

Measurement report: Impact of cloud processes on secondary organic aerosols at a forested mountain site in southeastern China

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Abstract. Aerosol particles play critical roles in climate and human health. However, aerosol composition and evolution,
15 particularly secondary organic aerosols (SOA), and its interaction with clouds in high-altitude background areas in China remain less understood. Here we conducted real-time measurements of submicron aerosols (PM₁) using aerosol mass spectrometers at a forested mountain site (1128 m a.s.l.) in southeastern China in November 2022. The average ($\pm 1\sigma$) PM₁ mass concentration was ~~4.45~~ ~~± 6.513~~ ± 4.8 $\mu\text{g m}^{-3}$, which was ubiquitously lower than those at other mountain sites in China. Organic aerosol (OA) constituted the largest fraction of PM₁ (42.9 %) and was dominantly secondary as indicated by the
20 high oxygen-to-carbon (O/C) ratio (0.85–0.96) and carbon oxidation state (0.21–0.49). Back trajectory analysis revealed that higher concentrations of PM₁ were mainly associated with the transport from the western and southwestern regions. Notably, the remarkably enhanced PM₁ concentrations observed during daytime on cloudless days were identified to be likely produced from cloud evaporation. Positive matrix factorization resolved two distinct OOA factors, i.e., less oxidized oxygenated OA (LO-OOA) and more oxidized OOA (MO-OOA). While MO-OOA was scavenged efficiently during cloud
25 events, cloud evaporation was found to release a significant amount of LO-OOA from ~~airmass~~ ~~air mass~~ transported from polluted regions. The distinct increase of ~~O/C with the decrease of~~ OA/ Δ CO (CO after subtracting the background level) ~~with the decrease of O/C~~ during the cloud evaporative period further demonstrates that OA remained in cloud droplets are generally in a moderate oxidation state. Moreover, organic nitrates were also estimated and showed a higher contribution to the total nitrate during the cloudy period (27 %) than the evaporative period (3 %). Overall, our results demonstrate the
30 importance of SOA and the influences of cloud processes in regional mountain areas in southeastern China.

1 Introduction

Aerosol particles play essential roles in regional and global climate (Ramanathan et al., 2001; Kanakidou et al., 2005), as well as air pollution (Huang et al., 2014) and public health (Kampa and Castanas, 2008). Submicron aerosol (PM₁) from both natural and anthropogenic sources can be transported to the upper level of atmospheric boundary layer or even lower free troposphere through convection and frontal uplift (Monks et al., 2009; Huang et al., 2020; Carbone et al., 2014). At high altitudes, aerosol species can be transported over longer distances, depending on their particle sizes and compositions (Pokorná et al., 2022; Tang et al., 2016; Zhong et al., 2022), and the aging processes during transport results in continuous changes in chemical and physical properties (Calvo et al., 2013; Hallquist et al., 2009). Aerosols can have an important impact on CCN properties at high altitudes where atmospheric conditions favour the formation of clouds, thereby affecting the lifetime and optical properties of clouds through aerosol-cloud interactions (Haywood and Boucher, 2000; Asmi et al., 2012; Rejano et al., 2021). On the other hand, aerosol-cloud interactions also modify the chemical composition of aerosols within cloud droplets. This modification, in turn, affects the characteristics of surrounding aerosols after the cloud has evaporated (Roth et al., 2016). As a result, measurements of high-altitude regional aerosols are of great importance for a better understanding of aerosol-cloud interactions and their impacts on air pollution and climate.

Mountain sites are crucial platforms for studying aerosol characteristics over regional scales and the influences of diverse emission sources (e.g., biomass burning, industrial process, and biogenic emissions) and atmospheric processes (e.g., in-cloud processing, new particle formation). Compared to balloon and aircraft observations, mountain sites have advantages in continuous and long-term observations because of low cost, stable geographic location, and accessibility for instrument maintenance. In addition, the different meteorological conditions at mountain sites can have great impacts on aerosol formation, aging, and scavenging patterns. For example, Gao et al. (2023) showed that cloud processes can promote the formation of secondary organic aerosol (SOA) by multiphase oxidation. Li et al. (2013) found that high relative humidity (RH) at Mt. Hua can lead to a reduction in particle acidity, and thus reduce the formation of biogenic SOA by suppressing the acid-catalysis reaction. Chen et al. (2021) reported that fog scavenging was more efficient towards particles with aerodynamic diameter larger than 700 nm in Taiwan mountain regions, while the majority of smaller particles remained growthunscavenged and could grow into larger particles through ~~the process of~~ gas-to-aqueous partitioning. Although several mountain stations have been established in China to gain more information on the chemical composition and sources (Zhang et al., 2018; Zhang et al., 2019; Du et al., 2015; Zhang et al., 2014), optical properties (Wang et al., 2015), and hygroscopic properties of aerosols (Ding et al., 2021), most of these studies are mainly conducted on Qinghai-Tibetan Plateau and in Northern China Plain (NCP), while the studies in southeastern China remain limitedscarce.

In this work, a high-resolution time-of-flight aerosol mass spectrometer (AMS hereafter) and a quadruple aerosol chemical speciation monitor (ACSM hereafter) were deployed at a forested mountain site in southeastern China. The chemical composition and properties of PM₁ are characterized, the elemental composition and oxidation state of OA are determined, and the potential transport pathways of PM₁ are also investigated using back trajectory analysis. Furthermore, the effect of

cloud processes on aerosol species is discussed. The sources of organic aerosol are investigated using positive matrix factorization (PMF) analysis of combined high-resolution organic and inorganic aerosol mass spectra. Particularly, particulate organic nitrates (ONs) are determined and quantified based on PMF results.

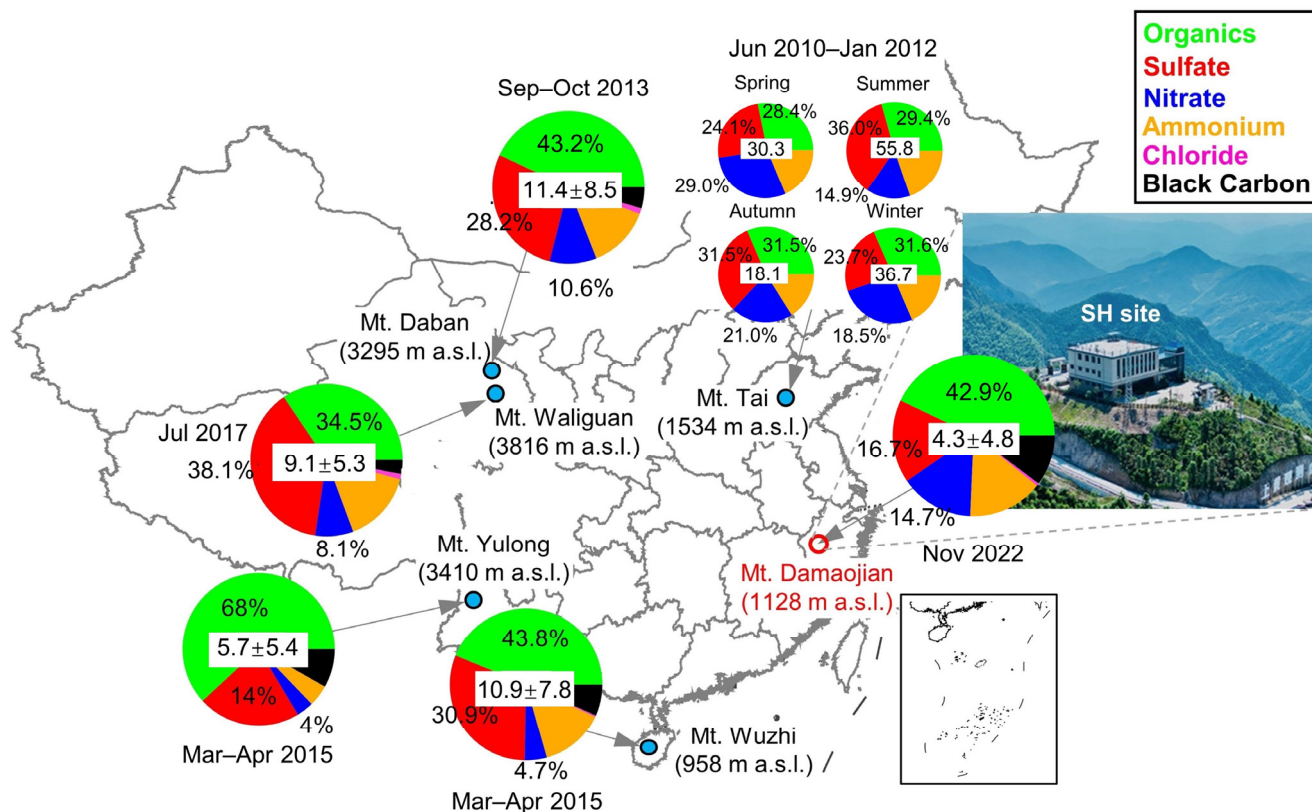


Figure 1. Location of the sampling site (Mt. Damaojian, red circle on the map). The mean concentration ($\mu\text{g m}^{-3}$) and chemical composition of submicron aerosols (NR-PM₁+ BC) measured at selected mountain sites in China are also shown. Note that BC and chloride are not accounted for Mt. Tai due to the lack of measurement or data in the relevant study measurements. Detailed information on these sampling sites is presented in Table S1S3 in the Supplement.

2 Methods

2.1 Site and instrumentation

The campaign was carried out from 1 November to 30 November 2022 at Shanghuang Eco-Environmental Observatory of Chinese Academy of Sciences (SH), located at Mt. Damaojian (119.51°E, 28.58°N, 1128 m a.s.l.) in Wuyi County, Zhejiang Province (Fig. 1). This site is a typical background site in southeastern China that is surrounded by mountains and forests, and there are no strong local anthropogenic sources nearby.

PM₁ species were measured using a suite of real-time instruments with 1–20 min time resolution, including an AMS operated under the V-mode and a quadrupole ACSM for non-refractory (NR)-PM₁ composition, together with a seven-wavelength Aethalometer (AE33, Magee Scientific Corp.) for equivalent black carbon (BC) mass concentration. The AMS measurements were only conducted during two periods (15–20 November and 24–28 November, respectively) in this study due to the malfunction of the instrument. Briefly, aerosol particles were sampled into an air-conditioned room through stainless steel tube (O.D.: 1/4 inch), and the residence time was estimated as 5 s. A nafion dryer was placed upstream of the ACSM and AMS to remove the moisture, after that, aerosol particles were sampled into AMS, ACSM, and AE33, respectively. Because there was no size cutoff in front of the sampling line, the AMS and ACSM may report slightly higher concentrations than expected because of measuring some cloud droplets larger than 2.5 μm. According to a previous study during three fog episodes at a rural site in North China Plain, the influence of fog droplets on submicron aerosol measurements was less than 20% (Kuang et al., 2024). Simultaneously, air pollutants including NO_x, O₃ were measured by a suite of gas analyzers (Thermo Scientific Inc., USA), PM_{2.5} and PM₁₀ were measured using continuous ambient particulate monitors with PM_{2.5} and PM₁₀ cutoff before the sampling inlet (Model 5014i, Thermo Scientific., USA), and CO was measured by a Picarro greenhouse gas analyzer (G2401, Picarro Inc., USA). Meteorological parameters containing temperature (*T*), relative humidity (RH_r), wind speed (WS), wind direction (WD), and pressure (*P*) were measured by an automatic weather station. In addition, ~~(PDR)~~the particle depolarization ratio (PDR) was measured by a particle monitoring lidar (Science of Light Technologies Co., Ltd) to analyze the vertical distribution of aerosol particles, and the hourly boundary layer height (BLH) was obtained from European Reanalysis 5 (ERA5) reanalysis dataset. More details and descriptions of the instruments and data are provided in Table S1.

2.2 Data analysis

2.2.1 ACSM and AMS

ACSM data were analyzed using ACSM standard data analysis software (v2.5.13) and AMS data were analyzed using SQUIRREL v1.65F and PIKA v1.25F. A composition-dependent collection efficiency (CDCE) was applied to the ACSM/AMS data according to Middlebrook et al. (2012). Elemental analysis of high-resolution mass spectra (HRMS) was performed using the “Improved-Ambient” (I-A) method (Canagaratna et al., 2015). The default relative ionization efficiency (RIE) values of 1.1, 1.4, and 1.3 were applied for nitrate, organics, and chloride (Canagaratna et al., 2007; Nault et al., 2023). According to the ion efficiency (IE) calibration results using ammonium sulfate, the RIE values of ammonium and sulfate were 5.05 and 0.73 for ACSM, and 5.26 and 1.28 for AMS, respectively. Additionally, the detection limits of ACSM and AMS for different NR-PM₁ species are determined as 3 times the standard deviation of mass concentrations during a period (16–20 November) with low particle concentration (Table S2). Detailed comparison of the concentrations of NR-PM₁ species measured by AMS and ACSM are shown in Fig. S1. ~~The ACSM data during the sampling period was corrected by using the regression coefficients between ACSM and AMS.~~The concentrations of NR-PM₁ species measured by ACSM

110 correlated well with those measured by AMS ($r^2 = 0.35\text{--}0.97$) except for chloride ($r^2 = 0.18$). This is likely due to the low ambient chloride masses at this background site, which are close to or below the ACSM detection limit. Also, the non-ideal vaporization behaviour of chloride in ACSM may be another reason (Crenn et al., 2015). Notably, similar slopes (0.62–0.74) were found for organics, sulfate, nitrate, and ammonium measured by AMS and ACSM, indicating a general overestimation of ACSM. The possible systematic bias in IE calibration of ACSM using ammonium sulfate could be a potential cause.

115 Therefore, the ACSM data during the sampling period was scaled to the AMS data using these linear regression slopes. As shown in Figs. S1g and h, after this adjustment, the concentrations of NR-PM₁ tracked well with PM_{2.5} ($r^2 = 0.60$ and slope = 0.48 for ACSM, $r^2 = 0.93$ and slope = 0.54 for AMS ~~respectively~~) and PM₁₀ ($r^2 = 0.53$ and slope = 0.27 for ACSM, $r^2 = 0.99$ and slope = 0.39 for AMS ~~respectively~~) measured by the particle monitor, suggesting that the AMS/ACSM quantification was reasonable.

120 PMF Evaluation Tool (PET v3.04) was employed to further deconvolve the HRMS derived from AMS into different source factors following the procedures reported by Ulbrich et al. (2009) and Zhang et al. (2011). In addition to organic fragment ions, the major fragment ions of inorganic species, i.e., SO⁺ (m/z 48), SO₂⁺ (m/z 64), SO₃⁺ (m/z 80), HSO₃⁺ (m/z 81), H₂SO₄⁺ (m/z 98) for sulfate, NO⁺ (m/z 30), NO₂⁺ (m/z 46) for nitrate, NH⁺ (m/z 15), NH₂⁺ (m/z 16), NH₃⁺ (m/z 17) for ammonium, and Cl⁺ (m/z 35), HCl⁺ (m/z 36) for chloride were also included into the HR data and error matrices for PMF.

125 detailed description of the procedures was given in Sun et al. (2012). After checking the key diagnostic plots (Fig. S2), mass spectra, and the correlations with related tracers, a four-factor solution was considered as the optimal solution in this study (Text S1).

2.2.2 Estimation of organic nitrates

ONs were estimated from the PMF results (Zhang et al., 2011; Xu et al., 2015). Briefly, NO_x⁺ (i.e., NO⁺ and NO₂⁺) are major

130 fragments of nitrate functionality (-ONO₂), which can be referred to as the total nitrate measured by AMS. Combining inorganic with organic mass spectra in PMF, NO⁺ and NO₂⁺ can be separated into different organic aerosol (OA) factors and an inorganic nitrate aerosol factor (~~NHAINA~~). According to previous studies, the ratios of NO⁺/NO₂⁺ for ONs are approximately 2.25–3.7 times higher than pure NH₄NO₃ (Fry et al., ~~2013~~2009; Fry et al., ~~2009~~2013). Consistently, the PMF results in our study (Fig. ~~7~~6) show that the average NO⁺/NO₂⁺ ratios of LO-OOA and MO-OOA were 13.19 and 11.2, falling

135 within the range of ONs. In contrast, a NO⁺/NO₂⁺ ratio of 3.56 was observed for ~~NHAINA~~, reflecting its characteristics of inorganic nitrates. Therefore, the PMF analysis of nitrate is reasonable, and based on this result, the mass concentration of ONs (NO_{3,org}) can be calculated by summing these two ion signals distributed in all OA factors as follows:

$$\text{NO}_{3,\text{org}} = \text{NO}_{\text{org}}^+ + \text{NO}_{2,\text{org}}^+ \quad (1)$$

$$\text{NO}_{\text{org}}^+ = \sum([\text{OA factor}]_i \times f_{\text{NO}^+,i}) \quad (2)$$

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$$\text{NO}_{2,\text{org}}^+ = \sum([\text{OA factor}]_i \times f_{\text{NO}_2^+,i}) \quad (3)$$

where [OA factor]_{*i*} represents the mass concentration of OA factor *i* resolved by PMF, $f_{\text{NO}^+,i}$ and $f_{\text{NO}_2^+,i}$ are the mass fractions of NO⁺ and NO₂⁺ in OA factor *i*, respectively.

2.2.3. Backward trajectory analysis

The Hybrid Single-Particle Lagrangian Integrated Trajectories (HYSPPLIT) model and meteorological data from the NOAA
145 Global Data Assimilation System (GDAS) were used to calculate 72 h backward trajectories at the SH site. The trajectory
arrival time was set from 0:00 to 23:00 at 1 h intervals, and the arrival height at the site was set as 1100 m. To further show
the aerosol particle concentration levels in different regions, the map contained the back trajectory (Fig. S5S7) was coloured
by the time-averaged organic carbon surface mass concentration from the M2T1NXAER v5.12.4 dataset ($0.5 \times 0.625^\circ$,
hourly). This dataset, part of the Modern-Era Retrospective analysis for Research and Applications version 2 (MERRA-2)
150 model (Gelaro et al., 2017), was sourced from the NASA Giovanni website (<https://giovanni.gsfc.nasa.gov>).

3. Results and discussion

3.1. Mass concentration and chemical composition of submicron aerosols

Figure 2 shows the time series of meteorological parameters (*T*, RH, WD, and WS), air pollutants (PM_{2.5} and CO) along with
PM₁ (NR-PM₁ + BC) species during the campaign. The total PM₁ concentration varied dynamically from 0.3 μg m⁻³ to 39.8
155 μg m⁻³ during the whole sampling period, with an average ($\pm 1\sigma$) of $4.3 \pm 4.8 \mu\text{g m}^{-3}$. As shown in Fig. 1, organics accounted
for the largest contribution (42.9 %) to the total PM₁ during the sampling period, followed by sulfate (16.7 %), ammonium
(15.0 %), nitrate (14.7 %), BC (10.2 %), and chloride (0.5 %). In addition, BC was observed to correlate well with PM₁ and
PM_{2.5} ($r^2 = 0.59$ and 0.62 , respectively), suggesting ~~that the aerosol particles at this site may be largely influenced by~~
~~the potential influences of~~ regional biomass burning plumes in the Yangtze River Delta (YRD) on aerosol particles at this site
160 (Zhang et al., 2015). The concentration and composition of PM₁ are quite different from those observed at other Chinese
mountain sites in different seasons (Fig. 1). For example, the average PM₁ concentration here ~~is was~~ much lower compared to
Mt. Wuzhi ($10.9 \pm 7.8 \mu\text{g m}^{-3}$) at a similar altitude, and ~~is was~~ also ~~slightly~~ lower than ~~those~~ at higher altitude mountains such
as Mt. Yulong ($5.7 \pm 5.4 \mu\text{g m}^{-3}$) and Mt. Waliguan ($9.1 \pm 5.3 \mu\text{g m}^{-3}$). Although PM₁ concentration has a strong seasonal
dependence, such as in Mt. Tai, with low concentration in autumn ($18.1 \mu\text{g m}^{-3}$) and high in summer ($33.5 \mu\text{g m}^{-3}$), the
165 average PM₁ concentration at our site ~~is was~~ still much lower than Mt. Daban ($11.4 \mu\text{g m}^{-3}$) and Mt. Tai in the same season
(autumn). Considering the occurrence of frequent cloud events during the sampling period, the low PM₁ concentration might
be mainly associated with cloud scavenging, which will be further validated in subsequent sections. Notably, a relatively
lower contribution of sulfate to PM₁ was observed (16.7 %) in this site compared to other mountain sites (~~44.0 except Mt.~~
~~Yulong (23.7 %–38.1 %)~~, which was likely attributed to the significant reduction of SO₂ emission in China during the past
170 decade (Wen et al., 2023). The decrease in sulfate contribution was associated with an elevation of nitrate contribution. This

shift suggests that most NH_4 will probably be in the form of ammonium nitrate which is more volatile than ammonium sulfate and thus unlikely to transport over a large scale. ~~Notably~~Additionally, at Mt. Tai, even though the contributions of sulfate were high, nitrate still accounted for considerable fractions (14.9 %–29.0 %) that were comparable to or higher than this site (20.9 %). Taken together, PM_{10} at this site is more likely influenced by anthropogenic emissions over a smaller regional scale, consistent with the high urban density in eastern China-(Li et al., 2018).

Figure ~~3a~~S3a shows the relative contribution of each PM_{10} component as a function of PM_{10} concentration together with the probability density of PM_{10} mass loading. The highest two ~~frequencies~~frequencies of PM_{10} concentrations were distributed within 0–3 $\mu\text{g m}^{-3}$ and 5–8 $\mu\text{g m}^{-3}$ (29.0 % and 29.8 %, respectively). The fraction of nitrate increased significantly with PM_{10} concentration, and meanwhile, the fraction of organics and BC exhibited a decreasing trend. We noted that a nitrate-dominant peak of PM_{10} mass loading was observed at the nighttime of 4 November (EP1, Fig. 2d), and the concentrations of organic and nitrate as a function of PM_{10} mass during this event and the rest of the campaign are shown in Fig. ~~S3~~S4. Almost all the data points with PM_{10} concentrations above 20 $\mu\text{g m}^{-3}$ were from this event. During EP1, a steeper slope for nitrate relative to PM_{10} was found than that for organics, which was contrary to slopes during the rest period-(Fig. S3)-, resulting in significant contributions of nitrate to PM_{10} across all PM_{10} concentrations (Figs. S3b and S4). These distinct differences in slopes for nitrate and organics implied different mechanisms of PM_{10} elevation during these two periods. Consequently, we also excluded EP1 from the statistic of Fig. ~~3a~~S3a, and the result is presented in Fig. ~~3b~~S3c. After removing EP1, organics became the dominant contributor (> 40 %) across all PM_{10} concentrations. Despite this, there was still an increasing trend for nitrate and a decreasing trend for sulfate along with increased PM_{10} , yet the contribution of ammonium remained relatively constant. This further supported our previous hypothesis that there was a conversion of ammonium sulfate to ammonium nitrate with the increase in PM_{10} concentration. Overall, these results suggested that high levels of PM_{10} at the SH site might be mainly attributed to the formation or transport of organics, meanwhile, nitrate also plays a nonnegligible role.

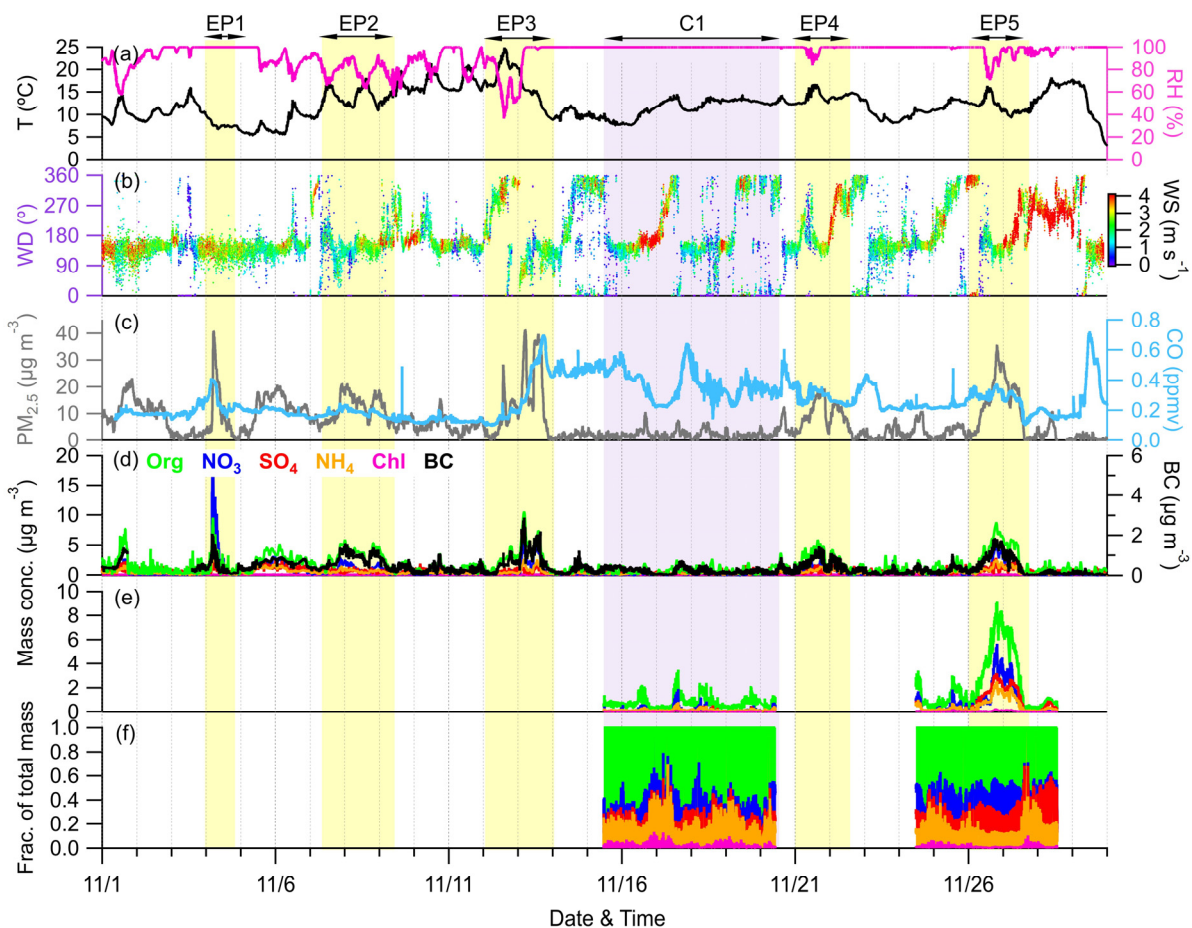


Figure 2. Time series of (a) T and RH; (b) WD coloured by WS; (c) mass concentration of $PM_{2.5}$ and mixing ratio of CO; (d) mass concentrations of NR- PM_1 species measured by ACSM together with BC measured by AE33; and (e, f) mass concentrations and contributions of NR- PM_1 species measured by AMS. The yellow and purple shaded areas represent six selected episodes (EP1–EP5 and C1, respectively).

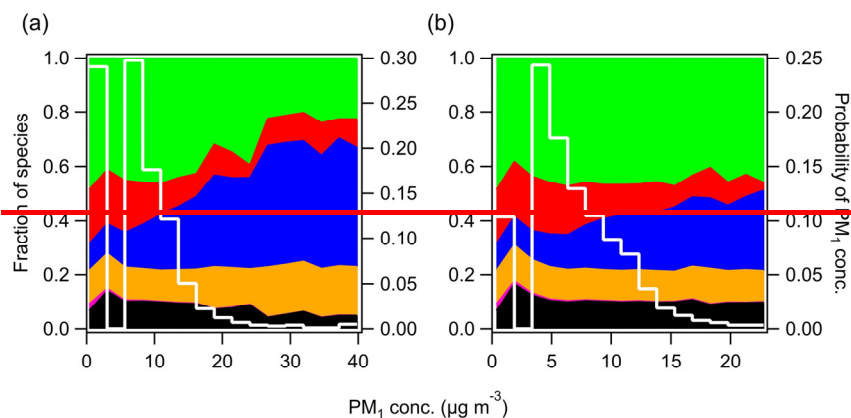
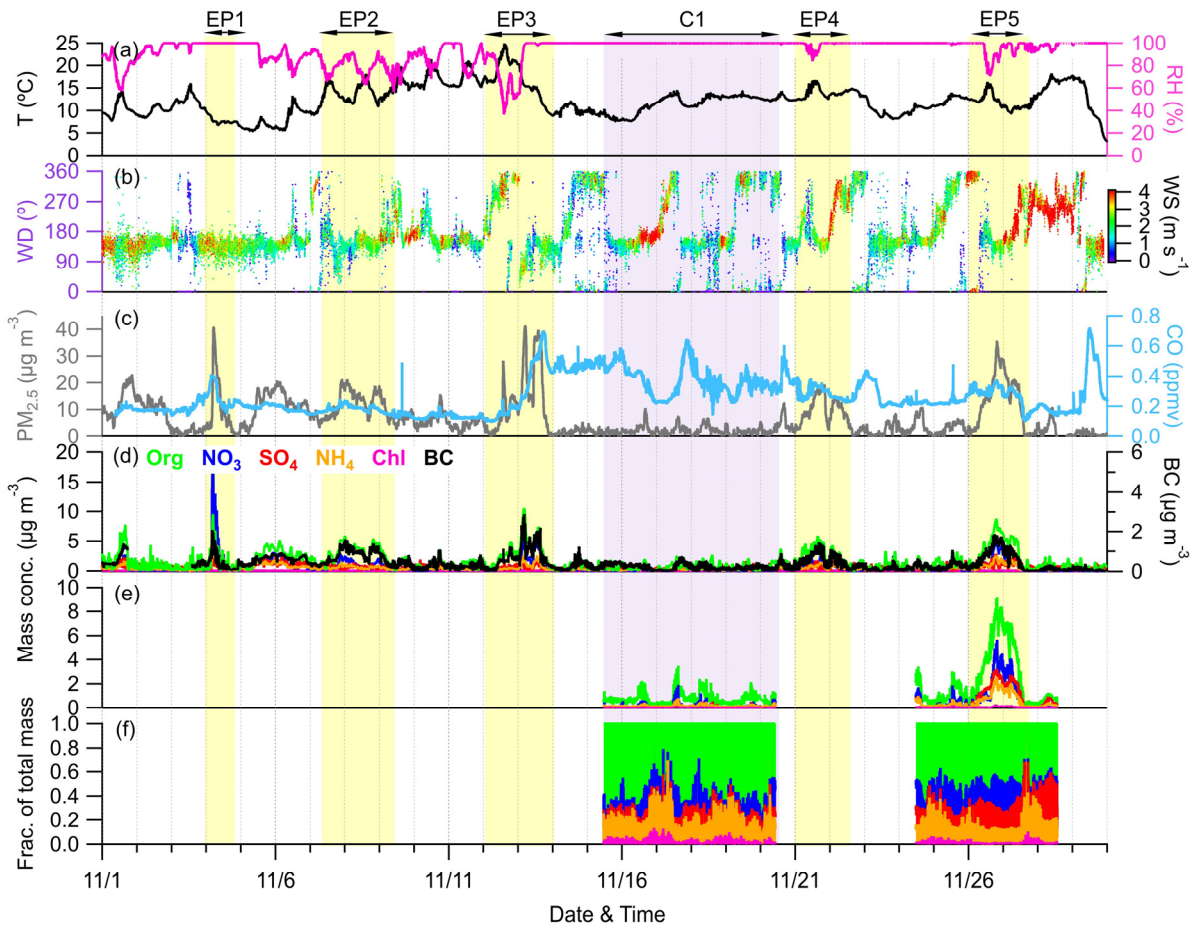


Figure 3. Variations of aerosol composition as a function of PM₁ mass concentration and the probability density of PM₁ during (a) the entire study period and (b) the period excluding EPI.

200 The diurnal cycles of PM₁ species, air pollutants, and meteorological parameters during the entire campaign are illustrated in Fig. S3S5. The mean and median values of organics and nitrate both showed distinct noon peaks at around 14:00, which could be attributed to the daytime photochemical production and the low wind speed (Tang et al., 2022; Xu et al., 2018b). In addition, ~~these noon peaks may as depicted in Fig. S6, these noon peaks coincided closely with the diurnal variation in BLH~~ (reached up to ~1450 m at around 13:00). Consequently, the relatively elevated concentrations of organics and nitrate at ~~noon might~~ also be related to the development of atmospheric boundary layer, which facilitates the mixing of air masses transported from polluted areas. The high nighttime peak of nitrate was mainly due to the influences of the nitrate event on 4 November. Comparatively, sulfate, chloride, and BC showed relatively flat diurnal variations, suggesting the regional characteristics of these species (Zhang et al., 2015).



210 **Figure 2.** Time series of (a) T and RH; (b) WD coloured by WS; (c) mass concentration of PM_{2.5} and mixing ratio of CO; (d) mass concentrations of NR-PM₁ species measured by ACSM together with BC measured by AE33; and (e, f)

mass concentrations and contributions of NR-PM₁ species measured by AMS. The yellow and purple shaded areas represent six selected episodes (EP1–EP5 and C1, respectively).

3.2 Meteorological conditions and air mass origin

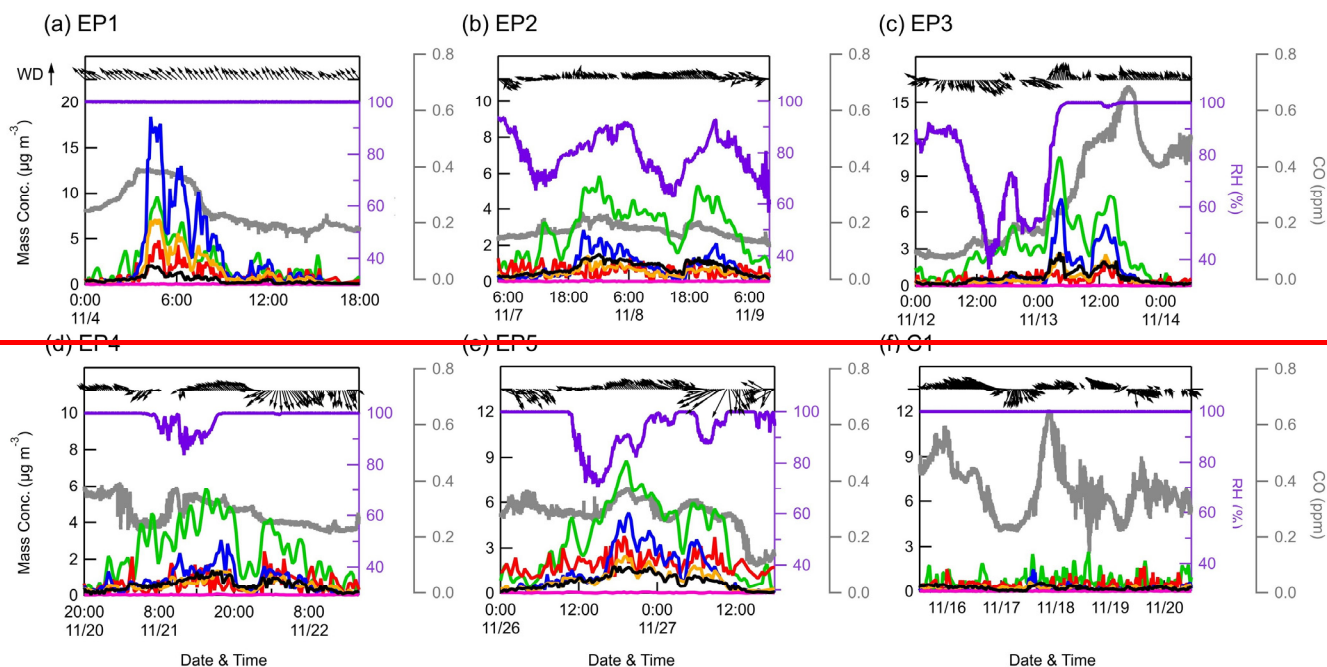
215 The entire study period was characterized by five elevated PM₁ episodes (EP1–EP5), as marked in Fig. 2-2. These five elevated PM₁ episodes exhibited mean PM₁ concentrations ranging from 5.2–9.8 μg m⁻³, which were close to or exceeded the 75th percentile of the overall PM₁ concentration distribution (5.6 μg m⁻³). Meanwhile, a clean period (C1) with low-PM₁ levels a mean PM₁ concentration of 1.3 μg m⁻³, falling below the 25th percentile of the distribution (1.6 μg m⁻³) was also selected for comparison. The relationship between the PM₁ species concentration, meteorology, and air mass transport
220 during these six periods is shown in Fig. 43 and Fig. S5S7. EP1 exhibited a nitrate-dominant PM₁ peak, with nitrate concentration rapidly increased by ~18 μg m⁻³ within an hour. In addition, a CO peak was also observed at that time, suggesting potential contributions of pollution transport to nitrate. Given the lack of anthropogenic activities near the sampling site, the enhanced nitrate concentrations likely originated from NO_x transported from adjacent towns in the northeast, as indicated by the nitrate wind polar plot (Fig. S6S8) showing high levels of nitrate mainly associated with
225 northeastern wind directions. This was further supported by the back trajectory analysis, in which air masses on 4 Nov November were also from the northeast. Moreover, the RH was stable at 100% during EP1, which could facilitate favor the heterogenous uptake of N₂O₅ and subsequent nitrate formation in the aqueous phase at night (Brown et al., 2006; Li et al., 2020).

In contrast to EP1, PM₁ in other episodes (EP2–5) were all dominated by organics. EP2 was associated with low
230 concentrations of CO, indicating relatively slight pollution transport influence. In addition, clear and similar diurnal variation patterns were found in aerosol species and RH, suggesting that PM₁ in this period was mainly affected by the evolution of the atmospheric boundary layer. EP3 was initiated by an organic-only increase under high temperature (~15–25 °C) and low RH (~40 %–80 %), while other aerosol species remained at very low levels. Considering the low CO levels in this period, the increasing trend of OA could be attributed to the local biogenic emissions. The concentrations of organic, together with
235 other PM₁ species and CO, were further elevated after air mass air mass sources shifted from southwest to northwest on 13 Nov November, which may bring a large amount of aerosols from megacities such as Zhengzhou and Hangzhou to the SH site (Fig. S5eS7c). EP4 and EP5 were two similar episodes, where PM₁ concentrations were generally associated with the changes in RH and WD. During these two periods, air masses both originated from the west and southwest, but the trajectory distances during EP5 were shorter, suggesting that the air masses transported more slowly in EP5.

240 Overall, although these episodes had different mechanisms of PM₁ variation, they were mostly influenced by the transport of western and southwestern air masses, indicating a substantial impact of anthropogenic emissions from big cities like Nanchang and Fuzhou. These areas are marked by higher pollution according to the organic carbon distribution in Fig. S5S7. However, we have also identified similar transport pathways and even higher CO concentrations during C1 (0.37 ppm on average) than EP2–5 (0.18–0.31 ppm on average), while the PM₁ concentration stayed at very low levels for the whole

245 period. It is worth noting that PM₁ peaks in EP2=, EP4, and EP5 all occurred along with RH below 100 %. When the RH returned to 100 %, PM₁ concentrations gradually diminished to levels comparable to those observed during the clean period (C1). Furthermore, as shown in Fig. S7S9, after omitting EP1 and EP3 to exclude the effect of air mass change, there were notable reductions in the mean and median mass concentrations of all PM₁ species over the ~~entire~~-campaign, ranging between ~~2.6 %-4415.5 %-63.4 %~~ and ~~26.8 %-50.461.0 %~~ when RH reached 100 % from conditions of lower RH, 250 respectively. Considering the frequently occurring cloud events at this site, it was most likely that these variations of RH were affected by the cloud process, which may further play an important role in PM₁ concentrations.

Figure S8S10 shows the particle depolarization ratio measured by the particle lidar during EP5 and C1. During EP5, clouds were identified at ~1 km above the sampling site from 2:00 to 12:00 on 26 ~~Nov~~November (Fig. S8aS10a). As expected, these clouds gradually disappeared afterward, possibly owing to strong solar radiation at noon on the mountaintop causing cloud droplet evaporation. Meanwhile, a large amount of aerosol particles (PDR = ~0.15) was released from the clouds, which exhibited a strong agreement with the timing of the PM₁ peak. These particles were then ~~scavenged~~removed by ~~the increased~~ ~~RH~~new cloud formation and strong wind (WS > 4 m s⁻¹). This cloud evaporation phenomenon was also found in urban Guangzhou, which caused remarkably enhanced nitrate mass concentration at noon (Tao et al., 2018). In contrast, no such evaporation process was observed during C1 (Fig. S8bS10b). Instead, high PDR values were found generally at around 0 km, 255 which means that clouds constantly existed near the sampling area during this period, coinciding with the constant 100% RH. Therefore, PM₁ transported to this site was likely to be scavenged by the cloud during C1. These findings were also evidenced by the photos taken at this site at local time ~15:00 on 19 Nov and 26 Nov, where severe cloud cover was observed in C1 (Fig. S9aS11a), yet the weather was generally sunny and cloudless in EP5 (Fig. S9bS11b). As a result, besides regional transport, cloud processes can also have significant impacts on aerosol particles at this site. 260



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However, there were several observations and questions raised that cannot be conclusively confirmed with the current data we have. First, it should be noted that there was a difference in cloud height between C1 (~ 0 km) and EP5 (~ 1 km, before evaporation), indicating that aerosol particles were sampled within and below the cloud during C1 and EP5, respectively.

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With no cloud evaporation occurring during C1 with low-level clouds, it raises the question of how evaporation processes affected by cloud height. Tao et al. (2018) found that cloud evaporation could occur at 500–1000 m above the ground, and the released aerosols can be transported to low levels through strong turbulent mixing. This finding agrees with our observation at EP5, where the development of the boundary layer likely promoted the vertical transport of released aerosols to this site (Fig. S12). In contrast, during C1, the severe cloud layer hindered solar radiation, leading to generally lower boundary layer heights. These clouds might be identified as orographic clouds formed by the moist air masses lifted and saturated under the influence of complex mountain terrain (Houze, 2012). If once the solar radiation and other environmental conditions are sufficient to evaporate the cloud at this site, the boundary layer would also develop correspondingly, enhancing the vertical mixing of released aerosols during daytime. Therefore, we hypothesized that clouds with heights lower than the developed boundary layer heights (up to ~ 2 km, Fig. S12) may all have the probability of evaporating and increasing the observed aerosol mass at this site.

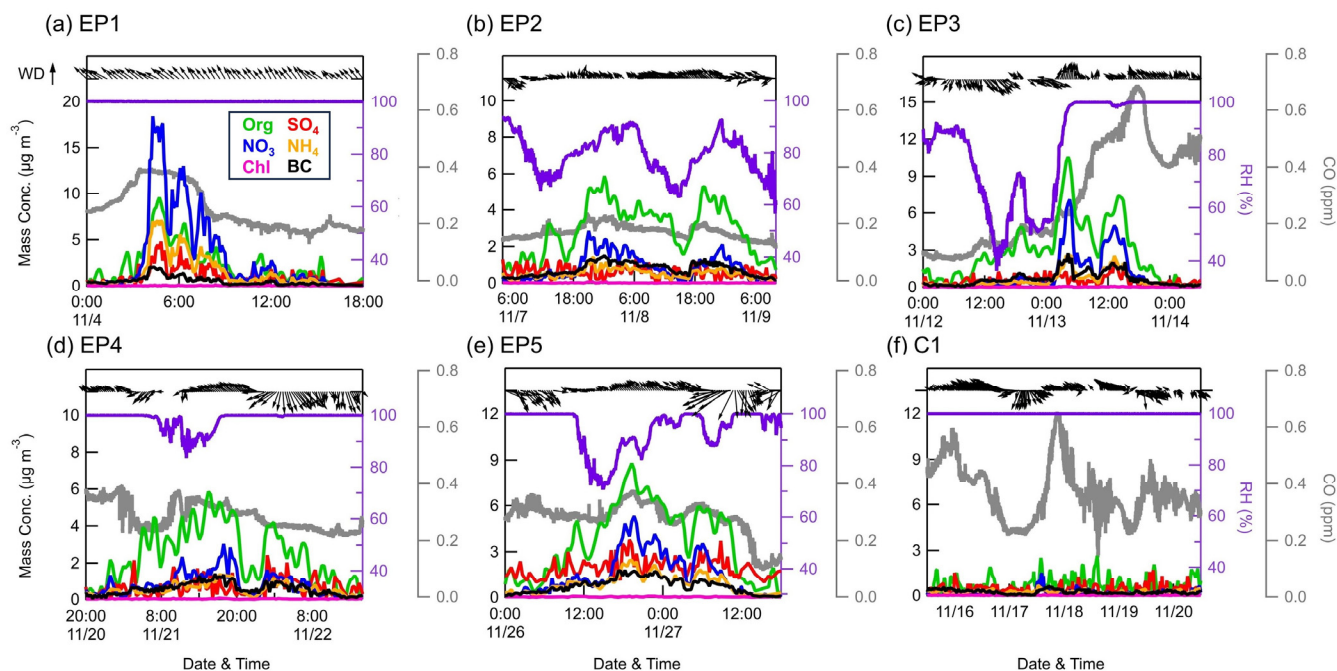
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Second, as discussed in Sect. 2.1, the sampling uncertainties caused by cloud droplets accounted for up to 20% in our study, yet significantly more aerosol particles were observed to release after the evaporation of the cloud above this site during EP5. This was likely attributed to the polluted air mass transported and entered the cloud at higher altitudes. In contrast, at the altitude near the sampling inlet, the continuous uplift of clean moist air masses from lower elevations might be unfavorable for aerosol accumulation. Moreover, the evaporation itself could also trigger some aqueous SOA formation processes such as oligomerization (Lim et al., 2010; Brégonzio-Rozier et al., 2016),

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285 thereby contributing additional aerosol mass within the cloud droplets. Third, these selected periods were all under the
influence of anthropogenic-emission-related air masses, while whether the evaporation of a clean air mass cloud can impact
the aerosol concentrations at this site remains unknown. However, we noticed a period (from 10 to 12 November) that seems
to contain a cloud evaporation process as indicated by the suddenly reduced RH (from 100 % to 70 %) during the daytime on
11 November, but still exhibited relatively low PM1 levels (2.6 $\mu\text{g m}^{-3}$ on average). The back trajectory analysis further
290 showed that the air mass during this period originated from the sea, which was less influenced by anthropogenic emissions.
As a result, it is likely that a small amount of aerosol can also be released even if a clean air mass enters the cloud and
evaporates. Despite these issues that need to be addressed through future long-term measurements and model studies, the
cloud evaporation phenomenon we observed might still be a plausible mechanism to explain certain aerosol episodes in this
and other similar regions.



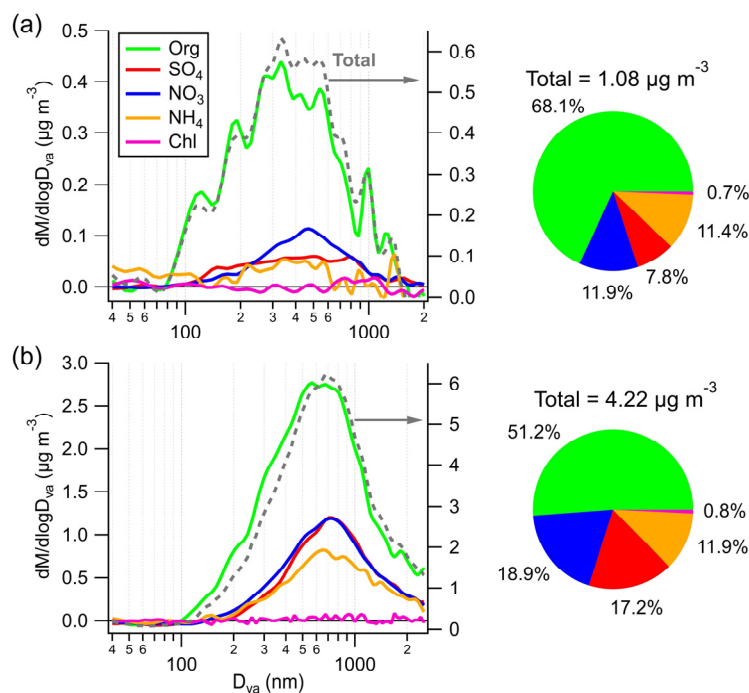
295 **Figure 43.** Temporal variations of ~~NR~~ the concentrations of PM₁ species measured by ACSM, as well as CO, RH, (grey lines), RH (purple lines), and WD, and WS (black arrows) during six ~~case~~ events.

3.3 Impacts of cloud processes on submicron aerosols

To further explore the impacts of cloud scavenging and cloud evaporation on aerosol characteristics, we ~~select~~selected C1
300 and EP5, ~~in~~during which NR-PM₁ ~~were~~was also measured by AMS, for ~~further~~additional investigation on aerosol size
distributions, elemental ratios, and HRMS in this section.

3.3.1 Size distributions and composition

The average chemically resolved size distributions of NR-PM₁ during C1 and EP5 are shown in Fig. S4. Generally, all species were distributed in accumulation mode in both periods. However, a smaller peak size (300–500 nm) and broader size distribution were observed during C1 when compared to those of EP5 (~700 nm), probably owing to the wet removal of larger and hygroscopic particles in C1 (Ge et al., 2012). Also, the complex and broad size distribution observed in C1 suggests that ~~these smaller particles are the aerosol is~~ likely externally mixed with small particles dominated by organics, which may further imply the potential formation of SOA from local biogenic sources. In contrast, the uniform size distribution across all particulate species in EP5 indicates their well-mixed and highly aged characteristics. Moreover, ammonium showed similar size distribution with sulfate and nitrate during EP5, while in C1, it was mixed mainly with sulfate and exhibited a quite different size distribution with nitrate, indicating the potential contribution of organic nitrate. Clear differences were also found in aerosol composition during C1 and EP5. Organics were the dominant contributor to total NR-PM₁ mass during C1 (68 %), followed by nitrate (12 %), ammonium (11 %), and sulfate (8 %). In contrast, despite the decreased contribution of organics (51 %), the mass fractions of sulfate and nitrate increased considerably during EP5 (by 9.4% and 7.0%, respectively). These changes in the mass fraction of species were attributed to the lower mass scavenging efficiency of organics than inorganic species (Gilardoni et al., 2014). The average HRMS of OA during C1 and EP5 are presented in Fig. S10S14. The HRMS of OA were quite similar for the two periods, with a significant peak m/z 44 (mainly CO₂⁺). The OA was highly oxidized, with C_xH_yO₁⁺ dominating the total OA in C1 and EP5 by 41 % and 40 %, followed by C_xH_y⁺ (31 % and 32 %), and C_xH_yO₂⁺ (20 % and 19 %). The contributions of the two major oxygen-containing ion fragments (C_xH_yO₁⁺ and C_xH_yO₂⁺) at the SH site were much higher than those at various urban or suburban sites in China, such as 37.4 % in urban Nanjing (Wang et al., 2016), and 52.9 % in suburban Lanzhou (Tang et al., 2022). Note that higher fraction of CO₂⁺ (3 % higher) was found in C1 than EP5 (Fig. S10eS14c), indicating a higher oxidation degree of OA in C1 (Xu et al., 2014). This is consistent with the higher O/C and OSc in C1 (0.96 and 0.49) than EP5 (0.85 and 0.21).



325 **Figure 54.** Averaged size distributions and chemical composition of NR-PM₁ during (a) C1 and (b) EP5.

3.3.2 Elemental ratios

Figure 65 shows the time series ~~and box plots~~ of N/C, H/C, OM/OC, O/C, and carbon oxidation state (OSc) of OA, as well as two ion ratios at the SH site. The hourly averaged ratios were only reported for periods with OA concentrations above 0.7 $\mu\text{g m}^{-3}$. The average N/C ratios were 0.018 for C1 and 0.027 for EP5, consistent with the higher fraction of $\text{C}_x\text{H}_y\text{N}_p^+$ in OA during EP5. OA had a slightly lower H/C ratio (1.44 vs. 1.48) and higher average ratios of O/C (0.96 vs. 0.85), OM/OC (2.42 vs. 2.29), and OSc (0.49 vs. 0.21) during C1 than those during EP5, indicating more oxidized OA during C1. The O/C ratios were overall within the range of 0.94 ± 0.18 at regional background sites (Zhou et al., 2020), yet much higher than those observed at urban and suburban sites. These results suggest that OA at the SH site was relatively aged.

The ratio of fragment ions NO^+ (m/z 30) to NO_2^+ (m/z 46) is a good indicator for identifying the presence of ONs (Farmer et al., 2010; Lin et al., 2021). The mean ratio of $\text{NO}^+/\text{NO}_2^+$ in C1 was 4.9, which exceeds the value of 3.88 for pure ammonium nitrate (NH_4NO_3) obtained from the AMS IE calibration, indicating a potential contribution of ONs. Conversely, the mean $\text{NO}^+/\text{NO}_2^+$ in EP5 (3.9) was almost identical to that of NH_4NO_3 , implying the dominance of inorganic nitrates (INs). Additionally, the CH_2SO_2^+ (m/z 79) and CH_3SO_2^+ (m/z 80) ions were used as signature fragments of methanesulfonate, a typical organic sulfur species generated in marine and remote coastal regions (Chen et al., 2019). However, the mean ratios of CH_3SO_2^+ and CH_2SO_2^+ during C1 and EP5 (0.98 and 1.99, respectively) were lower than the value of 2.9 reported in previous studies for MSA, indicating the minimal contribution of MSA (Song et al., 2019). The low contributions of MSA further suggest that aerosols over this region are likely to have negligible oceanic influences. This is also consistent with the

back trajectory results in section 3.2, which demonstrate that PM_{10} levels at this site are dominantly influenced by continental sources in the west and southwest.

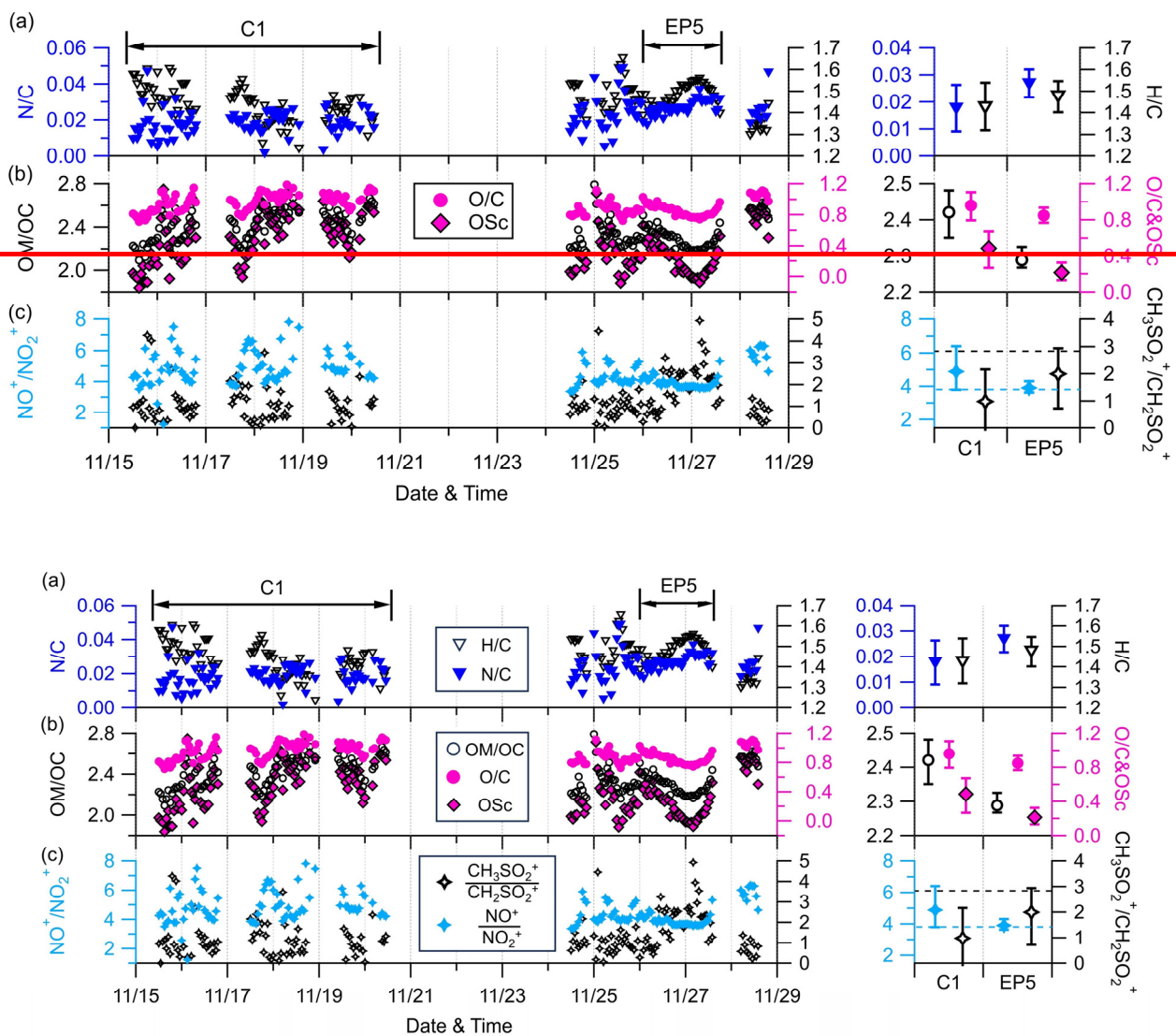
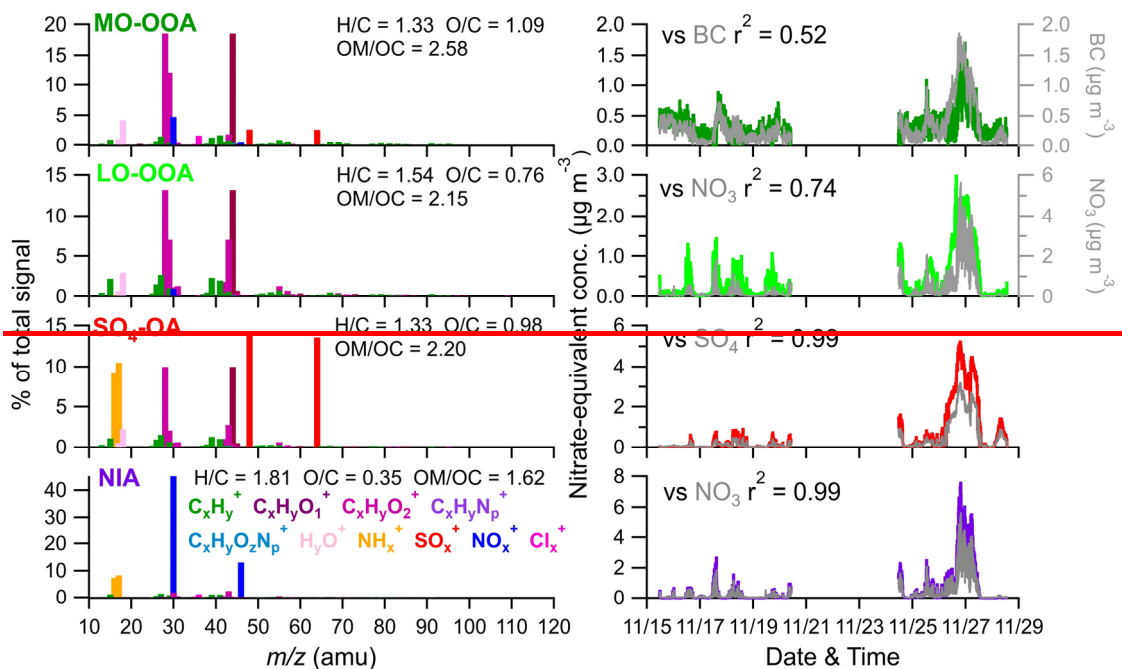


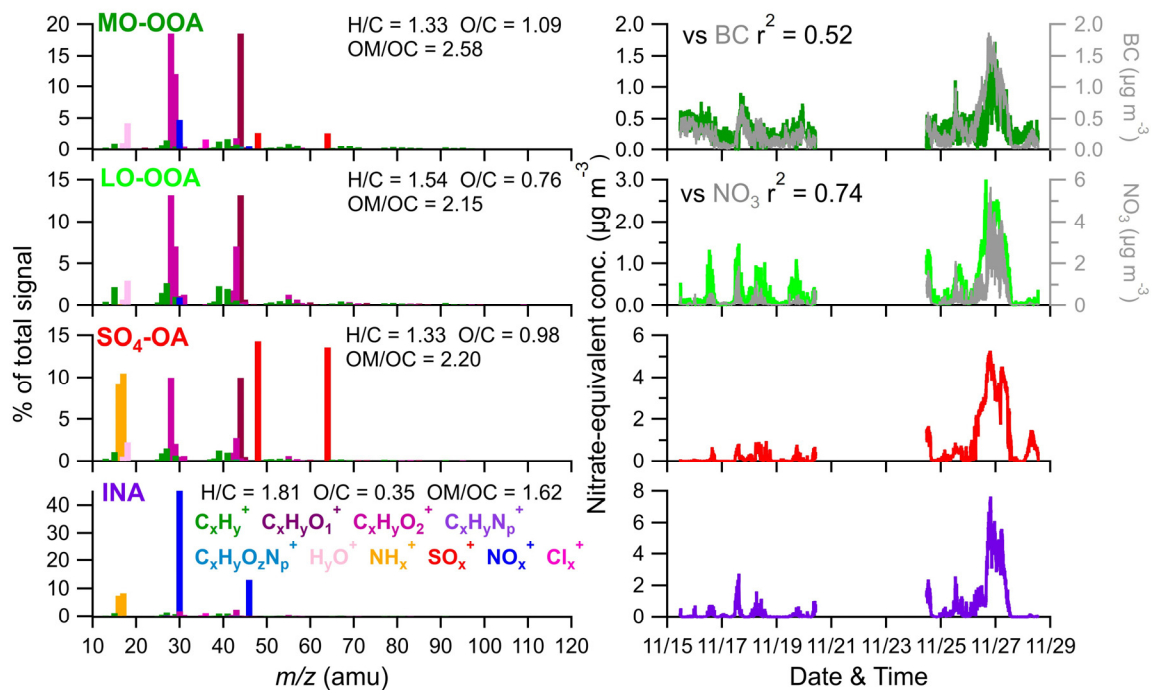
Figure 65. Time series of 1-hour averaged (a) N/C and H/C , (b) OM/OC , OSc , and O/C , and (c) $\text{NO}^+/\text{NO}_2^+$ and $\text{CH}_3\text{SO}_2^+/\text{CH}_2\text{SO}_2^+$ during the AMS sampling site (left) and their mean values in C1 and EP5 (right). Only ratios determined with good S/N (i.e., organics $> 0.7 \mu\text{g m}^{-3}$) are shown. The horizontal black and blue dash line in the bottom right plot represent the $\text{CH}_3\text{SO}_2^+/\text{CH}_2\text{SO}_2^+$ value for pure MSA (2.9) and the $\text{NO}^+/\text{NO}_2^+$ value for pure NH_4NO_3 (3.88), respectively.

3.4 Source apportionment of OA and contribution of organic nitrates

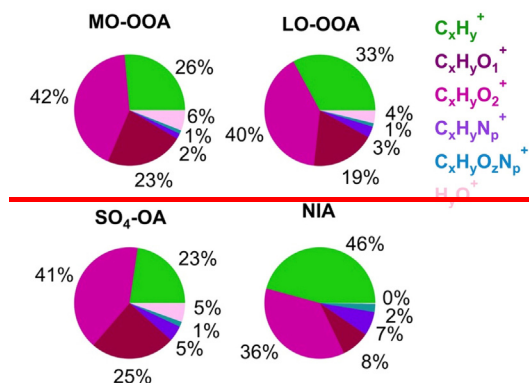
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Four factors were resolved by PMF, including three types of SOA and one inorganic factor: less oxidized oxygenated OA (LO-OOA), more oxidized oxygenated OA (MO-OOA), OA associated with sulfate ions (SO₄-OA), and inorganic nitrate aerosol (NIAINA). These four factors together on average accounted for 87.5 % of the total NR-PM₁ mass. The mass spectra profiles and OA ion family composition of the four factors are shown in Figs. 76 and 87.





360 **Figure 76.** High-resolution mass spectral profiles (left) and time series (right) of four factors. The correlations of four factors with corresponding tracers are also shown.



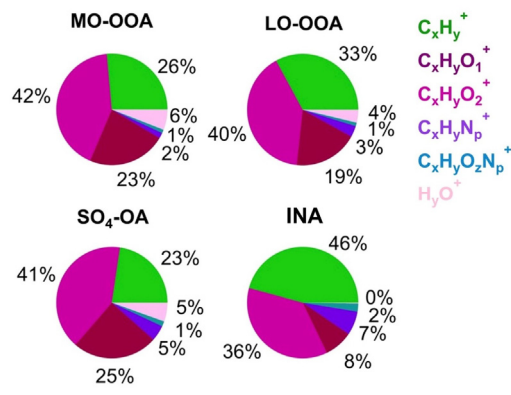
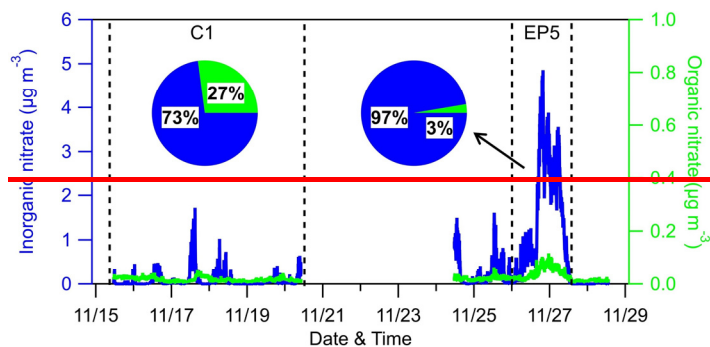


Figure 87. Mass fractional composition of OA ion families for the resolved four PMF factors.

365 LO-OOA and MO-OOA were identified by prominent peaks of CO^+ and CO_2^+ in the mass spectra. LO-OOA had a high fraction of $C_2H_3O^+$ (m/z 43, 6.8 % of the total signal), while MO-OOA, corresponding to more oxidized and aged components, had a higher abundance of CO_2^+ (17.6 % vs. 12.8 %), $C_xH_yO_2^+$ (23 % vs. 19 %), and O/C ratio (1.09 vs. 0.76) than LO-OOA. The average O/C ratios of these two OOA factors are similar to those at Mt. Bachelor Observatory, where the O/C ratios of SV-OOA and LV-OOA were 0.67 and 1.17, respectively (Zhou et al., 2019). LO-OOA correlate well with NO_3 ($r^2 = 0.74$), coinciding with their secondary nature. ~~Meanwhile, MO-OOA exhibit a moderate correlation with BC ($r^2 = 0.52$), indicating that they were likely coated on BC particles and underwent~~ However, unlike numerous previous studies, there was a limited correlation between MO-OOA and sulfate ($r^2 = 0.32$) during the sampling period. Instead, MO-OOA exhibited a moderate correlation with BC ($r^2 = 0.52$). This correlation between OOA and BC was also observed at a mountain site in southern China (Zhu et al., 2016), suggesting common air masses for MO-OOA and BC, such as biomass burning. Given that biomass burning plays an important role in aerosol composition in southeastern China (Fang et al., 2023; Liang et al., 2020), it is plausible that MO-OOA may have coated or mixed with BC particles emitted from biomass burning, undergoing extensive aging processes during long-range transport. Overall, the LO-OOA and MO-OOA components account for 39.1 % and 29.4 % of the total measured OA mass, respectively (Fig. S14-S15). The mass spectra of the SO_4 -OA factor had a large amount of NH_x^+ and SO_x^+ , together accounting for 48.5 % of the total mass of this factor. Meanwhile, organic components also made up a considerable fraction, yet on average contributing 51 %. It is also noticed that the O/C ratio of the organic fraction of this factor is 0.98, which ~~is was~~ even higher than LO-OOA and comparable to MO-OOA, suggesting that this factor has experienced aging processes during the regional transport. As expected, no primary OA factor (e.g., hydrocarbon-like OA, biomass burning OA, etc.) was resolved during this study due to the negligible influences of local emissions, which was consistent with the PMF results at other background sites (Zhou et al., 2019; Zhu et al., 2016).

385 An inorganic nitrate aerosol factor was also separated from these OA factors, with nitrates in this factor accounting for 92.6 % of the total NO_x^+ ions. Apart from ~~NIA~~INA, NO_x^+ ions were more assigned in MO-OOA (6.0 % of the total NO_x^+) than LO-OOA (1.4 % of the total NO_x^+), suggesting that ONs were more associated with MO-OOA. This is contrary to other studies

which reported that ONs were more correlated with less oxidized OA (Zhang et al., 2016; Yu et al., 2019). ~~One~~Since MO-OOA also correlated considerably with BC, one possible reason ~~is~~for this was that ONs formed and mixed with MO-OOA components during the aqueous aging processes of MO-OOA-coated BC particles. This hypothesis is supported by Cao et al. (2022), which demonstrated that ONs exhibit similar volatility to that of MO-OOA when coated on BC. Based on the PMF results above, the mass concentrations of ONs during the AMS sampling period were estimated (Fig. 98). Considering ONs as part of organics, we chose a RIE value of 1.4 for the estimated ONs, while a nitrate RIE value (1.1) was correspondingly applied for INs. The average mass concentration of ONs in C1 was $30 \pm 22 \text{ ng m}^{-3}$, which was lower than that in EP5 ($40 \pm 23 \text{ ng m}^{-3}$), which can also be explained by the evaporative release of ONs in EP5. However, considering the slightly elevated values (10 ng m^{-3}) between these two periods, we cannot rule out the possible formation of ON from aqueous phase processes (Xian et al., 2023) and gas phase reaction initiated by NO_3 during nighttime (Ayres et al., 2015). Also, the low levels of ON might cause uncertainties in its estimation. Since ONs at this site were close to the value (40 ng m^{-3}) reported by Hao et al. (2014) at a forest-urban mixed site in Finland, we considered our quantification of ONs reasonable. However, significant differences were observed between the INs mass concentrations during C1 and EP5 ($0.08 \mu\text{g m}^{-3}$ vs. $1.47 \mu\text{g m}^{-3}$). This could be attributed to that water-soluble HNO_3 or NH_4NO_3 was scavenged by cloud droplets and removed by precipitation before transporting to this site during C1, while in EP5, these INs dominated the elevation of total nitrate during cloud evaporation. These differences in INs also led to a greater contribution of ONs to total nitrate in C1 than in EP5 (27 % vs. 3 %).



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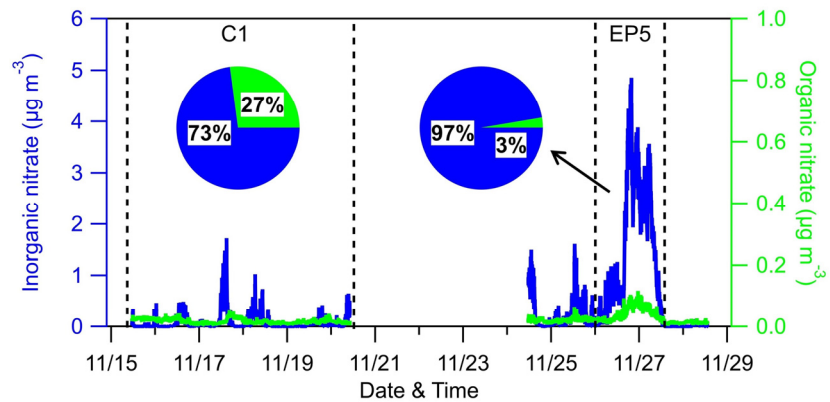


Figure 98. Time series and relative contributions of inorganic nitrate and organic nitrate during the AMS sampling period.

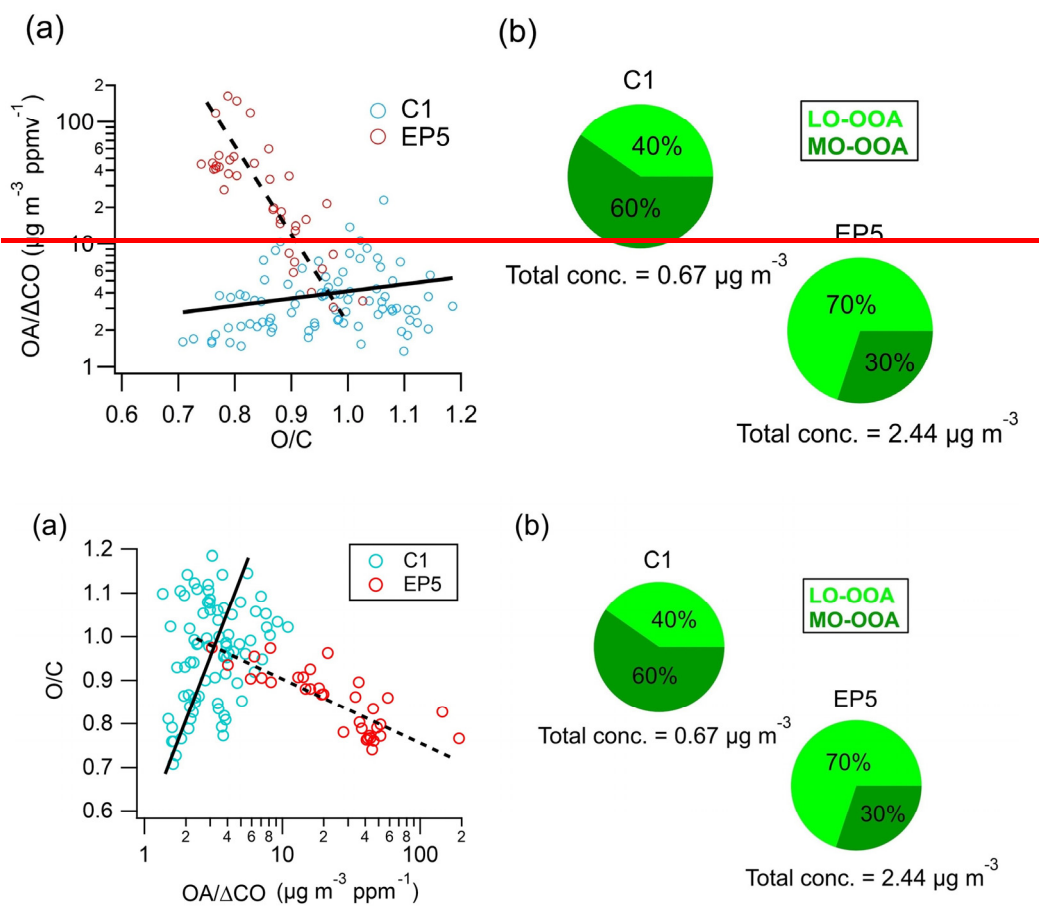


Figure 109. (a) Scatter plot of $OA/\Delta CO$ as a function of O/C ratios and (b) mass concentrations and proportions of MO-OOA and LO-OOA during C1 and EP5.

3.5 Evolution of OA

The formation and evolution of OA can be investigated using the ratio of OA to ΔCO (CO minus background CO) to remove the atmospheric dilution effects (Decarlo et al., 2010). In this study, 0.12 ppm (average of the lowest 5 % concentration) was used as the background mixing ratio of CO (Fig. S12S16), which is close to the 0.1 ppm used in Hu et al. (2013) and Yuan et al. (2013). OA/ ΔCO was $28.3 \pm 26.3 \mu\text{g m}^{-3} \text{ ppm}^{-1}$ during the study period, which is comparable to ($41.7 \pm 23.0 \mu\text{g m}^{-3} \text{ ppm}^{-1}$) in suburban Sichuan Basin (Hu et al., 2016) but much lower than the mean value ($70 \pm 20 \mu\text{g m}^{-3} \text{ ppm}^{-1}$) in worldwide urban air (De Gouw and Jimenez, 2009). The scatter plot of OA/ ΔCO as a function of O/C ratios during C1 and EP5 is shown in Fig. 10a9a. Interestingly, different OA/ ΔCO variations were found with the increasing O/C during C1 and EP5. During C1, OA/ ΔCO tended to increase with the increase of the O/C ratio, indicating aging process produced SOA (Hu et al., 2017). In contrast, a remarkable decrease trend of OA/ ΔCO was observed with the increasing O/C during EP5, suggesting that less oxidized OA may contribute more significantly to the high OA concentration during this period. Moreover, considering the cloud evaporation process in this period, (as discussed in Sect. 3.2), the negative correlation between OA/ ΔCO and O/C in EP5 also implies that the less oxidized OA previously formed or incorporated into cloud droplets can be released during cloud evaporation. Another possible mechanism was that after the cloud was evaporated, SOA formation from biogenic VOCs could then be strengthened by stronger solar radiation, which may also contribute to the increase of less oxidized OA. Conversely, more oxidized OA in cloud droplets may have already further undergone mass reduction through fragmentation reactions (that decrease the carbon content of particles, resulting in their mass loss (Kroll et al., 2009; Lee et al., 2012), which was far). This made MO-OOA much less likely to be reintroduced into the atmosphere through cloud evaporation. Similarly, significant increases in the LO-OOA concentration (0.27 to $1.70 \mu\text{g m}^{-3}$) and fraction (40 % to 70 %) were observed from C1 to EP5, while the MO-OOA concentration (0.40 vs. $0.74 \mu\text{g m}^{-3}$) did not show large variation (Fig. 10b9b), further supporting our conclusion.

Figure 11a10a shows the Van Krevelen diagrams of OA in this study. The slope of H/C to O/C in the present study was -0.66 ($r^2 = 0.64$), suggesting. Despite potential variations in the relative contributions of different types of OAs transported to this site, this slope still suggests the addition of carboxyl functional groups during OA evolution to some extent (Heald et al., 2010). This slope is slightly flatter than those (-0.7 to -1.0) of other remote/rural regions across the world (Chen et al., 2015), indicating the oxidation processes of OA at this site were more associated with fragmentation reactions. Consistent evolution trends are also shown in the f_{43} vs. f_{44} space (Fig. 11b10b). The SO_4 -OOA and MO-OOA showed similar high oxidation degrees, with f_{43} and f_{44} located at the upper part of the triangular region because of the larger fractional contribution of CO_2^+ in the organic mass spectrum. The LO-OOA was situated in the middle region of the triangle, while NIAINA resided near the bottom right. Moreover, the mass spectra of LO-OOA and MO-OOA resembles to those of the aged OAs observed at other elevated sites (Fig. S17) (Xu et al., 2018a; Zhou et al., 2019). These results together reveal that OA observed at SH site is representative of the background-aged SOA in the YRD region in China.

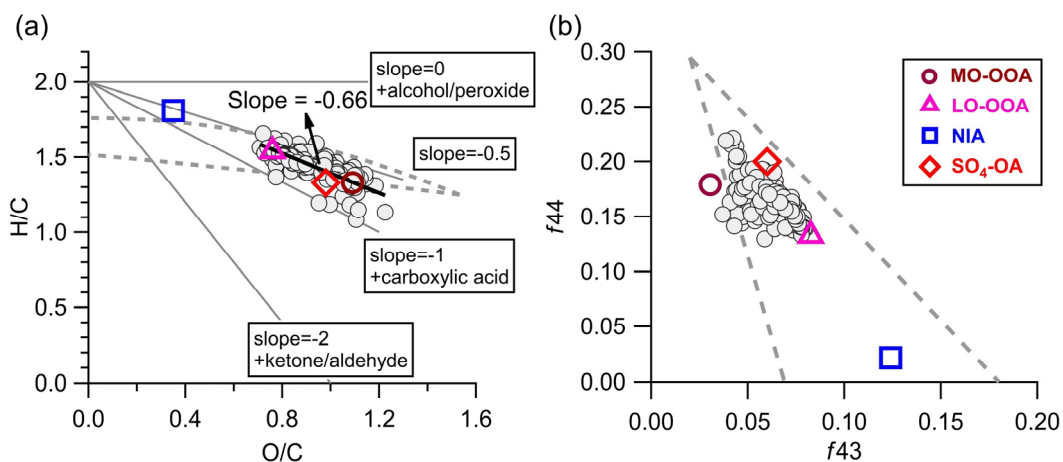


Figure 1110. (a) Van Krevelen diagram (H/C vs. O/C) and (b) f_{43} vs. f_{44} of OA (grey circles, hourly averaged) and four aerosol factors identified by the PMF analyses during the AMS sampling period.

4 Conclusions

450 Chemical composition and sources of PM_{10} at a forested mountain site in southeastern China in November 2022 were characterized by two different aerosol mass spectrometers. The average mass concentration of total PM_{10} ($4.45 \pm 6.513 \pm 4.8$ $\mu\text{g m}^{-3}$) was overall lower than those at other mountain sites in China, yet it was similarly dominated by organics (41.1 %). Remarkably, sulfate exhibited lower contributions (16.7 %) to PM_{10} compared to other mountain sites, in contrast to higher contributions of nitrate (14.7%), indicating the influences of anthropogenic emissions over a relatively small regional scale

455 in southeastern China. Back trajectory analysis revealed that higher concentrations of submicron aerosols at this mountain site were associated with the transport from the western and southwestern regions. OA was dominantly secondary and highly aged as suggested by the high O/C (0.85–0.96) and OSc (0.21–0.49) ratios. PMF analysis of combined organic and inorganic spectra identified two types of OOA and an OA factor associated with SO_4 . Most importantly, we found that cloud scavenging and evaporation processes influence LO-OOA and MO-OOA differently. The cloud scavenging showed a greater

460 efficiency in removing MO-OOA, consistent with the smaller size distributions of interstitial particles during cloud events. In contrast, the evaporation of cloud tended to release a large amount of LO-OOA, highlighting that SOA remained in cloud droplets was mostly in a moderate oxidation state. Overall, this study illustrates the importance of SOA in forested mountain site in southeastern China, where aerosol concentrations, composition, size distributions, and oxidation state are affected by aerosol-cloud interactions substantially. Moreover, our study provides valuable data and insights into understanding the

465 aerosol-cloud interactions in mountainous areas that are often under cloudy conditions, and cloud evaporation can be considered as a plausible mechanism to explain certain aerosol episodes in these regions.

Data availability. Data used in this study can be accessed at repository under: <https://doi.org/10.5281/zenodo.10312334> (Zhang et al., 2023).

Author contribution. WX and YS designed the research. WX, WZ, and HQ conducted the measurements. ZZ, YZ, WZ, XX, AD, YZ, XC and YS analysed the data. YK, XP, ZW, LL, QF, DRW, and YS reviewed and commented on the paper. ZZ and YS wrote the paper.

Competing interests. DRW is the manufacturer of AMS and ACSM utilized in this study.

~~utilized in this study. The authors declare that they have no conflict of interest.~~

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