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Measurement report: Molecular-level insights into the seasonal formation of organosulfates in a mountain atmosphere

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Abstract: Organosulfates (OSs) are important components of secondary organic aerosol, yet their molecular composition and formation processes in mountain atmospheres remain poorly constrained. In this study, $PM_{2.5}$ samples collected during summer and winter at a high-altitude background site at Wuyi Mountain, southeastern China, were analyzed using high-performance liquid chromatography coupled with orbitrap mass spectrometry (HPLC-Orbitrap MS) to examine how seasonal changes in precursor availability and atmospheric processing influence OS molecular composition. Clear seasonal differences were observed in the molecular composition and structural characteristics. Summer OSs were enriched in more highly oxygenated species (with $\overline{OS}_c$>-1 accounting for 72% of the total OS signal abundance), whereas winter OSs were characterized by larger (C_5–C_{23}), less oxygenated (O_4–O_7), and more unsaturated molecules, reflecting more diverse precursor influences. Among 35 structurally identified OSs, isoprene- and monoterpene-derived species were enriched in summer, while aliphatic-like OSs and monoterpene-derived nitrooxy-OSs were more abundant in winter, indicating seasonal variations in precursor availability and the relative importance of atmospheric processing. Mechanistic analysis suggested that summer OS formation was more closely associated with sulfate-rich and highly acidic conditions favorable for multiphase processing, whereas wintertime OS formation reflected greater influences from elevated atmospheric oxidants and nitrogen-related chemistry. Regional transport further contributed to wintertime OS composition by increasing anthropogenic precursor inputs and promoting atmospheric aging. These findings demonstrate that seasonal OS variability in mountain environments reflects the interplay between precursor availability and atmospheric processing, providing field-based molecular evidence for the coupling of biogenic and anthropogenic influences in secondary organic aerosol formation and evolution.

Keywords: Organosulfates, Molecular composition, Seasonal variation, Multiphase chemistry, Mountain atmosphere

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1. Introduction

Organosulfates (OSs) are an important class of sulfur-containing components in secondary organic aerosol (SOA), accounting for 3–30% of organic matter mass in ambient fine particles (Mazzoleni et al. 2010; Tolocka et al. 2012; Hettiyadura et al. 2018). They may also contribute up to 12% of particulate total sulfur, playing a significant role in the global biogeochemical sulfur cycle (Lukács et al. 2009). OSs are primarily formed through the oxidation of volatile organic compounds (VOCs), followed by sulfate esterification and related aqueous-phase or multiphase reactions involving SO_2 or H_2SO_4. Due to the presence of sulfate functional groups ($–OSO_3^-$), OSs generally exhibit high hygroscopicity and thermal instability, influencing aerosol water solubility and subsequent chemical transformations (Brüggemann et al. 2020). OSs may also modify particle surface properties and cloud condensation nuclei activity, thereby influencing aerosol-cloud interactions and climate-relevant processes (Lin et al. 2013; Hansen et al. 2015; Estillore et al. 2016). Despite important insights from laboratory and field studies, the processes governing the abundance and molecular composition of OSs in the ambient atmosphere remain incompletely understood.

OSs can be broadly classified into biogenic and anthropogenic types based on their precursor sources. Biogenic OSs are mainly derived from the oxidation of plant-emitted isoprene and terpenes (Stone et al. 2012; He et al. 2014), whereas anthropogenic OSs have been linked to aromatic compounds, alkanes and unsaturated fatty acids emitted from fossil fuel combustion, biomass burning, and cooking activities (Riva et al. 2015; Riva et al. 2016; Blair et al. 2017). A variety of formation processes have been proposed for different OS classes. For example, isoprene-derived OSs are closely linked to the acid-catalyzed ring-opening reactions of epoxide intermediates on sulfate-containing particles (Lin et al. 2013; Surratt et al. 2008). Chamber studies have also shown that monoterpenes (e.g., β-pinene) can be oxidized by O_3 to form alcohol intermediates that subsequently undergo sulfate esterification in the particle phase (Iinuma et al. 2007). Additional OS formation may occur through heterogeneous reactions of SO_2 with monoterpene-derived SOA, unsaturated fatty acids, or organic peroxides (Passananti et al. 2016; Yao et al. 2019; Zhu et al. 2019). For nitrooxy-OSs, nighttime NO_3 radical-initiated oxidation of VOCs has been identified as an important source of organic nitrate and peroxy intermediates that may subsequently undergo particle-phase processing (Wang et al. 2020; Yang et al. 2023). The relative importance of these processes depends

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on precursor composition and environmental conditions, including sulfate availability, aerosol acidity, aerosol liquid water content (ALWC), and atmospheric oxidants (e.g., O_3) (Duporté et al. 2020; Aoki et al. 2020; Duporté et al. 2016). However, only a limited number of laboratory-proposed processes have been evaluated through field observations, and a substantial fraction of ambient OSs remains structurally unresolved (Chen et al. 2021). Molecular-level investigation that link OS composition with precursor sources and atmospheric conditions are therefore needed to better understand their formation in the ambient atmosphere.

High-altitude mountain sites provide particularly useful natural environments for investigating OS formation, which are characterized by abundant biogenic VOC emissions, persistently high relative humidity (RH), and frequent influence from regional transport of anthropogenic pollutants (Zhao et al. 2020; Wang et al. 2021a; Cao et al. 2022). These conditions can promote photochemical aging, gas-particle partitioning, and aqueous-phase or heterogeneous reactions (Hallquist et al. 2009; McNeill 2015). Elevated ALWC associated with high RH may enhance the uptake and diffusion of gaseous and dissolved precursors, thereby facilitating sulfate esterification and related OS formation. Summer observations in the Qinling Mountains, for example, indicated that elevated RH favored the formation of monoterpene- and long-chain alkane-derived OSs and that aqueous-phase reactions contributed to multigenerational oxidation (Li et al. 2024). At the same time, the coexistence of biogenic precursors and regionally transported anthropogenic pollutants may increase the diversity of OS precursors and atmospheric processing. Consequently, OS molecular composition in mountain environments may reflect the combined influences of precursor emissions, atmospheric oxidation, multiphase chemistry, and regional transport. However, how the relative importance of these factors varies seasonally and influences OS formation remains poorly understood.

In this study, $PM_{2.5}$-bound OSs collected during summer and winter at a high-altitude background site on Wuyi Mountain were characterized using high-performance liquid chromatography coupled with orbitrap mass spectrometry (HPLC-Orbitrap MS). The objectives were to (1) characterize the molecular composition, structural characteristics, and seasonal variability of OSs; (2) identify major OS classes and determine their relative contributions in summer and winter; and (3) investigate the possible formation processes and regional transport influences of different OS groups

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by integrating molecular observations with sulfate, aerosol acidity, ALWC, oxidants, and backward-trajectory analysis.

2. Materials and methods

2.1 Sample Collection

Ambient $PM_{2.5}$ samples were collected from 3rd June to 26th July 2016 and from 27th December 2016 to 24th February 2017, representing summer and winter, respectively (Table S1 in the Supporting Information). The sampling site is located on the Wuyi Mountain (27.64°N, 117.99°E) in Fujian province. The samples were collected on pre-baked (450 °C, 3 h) quartz fiber filters (20.3 × 25.4 cm; Whatman, QM-A, Clifton, US) for 46 h using a high-volume $PM_{2.5}$ air sampler (Tisch TE-6070V-2.5-HVS, Cleveland, US) at a flow rate of 1.05 m^3 min^{-1}. After collection, the samples were wrapped in pre-baked (450 °C, 8 h) aluminum foil and stored at −20 °C until analysis.

2.2 Chemical Analysis

A portion of the filter sample was ultrasonically extracted with 3 mL of methanol for 20 min, repeated three times. The extracts were then filtered with a 0.22 μm PTFE pore syringe filter and concentrated to near dryness under a gentle stream of nitrogen. The residue was re redissolved in 70 μL of methanol, transferred into a 1 mL centrifuge tube, and centrifuged at 12000 rpm for 20 min. The supernatant was subsequently transferred into LC vials and analyzed within 24 h. The extracts were analyzed using a Dionex Ultimate 3000 UHPLC (Thermo Fisher Scientific, Waltham, US) coupled to an Orbitrap Exploris 120 mass spectrometer (Thermo Fisher Scientific, Waltham, US). A HSS T3 column (2.1 × 100 mm, 1.8 μm particle size) was used to separate the organics in the sample extracts. Details of the elution program are provided in the Supporting Information. The injection volume is 5 μL. The electrospray ionization source was operated in negative mode (ESI-) with a sheath gas flow of 35.0 L min^{-1}, capillary temperature of 320 °C, spray voltage of -3.2 kV, and mass resolution of 140,000. Full-scan data were acquired over the *m/z* range of 90–500.

2.3 Data Processing

The MS data were processed using Xcalibur 4.0 (Thermo Scientific) and the open-source software MZmine (http://mzmine.github.io/) to extract *m/z* values, molecular formulas, retention times, and peak areas of detected organic compounds. The

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molecular formula assignment was mainly based on detected *m/z* values with a mass tolerance of ±2 ppm, with elemental constraints set to 0.3 ≤ H/C ≤ 3.0, O/C ≤ 3.0, N/C ≤ 0.5, S/C ≤ 0.2. The double bond equivalent (DBE) of the assigned formulas (CcHhOoNnSs) was calculated as:

$$DBE=(2C+2-H+N)/2$$

where C, H, and N represent the number of carbon, hydrogen, and nitrogen atoms, respectively.

Potential OSs and nitrooxy-OSs were first screened based on elemental composition and oxygen content, and structural assignment was supported by diagnostic sulfate-related fragment ions, including HSO_4^- at *m/z* 96.9599, SO_4^- at *m/z* 95.9522, HSO_3^- at *m/z* 80.9651, and SO_3^- at *m/z* 79.9573, as well as characteristic neutral losses such as H_2SO_4. For nitrooxy-OSs, the additional presence of nitrogen-containing fragment ions (e.g., NO_2^-), together with sulfate-related fragments, provided supporting evidence for nitrogen-containing functional groups. Compound identities were assigned based on diagnostic spectral features and comparison with previously reported compounds in the literature. Because authentic standards were unavailable for most species, the reported abundances are based on relative signal intensities and are used primarily for comparative analysis across seasons and OS classes, assuming comparable ionization efficiencies among structurally related compounds.

2.4 Additional Measurements

Water-soluble inorganic ions were quantified by an ion chromatograph (IC, DIONEX, ICS2500/ICS2000); NO_2 was monitored by a NO_x analyzer (Thermo Fisher Scientific, model 42i). Meteorological parameters (RH and temperature) were provided by a weather station (Met one Instrument Inc.). In addition, the ALWC and aerosol pH were estimated using the thermodynamic model (ISORROPIA-II) with measured inorganic aerosol composition (NH_4^+, SO_4^{2-}, NO_3^-, and Cl^-), ambient temperature, and RH as input parameters (Fountoukis et al. 2007). The model was operated in metastable mode, assuming fully deliquescent particles without solid phases.

2.5 Backward-Trajectory and Cluster Analysis

Air-mass transport pathways were investigated using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory model (http://ready.arl.noaa.gov/HYSPLIT_traj.php). 72-h backward trajectories arriving at the Wuyi Mountain sampling site were calculated using the archived meteorological data obtained from the National Center for Environmental

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Prediction's Global Data Assimilation System (GDAS). The resulting trajectories were grouped using HYSPLIT trajectory cluster analysis, which classifies trajectories according to similarities in their spatial transport pathways. For each season, three trajectory clusters were retained, and the percentage assigned to each cluster represents the fraction of all calculated trajectories belonging to that cluster.

3. Results and discussion

3.1 Molecular Overview of Organic Sulfates

Fig. 1a summarizes the number of molecular formulas and signal abundance of organic compounds detected in $PM_{2.5}$ samples collected at Wuyi Mountain. A total of 2,283 and 3,253 molecular formulas were identified in summer and winter, respectively. Sulfur-containing compounds, mainly CHOS and CHONS species, accounted for more than 30% of all the detected formulas in both seasons, highlighting the importance of sulfur-containing organic compounds in mountain aerosol. Based on molecular formula counts, approximately 90% of CHOS and 30% of CHONS formulas satisfied the oxygen criterion required to accommodate the assignment of $-OSO_3H$ and $-ONO_2$ groups ($O/(4S + 3N) \geq 1$) (Fig. 1b, Fig. S1), suggesting that they were potential OSs or nitrooxy-OSs (Wang et al. 2016; Cai et al. 2020). In terms of signal abundance, potential OSs accounted for comparable fractions of total CHOS signals in summer and winter (> 92%), whereas nitrooxy-OSs contributed a larger fraction of CHONS signals in winter (~ 72%) than in summer (~ 61%). The total signal abundances of potential OSs and nitrooxy-OSs in winter were 1.4 and 2.1 times those in summer, respectively. These results indicate a greater wintertime contribution of sulfur- and nitrogen-containing organic compounds, particularly species associated with nitrogen-involved atmospheric processing.

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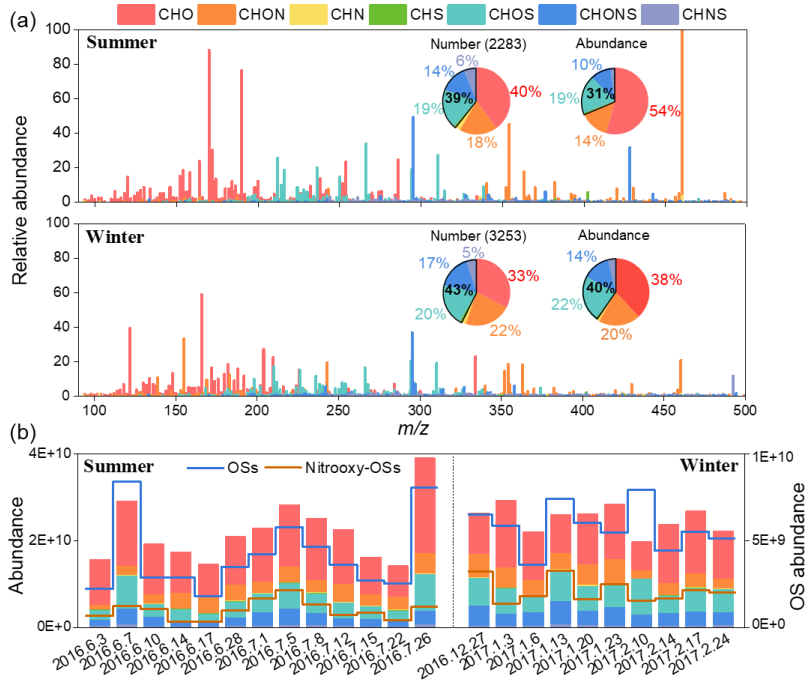

Figure 1. The number and abundance of detected organic compounds in summer and winter at Wuyi Mountain, with relative contributions of different elemental classes based on molecular number and signal abundance (a). Temporal variation in the abundance of the different elemental classes, potential OSs and nitrooxy-OSs (b).

Fig. 2a shows the molecular distributions of OSs and nitrooxy-OSs as functions of *m/z*, carbon number, and oxygen number. In both seasons, OS molecular mass generally increased with carbon number. However, clear seasonal differences were observed in molecular compositional space. Summer OSs were mainly distributed within the low-to-medium carbon-number range (C_5–C_{12}), whereas winter OSs extended to substantially higher carbon numbers, reaching C_{23}. As for oxygen-number distribution, summer OSs covered a wider range (O_4–O_{11}) and included a larger proportion of highly oxygenated species, whereas winter OSs were mainly concentrated between O_4 and O_7. The combination of higher carbon numbers and lower oxygen contents indicates that winter OSs occupied a distinct region of molecular compositional space characterized by larger and less oxygenated molecules. Nitrooxy-OSs exhibited comparatively narrower molecular distribution in both seasons, with carbon numbers mainly ranging from C_5 to C_{15} and oxygen numbers from O_7 to O_{11}. Compared with summer, winter nitrooxy-OSs displayed higher signal abundance and a more concentrated

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compositional distribution, suggesting enhanced formation and/or accumulation under winter conditions.

The Van Krevelen (VK) diagrams provide further information on the structural characteristics of OSs and nitrooxy-OSs (Fig. 2b). OSs occupied a broad region in H/C and (O−3S)/C space, indicating substantial compositional variability and the coexistence of aliphatic and unsaturated structures (Cai et al. 2020; Lin et al. 2012). In both seasons, OS signal abundance was dominated by compounds with H/C ratios greater than 1.0 and DBE values lower than 5, which accounted for 98% and 95% of the total OS signals in summer and winter, respectively. These features are consistent with sulfate esterification of relatively saturated or weakly unsaturated oxidation products, including those associated with biogenic precursors such as isoprene and monoterpenes (Thoma et al. 2025). Compared with summer, winter OSs showed a modest shift toward lower H/C and higher DBE values, indicating a greater contribution from more unsaturated molecular features (Mazzoleni et al. 2012). Nitrooxy-OSs occupied a narrower VK space and exhibited substantially higher signal abundances in winter, further supporting their formation under nitrogen-involved oxidation conditions. Moreover, OSs spanned a broad range of carbon oxidation state ($\overline{OS}_c$) from -2 to 2, reflecting substantial variability in molecular oxidation levels (Fig. S2). Highly oxidized OSs ($\overline{OS}_c$>-1) accounted for 80% of the molecular number and 72% of the signal abundance in summer, compared with 64% and 56%, respectively, in winter. This result is consistent with a higher degree of molecular oxidation and active sulfate esterification of biogenic oxidation products (Kroll et al. 2011). Nitrooxy-OSs exhibited moderate-to-high carbon oxidation states, with $\overline{OS}_c$ values mainly between -1 and 2, and showed higher molecular counts and signal abundances in winter than in summer.

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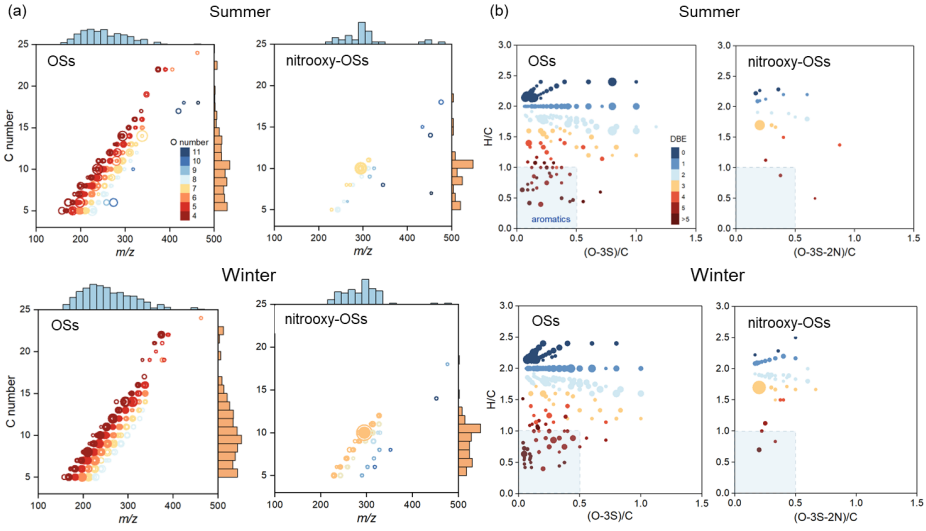

Figure 2. Distributions of molecular mass, carbon number, oxygen atom number (a) and Van Krevelen diagrams for OSs and nitrooxy-OSs in summer and winter.

3.2 Molecular Identification and Seasonal Variation

Based on HPLC-Orbitrap MS fragmentation patterns, 35 compounds with diagnostic sulfate ester features were structurally assigned (Fig. 3a, Table S1). These compounds were grouped into five classes according to molecular characteristics and likely precursor associations: OSs containing two or three carbon atoms (C_2/C_3 OS), isoprene-derived OSs (OSi), monoterpene-derived OSs (OSm), monoterpene-derived nitrooxy-OSs (NOSm), and aliphatic-like OSs (OSali). OSi was characterized by ions at *m/z* 198.9913 ($C_4H_7O_7S^-$) and *m/z* 197.0120 ($C_5H_9O_7S^-$), which are commonly attributed to products formed via reactive uptake and subsequent sulfate esterification of isoprene oxidation intermediates (Cai et al. 2020). OSm was mainly represented by *m/z* 249.0791 ($C_{10}H_{17}O_5S^-$), a species linked to monoterpene oxidation products such as those derived from limonene. The NOSm group was represented by *m/z* 294.0646 ($C_{10}H_{16}NO_7S^-$), which has been associated with monoterpene oxidation under NO_x-rich conditions or nighttime NO_3-radical chemistry, followed by incorporation of nitrate- and sulfate-containing functionalities (Wang et al. 2020; Wang et al. 2021b). For the C_2/C_3 OS group, the most abundant ions included *m/z* 152.9860 ($C_3H_5O_5S^-$), *m/z* 154.9651 ($C_2H_3O_6S^-$), and *m/z* 168.9808 ($C_3H_5O_6S^-$), corresponding to hydroxyacetone sulfate (HAS), glycolic acid sulfate (GAS), and lactic acid sulfate (LAS), respectively. These low-carbon-number OSs are generally considered products of highly oxidized biogenic and anthropogenic precursors, including carboxylic acids and hydroxyl-

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ketone, and their formation is often associated with aqueous-phase or multiphase reactions (Wang et al. 2023). OSali species were characterized by relatively high H/C ratios and low degrees of unsaturation, consistent with aliphatic-like molecular structures. They may originate from the oxidation of saturated or weakly functionalized organic precursors followed by sulfate esterification, although their exact sources remain uncertain. The molecular composition of OSali also differed between seasons: a species with *m/z* 209.0850 ($C_8H_{17}O_4S^-$) was more prominent in summer, whereas a species with *m/z* 153.0224 ($C_4H_9O_4S^-$) dominated in winter.

The relative contributions of the five OS groups varied substantially between seasons (Fig. 3b). OSi and OSm showed higher relative abundances in summer than in winter, consistent with enhanced emissions of isoprene and monoterpene under warmer and more irradiated conditions. The sulfur oxidation ratio (SOR, calculation shown in the Text S3) was higher in summer (0.83 ± 0.08) than in winter (0.42 ± 0.23; Fig. 3c, Fig. S3), indicating more efficient conversion of SO_2 to sulfate during summer despite lower anthropogenic SO_2 emissions (Duan et al. 2026). The resulting increase in sulfate levels likely enhanced sulfate-related transformations and multiphase processing. C_2/C_3 OS were strongly correlated with OSi in both seasons (R^2=0.90, Fig. S4), suggesting related precursor sources and/or similar atmospheric processing. However, the ratios of C_2/C_3 OS to OSi were approximately twice as higher in winter as in summer, indicating a greater relative contribution of C_2/C_3 OS under winter conditions. In contrast, NOSm exhibited higher abundances in winter and contributed substantially to total OS signals in several winter samples (Fig. S5). This enhancement coincided with higher NO_2 and particulate nitrate (NO_3^-) concentrations, as well as the elevated nitrogen oxidation rates in winter (NOR, average 0.54±0.10) than in summer (0.01±0.00), indicating a more nitrogen-rich secondary aerosol environment. Lower winter temperatures may further favor the partitioning and persistence of nitrate-containing species in the particle phase. Together, these observations suggest that NO_x-related oxidation and subsequent multiphase processing contributed more strongly to the formation and accumulation of nitrooxy-OSs during winter.

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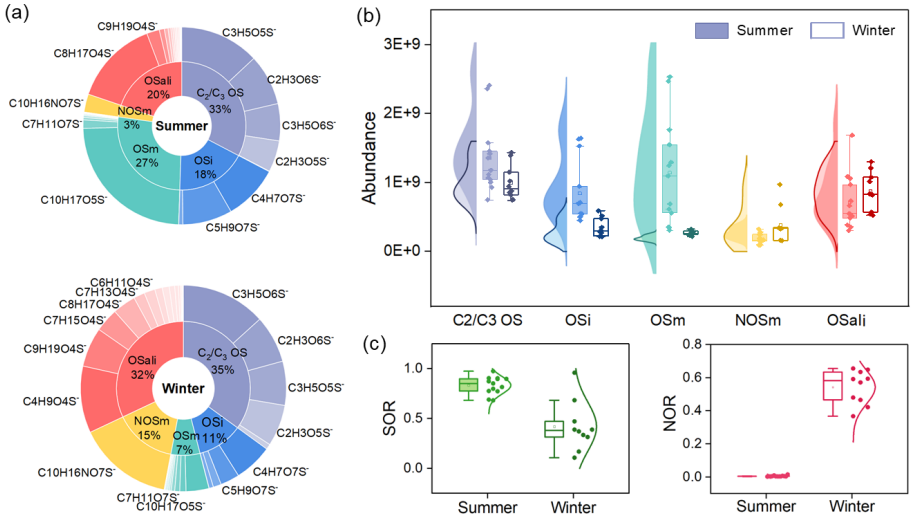

Figure 3. Seasonal variations in the chemical composition (a) and abundance (b) of five group OSs, together with SOR and NOR.

3.3 Possible Formation Processes and Transport Influence

Seasonal variations in atmospheric composition and meteorological conditions provide useful constraints on the processes associated with OS formation (Fig. 4). Difference in sulfate availability, aerosol acidity, oxidant levels, and ALWC indicated distinct atmospheric environments in summer and winter. Summer aerosols were more acidic and sulfate-rich, with an average pH of 2.8±0.7 and SO_4^{2-} concentration of 3.6±2.8 μg m^{-3}, compared with 3.7±0.7 and 1.58±0.8 μg m^{-3}, respectively, in winter (Fig. S6). Greater aerosol acidity during the warm season has also been reported in previous studies, where temperature-dependent ammonia partitioning and reduced particle neutralization contributed to the seasonal variation in aerosol pH (Ding et al. 2019; Ren et al. 2021). Although O_x (NO_2+O_3) concentrations were lower in summer (75.0±16.8 μg m^{-3}), the higher temperatures (26.3±2.2 °C) likely favored photochemical oxidation and the production of oxygenated biogenic intermediates. Most OS groups (except NOSm) closely tracked SO_4^{2-} concentrations and showed inverse relationships with aerosol pH (Fig. 4a), indicating preferential occurrence under sulfate-rich and acidic conditions. C_2/C_3 OSs exhibited a particularly strong positive correlation with SO_4^{2-} (r = 0.8, p < 0.001; Fig. 4b, Fig. S7), consistent with sulfate-

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associated aqueous-phase or multiphase processing. Previous studies have proposed that oxygenated precursors containing adjacent carbonyl and hydroxyl groups can facilitate the formation of reactive intermediates (e.g., cyclic sulfate structures), which readily react with sulfur trioxide or sulfuric acid to form low-carbon-number OSs (Wang et al. 2023). OSi also showed positive associations with SO_4^{2-} and acidity ($r > 0.7$, $p < 0.01$), consistent with the uptake and acid-catalyzed conversion of isoprene oxidation products on sulfate-containing particles (Piletic et al. 2013; He et al. 2018). These observations suggest that sulfate availability and aerosol acidity may play an important role in shaping OS distribution by influencing environments (Nguyen et al. 2014; Xu et al. 2015). In comparison, OSm and OSali exhibited weaker relationships on SO_4^{2-} and acidity, implying that their variability may additionally depend on precursor-specific processes, including heterogeneous reactions of SO_2 with monoterpene-derived SOA or relatively reduced organic compounds (Yao et al. 2019; Zhu et al. 2019).

Summer was also characterized by high RH (82.5 ± 7.7%), and approximately 80% of the samples had ALWC-to-dry $PM_{2.5}$ mass ratios greater than 0.3 (Fig. S8), suggesting that particles were predominantly in liquid or near-liquid states (Liu et al. 2023). Such conditions can enhance molecular diffusion and the uptake of soluble gaseous precursors, thereby facilitating aqueous-phase and heterogeneous reactions (Liu et al. 2017; Wu et al. 2018; Liu et al. 2019). However, the weak correlations between OS abundance and ALWC indicate that particle-phase water was not the primary factor governing the temporal variability of OSs. Instead, ALWC likely provided a reaction medium whose effect depended on the simultaneous availability of organic precursors, sulfate, and acidity (Ervens et al. 2011). The negative correlations between several OSs and RH ($r < -0.7$, Fig. S9) further indicate that RH mainly reflects meteorological variability (e.g., cloud/fog influence). This pattern suggests that, although elevated RH promotes multiphase processing, this effect may diminish under persistently high-humidity conditions approaching saturation. Similar behavior has been reported in urban regions such as Shanghai (Wang et al. 2021b), where higher RH

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was associated with increased ALWC and elevated aerosol pH due to enhanced neutralization, thereby suppressing acid-catalyzed OS formation (Riva et al. 2016; Duporté et al. 2016). In addition, enhanced ALWC may also introduce additional physical effects, including increased aqueous partitioning, dilution, and changes in particle phase state, which can reduce the apparent particulate concentrations of OSs (Guo et al. 2015; Shiraiwa et al. 2017).

Winter samples exhibited higher O_x concentrations (95.9 ± 20.9 μg m^{-3}) despite lower temperatures (10.0±3.8 °C), reflecting an atmospheric environment with elevated NO_2 and O_3. NOSm showed a positive relationship with O_x, particularly at higher organic aerosol loadings (r = 0.75), suggesting that oxidant availability and the amount of particle-phase organic material jointly affected its abundance. The coexistence of NO_2 and O_3 provides conditions favorable for nighttime NO_3-radical formation, which can initiate monoterpene oxidation and generate organic nitrate and peroxy intermediates. Subsequent partitioning and multiphase reactions of these products may contribute to nitrooxy-OS formation (Jiang et al. 2022; Wang et al. 2018). The absence of clear relationships between NOSm and SO_4^{2-}, aerosol acidity, or RH (Fig. S7) further suggests that its wintertime variability was more closely associated with NO_x-related oxidation conditions than with bulk sulfate or particle-water availability. OSali also exhibited higher abundances in winter showed a different pattern from NOSm. Its positive associations with SO_2 and NH_4^+ (r ~ 0.6) suggest an influence from anthropogenic precursor inputs and anthropogenically influenced secondary aerosol processing. Similar increases in the anthropogenic contribution to OSs have been observed during winter in Tianjin (Ding et al. 2022). OSali may form through the oxidation of relatively reduced organic precursors followed by sulfate esterification and particle-phase processing. Lower temperatures, enhanced regional pollution, and increased partitioning of low-volatility products may further favor their wintertime accumulation.

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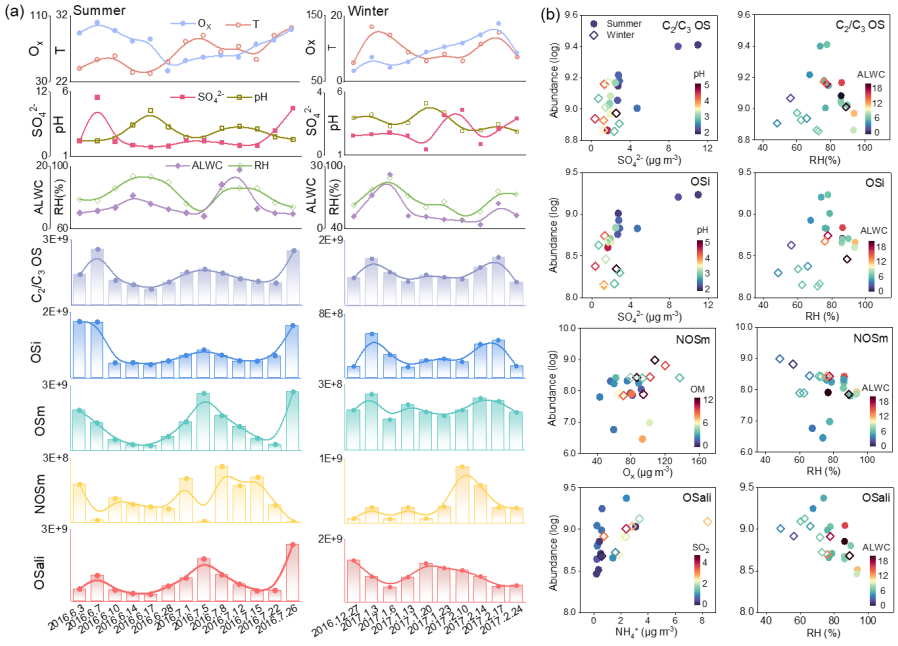

Figure 4. Temporal variations of O_x, T, SO_4^{2-}, pH, ALWC, RH and OSs (a), as well as the relationship between them during summer and winter (b).

To assess the influence of regional transport, 72 h-backward trajectories were analyzed (Fig. 5). In summer, 68 % of the trajectories were associated with air masses transported within Fujian Province, while a further 14 % originated over the South China Sea. The predominance of regional air masses from southeastern China, together with strong summertime biogenic emissions, is consistent with the high relative contributions of isoprene- and monoterpene-derived OSs and the importance of in situ photochemical and multiphase processing. Marine air masses may also modify aerosol water content and composition (Hodshire et al. 2019; Moore et al. 2024). However, chloride contributed only marginally to $PM_{2.5}$ mass even for these ocean-influenced air masses, suggesting that the contribution of marine-derived chloride to $PM_{2.5}$ was limited during the sampling periods. Winter air masses showed a greater influence from continental transport. Approximately 57% of trajectories passed over Jiangxi and Hunan provinces, while 27% arrived from northwestern inland regions. These pathways indicate increased exposure to anthropogenically influenced continental air masses

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before reaching Wuyi Mountain. During transport, oxidation, gas–particle partitioning, and multiphase aging may alter both precursor composition and the molecular characteristics of particulate OSs. This is consistent with the higher wintertime abundances of NOSm and OSali and their associations with nitrogen-rich and anthropogenically influenced aerosol conditions. These results suggest that OS composition at this mountain site reflects the combined influences of regional transport and in situ atmospheric processing, with regional transport playing a more prominent role in winter by modifying precursor availability and aerosol chemical environments.

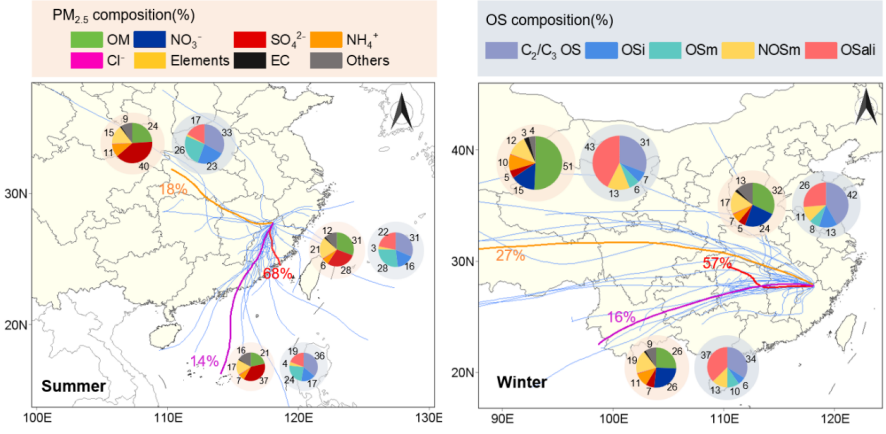

Figure 5. Seasonal 72 h-backward trajectory clusters, the associated $PM_{2.5}$ and OSs composition at the Wuyi Mountain site. Percentages indicate the fraction of trajectories in each cluster, and bold lines represent the corresponding mean trajectories. For each cluster, the left and right pie charts show $PM_{2.5}$ composition (OM, NO_3^-, SO_4^{2-}, NH_4^+, Cl^-, elements, EC, and others) and OS composition (C_2/C_3 OS, OSi, OSm, NOSm, and OSali), respectively. Pie chart sizes are proportional to the corresponding $PM_{2.5}$ mass concentration and the total OSs abundance, respectively.

4. Conclusions

This study investigated the molecular composition, seasonal variability, and possible formation processes of $PM_{2.5}$-bound OSs at a high-altitude background site on Wuyi Mountain using HPLC-Orbitrap MS. Among the detected sulfur-containing compounds, approximately 90% of CHOS and 30% of CHONS formulas met the compositional criteria for potential OSs and nitrooxy-OSs, respectively. Their total signal abundances in winter were 1.4 and 2.1 times those in summer. Nevertheless, summer OSs exhibited a higher overall degree of molecular oxidation, with species

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having $\overline{OS}_c$>-1 accounting for 72% of the total OS signal abundance compared with 56% in winter. Winter OSs were characterized by larger (C_5–C_{23}), less oxygenated (O_4–O_7), and more unsaturated molecules. Among the 35 structurally assigned OSs, isoprene- and monoterpene-derived species were relatively more abundant in summer, whereas monoterpene-derived nitrooxy-OSs and aliphatic-like OSs contributed more strongly in winter. The seasonal differences were associated with changes in both precursor availability and atmospheric processing. In summer, higher sulfate concentrations (3.6±2.8 μg m^{-3}) and greater aerosol acidity (2.8±0.7) were closely linked to the abundance of C_2/C_3 OSs and isoprene-derived OSs ($|r| > 0.7$, $p < 0.01$), supporting an important role for sulfate-related multiphase processing. Although particle water was abundant, its weak correlations with OS abundance suggest that it primarily provided a reaction medium rather than directly governing OS variability. In winter, higher O_x (95.9 ± 20.9 μg m^{-3}) and a more nitrogen-rich aerosol environment were associated with the enhanced contribution of nitrooxy-OSs, while the increase in aliphatic-like OSs was more closely related to anthropogenic precursor inputs and secondary aerosol processing. Backward-trajectory analysis further indicated a stronger influence of regional transport in winter, which likely altered precursor composition and promoted atmospheric aging. Our results suggest that seasonal OS composition in mountain environments reflects shifts in the relative importance of multiple coexisting precursor sources and atmospheric processes. These findings highlight how interactions among biogenic and anthropogenic precursor inputs, inorganic aerosol composition, atmospheric oxidation, multiphase processing, and regional transport regulate secondary organic aerosol composition and evolution in mountain environments. Further quantitative and experimental studies, including direct measurements of precursor VOCs and reactive oxidants, are needed to better constrain the seasonal contributions of individual OS formation processes.

ASSOCIATED CONTENT

Data availability. Raw data used in this study can be obtained from https://doi.org/10.5281/zenodo.22119962 (Wang et al., 2026). They are also available

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on request by contacting the corresponding authors.

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Competing interests. The contact author has declared that none of the authors has any competing interests.

Author contributions. YWH and WX designed the study. Data analysis was done by TW and YWH. TW and WX interpreted data, prepared the display items, and wrote the manuscript. All authors commented on and discussed the manuscript.

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