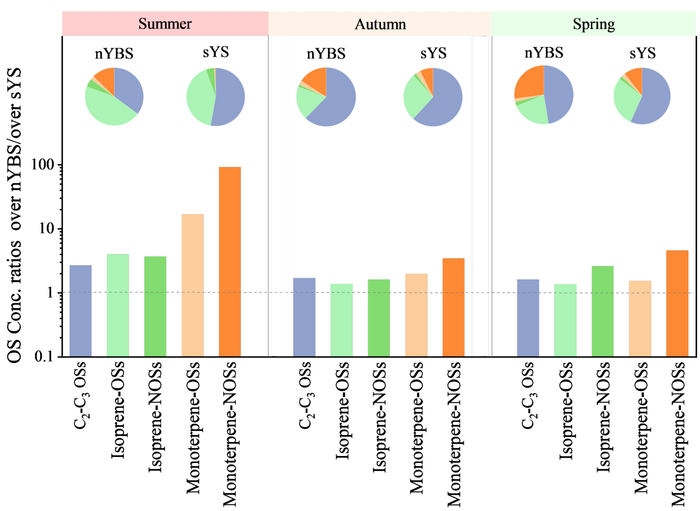


Figure 5. Concentration ratios of atmospheric OSs over the nYBS versus those over the sYS. The pie charts show the relative contribution of OS compound groups over the nYBS and the sYS during each season.

https://doi.org/10.5194/egusphere-2025-2154
Preprint. Discussion started: 28 May 2025

The concentration levels and compositions of OSs in atmospheric aerosols over the nYBS and the sYS are compared in Fig. 5. Among the quantified OSs derived from different VOC precursors, monoterpene-NOSs displayed the most obvious enhancement ratios over the nYBS compared to those over the sYS (Fig. 5). During the summer cruise, monoterpene-NOSs over the nYBS elevated to nearly two orders of magnitude higher than those over the sYS. The mass contributions of monoterpene-NOSs among the total OSs over nYBS were higher than those over the sYS, as shown in the pie charts of Fig. 5. Monoterpene-NOSs are usually formed via the interactions between anthropogenic NO_x, sulfate, and monoterpenes (Bryant et al., 2021, 2023; Wang et al., 2018). A recent study also suggested monoterpenes could be generated by biomass burning, besides the biogenic emissions (Wang et al., 2022a). The spatial difference of monoterpene-NOSs further indicated the more severe influence of anthropogenic pollutants over the nYBS.

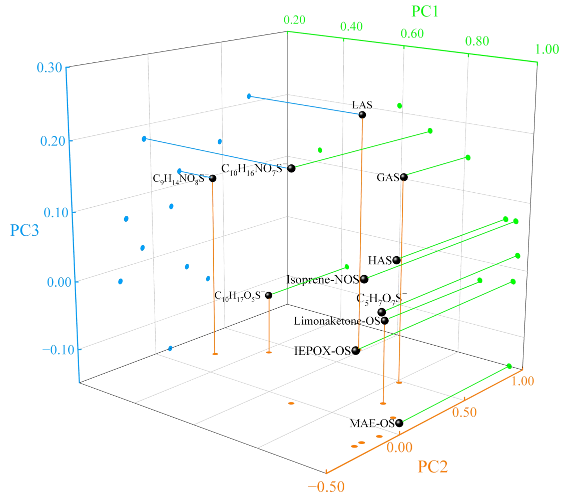
The OS abundance displayed the most obvious enhancement over the nYBS in the summer samples, in which the concentrations of C_2−C_3 OSs and isoprene-OSs/NOSs over the nYBS elevated to 2.4 and 3.9 times of those over the sYS. The biogenic emissions from marine phytoplankton were more abundant in summer than in the other seasons. Transported anthropogenic pollutants over the nYBS would promote the formation of biogenic OSs via anthropogenic-biogenic interactions in marine atmospheres. Previous observation has suggested that the formation of biogenic SOA, including isoprene OSs, could be obviously mediated by anthropogenic sulfate and NO_x in regions with substantial anthropogenic-biogenic interactions (Xu et al., 2015). We noted that isoprene-OSs were not observed in remote marine aerosols over the Southern Ocean or the southern Indian Ocean, where the influence of transported anthropogenic pollutants was likely limited (Claeys et al., 2010; Cui et al., 2019). Our results suggested the universal existence of biogenic OSs in marine aerosols over regions with anthropogenic-marine interactions, while the presence of OSs in different marine environmental conditions needs further observation evidence.

3.5 Origins and influence factors of atmospheric OSs

Principal Component Analysis (PCA) was performed to further understand the sources of atmospheric OSs over the YBS (Fig. 6, Table S2). A total of 18 particulate components, including OSs, water-soluble ions, EC, and MSA, were chosen to carry out the statistics. Three factors could explain 83% of the measurements. Majority of the OS and NOS compounds showed high loadings in Factor 1, which explained 52% of the measurements. Characterized by high loadings of nss-sulfate, Cl^-, and low loadings of anthropogenic species (e.g., EC, nss-K^+), Factor 1 represented the sulfate-catalyzed reactions with VOCs dominated by marine emissions. Factor 2 shows high loadings of EC, nss-K^+, and NO_3^-, suggesting the transported anthropogenic origins dominated by combustion emissions, which explained 21% of the measurements (Table S2). Factor 3, dominated by MSA, EC, and sea salts, was a mixed source of marine-anthropogenic interaction, which explained 10% of the

https://doi.org/10.5194/egusphere-2025-2154
Preprint. Discussion started: 28 May 2025

variance (Table S2).



295 **Figure 6.** PCA statistics of the measured OSs and NOSs during the cruise observations. PC1, PC2, and PC3 represent the source of sulfate-catalyzed reactions with biogenic VOCs, transported anthropogenic origin, and a mixed source of marine-anthropogenic interaction, respectively.

The majority of quantified OS compounds, especially the isoprene-derived ones (IEPOX-OS, MAE-OS, $C_5H_7O_7S^-$, isoprene-NOS, GAS, and HAS), were dominated by the source of sulfate-catalyzed reactions with biogenic VOCs (Factor 1), as displayed in Fig. 6 and Table S2. The homogeneous origin of C_2–C_3 OSs and isoprene-OSs/NOSs from the oxidation of isoprene has been approved in this work and previous observations(Surratt et al., 2008; Riva et al., 2016). This source factor was more related to the marine emissions, rather than anthropogenic pollutants, indicated by the low loadings of anthropogenic EC or nss-K^+. Isoprene could be largely emitted by phytoplankton and from photochemical processes in surface seawater, and then released into marine atmospheres (Brüggemann et al., 2018; Cui et al., 2023). The reactive uptake of isoprene by sulfate aerosols could be a vital reaction pathway for OS formation in marine aerosols (Wang et al., 2023b). Elevated OS concentrations were observed as the increasing of air temperature in summer or as the increasing of wind speed in spring (Fig. 7). Higher temperature or wind speed would promote the sea-to-air exchange of isoprene and favored the OS formation in marine atmospheres.

https://doi.org/10.5194/egusphere-2025-2154
Preprint. Discussion started: 28 May 2025

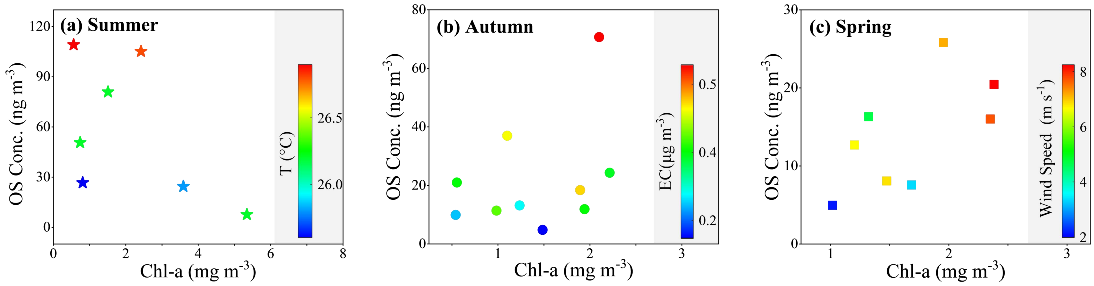


Figure 7. Variations of OS concentrations as a function of chlorophyll-a (Chl-a) in (a) summer, (b) autumn, and (c) spring. The markers are colored by air temperature, EC, and wind speed, respectively.

The loadings of monoterpene-OSs/NOSs in anthropogenic-related sources (Factor 2 and Factor 3) cannot be neglected, which was different from the main source of isoprene OSs from marine-dominated sulfate-biogenic VOC interaction (Factor 1). Lactic acid sulfate over the YBS showed comparable loadings in the transported anthropogenic origin (Factor 2, 0.70) and the marine-dominated sulfate-biogenic VOC interaction source (Factor 1, 0.59). The loadings of LAS in the mixed source of marine-anthropogenic interaction (Factor 3) were higher than other identified OS species (Fig. 6). A relatively high loading of GAS (0.48) was also observed in Factor 2 (Table S2). The PCA result provided observational evidence on the additional sources of monoterpene-OSs/NOSs and C_2−C_3 OSs from transported anthropogenic pollutants over marginal seas. During the autumn cruise, higher OS concentration levels were observed when higher EC concentrations occurred, which also indicated the additional contribution of OSs by anthropogenic sources (Fig. 7b).

This work quantified and characterized the atmospheric OSs derived from isoprene and monoterpenes over the Asia marginal seas. The chemical nature and distribution of OSs were modified by the joint influence of oceanic biological emissions and transported continental pollutants. The results highlight the abundant formation of airborne OSs in summer, which is promoted by the elevated biogenic VOC emissions from surface ocean. During high biological activity periods, atmospheric OS levels could surpass the MSA concentrations in marine aerosols, which is a vital species in the well-known climate regulation via oceanic phytoplankton sulphur emissions (CLAW hypothesis). Shipboard observations over open ocean areas are needed to gain further understanding on the roles of OSs in modifying the sulfur cycle, biogenic VOC oxidation and regulating climate in marine boundary layer.

Author contribution

Y.W. designed and supervised the research. M.H. supervised and provided the instrumentations. S.L., Y.Z., Y.Y., Y.G., C.Y. and Y.J. conducted the measurements. S.L. analyzed the data. Y.C.W synthesized the standards. S.L. and Y. W. wrote the

https://doi.org/10.5194/egusphere-2025-2154
Preprint. Discussion started: 28 May 2025

manuscript with contributions from all co-authors.

Declaration of competing interest

The authors declare no conflict of interest.

Data availability

The dataset is available upon request from the corresponding author.

Acknowledgments

This study was supported by the National Key Research and Development Program of China (2022YFF0803000; 2024YFC2815800), the National Natural Science Foundation of China (42205103; 42411540229; 22306059), the Taishan Scholars of Shandong Province, China (tsqn202306101), the Shandong Provincial Natural Science Foundation (ZR2022QD105), the Fundamental Research Funds for the Central Universities (202441011), special fund of State Key Joint Laboratory of Environmental Simulation and Pollution Control (22K01ESPCP), and Science and Technology Planning Project of Hunan Province (2023JJ40128).

Data acquisition and filter sample collection were performed onboard Lanhai No. 101, implementing the open research cruise NORC2021-01 and NORC2022-01 supported by the NSFC Shiptime Sharing Project (Project Number: 42049901 and 42149901).

References

Andreae, M. O.: Ocean-atmosphere interactions in the global biogeochemical sulfur cycle, Mar. Chem., 30, 1–29, https://doi.org/10.1016/0304-4203(90)90059-L, 1990.

Andreae, M. O. and Rosenfeld, D.: Aerosol–cloud–precipitation interactions. Part 1. The nature and sources of cloud-active aerosols, Earth-Sci. Rev., 89, 13–41, https://doi.org/10.1016/j.earscirev.2008.03.001, 2008.

Armstrong, N. C., Chen, Y., Cui, T., Zhang, Y., Christensen, C., Zhang, Z., Turpin, B. J., Chan, M. N., Gold, A., Ault, A. P., and Surratt, J. D.: Isoprene epoxydiol-derived sulfated and nonsulfated oligomers suppress particulate mass loss during oxidative aging of secondary organic aerosol, Environ. Sci. Technol., 56, 16611–16620, https://doi.org/10.1021/acs.est.2c03200, 2022.

Ayers, G. P. and Gras, J. L.: Seasonal relationship between cloud condensation nuclei and aerosol methanesulphonate in marine air, Nature, 353, 834–835, https://doi.org/10.1038/353834a0, 1991.

Ayers, G. P., Cainey, J. M., Gillett, R. W., and Ivey, J. P.: Atmospheric sulphur and cloud condensation nuclei in marine air in the Southern Hemisphere, Philos. Trans. R. Soc. B, 352, 203–211, https://doi.org/10.1098/rstb.1997.0015, 1997.

Bao, H., Niggemann, J., Luo, L., Dittmar, T., and Kao, S.-J.: Molecular composition and origin of water-soluble organic matter in marine aerosols in the Pacific off China, Atmos. Environ., 191, 27–35, https://doi.org/10.1016/j.atmosenv.2018.07.059, 2018.

https://doi.org/10.5194/egusphere-2025-2154
Preprint. Discussion started: 28 May 2025

Brüggemann, M., Hayeck, N., and George, C.: Interfacial photochemistry at the ocean surface is a global source of organic vapors and aerosols, Nat. Commun., 9, 2101, https://doi.org/10.1038/s41467-018-04528-7, 2018.

Brüggemann, M., Xu, R., Tilgner, A., Kwong, K. C., Mutzel, A., Poon, H. Y., Otto, T., Schaefer, T., Poulain, L., Chan, M. N., and Herrmann, H.: Organosulfates in ambient aerosol: State of knowledge and future research directions on formation, abundance, fate, and importance, Environ. Sci. Technol., 54, 3767–3782, https://doi.org/10.1021/acs.est.9b06751, 2020.

Bryant, D. J., Elzein, A., Newland, M., White, E., Swift, S., Watkins, A., Deng, W., Song, W., Wang, S., Zhang, Y., Wang, X., Rickard, A. R., and Hamilton, J. F.: Importance of oxidants and temperature in the formation of biogenic organosulfates and nitrooxy organosulfates, ACS Earth Space Chem., 5, 2291–2306, https://doi.org/10.1021/acsearthspacechem.1c00204, 2021.

Bryant, D. J., Nelson, B. S., Swift, S. J., Budisulistiorini, S. H., Drysdale, W. S., Vaughan, A. R., Newland, M. J., Hopkins, J. R., Cash, J. M., Langford, B., Nemitz, E., Acton, W. J. F., Hewitt, C. N., Mandal, T., Gurjar, B. R., Shivani, Gadi, R., Lee, J. D., Rickard, A. R., and Hamilton, J. F.: Biogenic and anthropogenic sources of isoprene and monoterpenes and their secondary organic aerosol in Delhi, India, Atmos. Chem. Phys., 23, 61–83, https://doi.org/10.5194/acp-23-61-2023, 2023.

Cai, D., Wang, X., Chen, J., and Li, X.: Molecular characterization of organosulfates in highly polluted atmosphere using ultra-high-resolution mass spectrometry, J. Geophys. Res., 125, e2019JD032253, https://doi.org/10.1029/2019JD032253, 2020.

Charlson, R. J., Lovelock, J. E., Andreae, M. O., and Warren, S. G.: Oceanic phytoplankton, atmospheric sulphur, cloud albedo and climate, Nature, 326, 655–661, https://doi.org/10.1038/326655a0, 1987.

Chen, Y., Zhang, Y., Lambe, A. T., Xu, R., Lei, Z., Olson, N. E., Zhang, Z., Szalkowski, T., Cui, T., Vizuete, W., Gold, A., Turpin, B. J., Ault, A. P., Chan, M. N., and Surratt, J. D.: Heterogeneous hydroxyl radical oxidation of isoprene-epoxydiol-derived methyltetrol sulfates: Plausible formation mechanisms of previously unexplained organosulfates in ambient fine aerosols, Environ. Sci. Technol. Lett., 7, 460–468, https://doi.org/10.1021/acs.estlett.0c00276, 2020.

Claeys, M., Wang, W., Vermeylen, R., Kourtchev, I., Chi, X., Farhat, Y., Surratt, J. D., Gómez-González, Y., Sciare, J., and Maenhaut, W.: Chemical characterisation of marine aerosol at Amsterdam Island during the austral summer of 2006–2007, J. Aerosol Sci., 41, 13–22, https://doi.org/10.1016/j.jaerosci.2009.08.003, 2010

Conte, L., Szopa, S., Aumont, O., Gros, V., and Bopp, L.: Sources and sinks of isoprene in the global open ocean: Simulated patterns and emissions to the atmosphere, J. Geophys. Res., 125, e2019JC015946, https://doi.org/10.1029/2019JC015946, 2020.

Cooke, M. E., Armstrong, N. C., Lei, Z., Chen, Y., Waters, C. M., Zhang, Y., Buchenau, N. A., Dibley, M. Q., Ledsky, I. R., Szalkowski, T., Lee, J. Y., Baumann, K., Zhang, Z., Vizuete, W., Gold, A., Surratt, J. D., and Ault, A. P.: Organosulfate formation in proxies for aged sea spray aerosol: Reactive uptake of isoprene epoxydiols to acidic sodium sulfate, ACS Earth Space Chem., 6, 2790–2800, https://doi.org/10.1021/acsearthspacechem.2c00156, 2022.

Cui, L., Xiao, Y., Hu, W., Song, L., Wang, Y., Zhang, C., Fu, P., and Zhu, J.: Enhanced dataset of global marine isoprene emissions from biogenic and photochemical processes for the period 2001–2020, Earth Syst. Sci. Data, 15, 5403–5425, https://doi.org/10.5194/essd-15-5403-2023, 2023.

Cui, T., Green, H. S., Selleck, P. W., Zhang, Z., O'Brien, R. E., Gold, A., Keywood, M., Kroll, J. H., and Surratt, J. D.: Chemical characterization of isoprene- and monoterpene-derived secondary organic aerosol tracers in remote marine aerosols over a quarter century, ACS Earth Space Chem., 3, 935–946, https://doi.org/10.1021/acsearthspacechem.9b00061, 2019.

https://doi.org/10.5194/egusphere-2025-2154
Preprint. Discussion started: 28 May 2025

Fu, T.-M., Jacob, D. J., Wittrock, F., Burrows, J. P., Vrekoussis, M., and Henze, D. K.: Global budgets of atmospheric glyoxal and methylglyoxal, and implications for formation of secondary organic aerosols, J. Geophys. Res., 113, D15303, https://doi.org/10.1029/2007JD009505, 2008.

Fu, P., Kawamura, K., and Miura, K.: Molecular characterization of marine organic aerosols collected during a round-the-world cruise, J. Geophys. Res., 116, D13302, https://doi:10.1029/2011JD015604, 2011.

Guo, T., Guo, Z., Wang, J., Feng, J., Gao, H., and Yao, X.: Tracer-based investigation of organic aerosols in marine atmospheres from marginal seas of China to the northwest Pacific Ocean, Atmos. Chem. Phys., 20, 5055–5070, https://doi.org/10.5194/acp-20-5055-2020, 2020.

Hawkins, L. N., Russell, L. M., Covert, D. S., Quinn, P. K., and Bates, T. S.: Carboxylic acids, sulfates, and organosulfates in processed continental organic aerosol over the southeast Pacific Ocean during VOCALS-REx 2008, J. Geophys. Res., 115, D13201, https://doi.org/10.1029/2009JD013276, 2010.

He, Q.-F., Ding, X., Wang, X.-M., Yu, J.-Z., Fu, X.-X., Liu, T.-Y., Zhang, Z., Xue, J., Chen, D.-H., Zhong, L.-J., and Donahue, N. M.: Organosulfates from pinene and isoprene over the Pearl River Delta, South China: Seasonal variation and implication in formation mechanisms, Environ. Sci. Technol., 48, 9236–9245, https://doi.org/10.1021/es501299v, 2014.

Hettiyadura, A. P. S., Stone, E. A., Kundu, S., Baker, Z., Geddes, E., Richards, K., and Humphry, T.: Determination of atmospheric organosulfates using HILIC chromatography with MS detection, Atmos. Meas. Tech., 8, 2347–2358, https://doi.org/10.5194/amt-8-2347-2015, 2015.

Hettiyadura, A. P. S., Jayarathne, T., Baumann, K., Goldstein, A. H., de Gouw, J. A., Koss, A., Keutsch, F. N., Skog, K., and Stone, E. A.: Qualitative and quantitative analysis of atmospheric organosulfates in Centreville, Alabama, Atmos. Chem. Phys., 17, 1343–1359, https://doi:10.5194/acp-17-1343-2017, 2017.

Hettiyadura, A. P. S., Al-Naiema, I. M., Hughes, D. D., Fang, T., and Stone, E. A.: Organosulfates in Atlanta, Georgia: anthropogenic influences on biogenic secondary organic aerosol formation, Atmos. Chem. Phys., 19, 3191–3206, https://doi.org/10.5194/acp-19-3191-2019, 2019.

Hu, Q.-H., Xie, Z.-Q., Wang, X.-M., Kang, H., He, Q.-F., and Zhang, P.: Secondary organic aerosols over oceans via oxidation of isoprene and monoterpenes from Arctic to Antarctic, Sci. Rep., 3, 2280, https://doi.org/10.1038/srep02280, 2013.

Huang, R.-J., Cao, J., Chen, Y., Yang, L., Shen, J., You, Q., Wang, K., Lin, C., Xu, W., Gao, B., Li, Y., Chen, Q., Hoffmann, T., O'Dowd, C. D., Bilde, M., and Glasius, M.: Organosulfates in atmospheric aerosol: synthesis and quantitative analysis of $PM_{2.5}$ from Xi'an, northwestern China, Atmos. Meas. Tech., 11, 3447–3456, https://doi.org/10.5194/amt-11-3447-2018, 2018.

Kanellopoulos, P. G., Kotsaki, S. P., Chrysochou, E., Koukoulakis, K., Zacharopoulos, N., Philippopoulos, A., and Bakeas, E.: $PM_{2.5}$-bound organosulfates in two Eastern Mediterranean cities: The dominance of isoprene organosulfates, Chemosphere, 297, 134103, https://doi.org/10.1016/j.chemosphere.2022.134103, 2022.

Kettle, A. J. and Andreae, M. O.: Flux of dimethylsulfide from the oceans: A comparison of updated data sets and flux models, J. Geophys. Res., 105, 26793–26808, https://doi.org/10.1029/2000JD900252, 2000.

Kloster, S., Feichter, J., Maier-Reimer, E., Six, K. D., Stier, P., and Wetzel, P.: DMS cycle in the marine ocean-atmosphere system – a global model study, Biogeosciences, 3, 29–51, https://doi.org/10.5194/bg-3-29-2006, 2006.

Li, H., Zheng, B., Lei, Y., Hauglustaine, D., Chen, C., Lin, X., Zhang, Y., Zhang, Q., and He, K.: Trends and drivers of anthropogenic NO_x emissions in China since 2020, Environ. Sci. Ecotechnology, 21, 100425, https://doi.org/10.1016/j.ese.2024.100425, 2024.

Li, J., Michalski, G., Davy, P., Harvey, M., Katzman, T., and Wilkins, B.: Investigating source contributions of size-aggregated aerosols collected in Southern Ocean and Baring Head, New Zealand using sulfur isotopes, Geophys. Res. Lett., 45, 3717–3727, https://doi.org/10.1002/2018GL077353, 2018.

Li, J., Carlson, B. E., Yung, Y. L., Lv, D., Hansen, J., Penner, J. E., Liao, H., Ramaswamy, V., Kahn, R. A., Zhang, P., Dubovik, O., Ding, A., Lacis, A. A., Zhang, L., and Dong, Y.: Scattering and absorbing aerosols in the climate system, Nat. Rev. Earth Environ., 3, 363–379, https://doi.org/10.1038/s43017-022-00296-7, 2022.

Liao, J., Froyd, K. D., Murphy, D. M., Keutsch, F. N., Yu, G., Wennberg, P. O., St. Clair, J. M., Crounse, J. D., Wisthaler, A., Mikoviny, T., Jimenez, J. L., Campuzano-Jost, P., Day, D. A., Hu, W., Ryerson, T. B., Pollack, I. B., Peischl, J., Anderson, B. E., Ziemba, L. D., Blake, D. R., Meinardi, S., and Diskin, G.: Airborne measurements of organosulfates over the continental U.S., J. Geophys. Res., 120, 2990–3005, https://doi.org/10.1002/2014JD022378, 2015.

Meade, L. E., Riva, M., Blomberg, M. Z., Brock, A. K., Qualters, E. M., Siejack, R. A., Ramakrishnan, K., Surratt, J. D., and Kautzman, K. E.: Seasonal variations of fine particulate organosulfates derived from biogenic and anthropogenic hydrocarbons in the mid-Atlantic United States, Atmos. Environ., 145, 405–414, https://doi.org/10.1016/j.atmosenv.2016.09.028, 2016.

Nguyen, Q. T., Christensen, M. K., Cozzi, F., Zare, A., Hansen, A. M. K., Kristensen, K., Tulinius, T. E., Madsen, H. H., Christensen, J. H., Brandt, J., Massling, A., Nøjgaard, J. K., and Glasius, M.: Understanding the anthropogenic influence on formation of biogenic secondary organic aerosols in Denmark via analysis of organosulfates and related oxidation products, Atmos. Chem. Phys., 14, 8961–8981, https://doi.org/10.5194/acp-14-8961-2014, 2014.

Ooki, A., Nomura, D., Nishino, S., Kikuchi, T., and Yokouchi, Y.: A global-scale map of isoprene and volatile organic iodine in surface seawater of the Arctic, Northwest Pacific, Indian, and Southern Oceans, J. Geophys. Res., 120, 4108–4128, https://doi.org/10.1002/2014JC010519, 2015.

Quinn, P. K. and Bates, T. S.: The case against climate regulation via oceanic phytoplankton sulphur emissions, Nature, 480, 51–56, https://doi.org/10.1038/nature10580, 2011.

Riva, M., Budisulistiorini, S. H., Zhang, Z., Gold, A., and Surratt, J. D.: Chemical characterization of secondary organic aerosol constituents from isoprene ozonolysis in the presence of acidic aerosol, Atmos. Environ., 130, 5–13, https://doi.org/10.1016/j.atmosenv.2015.06.027, 2016.

Schindelka, J., Iinuma, Y., Hoffmann, D., and Herrmann, H.: Sulfate radical-initiated formation of isoprene-derived organosulfates in atmospheric aerosols, Faraday Discuss., 165, 237–259, https://doi.org/10.1039/C3FD00042G, 2013.

Surratt, J. D., Gómez-González, Y., Chan, A. W. H., Vermeylen, R., Shahgholi, M., Kleindienst, T. E., Edney, E. O., Offenberg, J. H., Lewandowski, M., Jaoui, M., Maenhaut, W., Claeys, M., Flagan, R. C., and Seinfeld, J. H.: Organosulfate formation in biogenic secondary organic aerosol, J. Phys. Chem. A, 112, 8345–8378, https://doi.org/10.1021/jp802310p, 2008.

Surratt, J. D., Chan, A. W. H., Eddingsaas, N. C., Chan, M., Loza, C. L., Kwan, A. J., Hersey, S. P., Flagan, R. C., Wennberg, P. O., and Seinfeld, J. H.: Reactive intermediates revealed in secondary organic aerosol formation from isoprene, Proc. Natl. Acad. Sci., 107, 6640–6645, https://doi.org/10.1073/pnas.0911114107, 2010.

Wang, H., Ma, X., Tan, Z., Wang, H., Chen, X., Chen, S., Gao, Y., Liu, Y., Liu, Y., Yang, X., Yuan, B., Zeng, L., Huang, C., Lu, K., and Zhang, Y.: Anthropogenic monoterpenes aggravating ozone pollution, Natl. Sci. Rev., 9, nwac103, https://doi.org/10.1093/nsr/nwac103, 2022a.

Wang, Y., Hu, M., Guo, S., Wang, Y., Zheng, J., Yang, Y., Zhu, W., Tang, R., Li, X., Liu, Y., Breton, M. L., Du, Z., Shang, D., Wu, Y., Wu, Z., Song, Y., Lou, S., Hallquist, M., and Yu, J.: The secondary formation of organosulfates under interactions between biogenic emissions and anthropogenic pollutants in summer in Beijing, Atmos. Chem. Phys., 18, 10693–10713, https://doi.org/10.5194/acp-18-10693-2018, 2018.

Wang, Y., Hu, M., Wang, Y.-C., Li, X., Fang, X., Tang, R., Lu, S., Wu, Y., Guo, S., Wu, Z., Hallquist, M., and Yu, J. Z.: Comparative study of particulate organosulfates in contrasting atmospheric environments: Field evidence for the significant influence of anthropogenic sulfate and NO_x, Environ. Sci. Technol. Lett., 7, 787–794, https://doi.org/10.1021/acs.estlett.0c00550, 2020.

Wang, Y., Zhao, Y., Wang, Y., Yu, J.-Z., Shao, J., Liu, P., Zhu, W., Cheng, Z., Li, Z., Yan, N., and Xiao, H.: Organosulfates in atmospheric aerosols in Shanghai, China: seasonal and interannual variability, origin, and formation mechanisms, Atmos. Chem. Phys., 21, 2959–2980, https://doi.org/10.5194/acp-21-2959-2021, 2021.

Wang, Y., Ma, Y., Kuang, B., Lin, P., Liang, Y., Huang, C., and Yu, J. Z.: Abundance of organosulfates derived from biogenic volatile organic compounds: Seasonal and spatial contrasts at four sites in China, Sci. Total Environ., 806, 151275, https://doi.org/10.1016/j.scitotenv.2021.151275, 2022b.

Wang, Y., Liang, S., Le Breton, M., Wang, Q. Q., Liu, Q., Ho, C. H., Kuang, B. Y., Wu, C., Hallquist, M., Tong, R., and Yu, J. Z.: Field observations of C_2 and C_3 organosulfates and insights into their formation mechanisms at a suburban site in Hong Kong, Sci. Total Environ., 904, 166851, https://doi.org/10.1016/j.scitotenv.2023.166851, 2023a.

Wang, Y., Zhang, Y., Li, W., Wu, G., Qi, Y., Li, S., Zhu, W., Yu, J. Z., Yu, X., Zhang, H.-H., Sun, J., Wang, W., Sheng, L., Yao, X., Gao, H., Huang, C., Ma, Y., and Zhou, Y.: Important roles and formation of atmospheric organosulfates in marine organic aerosols: Influence of Phytoplankton Emissions and Anthropogenic Pollutants, Environ. Sci. Technol., 57, 10284–10294, https://doi.org/10.1021/acs.est.3c01422, 2023b.

Worton, D. R., Surratt, J. D., LaFranchi, B. W., Chan, A. W. H., Zhao, Y., Weber, R. J., Park, J.-H., Gilman, J. B., de Gouw, J., Park, C., Schade, G., Beaver, M., Clair, J. M. St., Crounse, J., Wennberg, P., Wolfe, G. M., Harrold, S., Thornton, J. A., Farmer, D. K., Docherty, K. S., Cubison, M. J., Jimenez, J.-L., Frossard, A. A., Russell, L. M., Kristensen, K., Glasius, M., Mao, J., Ren, X., Brune, W., Browne, E. C., Pusede, S. E., Cohen, R. C., Seinfeld, J. H., and Goldstein, A. H.: Observational insights into aerosol formation from isoprene, Environ. Sci. Technol., 47, 11403–11413, https://doi.org/10.1021/es4011064, 2013.

Wu, Y.-C., Li, J.-L., Wang, J., Zhuang, G.-C., Liu, X.-T., Zhang, H.-H., and Yang, G.-P.: Occurance, emission and environmental effects of non-methane hydrocarbons in the Yellow Sea and the East China Sea, Environ. Pollut., 270, 116305, https://doi.org/10.1016/j.envpol.2020.116305, 2021.

Xu, L., Guo, H., Boyd, C. M., Klein, M., Bougiatioti, A., Cerully, K. M., Hite, J. R., Isaacman-VanWertz, G., Kreisberg, N. M., Knote, C., Olson, K., Koss, A., Goldstein, A. H., Hering, S. V., de Gouw, J., Baumann, K., Lee, S.-H., Nenes, A., Weber, R. J., and Ng, N. L.: Effects of anthropogenic emissions on aerosol formation from isoprene and monoterpenes in the southeastern United States, Proc. Natl. Acad. Sci., 112, 37–42, https://doi.org/10.1073/pnas.1417609112, 2015.

Yan, S.-B., Xu, G.-B., Zhang, H.-H., Wang, J., Xu, F., Gao, X.-X., Zhang, J.-W., Wu, J.-W., and Yang, G.-P.: Factors controlling DMS emission and atmospheric sulfate aerosols in the western Pacific continental sea, J. Geophys. Res., 129, e2024JC020886, https://doi.org/10.1029/2024JC020886, 2024.

https://doi.org/10.5194/egusphere-2025-2154
Preprint. Discussion started: 28 May 2025

Ye, Y., Zhan, H., Yu, X., Li, J., Wang, X., and Xie, Z.: Detection of organosulfates and nitrooxy-organosulfates in Arctic and Antarctic atmospheric aerosols, using ultra-high resolution FT-ICR mass spectrometry, Sci. Total Environ., 767, 144339, https://doi.org/10.1016/j.scitotenv.2020.144339, 2021.