We thank the editor and referees for their comments. To guide the review process, we have copied the referee comments in **black text**. Our responses are in regular **blue font**. We have responded to all the referee comments and made alterations to our paper (in ***italic text***) and removed redundancies for clarification.

Response to Referee#1

We are thankful to referee#1 for the thoughtful comments that help improve the manuscript significantly. We have made the following key revisions to the manuscript:

1. To better utilize the ACSM data, analyses of six selected periods, including five PM episodes (EP1−EP5) and one clean period (C1), were added to the revised manuscript. **Note that the previous P1 and P2 are now referred to as C1 and EP5, respectively.** The relationship between the PM_1 species concentration, meteorology, and air mass transport during these periods has been clearly evaluated.
2. Prior to discussing the impacts of the cloud process on PM1, the air mass back trajectories during the selected periods have been further investigated to exclude the potential influence of air mass change.
3. To refine our analysis of cloud processes, we have analyzed the vertical distributions of particle depolarization ratios (PDR) measured by a particle lidar, further confirming the occurrence of cloud evaporation during the sampling period.

These discussions have been integrated into a new section (section 3.2) in the revised manuscript. Please see our detailed point-to-point responses below.

Specific comments:

RC1-1: This is contrary to many other studies that reported higher O/C ratios for OA in cloud/fog droplets than ambient OA, what could be the reasons?

Response: Thank the referee's comment. It is true that O/C values for OA have been previously observed to increase in cloud droplets via aqueous or heterogeneous oxidation. However, these results were mainly based on observations from relatively fresh OA with low O/C. For example, Hao et al. (2013) found a higher fraction of LV-OOA and a lower fraction of SV-OOA in cloud residential OA (43.6% and 46.3%) than interstitial OA (52.2% and 42.7%) at a semiurban station, but the O/C values of LVOOA and SV-OOA were only 0.41 and 0.74, respectively. While at background mountain sites with no anthropogenic emissions around, OAs are usually more highly oxidized because of aging during atmospheric transport. Previous observations at mountain sites only showed small and insignificant differences between the ambient/interstitial OA and droplet residual OA (Drewnick et al., 2007; Gao et al., 2023). In addition, further oxidation of these aged OA (O/C > 0.6) in cloud droplets will be dominated by fragmentation reactions instead of functionalization, resulting in reductions in mass (Lee et al. 2012). Therefore, in our study, the mass of MO-OOA with a high degree of oxidation (O/C = 1.09) may be decreased by cloud processing, while LO-OOA remained in the droplets and further being released after cloud evaporation.

References:

Hao, L. Q., Romakkaniemi, S., Kortelainen, A., Jaatinen, A., Portin, H., Miettinen, P., Komppula, M., Leskinen, A., Virtanen, A., Smith, J. N., Sueper, D., Worsnop, D. R., Lehtinen, K. E. J., and Laaksonen, A.: Aerosol Chemical Composition in Cloud Events by High Resolution Time-of-Flight Aerosol Mass Spectrometry. Environ. Sci. Techno., 47(6), 2645-53, https://doi.org/10.1021/es302889w, 2013.

Drewnick, F., Schneider, J., Hings, S. S., Hock, N., Noone, K., Targino, A., Weimer, S., and Borrmann, S.: Measurement of Ambient, Interstitial, and Residual Aerosol Particles on a Mountaintop Site in Central Sweden using an Aerosol Mass Spectrometer and a CVI. J. Atmos. Chem., 56(1), 1-20, https://doi.org/10.1007/s10874-006-9036-8, 2007.

Gao, M., Zhou, S. Z., He, Y., Zhang, G. H., Ma, N., Li, Y., Li, F. H., Yang, Y. X., Peng, L., Zhao, J., Bi, X. H., Hu, W. W., Sun, Y. L., Wang, B. G., and Wang, X. M.: In Situ Observation of Multiphase Oxidation-Driven Secondary Organic Aerosol Formation during Cloud Processing at a Mountain Site in Southern China. Environ. Sci. Techno. Lett., 10(7), 573-81, https://doi.org/10.1021/acs.estlett.3c00331, 2023.

Lee, A. K. Y., Hayden, K. L., Herckes, P., Leaitch, W. R., Liggio, J., Macdonald, A. M., and Abbatt, J. P. D.: Characterization of aerosol and cloud water at a mountain site during WACS 2010: secondary organic aerosol formation through oxidative cloud processing. Atmos Chem Phys. 2012;12(15):7103-16. https://doi.org/10.5194/acp-12-7103-2012.

RC1-2: This is confusing, please make the caption more clearer by mentioning exactly at which site the measurements were carried out for this study.

Response: We thank the referee for pointing this out. We have revised this caption to make it clearer.

Text modification: Line 71: "*Location of the sampling site (Mt. Damaojian, red circle on the map).*"

RC1-3: make it a superscript, check and rectify such discrepancies throughout the manuscript

Response: Checked and revised.

RC1-4: Please provide a table with all the instrumentation and corresponding periods

Response: We have added this table in the supplement (Table S1).

Table S1. Summary of the detailed descriptions for the instruments used during the sampling period, including the measured parameters, model, temporal resolution, and sampling time.

Instrument	Species	Model/Company	Time resolution	Sampling time
Q-ACSM	NR-PM_1 (Org, SO_4, NO_3, NH_4, Chl)	Aerodyne Research Inc.	20 min	11.1–30
HR-ToF-AMS	NR-PM_1 (Org, SO_4, NO_3, NH_4, Chl)	Aerodyne Research Inc.	1 min	11.15–20; 11.24–28
AE33	BC	Magee Scientific Corp.	1 min	11.1–30
CO analyzer	CO	Picarro G2401	1 min	11.1–30
Gas analyzers	O_3, NO_x	Thermo Scientific	1 min	11.1–30
Continuous ambient particle monitor	$PM_{2.5}$, PM_{10}	Thermo Scientific	1 min	11.1–30
Data logger	RH, T, WS, WD, P	CR1000, Campbell Scientific Inc.	1 min	11.1–30
Four-cup anemometers	WS, WD	Model O1OC and O2OC, Met One Instruments Inc.	1 min	11.1–30
P Sensor	P	CS106, Vaisala Ltd.	1 min	11.1–30
T/RH probe	T, RH	HC2S3, Campbell Scientific Inc.	1 min	11.1–30
Particle lidar	Depolarization ratio	Science of Light Technologies Co., Ltd	5 min	11.19–20; 11.25–27

RC1-5: What was the malfunction? and is the reported data checked and sanitized for possible errors?

Response: The AMS chopper servo failed to operate correctly between 21 and 24 November, requiring a few days for acquisition, installation, and adjustment of a new unit. As shown in Fig.S1, the NR-PM_1 measured by AMS correlated well with $PM_{2.5}$ measured by the particulate monitor ($r^2 = 0.70$, slope = 0.54), which suggests the reliability of our data.

RC1-6: confusing, PM measured by what?

Response: We are sorry for our incorrect writing. The $PM_{2.5}$ and PM_{10} were measured by continuous ambient particulate monitors (Model 5014*i*, Thermo Scientific). We have revised it in the text.

Text modification: Line 90: "*PM*$_{2.5}$ *and PM*$_{10}$ *were measured using continuous ambient particulate monitors with PM*$_{2.5}$ *and PM*$_{10}$ *cutoff (Model 5014i, Thermo Scientific., USA).*"

RC1-7: by what?

Response: These meteorological parameters were measured by an automatic weather station. Detailed information has been provided in Table S1 (see **RC1-4**).

Text modification: Line 92: "*Meteorological parameters containing temperature (T), RH, wind speed (WS), wind direction (WD), and pressure (P) were measured by an automatic weather station.*"

RC1-8: PM measured by gas analyzers? provide clear description in the methodology part.

Response: Revised as suggested. The updated text can be found in **RC1-6**.

RC1-9: and slopes?

Response: Added as suggested.

RC1-10: ON could also be estimated using NO+/NO2+ ratios as reported in some studies, so why PMF?

Response: Yes, ON can also be directly calculated using NO^+/NO_2^+, as by the following equations (Xu et al., 2015):

$$NO_{2,org} = \frac{NO_{2.meas} \times (R_{meas} - R_{AN})}{R_{ON} - R_{AN}}$$

$$NO_{org} = R_{ON} \times NO_{2,org}$$

However, this calculation needs R_{ON}, the NO^+/NO_2^+ ratio of organic nitrates, which is derived in the laboratory calibration. Unfortunately, we do not have the conditions to produce organic nitrates in the laboratory. Therefore, the PMF method may be more suitable for our study.

Reference:

Xu, L., Suresh, S., Guo, H., Weber, R. J., Ng, N. L.: Aerosol characterization over the southeastern United States using high-resolution aerosol mass spectrometry: spatial and seasonal variation of aerosol composition and sources with a focus on organic nitrates. Atmos. Chem. Phys., 15(13), 7307-36, https://doi.org/10.5194/acp-15-7307-2015, 2015.

RC1-11: why this height?

Response: Using 1100 m as the endpoint height stems from the altitude of our sampling location, which is 1128 m. Previous studies conducted at mountain sites also chose the altitude of the mountain to be the arrival height (Li et al., 2011; Li et al., 2015). To clarify this, we have corrected the term "height" to "*arrival height at the site*" in the text.

Reference:

Li, W. J., Zhang, D. Z., Shao, L. Y., Zhou, S. Z., and Wang, W. X.: Individual particle analysis of aerosols collected under haze and non-haze conditions at a high-elevation mountain site in the North China plain. Atmos. Chem. Phys., 11(22), 11733-44, https://doi.org/10.5194/acp-11-11733-2011, 2011.

Li, T., Wang, Y., Li, W. J., Chen, J. M., Wang, T., and Wang, W. X.: Concentrations and solubility of trace elements in fine particles at a mountain site, southern China: regional sources and cloud processing. Atmos. Chem. Phys., 15(15), 8987-9002, https://doi.org/10.5194/acp-15-8987-2015, 2015.

RC1-12: In fig. 2, it doesn't seem that PM2.5 ever crossed 50 ug/m3, so max PM1 levels of 57.95 ug/m3 seems off, please explain.

Response: We thank the referee for pointing out this problem. In the revised manuscript, we have included a comparison of the NR-PM_1 species masses as measured by both ACSM and AMS (Fig.S1). Results show that ACSM tends to overestimate the PM_1 concentrations by a factor of 0.69. This overestimation is likely due to the low ambient PM_1 masses in this background site, which are close to the ACSM detection limit. Therefore, the ACSM data was corrected by using the regression coefficients between ACSM and AMS. After correction, the maximum PM_1 level decreased to 39.8 μg m^{-3}, which is below the maximum concentrations of $PM_{2.5}$ (41.1 μg m^{-3}) and is thus more reasonable.

Text modification: Line 143: "*The total PM_1 concentration varied dynamically from 0.3 μg m^{-3} to 39.8 μg m^{-3} during the whole sampling period, with an average (± 1σ) of 4.3 ± 4.8 μg m^{-3}.*"

RC1-13: High PM1 even had 20.65 ug/m3 max conc. then when that 57.95 ug/m3 lPM1 level event happened?

Response: We are sorry for the confusion brought to the referee. The 20.65 μg m^{-3} refers to the maximum PM_1 concentration in EP5 measured by AMS, while the 57.95 μg m^{-3} (now 39.8 μg m^{-3} after correction) is the maximum PM_1 concentration during the whole campaign measured by ACSM. To clarify this, we have added an explanation for the PM_1 level of 39.8 μg m^{-3} in the relevant sentence. In addition, since we have reanalyzed and rewrote this section, the relevant sentence about the PM_1 level of 20.65 μg m^{-3} has been deleted.

Text modification: Line 143: "*The total PM_1 concentration varied dynamically from 0.3 μg m^{-3} to 39.8 μg m^{-3} during the whole sampling period, with an average (± 1σ) of 4.3 ± 4.8 μg m^{-3}.*"

RC1-14: I am not convinced with the identification/naming of P1 and P2 events, evidences are not clear to term them as cloud and evaporating cloud episodes. Rh 100% or 90% doesn't necessarily indicating cloud or its evaporation.

Response: Thank the referee for pointing this out. To further support our conclusions, we have incorporated an analysis of the vertical distribution of particle depolarization ratio measured by a particle lidar during C1 and EP5 in the revised text (Fig. S8). Figure S8 shows the particle depolarization ratio measured by 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 (Fig. S8a). As expected, these clouds gradually disappeared afterward due to strong solar radiation at noon on the mountaintop, causing cloud droplet evaporation. Meanwhile, a large amount of aerosol particles (PDR of ~0.15) was released from the clouds, which showed a strong agreement with the timing of the PM_1 episode. This cloud evaporation phenomenon was also found in urban Guangzhou, which caused remarkably enhanced nitrate mass concentration (Tao et al., 2018). In contrast, no such evaporation process was observed during C1 (Fig. S8b). Instead, high PDRs were found around 0 km, which means that clouds constantly existed near the sampling area during this period, coinciding with the constant 100% RH. Therefore, PM_1 transported to this site was likely to be scavenged by the cloud during C1.

In addition, we have also included two photos taken in these two periods in the supplement (Fig.S9), which clearly show the weather conditions at that time. Severe cloud cover was observed in C1 (Fig. S9a), yet the weather was generally sunny and cloudless in EP5 (Fig. S9b).

Following the referee's comments, we have added these discussions to the revised manuscript.

Text modification: Lines 227-239: "*Figure S8 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 (Fig. S8a).*

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 PM_1 *peak. These particles were then scavenged by the increased RH 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. S8b). Instead, high PDR values were found generally at around 0 km, which means that clouds constantly existed near the sampling area during this period, coinciding with the constant 100% RH. Therefore,* PM_1 *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. S9a), yet the weather was generally sunny and cloudless in EP5 (Fig. S9b). As a result, besides regional transport, cloud processes can also have significant impacts on aerosol particles at this site.*"

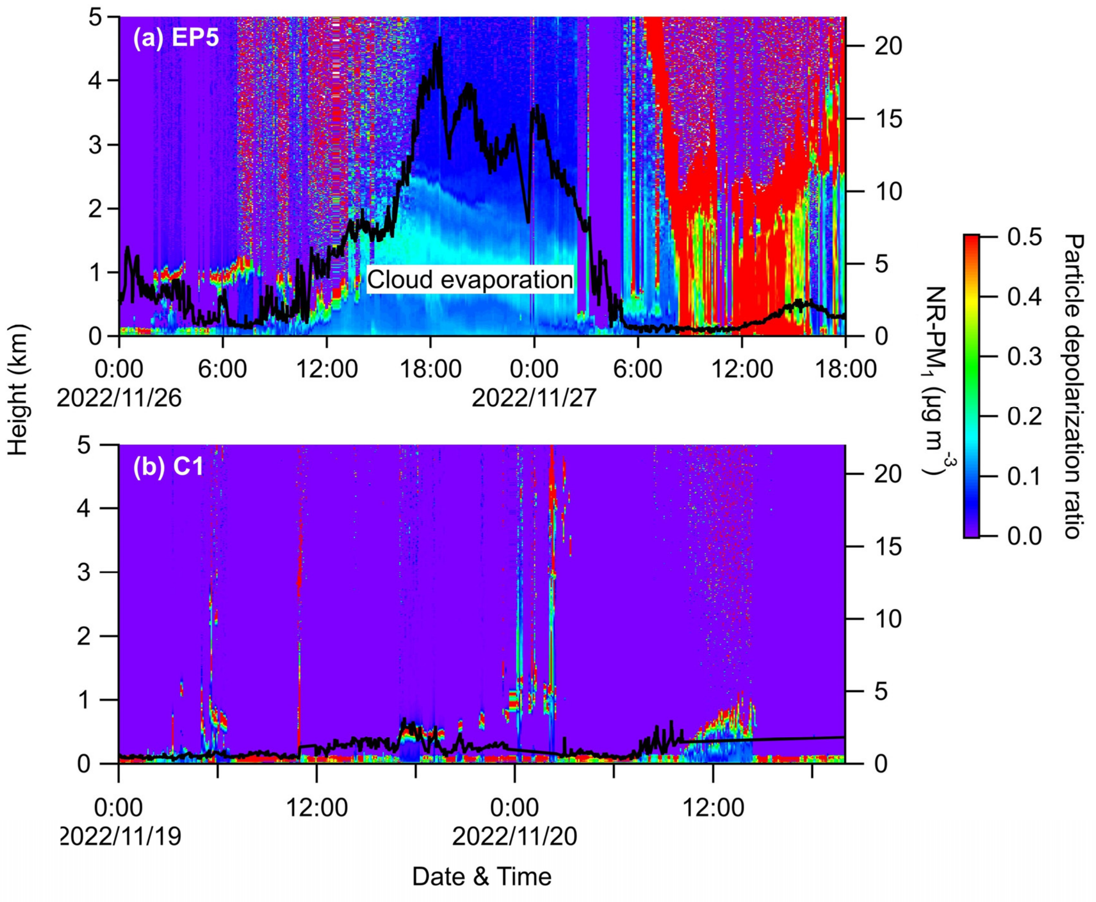

Figure S8. Vertical distribution of particle depolarization ratios in (a) EP5 and (b) C1.

Figure S9. The weather in (a) C1 and (b) EP5 (photos were taken on the roof of the sampling site at local time ~15:00 on 19 Nov and 26 Nov)

References:

Tao J, Zhang Z, Tan H, Zhang L, Wu Y, Sun J, et al. Observational evidence of cloud processes contributing to daytime elevated nitrate in an urban atmosphere. Atmospheric Environment. 2018;186:209-15. doi: https://doi.org/10.1016/j.atmosenv.2018.05.040.

RC1-15: please move the values in parenthesis after P1 and P2, respectively. Evaporating cloud residue organics in P2 was less oxidized than P1, why so?

Response: Revised as suggested. The possible reason for less oxidized OA in cloud residue can be found in **RC1-1**.

RC1-16: Remove "well" before "aged".

Response: Revised.

RC1-17: MSA is established as a good indicator for cloud/fog processing, why MSA was lower? an explanation is needed.

Response: Thank the referee for pointing this out. MSA is mainly generated in marine and remote coastal regions by the oxidation of dimethyl sulfide (DMS) (Chen et al 2019). However, the East China Sea is 180 km to the east of this site, and the low MSA suggests that aerosols over this region are likely to have negligible oceanic influences. This is further consistent with the back trajectory results, which show that PM_1 at this site is strongly influenced by continental sources at the west and southwest. Following the above suggestions, we have added an explanation in the revised text.

Text modification: Lines 285-288: "*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_1 levels at this site are dominantly affected by continental sources in the west and southwest.*"

References:

Chen, Y. L., Xu, L., Humphry, T., Hettiyadura, A. P. S., Ovadnevaite, J., Huang, S., Poulain, L., Schroder, J. C., Campuzano-Jost, P., Jimenez, J. L., Herrmann, H., O'Dowd, C., Stone, E. A., and Ng, N. L.: Response of the Aerodyne Aerosol Mass Spectrometer to Inorganic Sulfates and Organosulfur Compounds: Applications in Field and Laboratory Measurements. Environ. Sci. Techno., 53(9), 5176-86, https://doi.org/10.1021/acs.est.9b00884, 2019.

RC1-18: Use tracers not used as inputs in PMF, else these comparison doesn't properly justify the factors selection.

Response: We agree that external tracers are needed instead of PMF inputs. We thus used BC and nitrate for comparison with PMF factors. Results show that LO-OOA correlates well with NO_3 (r^2 = 0.74), coinciding with their secondary nature. Meanwhile, MO-OOA exhibits a moderate correlation with BC (r^2 = 0.52), indicating that they were likely coated on BC particles and underwent extensive aging processes during long-range transport.

Graph modification:

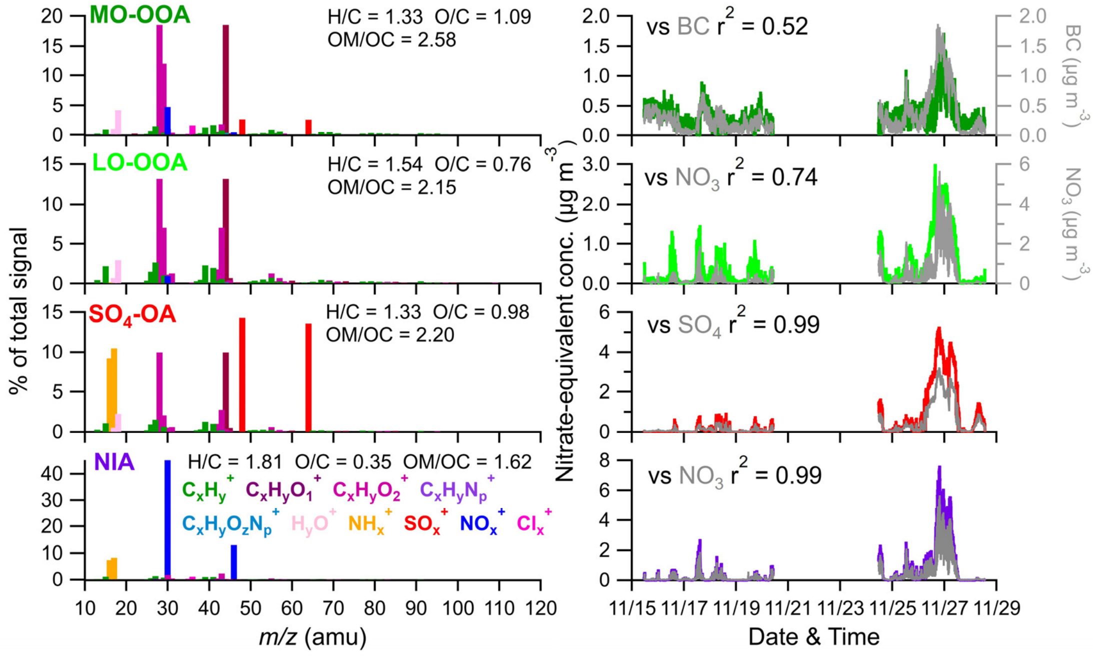

Figure 7. High-resolution mass spectral profiles (left) and time series (right) of four factors. The correlations of four factors with corresponding tracers are also shown.

Text modification: Lines 309-311: "*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 extensive aging processes during long-range transport.*"

RC1-19: Could such low values be detected by AMS/ACSM? please show some uncertainty estimates for ON.

Response: The HR-ToF-AMS has a detection limit of 3 ng m^{-3} for nitrate at 1 min resolution, so the ON with concentrations of 30–40 ng m^{-3} can be detected in our study. In addition, the ON concentrations at this site were close to the value (40 ng m^{-3}) obtained at a forest-urban mixed site in Finland using AMS (Hao et al., 2014), which could also prove that AMS has the ability to quantify ON at such low levels. Considering the referee's comment, we have included the standard deviations of ON mass concentrations.

References:

Hao, L. Q., Kortelainen, A., Romakkaniemi, S., Portin, H., Jaatinen, A., Leskinen, A., Komppula, M., Miettinen, P., Sueper, D., Pajunoja, A., Smith, J. N., Lehtinen, K. E. J., Worsnop, D. R., Laaksonen, A., and Virtanen, A.: Atmospheric submicron aerosol composition and particulate organic nitrate formation in a boreal forestland–urban mixed region, Atmos. Chem. Phys., 14, 13483-13495, https://doi.org/10.5194/acp-14-13483-2014, 2014.

RC1-20: What happened to the OA formed inside the cloud droplets? or no such formation happened?

Response: We thank the referee for pointing this out. According to our reply in **RC1-1**, OA with high oxidation degrees in cloud droplets can undergo fragmentation reactions, resulting in mass reductions. We have added a discussion about this to the revised manuscript However, we cannot because we did not separately measure the cloud residual particles, which is beyond the scope of our study. We hope, in the future, to employ a ground-based counterflow virtual impactor inlet (GC-VI) coupled with an aerosol mass spectrometer to further investigate the physical and chemical properties of aerosols inside the cloud droplets. Therefore, we seek the editor's tolerance and understanding.

Text modification: Lines 355-357: "*Conversely, more oxidized OA in cloud droplets may have already undergone mass reduction through fragmentation reactions (Lee et al., 2012), which was far less likely to be reintroduced into the atmosphere.*"

Response to Referee#2

General comments:

RC2: The study is basically about the interactions between SOA and clouds at a high-altitude background area in China as well as characterizing aerosol chemical composition at the site. The study involves real-time measurements conducted with aerosol mass spectrometry (AMS + ACSM) at 1128 m a.s.l. in southeastern China, in November 2022. The results from the AMS measurements are well presented and the paper is generally well written and the figures are mostly informative and clear. The weakness of the paper is the connection of the AMS data to the cloud events and thereby the fact that the conclusions of the paper are, as I see it, not very well supported by the rest of the paper. Only two time periods are chosen from the time series based on relative humidity (P1 and P2) to the analysis. RH stays at 100 % during P1 indicating in-cloud conditions while during P2, RH drops to about 80% several times. P2 is referred to as an "evaporative event". My main concern is that the authors draw rather strong conclusions on how droplet evaporation e.g., increases the abundance of less oxygenated organic aerosol, but a potential (perhaps likely) change in air mass is not discussed, which could also explain the finding. In addition, due to the inclusion of only two time periods in the analysis, the statistics are rather poor. More events/periods could certainly be included, if the ACSM data were utilized. I have added some comments below, which the authors can consider.

Response: We thank the referee for reading our paper carefully and giving the above valuable comments that helped improve the manuscript significantly. It is really true as the referee suggested that more case studies are needed to be included and the changes in air masses during these periods should be discussed to further support our conclusions. Considering the referee's comments, we have made the following key revisions to the manuscript:

1. To better utilize the ACSM data, analyses of six selected periods, including five PM episodes (EP1−EP5) and one clean period (C1), were added to the revised manuscript. **Note that the previous P1 and P2 are now referred to as C1 and EP5, respectively.** The relationship between the PM_1 species concentration, meteorology, and air mass transport during these periods has been clearly evaluated.
2. Prior to discussing the impacts of the cloud process on PM_1, the air mass back trajectories during the selected periods have been further investigated to exclude the potential influence of air mass change.
3. To refine our analysis of cloud processes, we have analyzed the vertical

distributions of particle depolarization ratios (PDR) measured by a particle lidar, further confirming the occurrence of cloud evaporation during the sampling period.

These discussions have been integrated into a new section (section 3.2) in the revised manuscript. Please see our detailed point-to-point responses below.

Specific comments:

RC2-1: The title of the paper does not reflect much the cloud processing angle that is brought up in the conclusions and abstract. A new title could perhaps be considered also after the authors work on providing more evidence supporting their conclusions.

Response: A very good point. We have changed our title to "**Measurement report: Impact of cloud processes on secondary organic aerosols at a forested mountain site in southeastern China**" to highlight the key point of this study.

RC2-2: L51: "grow via gas-to-aqueous partition" rephrase.

Response: Revised as suggested.

Text modification: Line 57: "*while smaller particles remained growth through the process of gas-to-aqueous partitioning*"

RC2-3: L82: Surely PM2.5 and PM10 were not measured with gas analyzers. See also L96. Please clarify.

Response: We are sorry for our incorrect writing. The $PM_{2.5}$ and PM_{10} were measured by continuous ambient particulate monitors (Model 5014i, Thermo Scientific). We have revised it in the text.

Text modification: Line 90: "*$PM_{2.5}$ and PM_{10} were measured using continuous ambient particulate monitors with $PM_{2.5}$ and PM_{10} cutoff before the sampling inlet (Model 5014i, Thermo Scientific., USA).*"

RC2-4: L76: Was ACSM really operated with <5 min time resolution?

Response: We are sorry for our incorrect writing. Q-ACSM was operated with a time resolution of 6 min during 1st–4th Nov, and 20 min during 4th–30th Nov. This information has been corrected in the text.

Text modification: Line 81: "*PM_1 species were measured using a suite of real-time instruments with 1−20 min time resolution.*"

RC2-5: L94: The RIE for sulfate is slightly low for the ACSM. Did you use a full scan mode (Freney et al., 2019) when calibrating for ammonium sulfate?

Response: Yes, we used a full scan mode during ACSM calibration. In addition, we have re-corrected the ACSM data by using the regression coefficients between ACSM and AMS. Therefore, our data quality in this study was ensured.

RC2-6: L97: PMF is not described appropriately. Figure S.2 is therefore not very useful, as the reader FPEAK,Q, Qexp, and residuals are not introduced anywhere.

Response: We have added a detailed description of the PMF diagnosis in the supplement (Text S1).

Text modification:

"***Text S1.*** *PMF diagnosis*

In this study, organic mass spectra together with several selected inorganic ions (SO^+, SO_2^+, SO_3^+, HSO_3^+, $H_2SO_4^+$, NO^+, NO_2^+, NH^+, NH_2^+, NH_3^+, Cl^+, and HCl^+) were analyzed by the PMF. The number of factors from 1 to 5 with fPeak varying from -1 to 1 were evaluated, and the diagnostic plots are shown in Figure S2. In the PMF analysis, the Q/Q_{exp} values represent the ratios between the actual sum of the squares of the scaled residuals (quality-of-fit parameter, Q) obtained from the PMF least square fit and the ideal Q (Q_{exp}) obtained if the fit residuals at each point were equal to the noise specified for each data point. The Q/Q_{exp} value should be approximate to unity if the number of factors is appropriate and with small errors (Ulbrich et al., 2009). As shown in Figure S2a, The Q/Q_{exp} value started to greatly decrease from one- (1.52) to three-factor solution (1.05). However, the three-factor solution (including one organic associated with sulfate ions factor (SO_4-OA), one inorganic nitrate factor (NIA), and one oxygenated OA (OOA)) cannot further differentiate OOA with different oxidation degrees. The four-factor solution, with a value of Q/Q_{exp} (1.0065) close to 1, can separate two OOA factors with different elemental ratios and temporal variations. Continuing to increase the factor number cannot significantly decrease the Q/Q_{exp}. Furthermore, with 4 factors, the reconstructed mass tracked well with the variations of measured mass, and the scaled residuals of all ion fragments were distributed between -4−4 (Figs. S2c and d), suggesting that they were well reproduced by the PMF model. Therefore, we considered the four-factor solution with fPeak = 0 to be the optimum solution for PMF analysis in this study."

References:

Ulbrich, I. M., Canagaratna, M. R., Zhang, Q., Worsnop, D. R., Jimenez, J. L.: Interpretation of organic components from Positive Matrix Factorization of aerosol mass spectrometric data. Atmos. Chem. Phys., 9(9), 2891-918, https://doi.org/10.5194/acp-9-2891-2009, 2009.

RC2-7: Sect. 2.2.3: How well does HYSPLIT capture actual airmass movements over a complex mountain terrain?

Response: Thank the referee for pointing this out. It is true that the complex terrain in this mountain may make meteorological modeling more difficult than in areas of flatter terrain. However, the aim of using HYSPLIT in this study is to evaluate the change in airmass transport over a large scale during different episodes rather than accurately quantify its contribution to aerosols. The sampling site is located on the top of one of the highest mountains in the region, and according to our results, air pollutants are observed to transport from higher altitudes (please see Fig. S8 in **RC2-20**). Therefore, the disturbance of airmass transport modeling caused by complex terrain may have minor impacts on our results. In addition, HYSPLIT used in other complex terrains such as Sichuan Basin (Lu et al., 2022), Tibetan Plateau (Pokharel et al, 2019), and Mt. Huang (Zhang et al., 2017) also obtained reliable results. In summary, we consider HYSPLIT results to be reasonable in our study.

References:

Lu, H., Xie, M., Liu, X. R., Liu, B. J., Liu, C., Zhao, X. L., Du, L., Wu, Z., Gao, Y, H., and Xu, L. P.: Spatial-temporal characteristics of particulate matters and different formation mechanisms of four typical haze cases in a mountain city. Atmos. Environ., 269, 118868. doi: https://doi.org/10.1016/j.atmosenv.2021.118868, 2022.

Pokharel, M., Guang, J., Liu, B., Kang, S., Ma, Y., Holben, B. N., Xia, X., Xin, J. Y., Ram, K., Rupakheti, D., Wan, X., Wu, G. M., Bhattarai, H., Zhao, C. F., and Cong, Z. Y.: Aerosol Properties Over Tibetan Plateau from a Decade of AERONET Measurements: Baseline, Types, and Influencing Factors. J. Geophys. Res. Atmos., 124(23), 13357-74, https://doi.org/10.1029/2019JD031293, 2019.

Zhang, X. R., Yin Y, Lin, Z. Y., Han, Y. X., Hao, J., Yuan, L., Chen, K., Chen, J. H., Kong, S. F., Shan, Y. P., Xiao, H., and Tan, W.: Observation of aerosol number size distribution and new particle formation at a mountainous site in Southeast China. Sci. Total Environ., 575, 309-20, https://doi.org/10.1016/j.scitotenv.2016.09.212, 2016.

RC2-8: L118: Maybe distinguish what is meant by “particle” here (aerosol particle vs the hybrid single-particle). I also suggest reformulating L117–L121. The sentence is slightly confusing to me.

Response: Revised as suggested.

Text modification: Lines 135-139: “*To further show the aerosol particle concentration levels in different regions, the map contained the back trajectory (Fig. 3) was colored by the time-averaged organic carbon surface mass concentration from* *the M2T1NXAER v5.12.4 dataset (0.5 x 0.625°, hourly). This dataset, part of the Modern-Era Retrospective analysis for Research and Applications version 2 (MERRA-2) (Gelaro et al., 2017), was obtained from the NASA Giovanni website (https://giovanni.gsfc.nasa.gov).*"

RC2-9: L131–L132: Maybe include the standard deviation of the PM1 concentrations from the other mountain sites (if possible) to make a fairer comparison.

Response: Added as suggested.

RC2-10: L135: What about the role of ammonia in nitrate formation?

Response: Thank the referee for pointing this out. Since we did not conduct additional measurements of ammonia (NH_3), we can only evaluate the role of ammonium in nitrate formation. The lower sulfate contribution at this site compared to other mountain sites was associated with higher nitrate contribution, suggesting that NH_4 will probably be in the form of ammonium nitrate that is much more volatile than ammonium sulfate. Also, as shown in Fig. 3b, after removing the nitrate episode (EP1), there was still an increasing trend for nitrate fraction and a decreasing trend fraction for sulfate, while the contribution of ammonium to PM_1 remained constant with increasing PM_1. This result further suggests that there was a conversion of ammonium sulfate to ammonium nitrate with the increase in PM_1 concentration. We have added these discussions to the revised manuscript.

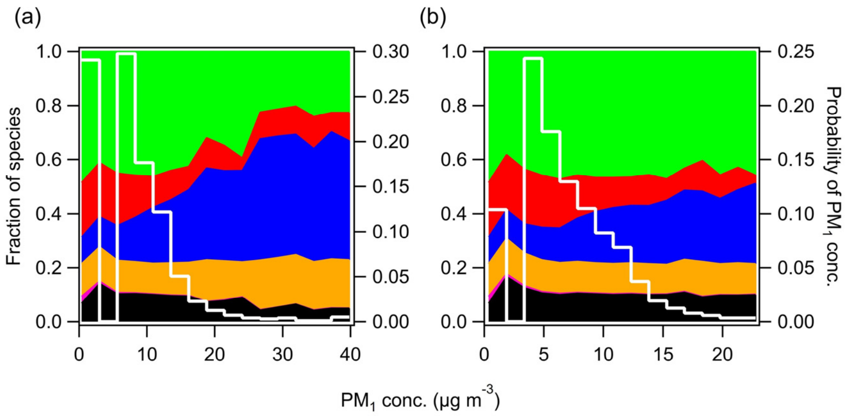

Figure 3. Variations of aerosol composition as a function of PM_1 mass concentration and the probability density of PM_1 during (a) the entire campaign and (b) the campaign without EP1.

Text modification: Lines 158-163: "*The decrease in sulfate contribution is associated with an elevation of nitrate contribution. This shift suggests that most ammonium 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, at Mt. Tai, even though the contribution of sulfate is high, nitrate still accounts for a considerable fraction (14.9 %−29.0 %) that is comparable to this site. Taken together, PM_1 at this site is more likely influenced by anthropogenic emissions over a smaller regional scale, consistent with the high urban density in eastern China.*"

Lines 175-179: "*Despite this, there was still an increasing trend for nitrate and a decreasing trend for sulfate along with increased PM_1, 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 of PM1 concentration.*"

RC2-11: L135: "impact on regional scale", please specify

Response: Revised. Please see **RC2-10** for the updated text.

RC2-12: L136: How does nitrate interact with clouds? Please specify.

Response: Thank the referee's comments. To avoid confusion, we have deleted this sentence. Instead, after careful evaluation, we found that high concentrations of nitrate (e.g., nitrate elevation during EP1) were more likely produced by the transport of NO_x followed by heterogeneous uptake of N_2O_5 under high RH at night. We have discussed this possible mechanism of nitrate formation in section 3.2 of the revised manuscript. Please see the following revised text:

Text modification: Lines 199-204: "*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. S6) showing high levels of nitrate mainly associated with northeastern wind directions. This was further supported by the back trajectory analysis, in which air masses on 4 Nov were also from the northeast. Moreover, the RH was stable at 100% during EP1, which could favor the heterogenous uptake of N_2O_5 and subsequent nitrate formation in the aqueous phase at night (Brown et al., 2006; Li et al., 2020).*"

RC2-13: L139: What do you mean by the highest two probabilities?

Response: As shown in the white line in Fig. 3, the highest two probabilities mean the two most frequently occurring PM_1 concentration ranges during the sampling period. To avoid confusion, we have added an explanation about it in the revised manuscript.

Text modification: Line 165: "*The highest two frequences of PM_1 concentrations* *were distributed within 0–3 μg* m^{-3} *and 5–8 μg* m^{-3} *(48.5 % and 30.1 %, respectively).*"

RC2-14: L144: do you mean "transport of nitrate"?

Response: We want to emphasize here that transport of both anthropogenic-emitted NO_x and nitrate may have a large impact on nitrate increase during the nitrate episode on 4th Nov. To avoid misunderstanding, we have revised this sentence.

Text modification: Line 177: "*Overall, these results suggested that high levels of* PM_1 *at the SH site might be mainly attributed to the formation or transport of organics, meanwhile, nitrate also plays a nonnegligible role.*"

RC2-15: L154 can't the OA afternoon peak be related to the development of a boundary layer (measuring air that has been in contact with airmasses below/potentially more polluted air masses?)

Response: It is really true that the development of atmospheric boundary layer could be another explanation. This discussion has been added to the text.

Text modification: Line 190: "*In addition, these noon peaks may also be related to the development of the atmospheric boundary layer, which facilitates the mixing of air masses transported from polluted areas.*"

RC2-16: L146: P1 and P2 are not explained in the caption.

Response: We have added the explanations of these periods in the caption.

Text modification: Line 182: "*The yellow and purple shaded areas represent six selected episodes (EP1-EP5 and C1, respectively).*"

RC2-17: L163: "Previous studies have shown that aerosol mass generally increases on foggy days" → What do you mean? Please add more references that support this statement. What is meant by foggy days? Overall or during fog? Note also that when comparing your results to others, things might be different in areas where fog formation is driven by radiative cooling.

Response: We thank the referee for pointing out this problem. Indeed, it is inappropriate to evaluate the RH dependence of aerosol species in this study. Also, this sentence is contradictory to the next sentence "This phenomenon could be due to the cloud scavenging effect under high RH at this site.". If fog increases aerosol mass, this cannot be caused by scavenging which would only lead to a mass reduction. In addition, the sampling site is dominated by cloud processes, which can have different influencing mechanisms on aerosol species from fog in other areas. Since we have re-written this section following the referee's general comments (**RC2**), this sentence is no longer needed and is now deleted.

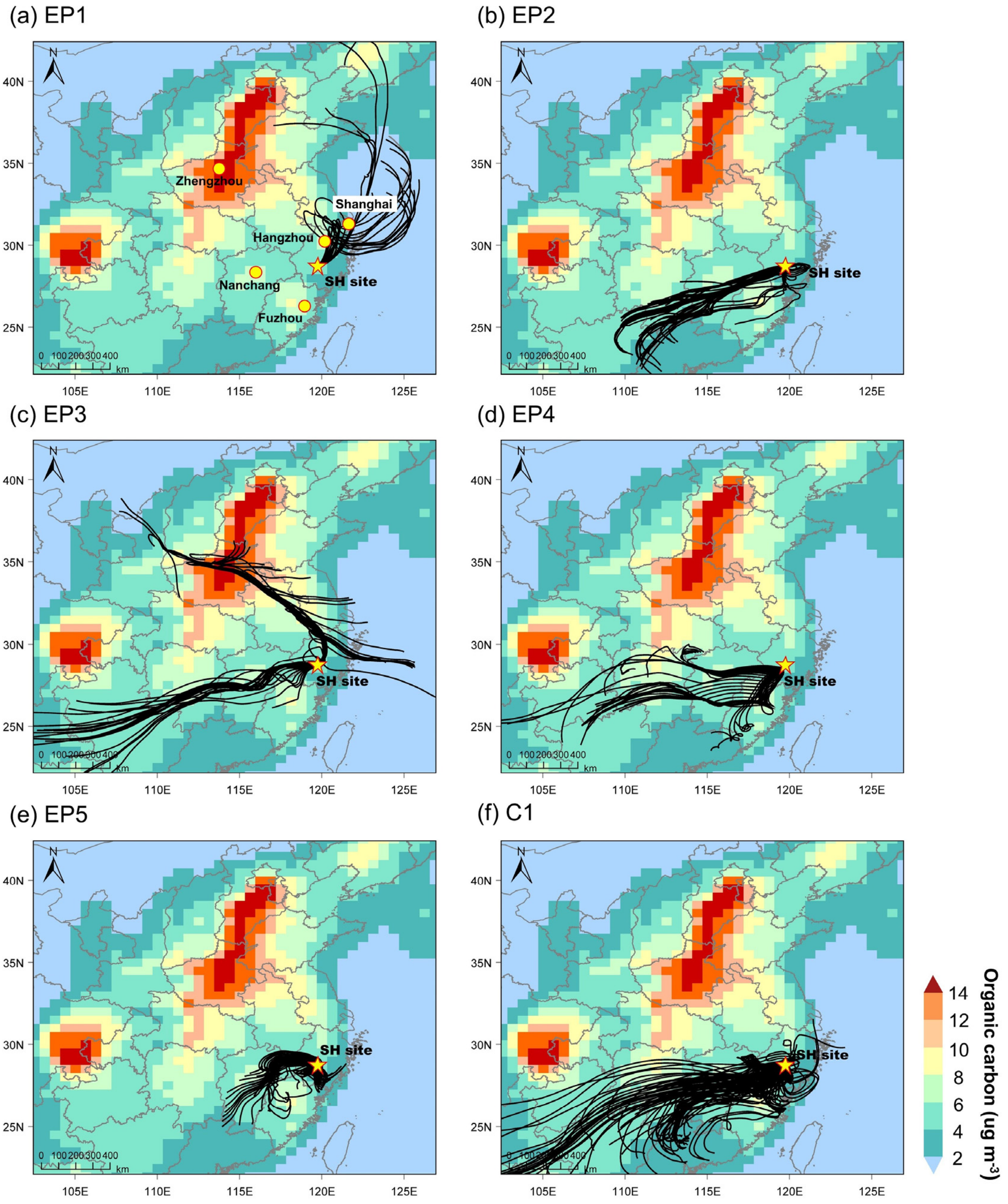

Figure S5. 72 h air mass back trajectories calculated at 1 h intervals at the SH site during six selected periods.

RC2-18: L168: What makes P1 “a typical cloud scavenging period”?

Response: We consider this period as a typical cloud scavenging period mainly for the following reasons:

First, the relative humidity was kept at 100% for the whole period, coinciding with the aqueous nature of cloud droplets. Second, as shown in Fig. S5 and Fig. 4 in the revised manuscript, the air mass trajectories in this period were generally similar to other high PM_1 episodes (EP2–5), and higher levels of CO were also observed (0.37 ppm on average) compared to other episode (0.18–0.31 ppm on average), suggesting the more significant influences of polluted air masses in C1. However, the aerosol concentrations did not increase correspondingly. Third, the cloudy weather can be clearly identified in a photo taken in P1 (Fig. S9a in the revised supplementary). Therefore, the most reasonable explanation is the influence of the cloud scavenging process.

Figure S9. The weather condition in (a) C1 and (b) EP5 (photos were taken on the roof of the sampling site at local time ~15:00 on 19 Nov and 26 Nov).

RC2-19: L169–L172: Description of P2 somewhat hard to follow. Could you rephrase?

Response: Rephrased as suggested. We have rewritten and reorganize this section, please see the detailed revised text.

Text modification: Lines 213-215: "*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.*"

RC2-20: L173: What would cause the evaporation? It looks like temperature goes slightly up. Why is that? Does your air mass change?

Response: Because of the high altitude (1128 m a.s.l.), this mountain site can receive strong solar radiation, which may cause cloud evaporation during EP5.

As shown in Fig. S8 (see **RC2-18**), the back trajectories in this period (EP5) were generally similar to other episodes except for EP1, indicating that the high levels of PM_1 at the SH site may be heavily influenced by the anthropogenic emissions from big cities like Nanchang and Fuzhou, which were relatively polluted according to the organic carbon distribution in Fig. S5 (see **RC2-18**). Therefore, we consider that the influence of air mass change during the cloud evaporation period (EP5) may be negligible. Furthermore, clear evidence has been provided by the vertical distribution of particle depolarization ratios measured by a particle lidar. As shown in Fig. S8, clouds were identified at ~1 km above the sampling site during EP5, which subsequently evaporated at ~12:00 on 26 Nov and released large amounts of aerosols (PDR ~0.15) coinciding with the PM_1 elevation. This cloud evaporation phenomenon was also found in urban Guangzhou, which caused remarkably enhanced nitrate mass concentration (Tao et al., 2018).

We have added the above discussion to the manuscript. Please see our revised text below.

Text modification: Lines 227-239: "*Figure S8 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 (Fig. S8a). 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* PM_1 *peak. These particles were then scavenged by the increased RH 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. S8b). Instead, high PDR values were found generally at around 0 km, which means that clouds constantly existed near the sampling area during this period, coinciding with the constant 100% RH. Therefore,* PM_1 *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. S9a), yet the weather was generally sunny and cloudless in EP5 (Fig. S9b). As a result, besides regional transport, cloud processes can also have significant impacts on aerosol particles at this site.*"

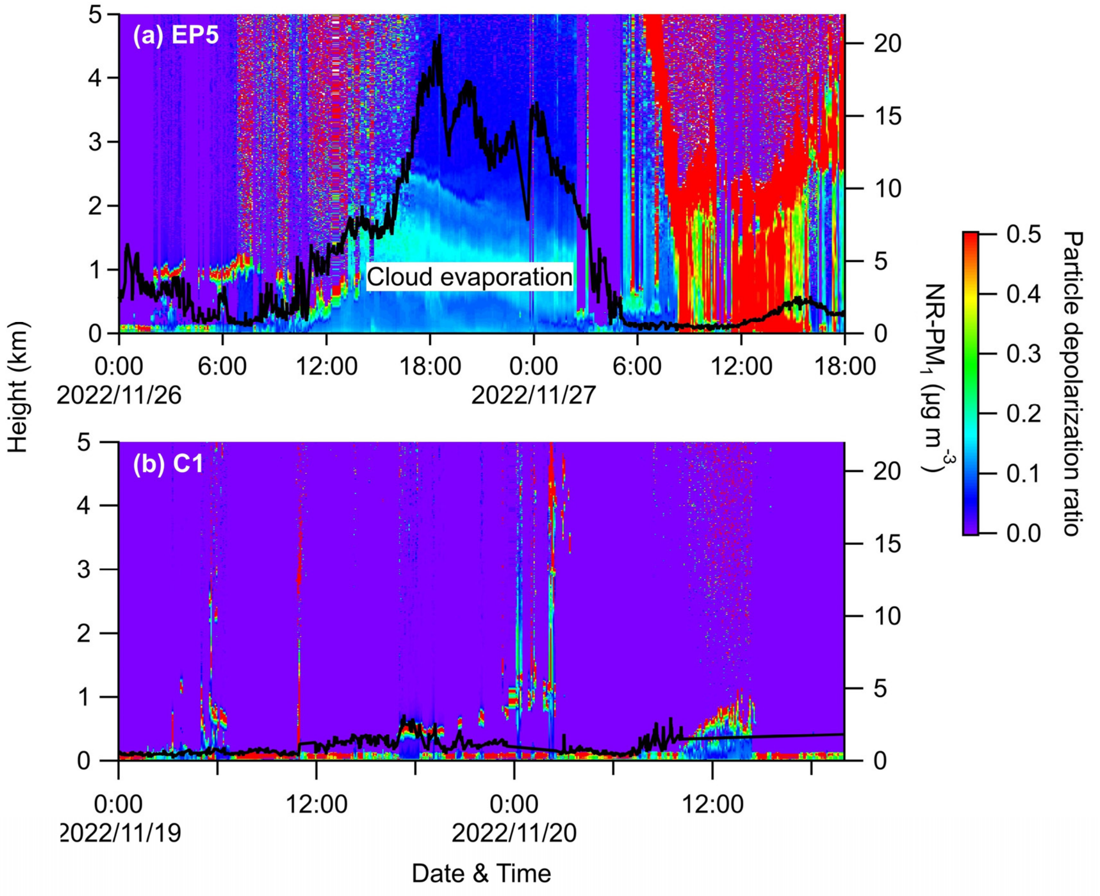

Figure S8. Vertical distribution of particle depolarization ratios in (a) EP5 and (b) C1.

References:

Tao J, Zhang Z, Tan H, Zhang L, Wu Y, Sun J, et al. Observational evidence of cloud processes contributing to daytime elevated nitrate in an urban atmosphere. Atmospheric Environment. 2018;186:209-15. doi: https://doi.org/10.1016/j.atmosenv.2018.05.040.

RC2-21: Sect. 3.2. It seems that an episode similar to P2 took place on 21.11. and conditions similar to P1 took place also before P2 and during the first week of November. Why are these not included? One could use ACSM data to test if your conclusions from the P1 and P2 comparisons hold outside the P1 and P2 time frames? Including more data into the cloudy and "evaporation" periods would make the paper stronger.

Response: We thank the referee for the valuable suggestions. We have added this episode together with the other 3 episodes into analysis in section 3.2 in the revised manuscript.

RC2-22: L182–183: did the mass fractions increase 17 and 19%?

Response: We are sorry for the incorrect writing. The mass factions of sulfate and nitrate in EP5 increased by 9.4% and 7.0%.

Text modification: Line 258: "*by 9.4% and 7.0%, respectively*"

RC2-23: L205: I wonder if what you see with NO+/NO2+ is mostly noise due to low concentrations during P1? Could you show the HR fit of NO+ in the for P1 and P2? I am just wondering if you also fit CH2O+ (see Graeffe et al., 2023).

Response: Thank the referee for pointing this out. Indeed, NO^{+}/NO_2^{+} can be noisy due to low concentrations during P1. However, as shown in Fig. 6 (see **RC2-35**), although NO^{+}/NO_2^{+} was slightly noisy in P1, most of them still exceeded the value of 3.88 for pure ammonium nitrate, suggesting large contributions of ON. Yes, we also fit CH_2O^{+} at *m/z* 30, and the HR fits of NO^{+} in P1 and P2 are shown in Fig. RC2-1.

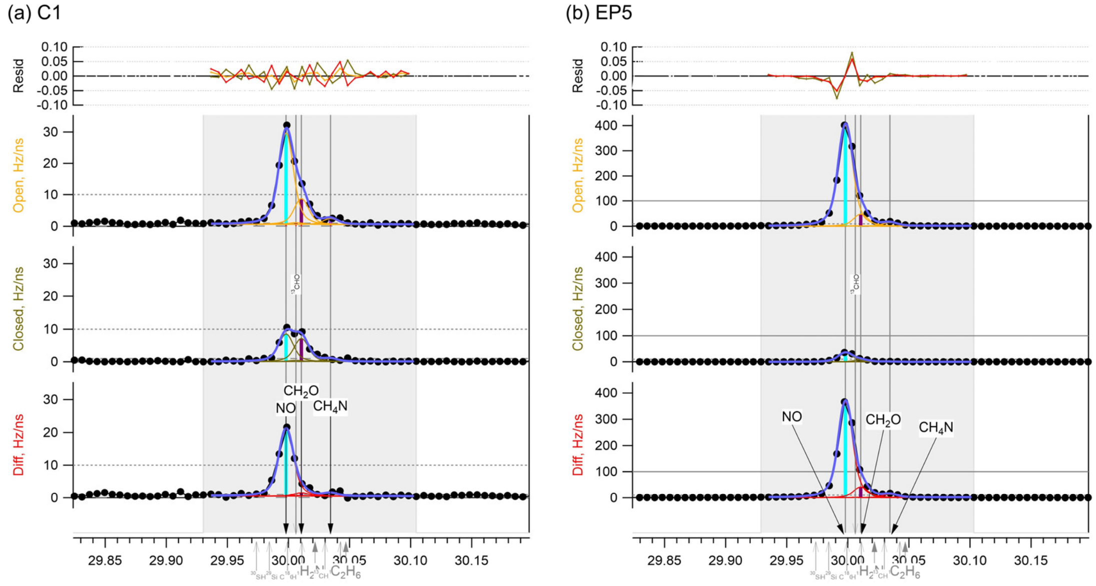

Figure RC2-1. HR fit at *m/z* 30 in (a) P1 and (b) P2.

RC2-24: L240: "Not surprise" could be reformulated

Response: We have revised it to "As expected".

RC2-25: L244: I am surprised to see ON to go with MO-OOA and not LO-OOA due to the semi-volatile nature of ONs. Could you comment on that?

Response: Thank the referee for pointing this out. The MO-OOA we observed at this background mountain station was highly oxidized and correlated well with BC (r^2 = 0.52), which suggested that MO-OOA may be coated on BC particles. Considering the high RH conditions over this mountainous region, ONs may form in aqueous phase of the MO-OOA-coated BC particles. This is further supported by Cao et al. (2022), which demonstrated that ONs exhibit similar volatility to that of MO-OOA when coated on BC (MO-OOA$_{BC}$).

Further studies with long-term measurements are needed to investigate this more deeply. Following the referee's comment, we added a brief discussion about this to the revised manuscript.

Text modification: Lines 322-326: "*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 possible reason is 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.*"

References:

Cao, L.-M., Wei, J., He, L.-Y., Zeng, H., Li, M.-L., Zhu, Q., and Yu, G. H.: Aqueous aging of secondary organic aerosol coating onto black carbon: Insights from simultaneous L-ToF-AMS and SP-AMS measurements at an urban site in southern China. J. Clean. Prod., 330, 129888, https://doi.org/10.1016/j.jclepro.2021.129888, 2021.

RC2-26: L256: How do you know it is evaporation and not a change in air mass?

Response: Thank the referee for pointing this out. The detailed explanation can be found in **RC2-20**.

RC2-27: L273–L274: Why would you only release LO-OOA from evaporating droplets and not MO-OOA, too? And what does "almost scavenged" mean?

Response: At background mountain sites with no anthropogenic emissions around, OAs are usually highly oxidized because of aging during atmospheric transport. Further oxidation of these aged OA (O/C > 0.6) in cloud droplets will be dominated by fragmentation reactions instead of functionalization, resulting in mass reductions (Lee et al. 2012). Therefore, in our study, the mass of MO-OOA with a high degree of oxidation (O/C = 1.09) may decrease by cloud processing, while LO-OOA remained in the droplets and further being released after cloud evaporation. Because we cannot give the exact number here, so "almost" was used here.

Considering the referee's comments, the above discussions have been added to the revised manuscript.

Text modification: Lines 355-357: "*Conversely, more oxidized OA being scavenged by clouds may further undergo mass reduction through fragmentation reactions (Lee et al., 2012), which was far less likely to be reintroduced into the atmosphere.*"

References:

Lee, A. K. Y., Hayden, K. L., Herckes, P., Leaitch, W. R., Liggio, J., Macdonald, A. M., and

Abbatt, J. P. D.. Characterization of aerosol and cloud water at a mountain site during WACS 2010: secondary organic aerosol formation through oxidative cloud processing. Atmos. Chem. Phys., 12(15), 7103-16, https://doi.org/10.5194/acp-12-7103-2012, 2012.

RC2-28: L278: Why do you say LO-OOA contains HULIS?

Response: We are sorry for this incorrect hypothesis. HULIS is usually identified as a compound in water-soluble organic matter (WSOM). However, there is no evidence of aqueous phase production of WSOM in this study. The relevant sentences have been deleted in the text.

RC2-29: L290: "high oxidative properties", consider reformulating

Response: Changed it to be "*high oxidation degrees*"

RC2-30: L300: how do you determine the air mass clusters? How cohesive are these clusters? Consider showing all trajectories in a plot per cluster in the SI.

Response: It is really true that these clusters have uncertainties in reflecting the air mass origins during different episodes in the whole sampling period. In our newly updated Fig.4, we have shown all trajectories during different episodes instead of simply showing clusters.

RC2-31: L322: Investigate further (using e.g. trajectory data if accurate enough) to see whether you can confirm that the air mass does not change during P2.

Response: We thank the referee's suggestions. We have further compared the air mass trajectories in six selected episodes, including five high PM_1 episodes and a clean period. Please see our detailed reply in **RC2-20**.

RC2-32: L324: "cloud evaporation released substantial amount of LO-OOA". Try to provide more evidence on this. I am not convinced this is the full story. Why not including ACSM data into this (+ ACSM PMF) and take at least 21.11. (no cloud), 15.–16.11. (no cloud), 13.–14.11 (cloud), 22.–26.11. (no cloud) into the analysis to improve statistics. Do your conclusions on the effects of droplet evaporation and LO-OOA change?

Response: We appreciate the referee's suggestion. Indeed, we performed the ACSM PMF analysis in this study (Fig. RC2-2). However, the results were not satisfactory mainly due to the limitation of the quadruple-ACSM with unit mass resolution and low sensitivity. Considering the low particle concentration at this background site, PMF results of ACSM have large uncertainties. This is also the reason why we conducted additional measurements using HR-ToF-AMS. However, according to our newly updated case studies in the revised manuscript (section 3.2), C1 and EP5 have been proven to represent typical cloudy and cloud evaporative periods, respectively. Therefore, we believe that cloud evaporation indeed released substantial amounts of LO-OOA at this site. Also, we agree that this is a potential limitation of the study. In the future work, the simultaneous measurements of aerosol composition and oxidation state of OA for interstitial aerosol and cloud droplets using a HR-ToF-AMS coupled to a ground-based counterflow virtual impactor (GCVI) inlet system is needed to fully address this question. Thanks again for your question.

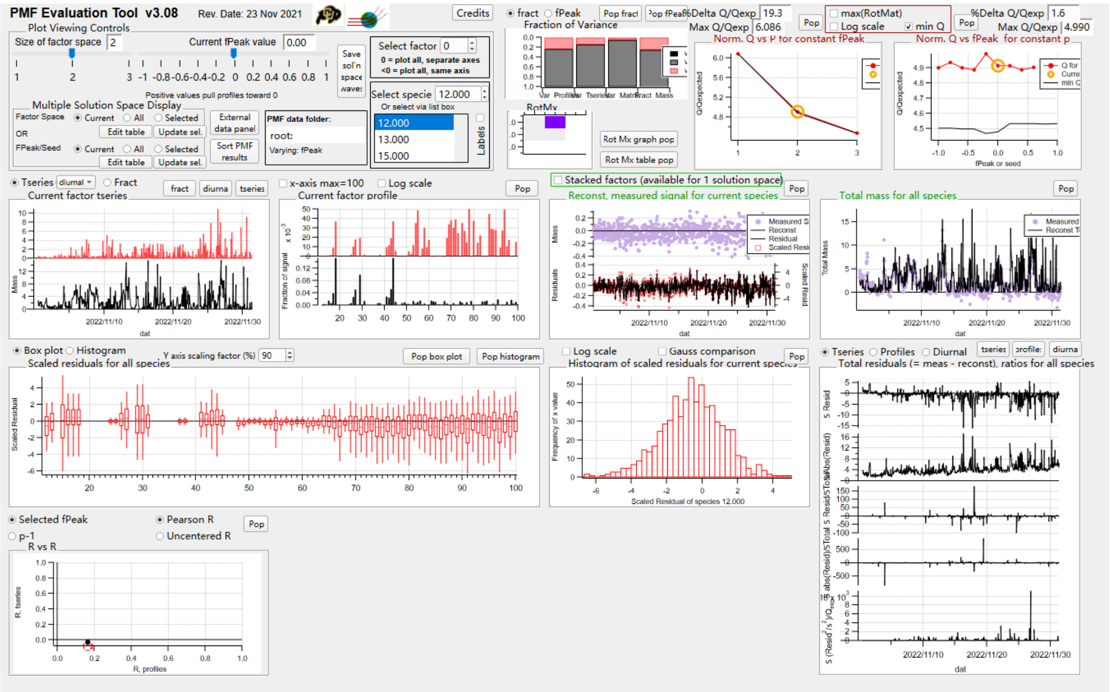

Figure RC2-2. PMF results of Q-ACSM data.

Reference:

Observational evidence of cloud processes contributing to daytime elevated nitrate in an urban atmosphere. Atmospheric Environment. 2018;186:209-15. doi: https://doi.org/10.1016/j.atmosenv.2018.05.040.

RC2-33: Figure 1: was there no BC at all in Mt. Tai?

Response: There was no BC measurement in their study. To avoid misunderstanding, we have added in the caption of Fig. 1.

Text modification: Lines 70-72: “*The mean concentration (in μg m*$^{-3}$*) and chemical composition of submicron aerosols (NR-PM*$_1$*+ 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.”

RC2-34: Figure 4: why does ammonium disappear from the size distribution after 500 nm in panel a? Sulfate also looks odd. How long average is this and how much variability is there within P1? I can see that right before P2 there is a period when AMS was measuring. How does the mass distribution look then?

Response: The abnormal size distributions of ammonium and sulfate may be attributed to the detection limit for pToF analysis. The average time is set as the whole C1 period, and the standard deviation of dM/dlogD$_{va}$ is 0.3–0.7 μg m^{-3}. To address this, we have re-calibrated the pToF data and re-checked the NH_4 signal, and the NH_4 size distribution in Fig. 5 has been revised. The comparison of the size distributions of ammonium and sulfate in C1 and before EP5 are also shown (Fig. RC2-3). Results show that these two periods have similar particle size distributions.

Graph modification:

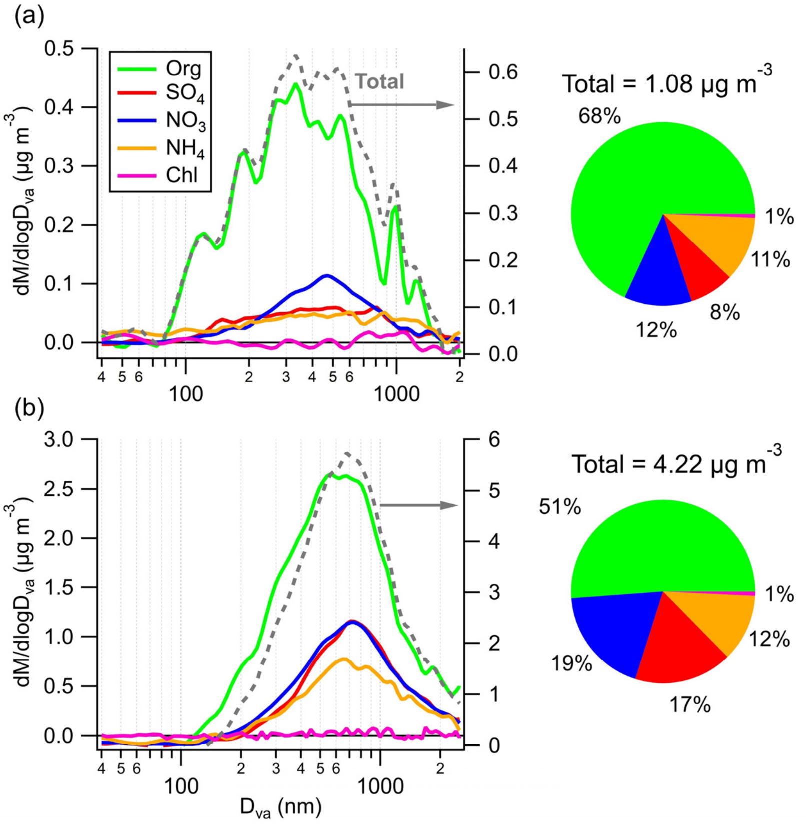

Figure 5. Averaged size distributions and chemical composition of NR-PM$_1$ during (a) C1 and (b) EP5.

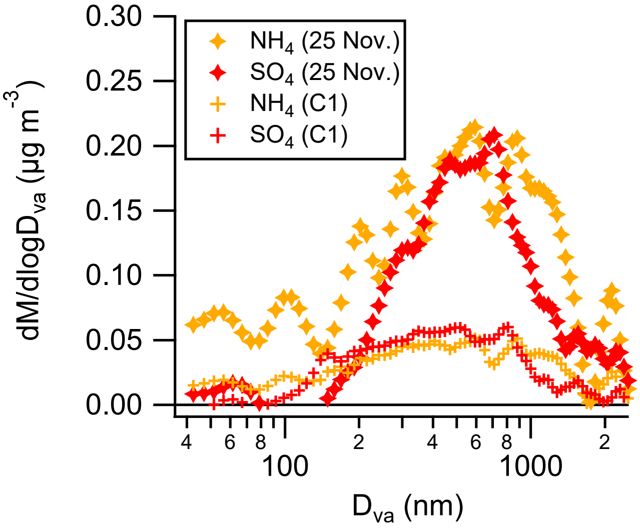

Figure RC2-3. Averaged size distributions of NH4 and SO4 during C1 and a short period before EP5

RC2-35: Figure 5: Could you explain for the bottom right plot what MSA and IN are in the caption? The arrow representation for those is also a bit hard to read. It is also stated in the text that 3.88 is obtained for AN calibration, but referred to as IN in the figure. Does the median for P1 equal the median of P2 in terms of the NOx+ ions?

Response: Thank the referee for pointing out this problem in this Figure. Considering the above comments, we have made the following corrections to this figure:

1. An explanation of MSA and AN in the bottom right plot has been added to the caption.
2. The arrow and the corresponding annotation have been deleted.
3. IN is now corrected to AN.

Graph modification:

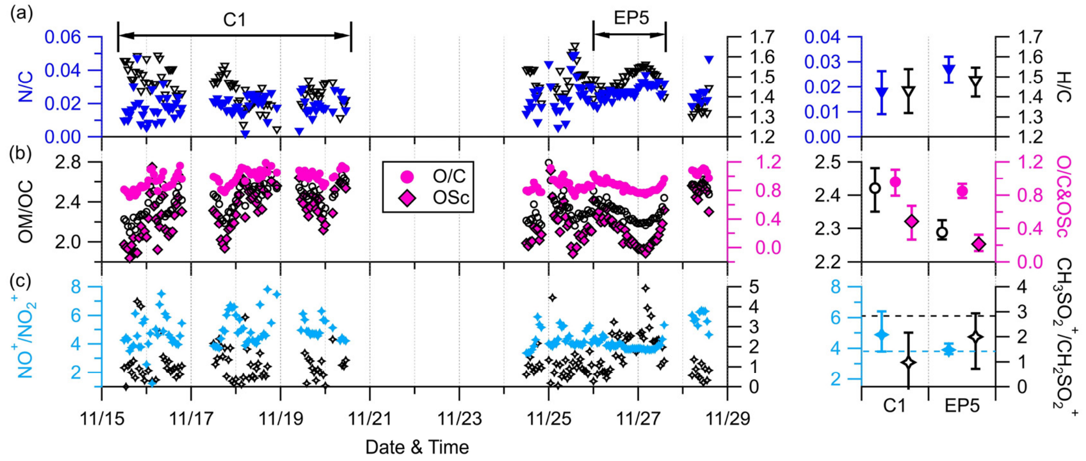

Figure 6. Time series of 1-hour averaged (a) N/C and H/C, (b) OM/OC, OSc, and O/C, and (c)

NO^+/NO_2^+ and $CH_3SO_2^+/CH_2SO_2^+$ 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 µg m^{-3}) are shown. The horizontal black and blue dash line in the bottom right plot represent the $CH_3SO_2^+/CH_2SO_2^+$ value for pure MSA (2.9) and the NO^+/NO_2^+ value for pure AN (3.88), respectively.

RC2-36: Figure 11: Why do you have "Events" here instead of P1 and P2? Event 3 seems to coincide with P2 and suggests an air mass change. I suggest you investigate this further to understand more what is due to the change of air mass and what due to droplet evaporation when you talk about differences in composition between P1 and P2.

Response: We agree that the naming of these periods should be unified. We have moved the back trajectory analysis part to section 3.2 in the revised manuscript, together with comprehensive case studies in six selected episodes. As shown in Fig. 4, although the trajectories during EP5 were shorter than the other period, they all brought relatively high levels of aerosol particles from two polluted regions in the west and southwest (yellow areas in Fig. S5). In addition, the average CO concentration in EP5 (0.28 ppm) was even lower than the clean period (C1, 0.37 ppm), indicating potentially more contribution of pollution transport during C1. These results together suggest negligible influence of air mass change, and droplet evaporation might be the reason. Fig. S5 and detailed discussions can be found in **RC2-20**.

Indeed, our statement about excluding the influence of air mass change on aerosol particles may still have uncertainties more or less. However, this study was not to quantify the contributions of regional transport from different source areas because such analyses need the involvement of modeling work which is beyond our current ability. Besides, we have further strengthened the evidence of cloud evaporation through the analysis of the particle lidar data. Therefore, we think our conclusion about the significant impacts of cloud evaporation on PM_1 at this site is reasonable. Thanks again for your valuable suggestions.

Text modification: Lines 216-226: "*Overall, although these episodes had different mechanisms of* PM_1 *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. S5. 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*

PM_1 concentration stayed at very low levels for the whole period. It is worth noting that PM_1 peaks in EP2−EP5 all occurred along with RH below 100 %. When the RH returned to 100 %, PM_1 concentrations gradually diminished to levels comparable to those observed during the clean period (C1). Furthermore, as shown in Fig. S7, there were notable reductions in the mean and median mass concentrations of all PM_1 species over the entire campaign, ranging between -2.6 %−44.4 % and 2.8 %−50.1 % when RH reached 100 % from conditions of lower RH, 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 PM_1 concentrations."

RC2-37: Figure S.1: How do AMS and ACSM measurements agree for the different chemical species?

Response: We have added a comparison of different aerosol species measured by AMS and ACSM in Fig. S1. Results show that there are generally good correlations between ACSM and AMS (r^2 = 0.35–0.97) except for chloride (r^2 = 0.18). This is likely due to the low ambient chloride masses in this background site, which are close to the ACSM detection limit. Also, the non-ideal vaporization behavior of chloride in Q-ACSM may be another reason (Crenn et al., 2015). Notably, ACSM tends to overestimate the concentrations of PM_1 and chemical species by a factor of 0.12–0.79 (slopes in Fig. S1). Therefore, we re-calibrated the ACSM data by multiplying the factors. After correction, the NR-PM_1 measured by AMS and ACSM correlated well with $PM_{2.5}$ and PM_{10} measured by the particulate monitor (r^2 = 0.53–0.99). In addition, the slope (0.48 and 0.54) was found to be slightly lower than the NR-PM_1/$PM_{2.5}$ ratio (0.6–0.77) derived by the same instruments at an urban site (Li et al., 2023), suggesting that fine aerosol particles existed more in 1–2.5 µm range at this background site because of aging during transport. Overall, these findings affirm the reliability of our data.

Graph modification:

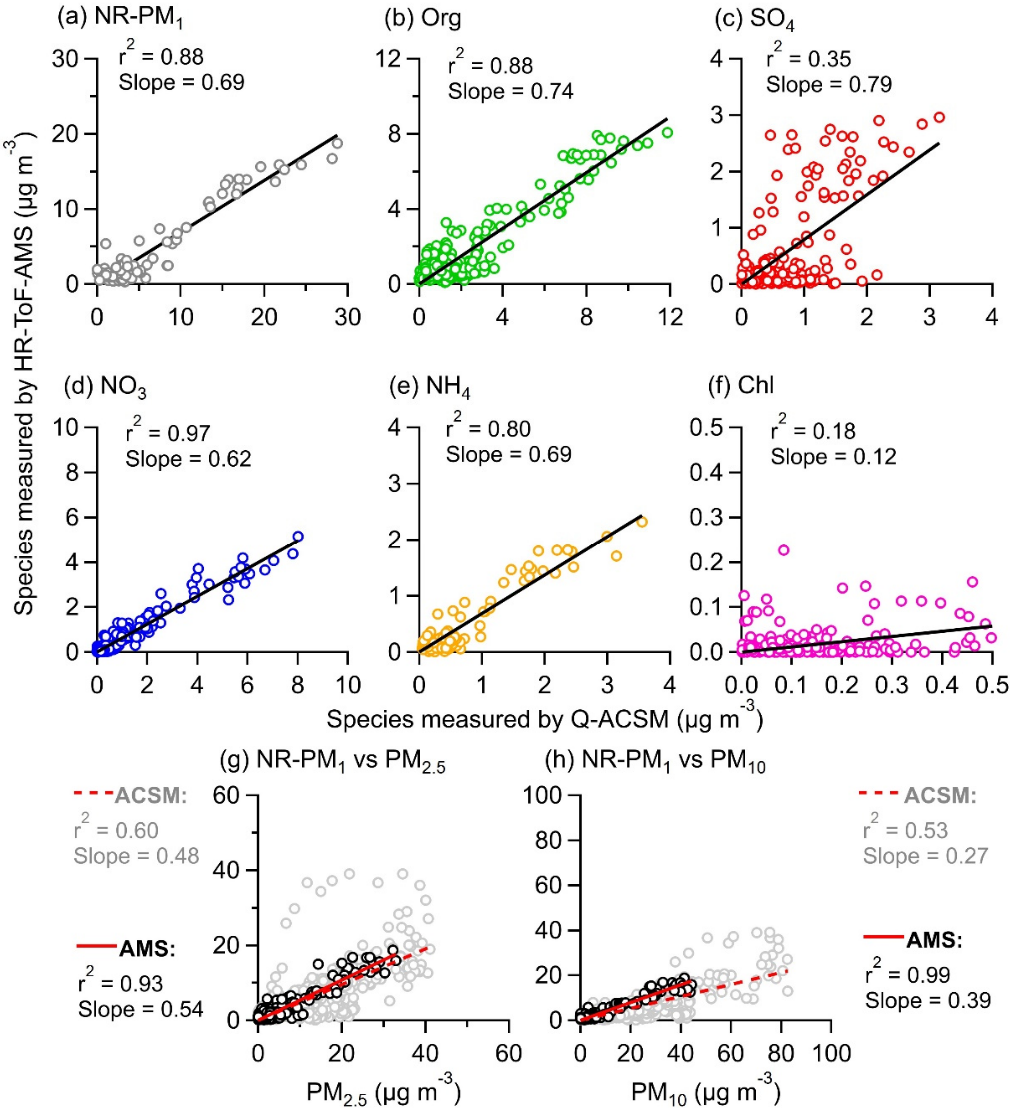

Figure S1. (a-f) Comparison of different NR-PM$_1$ species measured by ACSM and AMS during the AMS sampling period (15–20, 24–28 Nov); (g-h) Scatter plots of mass concentrations of NR-PM$_1$ measured by ACSM (grey) and AMS (black) vs. PM$_{2.5}$ and PM$_{10}$ measured by particulate monitor. The black and red lines are linear-fit lines.

Text modification: Lines 103-108: “*Detailed comparison of the concentrations of NR-PM$_1$ 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. As shown in Figs. S1g and h, after this adjustment, the concentrations of NR-PM$_1$ 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 PM$_{10}$ ((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.*"

References:

Crenn V, Sciare J, Croteau PL, Verlhac S, Fröhlich R, Belis CA, et al. ACTRIS ACSM intercomparison – Part 1: Reproducibility of concentration and fragment results from 13 individual Quadrupole Aerosol Chemical Speciation Monitors (Q-ACSM) and consistency with co-located instruments. Atmos Meas Tech. 2015;8(12):5063-87. doi: https://doi.org/10.5194/amt-8-5063-2015.

Li Z, Xu W, Zhou W, Lei L, Sun J, You B, et al. Insights into the compositional differences of PM1 and PM2.5 from aerosol mass spectrometer measurements in Beijing, China. Atmospheric Environment. 2023;301:119709. doi: https://doi.org/10.1016/j.atmosenv.2023.119709.

RC2-38: Figure S.3: are these all from the same exact measurement period? How come the diel cycle of PM2.5 is so much less in terms of mass than PM1? Almost as if the nitrate episode is not measured by the PM2.5 instrument?

Response: These diurnal variations are all derived from the same sampling period (1st 30th Nov). The reason for the higher PM_1 mass diel cycle than $PM_{2.5}$ is the overestimation of NR-PM_1 by ACSM (see **RC2-37**). After correction, the revised diel cycle of mean PM_1 mass is reasonably lower than $PM_{2.5}$ (Fig. S4). Furthermore, the diel cycle of median PM_1 mass, which mitigates the influence of the nitrate episode, remains lower than $PM_{2.5}$.

Graph modification:

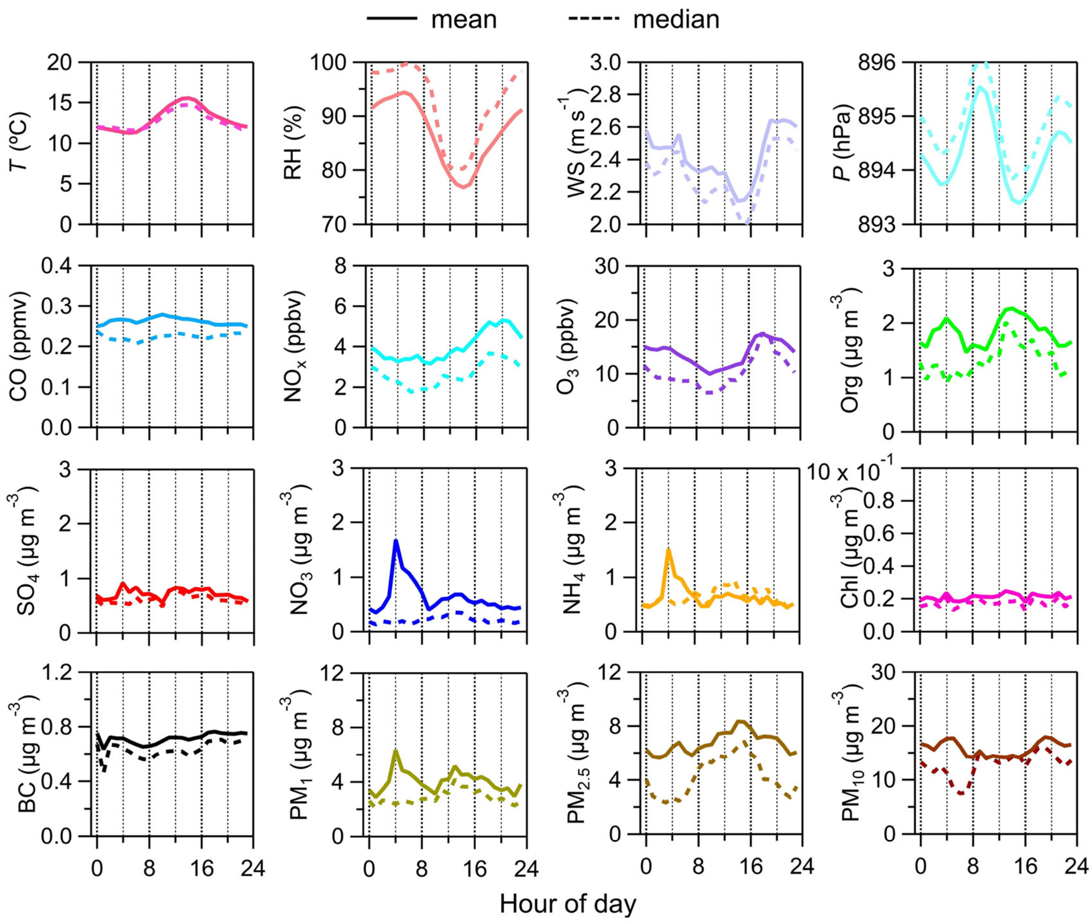

Figure S4. Diurnal variations of PM$_1$ species (BC + NR-PM$_1$ measured by ACSM), air pollutants, and meteorological parameters during the entire campaign (1–30 Nov).

RC2-39: Figure S.5 easier to read if both y-axes had the same scale

Response: We thank the referee's suggestion. Given that over half of the sampling period (55%) was under an RH of 100%, we have updated this figure to compare the scenarios of RH≠100% and RH=100%, which may further reflect the influence of the cloud process on aerosol particles.

Graph modification:

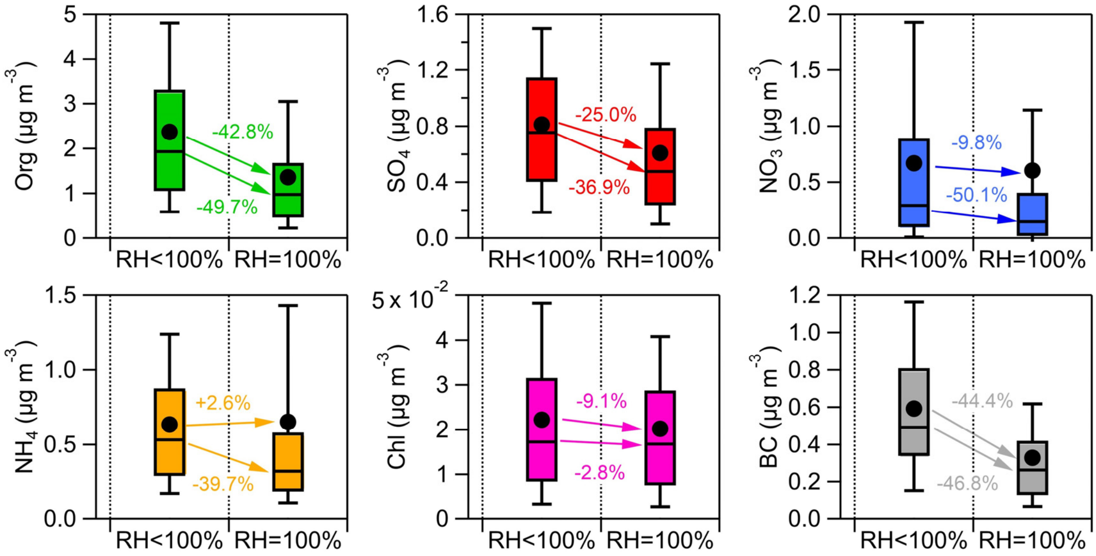

Figure S7. Comparison of mean and median mass concentrations of PM_1 species under conditions of RH below and at 100 % over the entire campaign.

Response to Referee#3

General comments:

RC3: Zhang et al. present a very interesting dataset of 1-month long ACSM measurements at a background mountain side in (south-)east China. Additionally, they include high resolution AMS measurements for a few days during that period. In their interpretation, they suggest a strong influence of cloud processing on the amount and composition of the aerosol at the site.

From the evidence they provide in the manuscript, I have to strongly disagree with their main conclusion that "aerosol composition, size distributions, and oxidation states are significantly influenced by aerosol-cloud interactions." (Line 328f). In my opinion, the authors did not sufficiently account for changes in airmass origin before looking at the effects of cloud/fog on aerosol composition. This is a systematic shortcoming in their analysis which needs to be rectified. I do think that this dataset can provide interesting insights into the aerosol properties over a mountain site in China and might even contain evidence of cloud processing/aerosol cloud interactions (ACI). But at the current state, the arguments are not convincing. To reveal such insight, the authors have to conduct some extensive reanalysis and reinterpretation of their data (I list specific points they should touch in the comments below). This means a significant rewrite of the manuscript and then another round of reviewing to evaluate the new data interpretation.

Response: We gratefully appreciate the referee for careful reading and valuable comments that helped improve the manuscript significantly. We agree with the referee that our manuscript needs improvement in our analysis of aerosol-cloud interactions. Following the reviewer's suggestions, we have revised the manuscript as much as we can. Below are the key revisions in our updated manuscript:

1. To better utilize the ACSM data, analyses of six selected periods, including five PM episodes (EP1−EP5) and one clean period (C1), were added to the revised manuscript. **Note that the previous P1 and P2 are now referred to as C1 and EP5, respectively.** The relationship between the PM_1 species concentration, meteorology, and air mass transport during these periods has been clearly evaluated.
2. Prior to discussing the impacts of the cloud process on PM_1, the air mass back trajectories during the selected periods have been further investigated to exclude the potential influence of air mass change.
3. To refine our analysis of cloud processes, we have analyzed the vertical distributions of particle depolarization ratios (PDR) measured by a particle lidar,

further confirming the occurrence of cloud evaporation during the sampling period. These discussions have been integrated into a new section (section 3.2) in the revised manuscript. Please see our detailed point-to-point responses below.

Major comments:

RC3-1: The authors must first identify the effect of airmass origin before interpreting their data with respect to ACI etc. I.e., they need to utilize the back trajectory data (and meteorological information) to identify comparable time periods in their data set. Then they can select cloud/fog formation cases and investigate the composition and mass changes. Note that for this analysis it is not sufficient to use the 4 back trajectory clusters they show currently in section 3.5. Examples:

a. The authors already identified "Event 3" (=P2 period) in Fig 11b as a plume from somewhere with very different starting composition than the P1 period that is representative for the typical background level. But they claim that the key difference between P1 and P2 (=Event 3) is the cloudiness. The total NR-PM1 concentration increases from ~1 ug/m^3 to over 25 ug/m^3 during the peak of Event 3/P2.

b. The 6 h spike in NO3 concentration on Nov 4th completely skews the NO3 averages and the relation of NO3/NR-PM1 (Fig 3) as well as the diurnal trends (Fig S3). See also specific comment #19.

Response: We appreciate the referee for the above suggestion. We agree that both airmass transport and cloud/fog can lead to aerosol composition and mass change, and the airmass origin needs to be discussed first. However, this study was not to quantify the contributions of regional transport from different source areas because such analyses need the involvement of modeling work which is beyond the scope of this study. The best we can do is to compare the back trajectories between different periods. Considering the referee's suggestion, we have deleted the previous section of the back trajectory analysis. Instead, a detailed investigation of the relationship between the PM_1 species concentration, meteorology, and air mass transport during six selected periods has been provided in section 3.2 in the revised manuscript. In addition, the back trajectories were separately plotted for each episode for comparison, and the clusters were no longer used. Please see the **text modifications** for detailed revision.

Below are detailed explanations for the examples given by the referee:

a. After careful re-analysis, we still found that the key difference between C1 and EP5 is the cloudiness. There is a series of evidence supporting our hypothesis:

First, as shown in Fig. S5, except for the nitrate episode (EP1), air masses in EP2−EP5 and C1 were similar and mainly originated from the west and southwest. Although the back trajectories in EP5 were shorter than C1, they can both bring relatively large amounts of anthropogenic-emitted pollutants from big cities like Nanchang and Fuzhou. Therefore, the potential impacts of air mass change were likely to be negligible.

Second, high levels of CO were observed during C1 (0.37 ppm), which were even higher than EP5 (0.28 ppm), suggesting severe influences of polluted air masses in C1. However, the aerosol concentrations did not increase correspondingly. Thus, the aerosol might be removed by some factors in C1.

Third, as shown in Fig. 4, a similar wind direction variation pattern was identified in EP4, EP5, and C1, which varied from northwest wind to southerly wind. Hence, the effects of wind direction can also be excluded.

Fourth, we found that the increasing PM_1 in EP2−EP5 was all associated with the decrease of RH. In contrast, RH was stable at 100 % during C1. Figure S7 also showed that the median mass of all PM_1 species decreased by 2.8 %−50.1% when RH reached 100% from lower RH conditions., suggesting the critical role of RH in the variability of PM_1 concentrations. Considering the occurrence of frequent cloud events during the sampling period, RH may be associated with cloud processes.

Taken together, the most likely reason for the different PM_1 concentrations in EP5 and C1 was cloudiness.

b. Indeed, the nitrate-dominant feature and distinct air mass back trajectories in EP1 may suggest a different mechanism of PM_1 elevation from other periods. 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. In addition, RH was stable at 100% during this period, which can favor the heterogeneous uptake of N_2O_5 and subsequent nitrate formation in the aqueous phase at night. Therefore, the spike in nitrate concentration was likely attributed to the rapid formation of nitrate at night under high RH. Also, to address the skew in Fig. 3 caused by this nitrate spike, we have re-analyzed the data after removing data during EP1 (Please see the revised Fig. 3 in **RC3-2**).

Thanks again for the referee's kind help, and the revised text is as follows:

Text modification:

Lines 195-226: "*The entire study period was characterized by five PM_1 episodes (EP1–EP5), as marked in Fig. 2. Meanwhile, a clean period (C1) with low PM_1 levels* *was also selected for comparison. The relationship between the* PM_1 *species concentration, meteorology, and air mass transport during these six periods is shown in Fig. 4 and Fig. S5. EP1 exhibited a nitrate-dominant* PM_1 *peak, with nitrate concentration rapidly increased by ~18* $\mu g\ m^{-3}$ *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. S6) showing high levels of nitrate mainly associated with northeastern wind directions. This was further supported by the back trajectory analysis, in which air masses on 4 Nov were also from the northeast. Moreover, the RH was stable at 100% during EP1, which can favor the heterogenous uptake of* N_2O_5 *and subsequent nitrate formation in the aqueous phase at night (Brown et al., 2006; Li et al., 2020).*

In contrast to EP1, PM_1 *in other episodes (EP2–5) were all dominated by organics. EP2 was associated with low 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* PM_1 *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 other* PM_1 *species and CO, were further elevated after airmass sources shifted from southwest to northwest on 13 Nov, which may bring a large amount of aerosols from megacities such as Zhengzhou and Hangzhou to the SH site (Fig. S5c). EP4 and EP5 were two similar episodes, where* PM_1 *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.*

Overall, although these episodes had different mechanisms of PM_1 *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. S5. 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 PM_1 concentration stayed at very low levels for the whole period. It is worth noting that PM_1 peaks in EP2−EP5 all occurred along with RH below 100 %. When the RH returned to 100 %, PM_1 concentrations gradually diminished to levels comparable to those observed during the clean period (C1). Furthermore, as shown in Fig. S7, there were notable reductions in the mean and median mass concentrations of all PM_1 species over the entire campaign, ranging between -2.6 %−44.4 % and 2.8 %−50.1 % when RH reached 100 % from conditions of lower RH, respectively. Considering the frequently occurring cloud events at this site, it is most likely that these variations of RH were affected by the cloud process, which may further play an important role in PM_1 concentrations.*"

Graph modification:

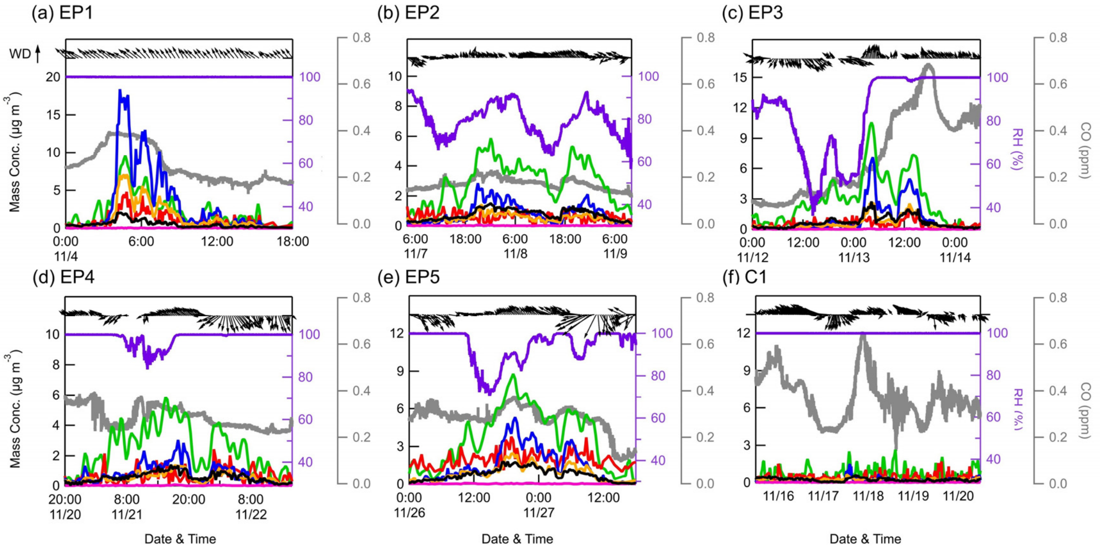

Figure 4. Temperial variations of PM_1 species measured by ACSM (organics, sulfate, nitrate, ammonium, and chloride) and AE-33 (BC), as well as CO, RH, WD, and WS during six selected periods.

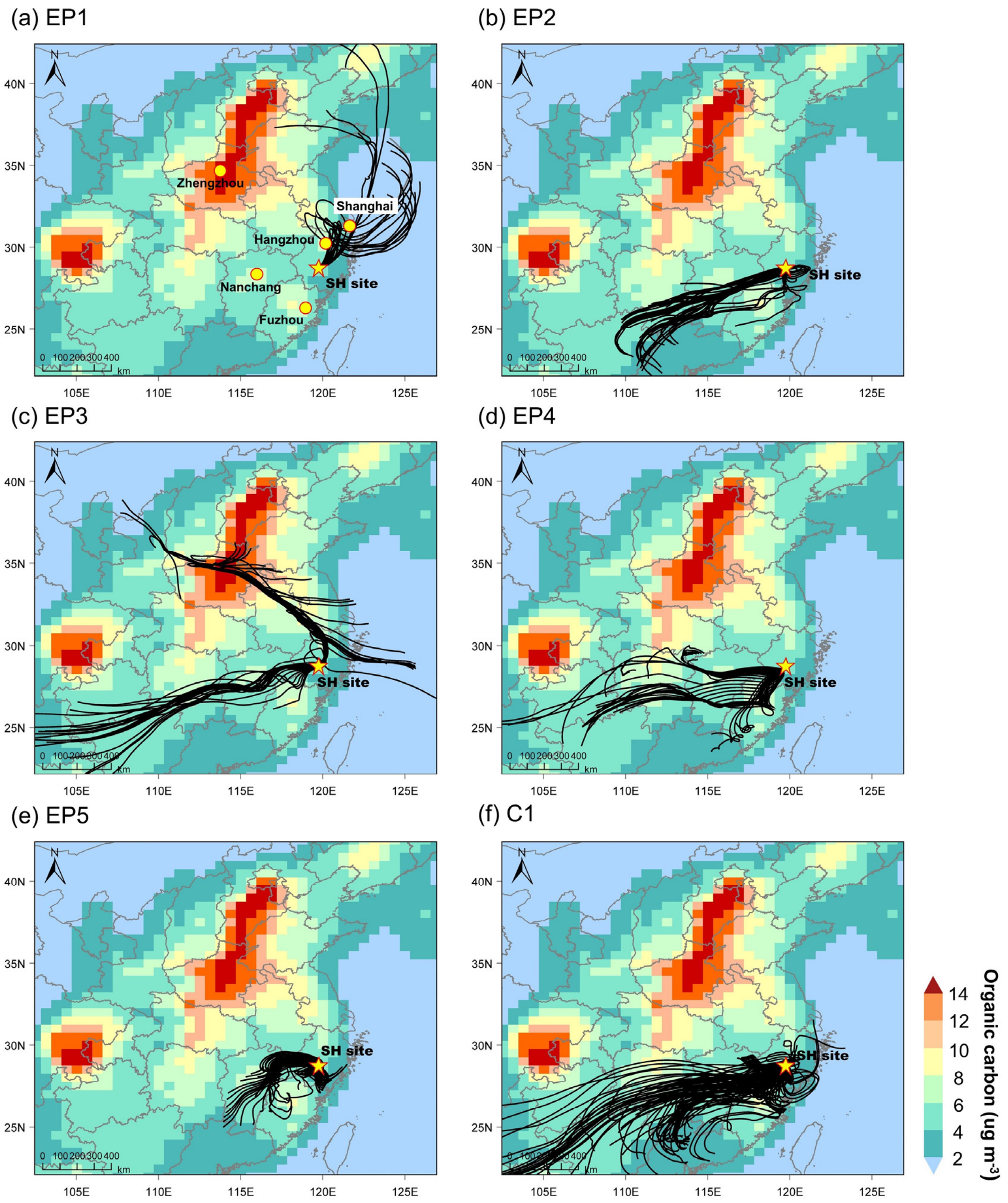

Figure S5. 72 h air mass back trajectories calculated at 1 h intervals at the SH site during six selected periods.

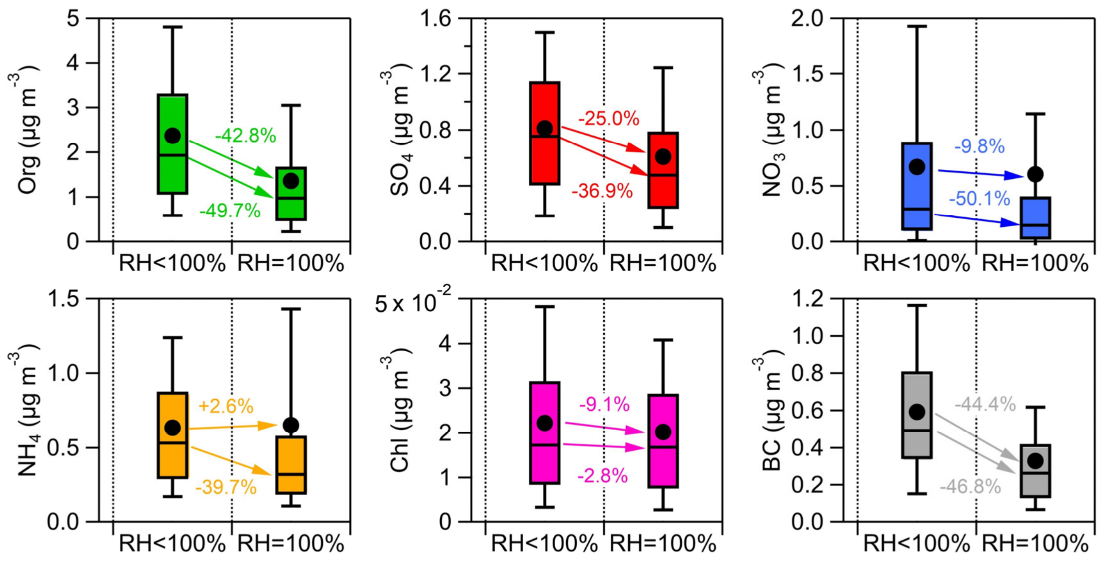

Figure S7. Comparison of mean and median mass concentrations of PM$_1$ species under conditions of RH below and at 100 % over the entire campaign.

RC3-2: Even after excluding the airmass origin as a driver for the observed differences, the authors need to look into other options than just cloud scavenging/evaporation. As an example, the station is at a mountain site at 1100 a.s.l. in a region with apparently complex topography (estimated from the photo in Fig 1). What are typical boundary layer heights at this location? Is the station high enough to always be in the free troposphere? Or are there effects of changes in boundary layer height? E.g. some day/night effects or something related to temperature trends?

Response: Thank the referee's comments. This site is located in Jinhua, Zhejiang Province, where the mean atmospheric boundary layer height (BLH) is ~1.3 ± 0.4 km, as reported by Zhang et al. (2016). Therefore, this station is not high enough to always be in the free troposphere. However, we did not measure the real-time BLH at this site. Thus, we cannot quantify the accurate effects of BLH changes on PM$_1$. Among the selected episodes, we only found that EP2 may be affected by the variation of BLH, as evidenced by the clear and similar diurnal variation patterns of PM$_1$ species, RH, and temperature. The PM$_1$ peaks in EP3, EP4, and EP5 all lasted for at least 1.5 days, which was unlikely related to BLH and temperature.

References:

Zhang, W., Guo, J., Miao, Y., Liu, H., Zhang, Y., Li, Z., and Zhai, P.: Planetary boundary layer height from CALIOP compared to radiosonde over China. Atmos. Chem. Phys., 16(15), 9951-63, https://doi.org/10.5194/acp-16-9951-2016, 2016.

RC3-3: The authors must provide a clear definition of what is considered a cloud/fog case and give more information about the type of cloud/fog.

a. It seems that only RH=100% was used as a criterion for cloud/fog conditions. This may be problematic. Many RH sensor do not like to be in condensing conditions (RH>=100%). Depending on the RH sensor type, there can be a significant delay before trustworthy values are reported again after condensing conditions. Is this relevant for the definition of "evaporation" of cloud/fog at this site?

Since the full description of the station instrumentation is missing, I can only speculate if additional instrumentation was present that might be helpful for definition of in cloud/fog conditions (e.g., liquid water path measurements, cloud droplet number measurements, visibility sensors). If any of such measurements are available, they should be included in the definition of in cloud/fog cases and especially the "evaporation" cases.

b. They do not specify if, e.g., a cloud was formed at the mountain top or fog was present in both valley and elevated area. It would be beneficial to learn more about the general weather situation at this location. E.g., how often is the station in cloud/fog. Is this a seasonal thing? Also, what causes the "evaporation" of the cloud/fog at the site?

Response: Thank the referee for the above suggestions. We indeed defined the cloud events mainly based on RH. We agree with the referee that RH sensor might have a delay in reporting RH value of 100%. However, our sampling site is a new site that was just built in half year, and the instruments, particularly for cloud measurements, e.g., fog monitor, GCVI, particulate volume monitor, and etc. are very limited. In fact, this is the first field campaign at this site. Therefore, the general weather situation at this location including the frequency and seasonality of cloud/fog, and the causes of evaporation is not known yet. It certainly shall be characterized in future studies.

To address the referee's comments, we added more analysis of the available measurements during this study.

a. There are a few days of particle depolarization ratio (PDR) data measured by a particle lidar at this site, which can provide some evidence of cloud evaporation (Fig. S8). In EP5 (evaporation case), clouds were identified at ~1km above the sampling site prior to the PM_1 peak. These clouds subsequently disappeared at noon and released a large amount of aerosol particles (PDR of ~0.15), coinciding with the PM_1 peak. Conversely, high PDRs were found around 0 km in C1 (cloud case), which means that clouds constantly existed near the sampling area during this period. Therefore, we defined EP5 and C1 as typical cloudy and cloud evaporative periods.

b. The clouds generally exist from near the sampling site (around the mountaintop) to ~1km above it. Photos taken on cloudy and sunny days have been also added to supplementary (Fig. S9), which can help readers know the weather situation at this site. Cloudiness is not a seasonal thing at this station, instead, it is a common occurrence here (about 20 days in a month), especially in June during the plum rain season. Given the high altitude (1128 m a.s.l.), this mountain site can receive strong solar radiation at noon, which may cause cloud evaporation. These discussions have been added to the revised manuscript:

Text modification: Lines 227-239: "*Figure S8 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 (Fig. S8a). 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* PM_1 *peak. These particles were then scavenged by the increased RH 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. S8b). Instead, high PDR values were found generally at around 0 km, which means that clouds constantly existed near the sampling area during this period, coinciding with the constant 100% RH. Therefore,* PM_1 *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. S9a), yet the weather was generally sunny and cloudless in EP5 (Fig. S9b). As a result, besides regional transport, cloud processes can also have significant impacts on aerosol particles at this site.*"

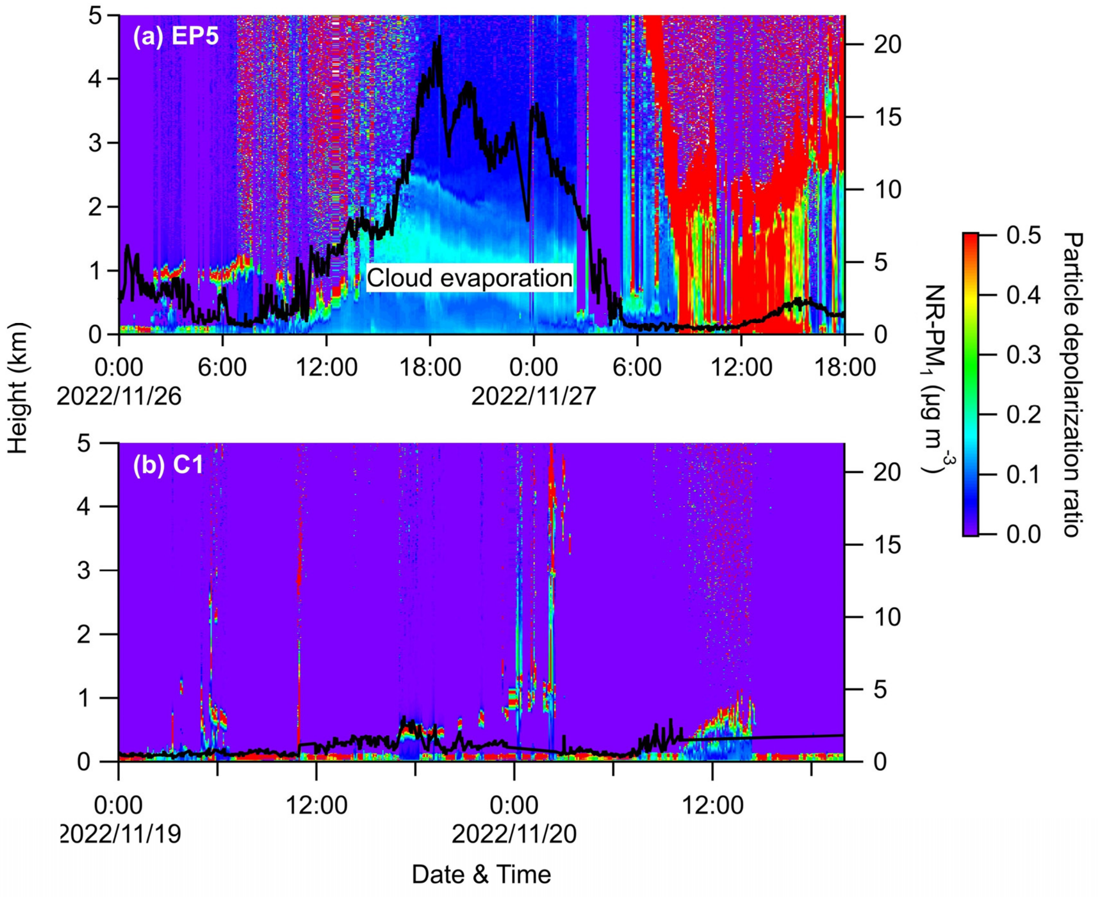

Figure S8. Vertical distribution of particle depolarization ratios in (a) EP5 and (b) C1.

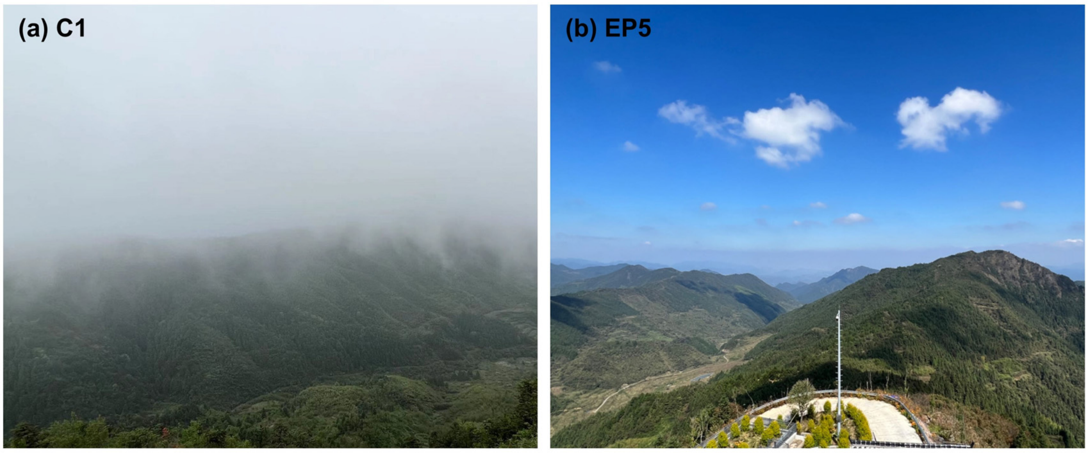

Figure S9. The weather condition in (a) C1 and (b) EP5 (photos were taken on the roof of the sampling site at local time ~15:00 on 19 Nov and 26 Nov).

RC3-4: It is not clear what fraction of the total aerosol (interstitial & cloud droplets) was measured during cloudy conditions. The crucial information about the inlet line is missing. The authors state that the sample line was ¼ inch OD and the sample was dried prior to entering the ACSM and AMS which have PM$_1$ inlets leading to the detection of dried PM_1 aerosol. But they do not report the cutoff size of their inlets at ambient conditions. This fact is of utmost importance for cloudy/foggy conditions: If there is a PM1 cutoff (or PM2.5 or whatever) at ambient conditions (e.g., a impactor at the tip of the sampling line outside of the building), it means that at cloudy conditions, none of the cloud droplets are sampled, only the interstitial aerosol (i.e. the particles that did not activate to cloud droplets). On the other hand, if there is no such cutoff at ambient conditions, cloud droplets will enter the sample line, get dried, and their residuals are measured together with the interstitial particles.

The authors must clarify this because, the way the authors use the scavenging and droplet evaporation as an explanation for measured composition differences does only hold true if AMS and ACSM are only seeing interstitial aerosol particles. Then the ACSM and AMS data will indeed not detect the material of the particles that activated to cloud droplets and compounds scavenged into cloud droplets. But if both interstitial and dried cloud droplets are entering the instruments, the composition measurement will detect the material scavenged by cloud droplets together with the interstitial aerosol. I.e., the instruments see the same "dry" aerosol distribution as if the cloud evaporated outside before the sample was taking. This would contradict the explanation of how scavenging and release upon (outside) cloud droplet evaporation impacts the measured composition-and thus the effect of ACI.

Response: Thank the referee for pointing this out. There was no size cutoff at the inlet of AMS and ACSM. We agree with the reviewer that the AMS/ACSM might sample some cloud droplets that were dried to small size less than 1 μm. However, we estimate that such influences might not be important. The reason is: 1) the sampling line was relatively long (~4 m), and cloud droplets, particularly large droplets (e.g., > 7 μm) entered the sampling line with high moisture may condense on the inner wall of the sampling line. The dryer was placed near the inlet of AMS and ACSM, and meanwhile, the sampling flow was relatively low (~6 L m^{-3}), which may not be enough for the condensed droplets to reach the dryer to get dried. This is further supported by the good correlations of PM_1 with $PM_{2.5}$ and PM_{10} measured by the continuous particle monitors with $PM_{2.5}$ and PM_{10} cutoffs (Fig. S1 in **RC3-19**); 2) One of previous studies at a rural site (Gucheng, Hebei province) showed that the supermicron aerosol particles ($PM_{2.5}$–TSP, both interstitial and fog droplets) accounted for ~10 – 20% of total particle mass during three fog episodes (Kuang et al., GRL, 2024). Considering this study dried aerosol particles and fog droplets immediately after entering the sampling line, we expect that it sampled more fog droplets than our study. Therefore, we conclude that the influence of the absence of size cutoff on our PM_1 measurements would not be significant. Considering the referee's comment, we have clarified this uncertainty in the revised manuscript. As it should be, we will equip such cutoffs at the tip of the sampling line in future measurements to avoid this uncertainty.

Text modification: "*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)*"

Reference:

Kuang, Y., Xu, W., Tao, J., Luo, B., Liu, L., Xu, H., Xu, W., Xue, B., Zhai, M., Liu, P., and Sun, Y.: Divergent Impacts of Biomass Burning and Fossil Fuel Combustion Aerosols on Fog-Cloud Microphysics and Chemistry: Novel Insights From Advanced Aerosol-Fog Sampling, Geophys. Res. Lett., 51, e2023GL107147, https://doi.org/10.1029/2023GL107147, 2024.

RC3-5: PMF analysis is only done with the few days of AMS. Yes, HR AMS data contains much more details for the organics and is more reliable in separating NO3 and SO4 from overlapping organic fragments. But since only 2 OA factors are identified anyway, why not run a PMF analysis with the whole month of ACSM data. This will include many more "interesting" periods providing a better chance to actually find examples of ACI or interesting airmass changes.

Response: We appreciate the referee's suggestion. Indeed, performed PMF analysis with ACSM OA mass spectra. Unfortunately, the ACSM in this study is a quadruple-ACSM with low mass resolution and relatively high detection limits. Considering the low particle concentration at this background site, large uncertainties may exist in PMF results. The results of ACSM PMF analysis are shown in Fig. RC3-1. It can be seen that the PMF results were indeed not satisfying. This is also the reason why we conducted additional measurements using HR-ToF-AMS. Therefore, we seek the editor and referee's tolerance and understanding. Also, we agree that this is a potential limitation of the study. Long-term AMS measurements are needed in the future to better characterize SOA at this site.

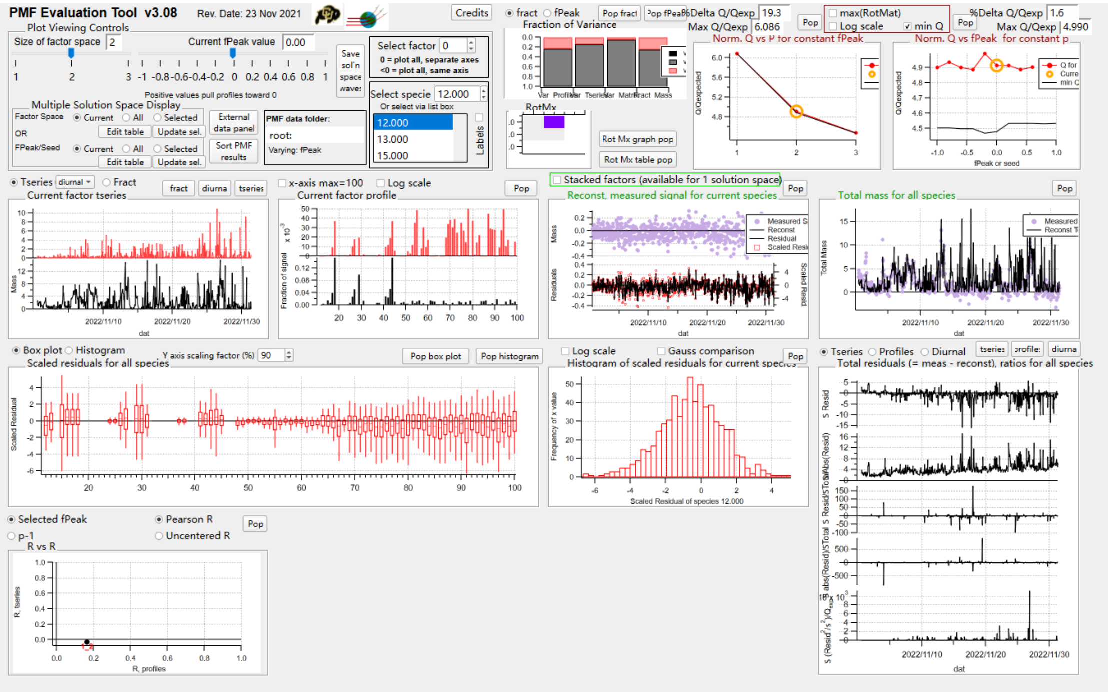

Figure RC3-1. PMF results of Q-ACSM data.

RC3-6: The role of clouds for the contribution of organonitrates (ON) needs to be re-examined. The abstract suggests that ON is actively formed/enhanced by the presence of clouds and that increases the contribution to the total nitrate significantly. But the data in Fig 8 shows a different picture with a massive increase of inorganic NO_3 during P2 automatically leading to the decreased contribution of ON. Note that the ON amount also increase from the background level more during P2 than duringP1. Hence, it is not an enhancement of ON during P1 potentially due to the presence of clouds, but rather a change in the strength of the inorganic NO3 source causing the different ON contribution values.

If the airmasses during P1 were coming from the same region as during P2 (i.e., there should have been similar inorganic NO_3 source), one could start to interpret the reduced inorganic NO_3 in the context of the clouds. One possible interpretation could be that the very water soluble HNO_3 or ammonium nitrate (AN) are indeed scavenged by water droplets and removed by precipitation before reaching the site. (Note that for this interpretation it is paramount to know if the sampling line had a PM_1 cutoff at ambient conditions, i.e., if only interstitial particles are sampled, see comment #4)

Response: Thank the referee's comments. Indeed, the contributions of ON large absolute mass changes of AN instead of ON itself, and both the relative and absolute changes should be considered. We have thus revised the relevant sentences in the abstract and section 3.4 in the updated manuscript. In addition, the discussion about

NO_3 scavenging by clouds in the referee's comments has also been added to the manuscript. Please see the detailed revisions below.

Text modification: Lines 328-338: "*The average mass concentration of ONs in C1 was 30 ± 22 ng* m^{-3}*, which was lower than that in EP5 (40 ± 23 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 μg* m^{-3} *vs.1.47 μg* 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 %).*"

RC3-7: Overall, the description of the station instrumentation is insufficient. Beside obvious mistakes (PM measurements with gas analyzers?), vital information is missing and no reference to a previous description is provided.

Response: We thank the referee for pointing this out. We have added a table describing the instruments in the Supplementary.

Table S1. Summary of the detailed descriptions for the instruments used during the sampling period, including the measured parameters, model, temporal resolution, and sampling time.

Instrument	Species	Model/Company	Time resolution	Sampling time
Q-ACSM	NR-PM_1 (Org, SO_4, NO_3, NH_4, Chl)	Aerodyne Research Inc.	20 min	11.1−30
HR-ToF-AMS	NR-PM_1 (Org, SO_4, NO_3, NH_4, Chl)	Aerodyne Research Inc.	1 min	11.15–20; 11.24–28
AE33	BC	Magee Scientific Corp.	1 min	11.1−30
CO analyzer	CO	Picarro G2401	1 min	11.1−30
Gas analyzers	O_3, NO_x	Thermo Scientific	1 min	11.1−30
PM analyzer	$PM_{2.5}$, PM_{10}	Thermo Scientific	1 min	11.1−30

Data logger	RH, T, WS, WD, P	CR1000, Campbell Scientific Inc.	1 min	11.1−30
Four-cup anemometers	WS, WD	Model O1OC and O2OC, Met One Instruments Inc.	1 min	11.1−30
P Sensor	P	CS106, Vaisala Ltd.	1 min	11.1−30
T/RH probe	T, RH	HC2S3, Campbell Scientific Inc.	1 min	11.1−30
Particle lidar	Depolarization ratio	Science of Light Technologies Co., Ltd	5 min	11.19–20; 11.25–27

RC3-8: I did not note a discussion of detection limits for the ACSM (and AMS) for the reported species nor do I see any measurement errors. Since P1 seems to be an extremely low concentration period such limits become relevant for things like PMF, ON estimation and elemental ratios. E.g., Line 256 states 0.08ug/m^3 for inorganic NO_3 which seems very close to the detection limit for an AMS running with 6 min time resolution switching v mode and pTof. The authors need to carefully check the low concentration times and exclude signals which are too low to be interpreted or used to derive parameters like ON or O/C ratios.

Response: We totally understand the referee's concern. The AMS detection limits (DLs, 1 min average) are 22, 5.2, 2.9, 38, and 12 ng m^{-3} for organics, sulfate, nitrate, ammonium, and chloride, respectively (DeCarlo et al., 2006). Since AMS was operated with 1 min time resolution in this study, the inorganic NO_3 with 0.08 µg m^{-3} can be quantified accurately.

The ACSM DLs (30 min average) are 540, 60, 70, 250, and 30 ng m^{-3} for organics, sulfate, nitrate, ammonium, and chloride, respectively (Sun et al., 2012). As shown in Fig. S1 (can be found in **RC3-19**), the organics, nitrate, and ammonium measured by ACSM correlated well with those measured by AMS (r^2 = 0.62−0.74), while the sulfate and chloride showed relatively poor correlation (r^2 = 0.35 and 0.18, respectively). This result suggests that the concentrations of sulfate and chloride may be close to the DLs of ACSM. However, after we corrected the ACSM data by using the regression coefficients between ACSM and AMS, the concentrations of NR-PM_1 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 PM_{10} ((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.

We have added these DLs as a table to the revised supplement (Table S2) and a discussion is now given in the revised text.

For AMS PMF, as shown in Fig. S2c and d, the reconstructed mass tracked well with the variations of measured mass, and the scaled residuals of all ion fragments were distributed between -4–4, suggesting that the PMF result was reasonable and with small errors.

For ON estimation, the ON concentrations at this site (30–40 ng m^{-3}) were close to the value (40 ng m^{-3}) obtained at a forest-urban mixed site in Finland using AMS (Hao et al., 2014), which could prove that AMS has the ability to quantify ON at such low levels. Considering the referee's comment, we have included the standard deviations of ON mass concentrations.

For elemental ratio analysis, to minimize noises, we have already excluded data points with OA concentrations lower than 0.7 μg m^{-3} when analyzing the elemental ratios, following the method used by Sun et al. (2009).

Table S2. The reference detection limits (in ng m^{-3}) of AMS (1 min average time) and ACSM (30 min average time) for different NR-PM_1 chemical species.

Species	**AMS (DeCarlo et al., 2006)**	**ACSM (Sun et al., 2012)**
Org	22	540
SO_4	5.2	60
NO_3	2.9	70
NH_4	38	250
Chl	12	30

References:

DeCarlo, P. F, Kimmel, J. R., Trimborn, A., Northway, M. J., Jayne, J. T., Aiken, A. C., Gonin, M., Fuhrer, K., Horvath, T., Docherty, K. S., Worsnop, D. R., and Jimenez, J. L.: Field-Deployable, High-Resolution, Time-of-Flight Aerosol Mass Spectrometer. Anal. Chem., 78(24), 8281-9, https://doi.org/10.1021/ac061249n, 2006.

Sun, Y., Zhang, Q., Macdonald, A. M., Hayden, K., Li, S. M., Liggio, J., Liu, P. S. K., Anlauf, K. G., Leaitch, W. R., Steffen, A., Cubison, M., Worsnop, D. R., van Donkelaar, A., and Martin, R. V.: Size-resolved aerosol chemistry on Whistler Mountain, Canada with a high-resolution aerosol mass spectrometer during INTEX-B. Atmos. Chem. Phys., 9(9), 3095-111, https://doi.org/10.5194/acp-9-3095-2009, 2009.

Sun. Y. L, Wang. Z. F, Dong H, Yang T, Li J, Pan X, et al. Characterization of summer organic and inorganic aerosols in Beijing, China with an Aerosol Chemical Speciation Monitor. Atmos.

Environ., 51, 250-9, https://doi.org/10.1016/j.atmosenv.2012.01.013, 2012.

RC3-9: The authors should reevaluate the title of their manuscript. The title is very generic and currently does not fully reflect the content of the paper. If after the necessary reanalysis ACI are indeed found to be relevant in this location, it is worth including that in the title.

Response: Thanks for the referee's suggestion. According to our analysis, ACI can have a critical impact on SOA at this mountain site. We thus have revised the title as "**Measurement report: Impact of cloud processes on secondary organic aerosols at a forested mountain site in southeastern China**".

Specific comments:

RC3: I recommend a major reanalysis and rewrite of the manuscript – some of these specific comments may then no longer be relevant. However, I still decided to provide detailed specific comments to provide the authors with a more in-depth analysis of their current manuscript. The comments are in order of appearance and not sorted by relevance.

Were relevant I assume that during cloudy periods only interstitial aerosol was sampled (see major comment #4). Note that the interpretation of the ACSM/AMS results will be different if interstitial + cloud droplets were sampled.

Responds: We thank the referee for providing these valuable and detailed comments. Below are our detailed point-to-point responses.

RC3-10: Line 22f: I do not agree with the statement about scavenging of MO-OOA and cloud droplet evaporation as a source for LO-OOA (see XXX). However, the way this sentence is written, it implies to me that LO-OOA is actively formed in the cloud droplets (e.g. from conversion of MO-OOA). Is that really the case?

Response: Thank the referee for pointing this out. According to our results, LO-OOA is physically released from the cloud droplet but not activity formed in them. To avoid misunderstanding, we have replaced the word "produce" with "release" and explained the source of LO-OOA, which is "from airmass transported from polluted regions".

Text modifications: Line 27: "*While MO-OOA was scavenged efficiently during cloud events, the cloud evaporation was found to release a significant amount of LO-OOA from airmass transported from polluted regions.*"

RC3-11: Line 23: I do not understand this sentence. I cannot follow the argument of how the O/C change with OA/ΔCO values implies things about the cloud processing.

Also, ΔCO is not introduced.

Response: We feel sorry for the inconvenience brought to the referee. We recognize that the sentence was not clearly articulated and may lead to confusion. We now introduce ΔCO in this section, defining it as "CO after subtracting the background level" that is 0.12 ppm in this study. Furthermore, we have combined it with the next sentence to make the argument more understandable to readers.

Text modification: Line 29: "*The distinct increase of OA/ΔCO (CO after subtracting the background level) with the decrease of O/C during the evaporative period further demonstrates that OA remained in cloud droplets are generally in a moderate oxidation state.*"

RC3-12: Line 37: It is not just high-altitude aerosol that can serve as CCN. Or are the authors implying that high-altitude particles are even better CCN?

Response: Thank the referee for highlighting the need for clarity regarding our discussion of high-altitude aerosols as CCN. In our manuscript, we intended to suggest that high-altitude aerosols can be effective as CCN because the atmospheric conditions at high altitudes can favor the cloud formation (Rejano et al., 2021). This was also based on the finding from Asmi et al. (2012), which demonstrates that anthropogenic emissions from the ground can enhance the CCN activation at a mountain site, where initial aerosol concentrations are low. To address the referee's concern, we have revised the sentence.

Text modification: Lines 41-44: "*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).*"

References:

Rejano, F., Titos, G., Casquero-Vera, J. A., Lyamani, H., Andrews, E., Sheridan, P., Cazorla, A., Castillo, S., Alados-Arboledas, L., and Olmo, F.: Activation properties of aerosol particles as cloud condensation nuclei at urban and high-altitude remote sites in southern Europe. Sci. Tot. Environ., 762, 143100, https://doi.org/10.1016/j.scitotenv.2020.143100, 2021.

Asmi, E., Freney, E., Hervo, M., Picard, D., Rose, C., Colomb, A., and Sellegri, K., Aerosol cloud activation in summer and winter at puy-de-Dôme high altitude site in France. Atmos. Chem. Phys., 12(23), 11589-607, https://doi.org/10.5194/acp-12-11589-2012, 2012.

RC3-13: Line 44f: what are the advantages. If the authors mention advantages of a method over another, they should give at least some examples of said advantages.

Response: Thank the referee for the above suggestions. We have added some examples that discuss the advantages of mountain sites for aerosol observation.

Text modification: Line 50: "*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.*"

RC3-14: The introduction does not mention ACI. If after the reanalysis, ACI is indeed found as a driver for composition changes etc., it should be introduced in the Introduction.

Response: Thank the referee's comments. We revise the introduction to include a section on ACI in the first paragraph.

Text modification: Line 44: "*On the other hand, ACI 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).*"

RC3-15: Section 2.1: I assume the SH site is an established station with some documentation. This manuscript has no information about the "standard" measurements at this location (e.g., meteorological parameters and instrumentation). The authors either have to mention all instruments from which they use data (can be in the supplement) or they must provide a reference to the full description of the station.

Response: It is really true as the referee suggested that the detailed information of the sampling site and instruments needs to be introduced. We therefore have provided a description of all instruments in the supplement.

Text modification: Line 94: "*More details and descriptions of the instruments and data are provided in Table S1.*"

RC3-16: Definitions and measurements of PM1, PM2.5, PM10. The authors introduce non-refractory PM1 (NR-PM1) as the result from AMS and ACSM. Then in the later text they mostly use PM1. I assume in most cases, the authors actually mean NR-PM1 from AMS/ACSM potentially including the Black\Carbon (BC) mass from the aethalometer. Either change to NR-PM1 at the appropriate places or clearly state that PM1 here means the value from AMS/ACSM + BC.

Response: Thank the referee for the careful check. The clarification of PM_1 ($NR-PM_1$ + BC) has been included in the manuscript in Section 3.1.

RC3-17: Line 93: Why were the default values used for RIE? Later they state that calibrations were conducted.

Response: We thank the referee for pointing this out. The default value was used based on two reasons. First, the default RIEs for organics, nitrate, and chloride are based on many calibrations of laboratory-generated aerosols previously, and have been proven to be representative in ambient conditions (Canagaratna et al., 2007; Nault et al., 2023). Second, we only use ammonium sulfate to calibrate the RIE values of ammonium and sulfate. We have added these references to the manuscript.

Text modification: Line 100: "*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).*"

References:

Canagaratna, M. R., Jayne, J. T., Jimenez, J. L., Allan, J. D., Alfarra, M. R., Zhang, Q., Onasch, T. B., Drewnick, F., Coe, H., Middlebrook, A., Delia, A., Williams, L. R., Trimborn, A. M., Northway, M. J., DeCarlo, P. F., Kolb, C. E., Davidovits, P., and Worsnop, D. R.: Chemical and microphysical characterization of ambient aerosols with the aerodyne aerosol mass spectrometer. Mass Spectrom. Rev., 26(2), 185-222, https://doi.org/10.1002/mas.20115, 2007.

Nault, B. A., Croteau, P., Jayne, J., Williams, A., Williams, L., Worsnop, D. R., Katz, E. F., DeCarlo, P. F., and Canagaratna, M.: Laboratory evaluation of organic aerosol relative ionization efficiencies in the aerodyne aerosol mass spectrometer and aerosol chemical speciation monitor. Aerosol Sci. Techno., 57(10), 981-97, https://doi.org/10.1080/02786826.2023.2223249, 2023.

RC3-18: Line 94: Which compounds were used for the calibration? Ammonium nitrate? Ammonium sulfate? Both? Individually? As mixtures?

Response: We only used ammonium sulfate for calibration of RIE values of ammonium and sulfate. This clarification has been included in the manuscript.

Text modification: Line 102: "*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.*"

RC3-19: Figure S1: the y axis is probably NR-PM1 from AMS and/or ACSM? What are the sources for PM2.5 and PM10? Was PM2.5 and PM10 also measured after drying or from ambient conditions?

Response: We have revised Fig.S1 to include NR-PM_1 from both AMS and ACSM. Also, we added a description of the sources of NR-PM_1, $PM_{2.5}$, and PM_{10} in the capture of Fig. S1. $PM_{2.5}$ and PM_{10} were measured directly from ambient conditions (without drying). Also, a detailed comparison of AMS and ACSM data has been added to the revised manuscript.

Text modification: Line 103−108: "*Detailed comparison of the concentrations of NR-PM$_1$ 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. As shown in Fig. S1g and h, after this adjustment, the concentrations of NR-PM$_1$ 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 PM$_{10}$ ((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.*"

Graph modification:

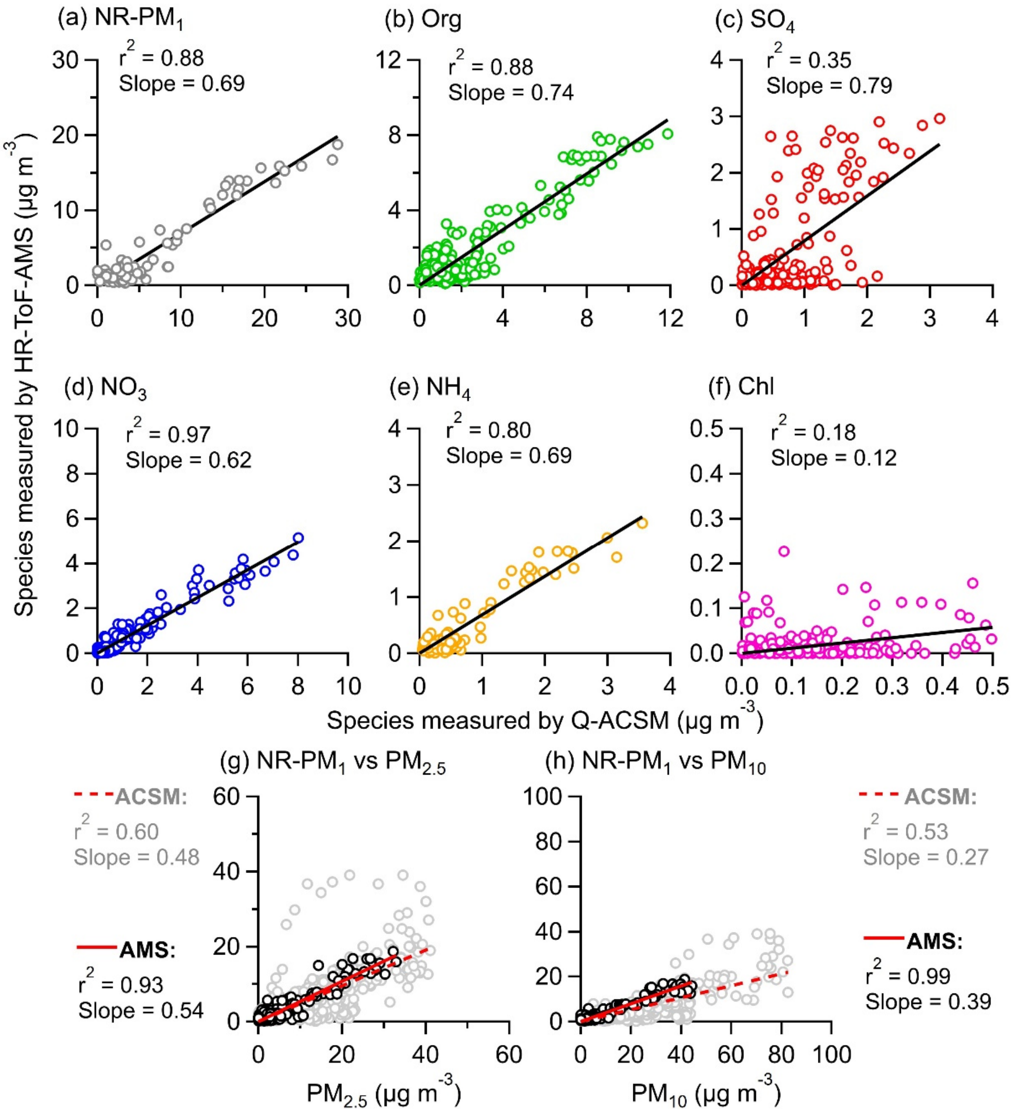

Figure S1. (a-f) Comparison of different NR-PM$_1$ species measured by ACSM and AMS during the AMS sampling period (15–20, 24–28 Nov); (g-h) Scatter plots of mass concentrations of NR-PM_1 measured by ACSM (grey) and AMS (black) vs. $PM_{2.5}$ and PM_{10} measured by particulate monitor. The black and red lines are linear-fit lines.

RC3-20: Figure S2: While it is good to provide the PMF diagnostics for the chosen solution, a non-PMF expert cannot do anything with this. The authors should provide at least the explanation of the parameters (e.g. Q/Qexp) and ideally write a short paragraph in the supplement about their indication for the validity of the chosen solution.

Response: We thank the referee for pointing this out. We have added a paragraph (Text S1) to explain these parameters and the determination of factor number in the supplement.

Text modification:

"***Text S1.*** *PMF diagnosis*

In this study, organic mass spectra together with several selected inorganic ions (SO^+, SO_2^+, SO_3^+, HSO_3^+, $H_2SO_4^+$, NO^+, NO_2^+, NH^+, NH_2^+, NH_3^+, Cl^+, and HCl^+) were analyzed by the PMF. The number of factors from 1 to 5 with fPeak varying from -1 to 1 were evaluated, and the diagnostic plots are shown in Figure S2. In the PMF analysis, the Q/Q_{exp} values represent the ratios between the actual sum of the squares of the scaled residuals (quality-of-fit parameter, Q) obtained from the PMF least square fit and the ideal Q (Q_{exp}) obtained if the fit residuals at each point were equal to the noise specified for each data point. The Q/Q_{exp} value should be approximate to unity if the number of factors is appropriate and with small errors (Ulbrich et al., 2009). As shown in Figure S4a, The Q/Q_{exp} value started to greatly decrease from one- (1.52) to three-factor solution (1.05). However, the three-factor solution (including one organic associated with sulfate ions factor (SO_4-OA), one inorganic nitrate factor (NIA), and one oxygenated OA (OOA)) cannot further differentiate OOA with different oxidation degrees. The four-factor solution, with a value of Q/Q_{exp} (1.0065) close to 1, can separate two OOA factors with different elemental ratios and temporal variations. Continuing to increase the factor number cannot significantly decrease the Q/Q_{exp}. Furthermore, with 4 factors, the reconstructed mass tracked well with the variations of measured mass, and the scaled residuals of all ion fragments were distributed between -4−4 (Figs. S2c and d), suggesting that they were well reproduced by the PMF model. Therefore, we considered the four-factor solution with fPeak = 0 to be the optimum solution for PMF analysis in this study."

RC3-21: Section 2.2.2 and 3.3 The derivation of organonitrates (ON). This simple approach assumes that all NO3 that correlates with organic compounds is indeed organic nitrate. This assumption cannot be made ab initio and only is validated when the PMF factors are discussed.

Response: Following the referee's comments in **RC3-21** and **RC3-22**, we have revised section 2.2.2 to the validation of this method.

Text modification: Lines 118-126: "*ONs were estimated from the PMF results (Zhang et al., 2011; Xu et al., 2015). Briefly, NO_x^+ (i.e., NO^+ and NO_2^+) are major fragments of nitrate functionality ($-ONO_2$), which can be referred to as the total nitrate measured by AMS. Combining inorganic with organic mass spectra in PMF, NO^+ and NO_2^+ can be separated into different organic aerosol (OA) factors and an inorganic nitrate aerosol factor (NIA). According to previous studies, the ratios of NO^+/NO_2^+ for ONs are approximately 2.25–3.7 times higher than pure NH_4NO_3 (Fry et al., 2013; Fry et al., 2009). Consistently, the PMF results in our study (Fig. 7) show that the average NO^+/NO_2^+ ratios of LO-OOA and MO-OOA were 13.19 and 11.2, falling within the range of ONs. In contrast, a NO^+/NO_2^+ ratio of 3.56 was observed for NIA, 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:*"

RC3-22: Section 2.2.2 and 3.3. The method described in section 2.2.2 assumes that any NOx that correlates with organic is organic nitrate. This cannot be assumed a priori as NO3 can correlate with organic also if it is emitted from the same source. In section 3.3, the authors finally bring up the NO2+/NO+ fragmentation method (note the benefits of using the revers ratio (Kiendler-Scharr et al., 2016)). This method needs to be introduced already in the methods section. This provides the evidence that the simplification is indeed valid.

Response: We thank the referee for further suggestions about this issue. Since the **RC3-21** and **RC3-22** are pointed to the same issue, the revised text has been provided together in **RC3-21**.

RC3-23: Continuing on this topic: The NO2+/NO+ fragmentation method can also be used for ACSM data (using adjusted fragmentation values). Why was that not done? This would provide a more holistic picture for the ON during the whole measurement period and provide more cases of "evaporating clouds" to test their hypothesis of enhancement of ON contribution due to the presence of clouds.

Response: We appreciate the referee's suggestion. Unfortunately, due to the limitations outlined in **RC3-5**, the ACSM PMF results were not utilized in this study. Consequently, it is not feasible for us to estimate the ON throughout the entire measurement period. We respectfully request the understanding and leniency of both the editor and the referee in this matter.

RC3-24: Section 2.2.3: This section is not clear.

- What map was coloured?
- What does ENSEMBLE stand for? Is it a data product from some measurement? Is it a modelling product?
- The MERRA-2 model is not general knowledge and needs a reference.
- The Giovanni link did not work for me. When I found it eventually, I realized that this section does not contain the necessary information to track what data was used.

Response: Thank the referee for pointing this out.

-The map that is being colored is the map containing the back trajectory (Fig. S5).

-The "Time-averaged organic carbon surface mass concentration (ENSEMBLE)" is the name variable used from the MERRA-2 reanalysis dataset. This dataset is named "MERRA-2 tavg1_2d_aer_Nx: 2d, 1-Hourly, Time-averaged, Single-Level, Assimilation, Aerosol Diagnostics V5.12.4 (M2T1NXAER)", which is an hourly time-averaged 2-dimensional data collection in MERRA-2. This collection consists of assimilated aerosol diagnostics, such as column mass density of aerosol components (black carbon, dust, sea salt, sulfate, and organic carbon), surface mass concentration of aerosol components, and total extinction (and scattering) aerosol optical thickness (AOT) at 550 nm.

-Modern-Era Retrospective analysis for Research and Applications version 2 (MERRA-2) model is the latest version of global atmospheric reanalysis for the satellite era produced by NASA Global Modeling and Assimilation Office (GMAO) using the Goddard Earth Observing System Model (GEOS) version 5.12.4 (Gelaro et al., 2017).

-The available Giovanni link is revised as https://giovanni.gsfc.nasa.gov.

We have added the above information in section 2.2.3.

Text modification: Lines 135-139: "*To further show the particle concentration levels in different regions, the map contained the back trajectory (Fig. S5) was colored by the time-averaged organic carbon surface mass concentration from the M2T1NXAER v5.12.4 dataset (0.5 x 0.625°, hourly). This dataset, part of the Modern-Era*

Retrospective analysis for Research and Applications version 2 (MERRA-2) model (Gelaro et al., 2017), was sourced from the NASA Giovanni website (https://giovanni.gsfc.nasa.gov)."

References:

Gelaro, R., McCarty, W., Suárez, M. J., Todling, R., Molod, A., Takacs, L., Randles, C. A., Darmenov, A., Bosilovich, M. G., Reichle, R., Wargan, K., Coy, L., Cullather, R., Draper, C., Akella, S., Buchard, V., Conaty, A., de Silva, A. M., Gu, W., Kim, G.-K., Koster, R., Merkova, D., Nielsen, J. E., Partyka, G., Pawson, S., Putman, W., Rienecker, W., Rienecker, M., Schubert, S. D., Sienkiewicz, M., and Zhao, B.: The Modern-Era Retrospective Analysis for Research and Applications, Version 2 (MERRA-2). J. Climate, 30(14), 5419-54, https://doi.org/10.1175/JCLI-D-16-0758.1, 2017.

RC3-25: Line 130ff, Fig 1, Table S1: It is great that the authors compare their measurements with other sites. But they need to be more specific with their selection and interpretation. Particle concentrations can be highly variable between seasons (times of year). Table S1 reveals that only the Mt Daban case is from the same season (October - autumn) while the others are from spring, summer or full year average.

The Mt Tai data from set does contain data for each season, showing that fall has much lower NR-PM1 concentrations than any other season with NR-PM1 of 18.1 ug/m^3 instead of 33.5 ug/m^3 (org+SO4+NO3+NH4 from Table 1 in (Zhang et al., 2014)). It is highly likely that the other selected mountain site may have a similar level of seasonal trends. The seasonal differences need to be indicated in Figure 1 and mentioned/considered in the text when the comparisons are done.

Response: We thank the referee's suggestion. We have indicated the seasonal differences in Fig. 1, Table S1, and discussed them in the text.

Text modification: Lines 148-154: "*The concentration and composition of PM_1 are quite different from those observed at other Chinese mountain sites in different seasons (Fig. 1). For example, the average PM_1 concentration here is much lower compared to Mt. Wuzhi (10.9 ± 7.8 $\mu g\ m^{-3}$) at a similar altitude, and is also lower than at higher altitude mountains such as Mt. Yulong (5.7 ± 5.4 $\mu g\ m^{-3}$) and Mt. Waliguan (9.1 ± 5.3 $\mu g\ m^{-3}$). Although PM_1 concentration has a strong seasonal dependence, such as in Mt. Tai, with low concentration in autumn (18.1 $\mu g\ m^{-3}$) and high in summer (33.5 $\mu g\ m^{-3}$), the average PM_1 concentration at our site is still much lower than Mt. Daban (11.4 $\mu g\ m^{-3}$) and Mt. Tai in the same season (autumn).*"

Graph modification:

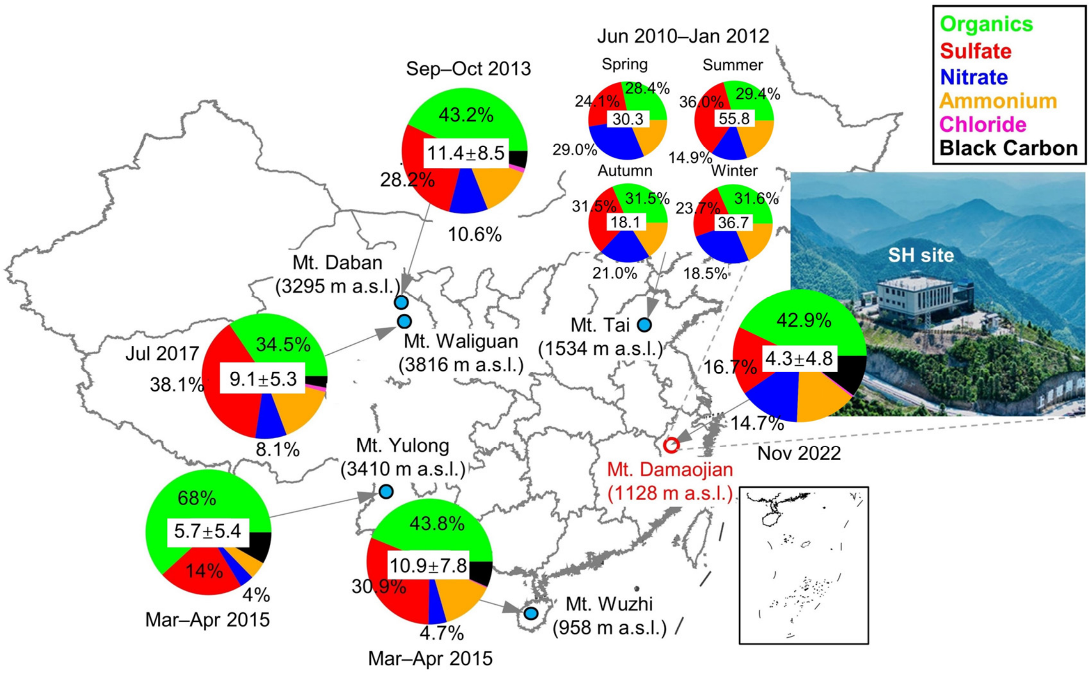

Figure 1. Location of the sampling site. The mean concentration (in µg m^{-3}) and chemical composition of submicron aerosols (NR-PM$_1$+ BC) measured at selected mountain sites in China are also shown. Note that BC and chloride are not accounted for Mt. Tai due to the lack of measurement or data in the relevant study. Detailed information of these sampling sites is presented in Table S1 in the Supplement.

Table modification:

Table S2. Summary of mean mass concentrations (in µg m^{-3}) and chemical composition of submicron aerosols measured at selected mountain sites in China.

Location		**Mt. Wuzhi**	**Mt. Tai**				**Mt. Yulong**	**Mt. Daban**	**Mt. Waliguan**
Time		3/19/2015	2011				3/22/2015	9/5/2013	7/1/2017
		4/15/2015	Spr	Sum	Aut	Win	4/14/2015	10/15/2013	7/31/2017
Org	Mass	4.8	8.6	16.4	5.7	11.6	3.9	4.9	3.5
	Frac.	43.8 %	28.4 %	29.4 %	31.5 %	31.6 %	68 %	43.2 %	38.1 %
SO_4	Mass	3.4	7.3	20.1	5.7	8.7	0.8	3.2	3.1
	Frac.	30.9 %	24.1 %	36.0 %	31.5 %	23.7 %	14 %	28.2 %	34.5 %
NO_3	Mass	0.5	8.8	8.3	3.8	9.6	0.2	1.2	0.7
	Frac.	4.7 %	20.9 %	14.9 %	21.0 %	26.2 %	4 %	10.6 %	8.1 %
NH_4	Mass	1.5	5.6	11.0	2.9	6.8	0.3	1.4	1.4

	Frac.	13.7 %	18.5 %	19.7 %	16.0 %	18.5 %	5 %	12.3 %	15.2 %
Chl	Mass	0.03						0.14	0.1
	Frac.	0.3 %						1.2 %	1.1 %
BC	Mass	0.7					0.5	0.51	0.3
	Frac.	6.6 %					9 %	4.5 %	3.0 %
NR-PM$_1$		10.2	30.3	55.8	18.1	36.7	5.2	10.9	8.8
PM$_1$		10.9					5.7	11.4	9.1
References		(Zhu et al., 2016)	(Zhang et al., 2014)				(Zheng et al., 2017)	(Du et al., 2015)	(Zhang et al., 2019)

RC3-26: Figure 1: "BC if it was available": Do the authors really mean "available" as in there were measurements of BC. Or do they mean that they did not detect BC? (I have to ask as the authors have been using terms in unusual ways throughout the manuscript)

Response: The "available" means that there were measurements of BC in their studies. To avoid misunderstanding, we added an explanation to the caption.

Text modification: Lines 71-73: "*The mean concentration (in $\mu g\ m^{-3}$) and chemical composition of submicron aerosols (NR-PM$_1$ + 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.*"

RC3-27: The authors decided to focus on the organic compounds and nitrate for their deeper analysis. But I found the fact that the SO4 contribution is much lower than at any other station even more interesting. Typically, SO4 is associated with anthropogenic sources (or volcanoes) as is most NO3. For multiple periods, NO3 will increase drastically while SO4 is less affected. What could this mean? That the source regions do only have NOx as a pollutant but no SOx?

Response: We thank the referee for pointing this out. Indeed, the SO4 contributions in our study are noticeably lower compared to other mountainous locations. This discrepancy partly results from the successful implementation of SO$_2$ emission reduction initiatives in China, such as the "Action Plan on Prevention and Control of Air Pollution" in 2013 and the "Three-Year Action Plan" in 2018. Supporting our hypothesis, Wen et al. (2023) reported an 89.7% decrease in particulate SO$_4$ on Mt. Tai over a decade (2008-2018). Consequently, the lower SO$_4$ contribution at our station in 2022, in comparison to other sites during 2011−2017, can be likely attributed to these ongoing SO_x emission reductions. Additionally, the low SO_4 contribution at this site may suggest that aerosols at this site are more likely influenced by emissions transported from small regional areas. This is further supported by the high NO_3 contribution in this site. We have added the above discussion to the text.

Text modification: Line 156-163: "*Notably, a ubiquitously lower contribution of sulfate to* PM_1 *was observed (16.7 %) in this site compared to other mountain sites (14.0 %−38.1 %), which was likely attributed to the significant reduction of* SO_2 *emission in China during the past 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, at Mt. Tai, even though the contribution of sulfate were high, nitrate still accounted for considerable fractions (14.9 %−29.0 %) that were comparable to this site. Taken together,* PM_1 *at this site is more likely influenced by anthropogenic emissions over a smaller regional scale, consistent with the high urban density in eastern China.*"

References:

Wen, L., Xue, L. K., Dong, C., Wang, X. F., Chen, T. S., Jiang, Y., Gu, R. R., Zheng, P. G., Li, H. Y., Shan, Y., Zhu, Y. J., Zhao, Y., Yin, X. K., Liu, H. D., Gao, J., Wu, Z. J., Wang, T., Herrmann, H., and Wang, W. X.: Reduced atmospheric sulfate enhances fine particulate nitrate formation in eastern China. Sci. Total Environ., 898, 165303, https://doi.org/10.1016/j.scitotenv.2023.165303, 2023.

RC3-28: Also, because the SO4 is so low and NO3 high, most NH4 will probably be in the form of ammonium nitrate which is much more volatile than ammonium sulphate. This should be brought up in the discussion somewhere.

Response: We have added this discussion in the text following the comment (see **RC3-27**).

RC3-29: Line 132ff: Here the authors claim that the low PM1 concentrations are linked to the cloud scavenging. But they do not provide supporting arguments.

I played around with the data set. Using the "RH=100% = cloud" criterion, I get an average total aerosol concentration (NR-PM1+BC) of 3.6+/-6.1 ug/m^3 for cloudy periods and 5.9+/-5.0 for the non-cloudy ones. But the highest concentration (Nov 4th) occurs during a "cloudy" time. This circles back to first needing to investigate the airmass origin and other meteorological conditions before making such a claim. What are the characteristics during RH=100% periods? Is the air more stagnant with less likelihood of long-range transport plumes? Where are the plumes coming from? What local conditions create the TH=100% conditions? Is there coincidence (instead of causation) e.g. that a certain wind direction favours RH=100% which is also the “cleaner” wind sector?

Response: We thank the referee’s suggestions. The discussion of airmass origin and other meteorological conditions have been added in section 3.2. Please see **RC3-1** for the revised text.

Text modification: Lines 154-156: “*Considering the occurrence of frequent cloud events during the sampling period, the low PM_1 concentration might be mainly associated with cloud scavenging, which will be further validated in subsequent sections.*”

RC3-30: Line 135ff: “One explanation was that nitrate formed in polluted regions interacted with clouds and affected the regional nitrate level as cloud evaporates (Tao et al., 2018)” Why are clouds needed for this? At Rh >70% aerosol particles have a considerable aqueous phase and thus also take up considerable amounts of HNO3 – especially if NH3 or other bases are available.

Response: This sentence has been deleted. It is really true that during several episodes in this study (e.g., EP1), high concentrations of nitrate were more likely produced by the anthropogenic-emitted NO_x transported from big cities followed by heterogeneous uptake of N_2O_5 under high RH at night. Thus, we have deleted this sentence and further discussed this possible mechanism of nitrate formation during a nitrate episode in section 3.2 of the revised manuscript. Please see the revised text below:

Text modification: Line 203−208: “*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. S6) showing high levels of nitrate mainly associated with northeastern wind directions. This was further supported by the back trajectory analysis, in which air masses on 4 Nov were also from the northeast. Moreover, the RH was stable at 100% during EP1, which can favor the heterogenous uptake of N_2O_5 and subsequent nitrate formation in the aqueous phase at night.*”

RC3-31: The link of increased NO3 to the “evaporation” of cloud droplets is also not justified as such a general statement. I looked into two periods with decreasing RH trends. Nov 26-27th (=P2 top panel in Fig 1below) and Nov 21st midday (bottom panel in Fig 1). In both cases, the organic signal starts to increase well before the RH decreases. The increase coincides with a change in wind direct. Importantly, NO3 does not increase at the same time. For the 26th, the NR-PM1 further increase in the afternoon (16:00) even further, this time also for NO3. Note that this is when the RH increases – potentially due to decreasing temperatures. On the 27th, the decrease in concentration of all species does not coincide with RH reaching 100% or decreasing from 100%. Instead, it again coincides with a change in wind direction (and this time also wind speed). Similarly, the 21st has a mysterious abruptly lower NR-PM1 concentration before midnight during a RH=100% period. But the decreasing trend starts again together with a change in wind direction.

The authors need to look further into such events and include more details (e.g. air mass origin, temperature) before making such generalised statements as they did.

Response: We thank the referee's comments. After careful re-analysis, we strongly agree that there were different causes for nitrate elevation in different episodes. Detailed discussions have been added in the revised manuscript (please see **RC3-1** for the text).

RC3-32: Line 141ff and Fig 3: the authors state that the high levels of PM1 are mainly caused by (inorganic) nitrate formation. Fig 3 seems to indeed suggest that. But it is an example of how the inclusion of the 6h NO3 event from Nov 4th can completely skew an interpretation. All datapoint with PM1 >35 ug/m^3 are from that single event which is completely dominated by inorganic NO3. Plotting the concentration of organic and NO3 as a function of total aerosol mass (NR-PM1+BC) shows that the slope is steeper for organic than for NO3 when omitting the data points from the Nov 4th plume (i.e. the points marked with red and black circles in Fig 2). This means that the majority of NR-PM1 mass stems from the increase in organic compounds and not from NO3. Only for that one short period, NO3 becomes the dominant driver for aerosol mass formation.

Fig 3 below shows the species contribution to NR-PM1 after removing the Nov 4th plume. There is still an increasing trend for NO3 and a decrease for organics with increasing NR-PM1. But the main difference is now between the first bin (<5 ug/m^3) - which represents the typical low concentration background level at this site- and the higher concentration cases which are probably dominated by long-range transport of anthropogenic emissions. It would also be interesting to check the ion balance to see if the rather constant NH4 contribution is converted from ammonium sulphate to nitrate with increasing aerosol concentration.

Response: Thank the referee for the valuable suggestions. Following the suggestions, we have added a scatter plot according to Fig. 2 in the referee's comments to our revised manuscript (Fig. S3). It is true that the increase in organic dominated the NR-PM_1 mass after excluding the nitrate episode on 4th Nov. Moreover, we have updated Fig. 3 to include the variations of aerosol composition as a function of PM_1 mass without the nitrate episode (Fig. 3b). As shown in Fig. 3b, while the fraction of nitrate exhibited an upward trend along with increasing PM_1, the fraction of ammonium remains consistent, indicating their transition from ammonium sulfate to nitrate with increasing aerosol concentration.

Text modification: Lines 167-178: "*We noted that a nitrate-dominant peak of PM_1 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_1 mass during this event and the rest of the campaign are shown in Figure S3. Almost all the data points with PM_1 concentrations above 20 μg m^{-3} were from this event. During EP1, a steeper slope for nitrate relative to PM1 was found than that for organics, which was contrary to slopes during the rest period (Fig. S3). These distinct differences in slopes for nitrate and organics implied different mechanisms of PM_1 elevation during these two periods. Consequently, we also excluded EP1 from the statistic of Fig. 3a, and the result is presented in Fig. 3b. After removing EP1, organics became the dominant contributor (> 40 %) across all PM_1 concentrations. Despite this, there was still an increasing trend for nitrate and a decreasing trend for sulfate along with increased PM_1, 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_1 concentration. Overall, these results suggested that high levels of PM_1 at the SH site might be mainly attributed to the formation or transport of organics, meanwhile, nitrate also plays a nonnegligible role.*"

Graph modification:

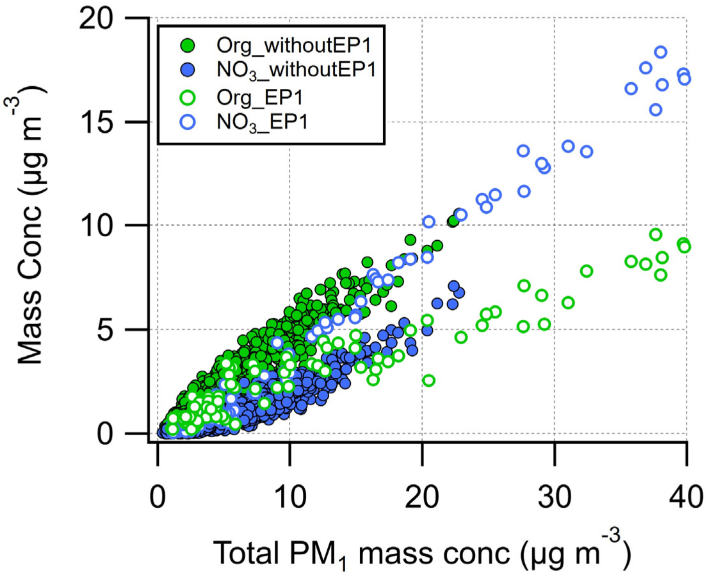

Figure S3. Scatter plot of organic and nitrate concentrations versus PM$_1$ concentrations during the entire campaign and the campaign without EP1.

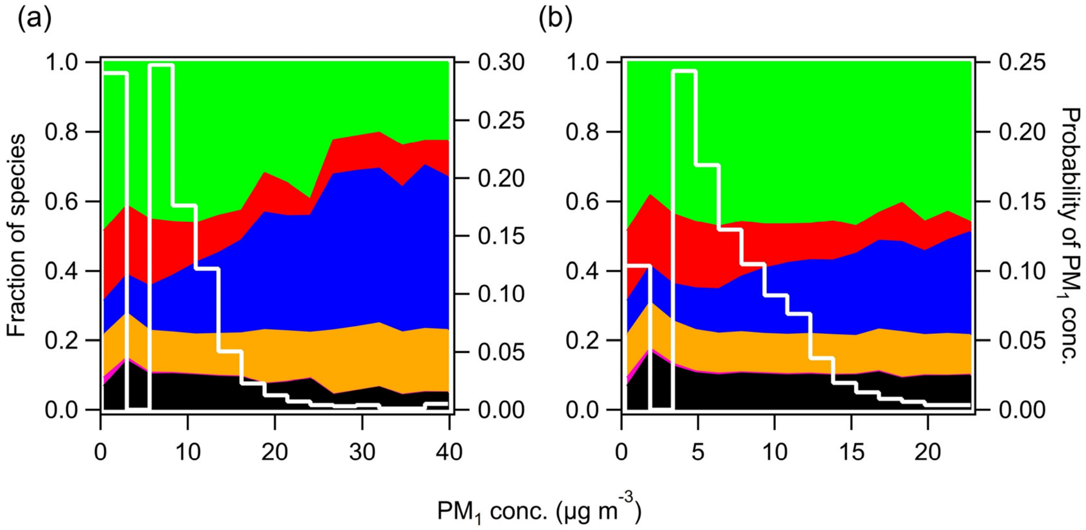

Figure 3. Variations of aerosol composition as a function of PM$_1$ mass concentration and the probability density of PM$_1$ during (a) the entire campaign and (b) the campaign without EP1.

RC3-33: The time series of PM2.5 seems to correlate with BC? Is that the case? Does that mean something?

Response: Thank the referee for pointing this out. Indeed, PM$_1$ and PM$_{2.5}$ correlated well with BC (r^2 = 0.59 and 0.62, respectively). These correlations suggest that the PM pollution at this site may be largely influenced by the regional biomass burning plumes in the Yangtze River Delta (Zhang et al., 2015). We have added this discussion in section 3.1.

Text modification: Lines 146-148: "*In addition, BC was observed to correlate well* *with PM_1 and $PM_{2.5}$, suggesting that the aerosol particles at this site may be largely influenced by the regional biomass burning plumes in the Yangtze River Delta (Zhang et al., 2015).*"

References:

Zhang, Y. J., Tang, L. L., Wang, Z., Yu, H. X., Sun, Y. L., Liu, D., Qin, W., Canonaco, F., Prévôt, A. S. H., Zhang, H. L., and Zhou, H. C.: Insights into characteristics, sources, and evolution of submicron aerosols during harvest seasons in the Yangtze River delta region, China, Atmos. Chem. Phys., 15, 1331-1349, https://doi.org/10.5194/acp-15-1331-2015, 2015.

RC3-34: Fig 5: I do not understand the right-side panels. There is supposed to be a box plot? Also, why is this analysis only conducted for the AMS time period, most of these values can be derived from ACSM data as well.

Response: We are sorry for the incorrect writing of the caption. The right-side panels are the average values of elemental ratios in P1 and P2.

The H/C and O/C of OA measured by ACSM can indeed be estimated based on f_{43} and f_{44}. However, these parametrizations are approximations and ACSM f_{44} measurements have been shown to vary between instruments (Fröhlich et al., 2015). Uncertainties may also be introduced especially in this background site with low aerosol concentrations. In addition, in the revised manuscript, we have validated the representativeness of the two AMS periods as typical cloudy and evaporative periods. Therefore, we think that using only AMS data for the analysis may be suitable.

Text modification: Line 290-294: "*Time series of 1-hour averaged (a) N/C and H/C, (b) OM/OC, OSc, and O/C, and (c) NO^+/NO_2^+ and $CH_3SO_2^+/CH_2SO_2^+$ during the AMS sampling site (left) and their mean values in P1 and P2 (right). Only ratios determined with good S/N (i.e., organics > 0.7 μg m^{-3}) are shown. The horizontal black and blue dash line in the bottom right plot represent the $CH_3SO_2^+/CH_2SO_2^+$ value for pure MSA (2.9) and the NO^+/NO_2^+ value for pure AN (3.88), respectively.*"

References:

Fröhlich, R., Crenn, V., Setyan, A., Belis, C. A., Canonaco, F., Favez, O., Riffault, V., Slowik, J. G., Aas, W., Aijälä, M., Alastuey, A., Artiñano, B., Bonnaire, N., Bozzetti, C., Bressi, M., Carbone, C., Coz, E., Croteau, P. L., Cubison, M. J., Esser-Gietl, J. K., Green, D. C., Gros, V., Heikkinen, L., Herrmann, H., Jayne, J. T., Lunder, C. R., Minguillón, M. C., Močnik, G., O'Dowd, C. D., Ovadnevaite, J., Petralia, E., Poulain, L., Priestman, M., Ripoll, A., Sarda-Estève, R., Wiedensohler, A., Baltensperger, U., Sciare, J., Prévôt, A. S. H. ACTRIS ACSM intercomparison – Part 2: Intercomparison of ME-2 organic source apportionment results from 15 individual, co-located aerosol mass spectrometers. Atmos. Meas. Tech., 8(6), 2555-76,

https://doi.org/10.5194/amt-8-2555-2015, 2015.

RC3-35: Section 3.2 and Fig S4: The authors conflate two things in their RH dependency analysis: 1) the actual RH dependency for RH <100% and 2) the cloud/no cloud condition. The box plots in Fig S4 also use 90-100% as the highest bin. I.e., grouping saturated and subsaturated conditions together.

I do not think that this approach is suitable to identify true RH dependencies in the data. Yes, usually a statistical approach is better to reveal general trends in long term ambient data. But here we are looking at 30 days of measurements with 55% of the data points are "cloudy". During both cloudy and non-cloudy periods distinct plumes of different origin arrive at the site overlapping any local meteorological trend. There are very likely interesting dependencies as the detailed look in Fig 1 above and specific comment #20 show.

Response: Thank the referee's valuable suggestions. We have updated this figure to compare the scenarios of RH≠100% and RH=100%, which may further reflect the influence of the cloud process on aerosol particles.

Text modification: Lines 221-226: "*It is worth noting that PM_1 peaks in EP2−EP5 all occurred along with RH below 100 %. When the RH returned to 100 %, PM_1 concentrations gradually diminished to levels comparable to those observed during the clean period (C1). Furthermore, as shown in Fig. S7, there were notable reductions in the mean and median mass concentrations of all PM_1 species over the entire campaign, ranging between -2.6 %−44.4 % and 2.8 %−50.1 % when RH reached 100 % from conditions of lower RH, respectively. Considering the frequently occurring cloud events at this site, it is most likely that these variations of RH were affected by the cloud process, which may further play an important role in PM_1 concentrations.*"

Graph modification:

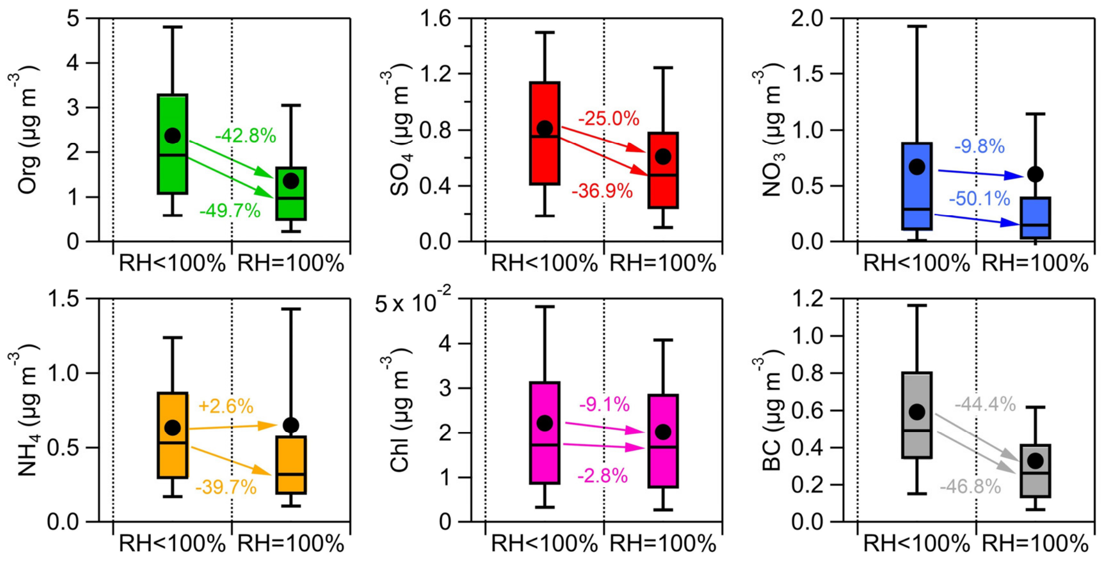

Figure S7. Comparison of mean and median mass concentrations of PM_1 species under conditions of RH below and at 100 % over the entire campaign.

RC3-36: Fig S4: why are there two separate y axis? They made comparison difficult.

Response: Sorry for the inconvenience brought to the referee. In the newly updated figure, we only use one y axis (see **RC3-35**).

RC3-37: Line 161: What test was used to identify the “significant” decrease of organics with RH? In this context “significant” refers to statistical tests and cannot be assigned “by eye”

Response: We thank the referee’s comments. The specific values have been added to the newly updated Figure (see **RC3-35**).

RC3-38: Line 163f: “Previous studies have shown that aerosol mass generally increases on foggy days (Chen et al., 2021). This phenomenon could be due to the cloud scavenging effect under high RH at this site.” These two sentences are contradictory. If fog increases aerosol mass, this cannot be caused by scavenging which would only lead to a mass reduction.

Response: We thank the referee for pointing out this problem. Since we have re-plot Fig. S4 and re-write this section, this sentence is no longer needed.

RC3-39: Line 165 “submicron aerosols grow to larger sizes under high RH that AMS aerodynamic lens cannot transmit”. Since there is a drier in the sample line, the aerosol will always be at low enough RH at the AMS/ACSM inlet. (If it isn’t the authors should have highlighted that problem already in the methods section.) But it is relevant if there was a size cutoff for the sample line at ambient conditions (see Major

Comment #4). That would indeed exclude the cloud and fog droplets from measurements only leaving the interstitial aerosol. The authors really need to clarify their sampling assembly as again things really depend on the setup.

Response: Thank the referee's comment. Indeed, the dryer before the AMS/ACSM inlet made aerosols always be at low RH. We have clarified our sampling assembly in the method section of revised manuscript (see **RC3-4**) and deleted this incorrect sentence.

RC3-40: Section 3.2: The text suggests that P1 and P2 were selected as typical cloud/non-cloudy periods. The authors do not justify this selection: What does make these periods typical/representative? Nov 6th-11thseems to be a much more stable non-cloud/fog period than the few hours in Period P2. Why was that not included? Why does P1 start on midday Nov 15th? RH is at 100% already for the two previous days. If the reason for selecting these time periods was mainly the availability of AMS data, then this should be stated as such – and arguments provided why AMS data is needed for the interpretation. At the current state only the general composition and oxidation state is utilised (both available from ACSM as well). The particle size information (only available from AMS) is only a minor supporting argument in this.

Response: We thank the referee for pointing these out. Below are our point-to-point replies:

- Validation of this selection has been included in our revised paragraph in **RC3-1**.
- An episode (EP2) from Nov 6th–11th has been included in our analysis now.
- An important reason for only using AMS data is that we cannot perform PMF analysis on ACSM data (we apologize again for this) in this study.

RC3-41: Section 3.2.1 and Fig 4: for me, the key difference is that P1 seems to be externally mixed with organics dominating smaller particles. This could be an indication for local SOA formation from biogenic sources (There is a lot of forest around the site in the photo in Fig 1).

Response: Thank the referee for the valuable suggestion. We have added this discussion in the manuscript.

Text modification: Line 249-253: "*Also, the complex and broad size distribution observed in C1 suggests that these smaller particles are likely externally mixed with organics, which may further imply the potential formation of SOA from local biogenic sources.*"

RC3-42: Fig 4: The authors should check if the NH4 signal can be interpreted or is below the detection limit for pToF analysis.

Response: We have re-calibrated the pToF data and re-checked the NH_4 signal, and the NH_4 size distribution in Fig. 5 has been revised.

Graph modification:

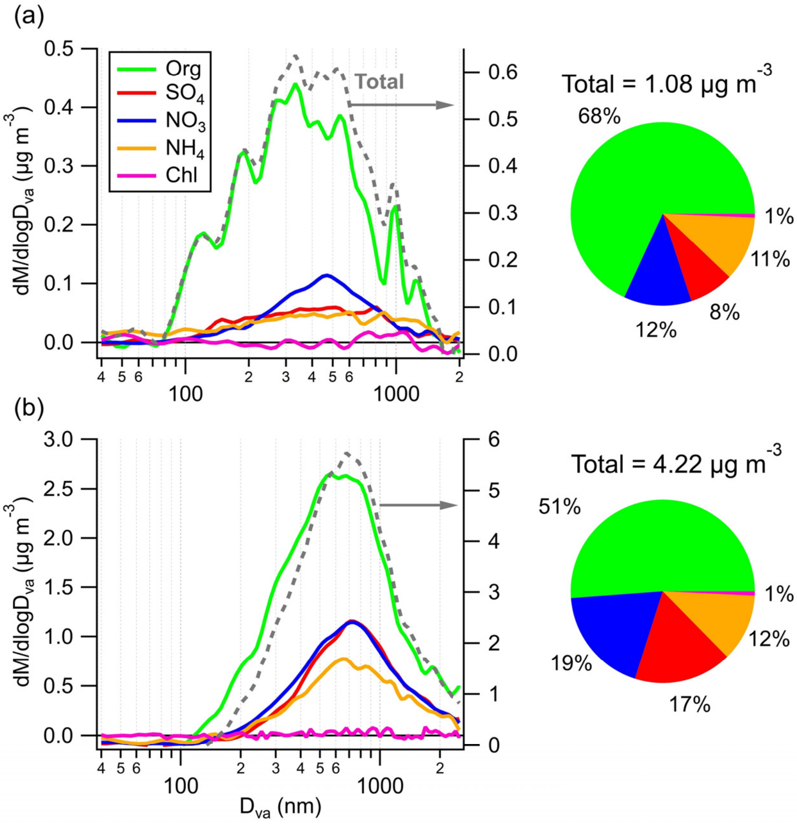

Figure 5. Averaged size distributions and chemical composition of NR-PM_1 during (a) C1 and (b) EP5.

RC3-43: Line 184f: "attributed to the lower mass scavenging efficiency of organics than inorganic species" What is meant by this phrase?

Response: In our study, we identified C1 as a cloud scavenging period. We found that the contribution of organics to NR-PM_1 in C1 (68.1%) was higher than that in EP5 (51.2%). Correspondingly, the contributions of inorganic species decreased in C1. Since C1 was identified as a typical cloudy period, indicating that inorganic species were more effectively scavenged than organics.

RC3-44: Line 209: Methane sulphonate is a typical compound in marine and remote coastal boundary layer (Chen et al 2019). Are they really expecting that compound on an inland mountain?

Response: Thank the referee for pointing this out. Indeed, methanesulfonic acid

(MSA) is mainly formed from DMS and generally exists in marine aerosols, which are unlikely to be abundant in inland mountains. However, the East China Sea is 180 km to the east of this site. We therefore used MSA as a tracer to see whether the air mass transport from the East China Sea can have an impact on PM_1 at this site. According to the low contributions of MSA, aerosols over this region are likely to have negligible oceanic influences. This is consistent with the back trajectory results, which show that PM_1 at this site is dominantly influenced by continental sources in the west and southwest. Following the above suggestions, we have added an explanation in the revised text.

Text modification: Lines 285-288: "*The low contribution of MSA further suggests that aerosols over this region is likely to have negligible oceanic influences. This is also consistent with the back trajectory results in section 3.2, which demonstrate that* PM_1 *levels at this site are dominantly influenced by continental sources in the west and southwest.*"

RC3-45: Line 219: Using the term 3 types of SOA is tricky in this context and strongly depends on the definition of SOA that the authors use. Is POA that gets oxidised in the particle phase still POA or SOA? This issue can be easily avoided by speaking of 3 types of OOA.

Response: Thank the referee for pointing this out. We have revised "SOA" to "OOA" as suggested.

RC3-46: Fig 6 right panels: Comparting individual AMS ions to the timeseries of factors only shows how well a factor could be represented by that ion. Or in other words if that ion is mostly represented by that factor. C2H3O+ looks good because most of it is in Factor LO_OOA. CO2+ looks worse because that ion is split between MO-OOA and LO-OOA. For me, there is no scientific value in this correlation. Instead, the authors should check if there are correlations with other trace gases or parameters. E.g. NOx, CO, CO2 etc.)

Response: We agree that external tracers are needed instead of PMF inputs. We thus used BC and nitrate for comparison with PMF factors. Results showed that LO-OOA correlated well with NO_3 ($r^2 = 0.74$), confirming their secondary nature. Meanwhile, MO-OOA exhibited a moderate correlation with BC ($r^2 = 0.52$), indicating that they were likely coated on BC particles and underwent extensive aging processes during long-range transport.

Graph modification:

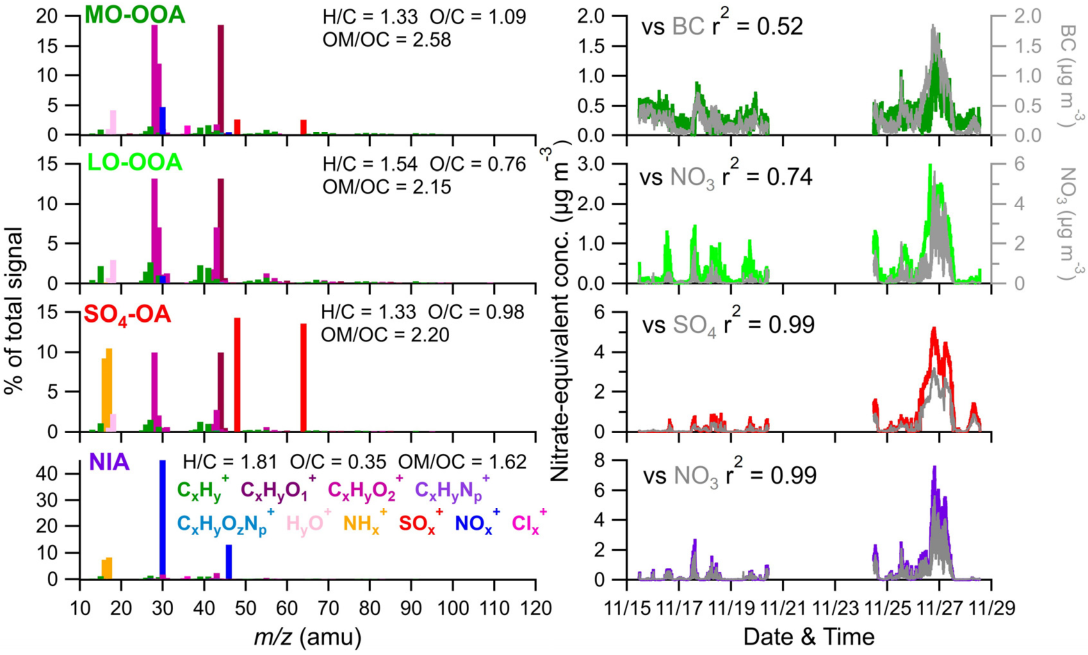

Figure 7. High-resolution mass spectral profiles (left) and time series (right) of four factors. The correlations of four factors with corresponding tracers are also shown.

RC3-47: Line 250ff. I find their conclusions about ON vs inorganic NO3 confusing. The ON concentration is clearly much higher during the peak in P2 (the “non-cloudy” period). The only reason the relative contribution is lower is because the inorganic NO3 is increasing orders of magnitude at the same time. But it seems that the authors are more focused on the relative contribution pointing to a reference (Huang et al) that suggests higher ON formation at higher RH (so during P1 in their interpretation). But this does not apply because most of P2 is also >70%RH and if that would matter, P1 should show and increase in the concentration of ON not just the relative contribution.

Response: Thank the referee for pointing out this problem. We agree that our previous speculation is quite weak and we have deleted this sentence.

RC3-48: Instead of the RH dependence, one could speculate that the higher inorganic NO3 contributed to more formation of ON during the aging (most likely in the aqueous phase) or that high gas phase NOx concentrations close to the source region led to direct gas phase reaction during nighttime (NO3 radical chemistry).

Response: We thank the referee for the valuable suggestions. We agree with the referee that besides cloud evaporation, the formation in both aqueous and gas phases could also contribute to ON. These discussions have been added to the revised manuscript. Future studies are absolutely needed for further investigations.

Text modification: Lines 328-332: “*The average mass concentration of ONs in C1* *was 30 ng m^{-3}, which was lower than that in EP5 (40 ng m^{-3}). The observation can also be explained by the evaporative release of ON 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).*"

References:

Ayres, B. R., Allen, H. M., Draper, D. C., Brown, S. S., Wild, R. J., Jimenez, J. L., Day, D. A., Campuzano-Jost, P., Hu, W., de Gouw, J., Koss, A., Cohen, R. C., Duffey, K. C., Romer, P., Baumann, K., Edferton, E., Takahama, S., Thornton, J. A., Lee, B. H., Lopez-Hilfiker, F. D., Mohr, C., Wennberg, P. O., Nguyen, T. B., Teng, A., Goldstein, A. H., Olson, K., and Fry, J. L.: Organic nitrate aerosol formation via NO3 + biogenic volatile organic compounds in the southeastern United States. Atmos. Chem. Phys., 15(23), 13377-92, https://doi.org/10.5194/acp-15-13377-2015, 2015.

Xian, J., Cui, S., Chen, X., Wang, J., Xiong, Y., Gu, C., Wang, Y., Zhang, Y. J., Li, H. W., Wang, J. F., and Ge, X. L.: Online chemical characterization of atmospheric fine secondary aerosols and organic nitrates in summer Nanjing, China. Atmos. Res., 290, 106783, https://doi.org/10.1016/j.atmosres.2023.106783, 2023.

RC3-49: Line 254f: Why do the authors bring the ON values from Hyytiälä? Is there site similar to a boreal forest? I did not expect that that far south at only 1100m asl.

Response: We thank the referee's comments. The sampling site we brought here to compare is located on the top of a tower (224 m height) in Kuopio, Finland (not in Hyytiälä), where aerosol particles are influenced by both the local forest and the nearby urban center. Likewise, aerosol particles at the SH site are also affected by local biogenic emissions and transport anthropogenic emissions. Therefore, we believe that this comparison of ON values between these two sites is reasonable, and could also suggest that AMS has the ability to quantify ON at such low levels.

Text modification: Lines 332-334: "*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.*"

RC3-50: Line 264: the right reference for the CO method should be (Decarlo et al., 2010).

Response: Cited as suggested.

RC3-51: Line 273: "negative correlation between OA/ΔCO and O/C in P2 also implies that more oxidized OA had almost been scavenged". This statement does not fit the data. A relative contribution of compound X can change by two ways: A) the concentration of compound X decreases or B) X stays constant and all other compounds increase. The P2 period as 10 more OA than P1. Even if none of the general background OA that is present in P1 is removed, the fresh OA will still dominate the overall properties. This indicates that the main mechanism of increasing OA mass is a source of fresh OA that does not correlate with CO emissions for this specific period. E.g., fresh SOA from biogenic VOCs can have low O/C ratios also anthropogenic VOC emissions are not necessarily correlated with CO.

It can also be that a different POA source is encountered along the trajectory. E.g., biomass burning POA can have a huge range of OA/ΔCO values (50-200 ug m^-3 ppm^-1, (Decarlo et al., 2010)). Such a source between the original source region and the station would through off the original correlation as is stated in (Decarlo et al., 2010).

Response: Thank the referee for the valuable comments. We agree that the increasing OA mass did not correlate with CO emissions in this period. While we appreciate the alternative explanations that the referee provided, we believe that our original interpretation remains consistent with our dataset and the broader context of our study. Our stance is based on the following considerations:

1. Our revised manuscript has presented clear evidence of cloud evaporation in Episode 5 (please see **RC3-3**), which we identify as the main cause of elevated PM_1 levels in this period.
2. The negative correlation between OA/ΔCO and O/C thus indicates that the OA released into PM_1 primarily consisted of less oxidized OA. This release of OA is a process independent of CO emissions, which aligns with the comment "the main mechanism of increasing OA mass is a source of fresh OA".
3. In contrast, the low OA/ΔCO values at high O/C suggest a significantly reduced production of more oxidized OA compared with less oxidized OA. Therefore, this result demonstrates that more oxidized OA had almost been scavenged by clouds, which cannot be released after evaporation.

Indeed, after the cloud was evaporated, fresh SOA formation from biogenic VOCs can then be strengthened by stronger solar deviation, which may also contribute to the increase of less oxidized OA. Considering the referee's suggestions, we have added this possible reason to the manuscript.

We did not investigate the POA source because such analyses need the involvement of emission inventory in China and modeling work, which is beyond the scope of this study. However, through case studies (see **RC3-1**), we have evaluated the negligible influence of pollution source change in EP5.

Text modification: Lines 351-357: "*Moreover, considering the cloud evaporation process in this period, 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 undergone mass reduction through fragmentation reactions (Lee et al., 2012), which was far less likely to be reintroduced into the atmosphere.*"

RC3-52: Line 277f: The conclusion that those "released" compounds must be HULIS is completely baseless as there is no reason to suggested that there is indeed increased aqueous phase production of WSOC which would then partially be of HULIS type.

Response: We are very sorry for our incorrect hypothesis. This sentence has been deleted following the suggestion.

RC3-53: Line 285: where is the other mountain site data? I see the coloured symbols for the factors and the grey 1h averages of the data for this study.

Response: We are very sorry for our incorrect introduction of this figure. We have changed this sentence to "*Figure 11a shows the Van Krevelen diagrams of OA in this study.*".

RC3-54: Line 290 and Fig 10: the alinement of the two factors with the total trend line is indeed not a surprise. Most of the OA is expressed by these two factors which were derived from the data itself by PMF. It is then a logical consequence that the H/C and OC ratios of the factors will be in the range of the original data and that if a essential 2 factor linear combination is the case, the characteristics of those two factors will be at the extreme of the data range (so lowest OC&highest HC for one and revers for the other). This is caused by the underlying math of PMF.

Response: Considering the referee's suggestion, we have deleted the related sentences in the text. Instead, a simple comparison of the slope in our study with other remote/rural sites across the world has been added to the manuscript.

Text modification: Lines 364-366: "*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.*"

RC3-55: Line 292: what is meant by "overlap with the aged OAs observed at other sites"?

Response: We are sorry for the confusion caused by this sentence. The original meaning of this sentence is that LO-OOA and MO-OOA measured in this study have a similar distribution in the f_{43} vs. f_{44} graph with other elevated sites. To avoid misunderstanding, we have deleted this sentence.

RC3-56: Line 294: "These results together reveal…" A simple comparison of the factor mass spectra (or the total mass spectra) with reference data from other site would have revealed that much easier and more clearly.

Response: We thank referee for the above suggestions. A comparison of the factor mass spectra with data from other two mountain sites has been added to the supplementary (Fig. S13).

Graph modification:

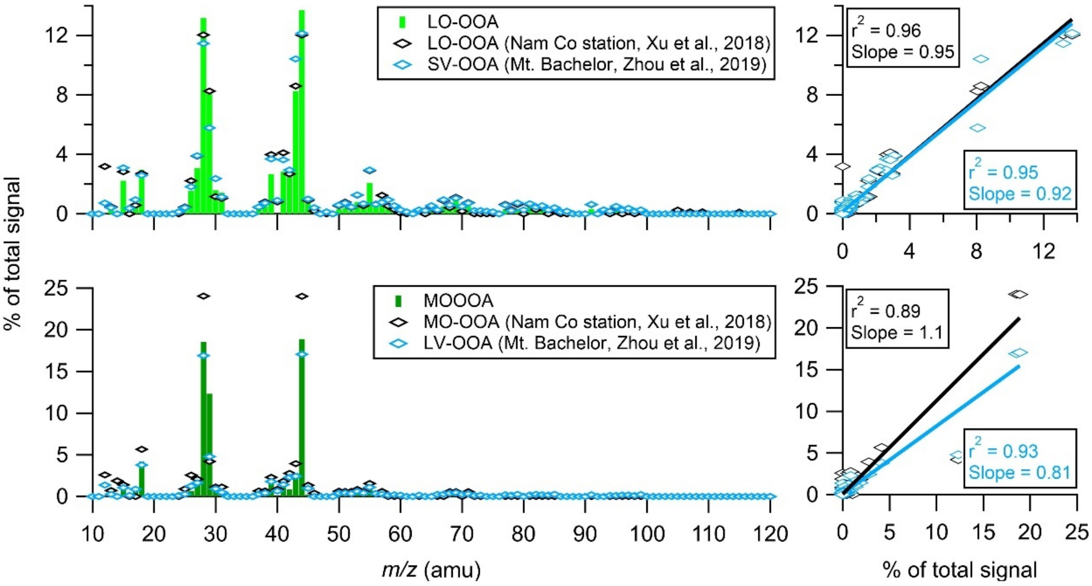

Figure S13. Comparison of the mass spectra of OA in this study with other two mountain station. The correlation of the mass spectra of these OA factors are also shown.

References:

Xu, J., Zhang, Q., Shi, J., Ge, X., Xie, C., Wang, J., Kang, S., Zhang, R., and Wang, Y.: Chemical characteristics of submicron particles at the central Tibetan Plateau: insights from aerosol mass spectrometry, Atmos. Chem. Phys., 18, https://doi.org/10.5194/acp-18-427-2018, 2018.

Zhou, S., Collier, S., Jaffe, D. A., and Zhang, Q.: Free tropospheric aerosols at the Mt. Bachelor

Observatory: more oxidized and higher sulfate content compared to boundary layer aerosols, Atmos. Chem. Phys., 19, 1571-1585, https://doi.org/10.5194/acp-19-1571-2019, 2019.

RC3-57: Line 301: what clustering mechanism was used? Where the trajectories treated as time series of data pairs? How tight were the clusters? The authors need to show the range of trajectories in each cluster (e.g., mapping all trajectories in a cluster and indicating the mean.

Response: Thank the referee for pointing this out. Since we rewrite the back trajectory analysis section (see **RC3-1**), the information about cluster is thus no longer needed.

RC3-58: Everything suggests very long aging times for the general background aerosol during P1. Are 72h trajectories long enough to identify the source regions? Especially, Type C4 seems to be "short" and may need an extra day or two to get more differentiation.

Response: We thank the referee for the above comments. According to Xu et al. (2022), OA in urban ambient air can reach its highest oxygenation degree after approximately 2-3 days of photooxidation. Previous studies at mountain sites also used 72h as trajectory arrival time (Xu et al., 2018; Sun et al., 2024). In addition, C4 was replaced by individual trajectories, and a clear differentiation is observed from other trajectories. Taken together, we think 72h trajectories can be long enough to identify the source regions.

Reference:

Xu, W. Q., Sun, Y. L., Chen, C., Du, W., Han, T. T., Wang, Q. Q., Fu, P. Q., Wang, Z. F., Zhao, X. J., Zhou, L. B., Ji, D. S., Wang, P. C., and Worsnop, D. R.: Secondary organic aerosol formation and aging from ambient air in an oxidation flow reactor during wintertime in Beijing, China. Environ. Res., , 209, 112751, https://doi.org/10.1016/j.envres.2022.112751, 2022.

Xu, J. Z., Zhang, Q., Shi, J. S., Ge, X. L., Xie, C. H., Wang, J. F., Kang, S. C., Zhang, R. X., and Wang, Y. H.: Chemical characteristics of submicron particles at the central Tibetan Plateau: insights from aerosol mass spectrometry, Atmos. Chem. Phys., 18, 427-443, https://doi.org/10.5194/acp-18-427-2018, 2018.

Sun, Y., Zhu, Y., Qi, Y., Chen, L., Mu, J., Shan, Y., Yang, Y., Nie, Y., Liu, P., Cui, C., Zhang, J., Liu, M., Zhang, L., Wang, Y., Wang, X., Tang, M., Wang, W., Xue, L.: Measurement report: Atmospheric ice nuclei in the Changbai Mountains (2623 m a.s.l.) in northeastern Asia. Atmos Chem. Phys., 24(5), 3241-56, https://doi.org/10.5194/acp-24-3241-2024, 2024.

RC3-59: For readers not so familiar with this specific region of China, it will be helpful to provide some more general information about population and industry centres relevant for the trajectory groups. E.g. is C4 only over rural areas?

Response: Thank the referee for pointing this out. Actually, the usage of the organic carbon mass concentration reanalysis dataset in Fig. S5 is to help the readers know the pollution levels of regions. Also, some big and mega cities with large populations that these trajectories pass have been marked in Fig. S5.

RC3-60: Line 318f: "Remarkably, nitrate (20.9 %) and ammonium (17.0 %) exhibited unexpectedly high contributions to PM_1 in this forested mountain area" Compared to the other mountain sites I found the SO_4 signal remarkably LOW! Fig 1 has 50 – 66% of Inorg for all but Mt Yulong.

Response: We are really sorry for our negligence of the remarkably low SO_4 contribution in this site.

Text modification: Lines 380-384: "*Remarkably, sulfate exhibited lower contributions (16.7 %) to PM_1 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 in southeastern China.*"

RC3-61: Line 322: PMF does not identify two types of SOA. It identifies 2 OOA factors and some OA associated with SO_4.

Response: Revised as suggested.

RC3-62: If the authors still find aerosol cloud interactions after the reanalysis, they need to put that into the wider context. Who representative will those periods be for this region/location. How often are such conditions met in that season (November) and how likely are they for other times of the year? Is this site representative for the wider region?

Response: Thank the referee for the valuable suggestion. Following the referee's comments above, we have reassessed and reanalyzed our data, confirming the presence of aerosol-cloud interactions at this site. The aerosols transported from polluted regions need to first enter the cloud droplets and can then be released during cloud evaporation. This means the aerosol release may only occurred under cloudy conditions (RH = 100 %). Therefore, we consider EP4 and EP5 in the sampling period as cloud evaporation periods, i.e. 2 times in November. Since cloud is a common occurrence here, this process can also happen at other times of the year. Therefore, our study can provide valuable insights into understanding the aerosol-cloud interaction in mountainous areas that are often under cloudy conditions. Additionally, it can be considered as a plausible mechanism to explain certain aerosol episodes in these regions.

We also agree with the reviewer's several critical and important questions. Because the site is very new (built in one-year), the collocated measurements are still limited. This study presents the first measurements of aerosol composition using AMS/ACSM showing the importance of aerosol-cloud interactions. To address the reviewer's questions, highly time-resolved long-term aerosol composition measurements along with collocated measurements, such as liquid water content, size distributions of cloud droplets, volatile organic compounds, and etc. are needed in the future.

Text modification: Lines 391-393: "*Moreover, our study provides valuable data and insights into understanding the aerosol-cloud interaction 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.*"

Language comments:

RC3-63: Line 23: ratio of OA/delta CO. OA/ΔCO is already a ratio. Should be "value of OA/delta CO" or "ratio of OA to delta CO". Since the authors also use the term "O/C ratio" (which technically also contains the ratio aspect twice), they could use the term "OA/ΔCO ratio"

Response: Revised.

RC3-64: Line 33: "transported to the upper layer of atmospheric boundary layer or even lower free Troposphere" – layer of the atmospheric boundary layer

Response: We have changed "upper layer" to "upper level".

RC3-65: Line 40: Sentence in Line 40 (As a result…) points to previous sentence which has nothing to do with ACI.

Response: We have added an introduction of ACI before this sentence (see **RC3-14**).

RC3-66: Line 42: characteristics over regional scales

Response: Revised.

RC3-67: Line 41 What impacts are referred to here?

Response: We have added the specific impacts of ACI to this sentence.

RC3-68: Line 46 aerosol formation is there twice

Response: Deleted.

RC3-69: Line 51ff “Although…” this sentence is grammatically broken – rephrase

Response: Rephrased.

RC3-70: Line 51 “several studies over mountain sites”: probably at mountain sites. Or do they mean measurements literally above those mountain stations?

Response: We have changed “several studies over mountain sites” to “several mountain stations have been established in China”.

RC3-71: Line 52: Is there a comma missing after sources? That would provide some meaning to the sentence.

Response: We have deleted “several studies over mountain sites have been conducted to characterize chemical composition and sources”.

RC3-72: Line 53 “stations have been settled “ - what do they mean? Stations were established?

Response: We have changed the word “settled” to “established”.

RC3-73: Line 54f: “most of them are “

Response: Changed it to “most of these studies”.

RC3-74: Line 55: “mainly conducted on Qinghai-Tibetan Plateau” – the verb refers back to “stations as a subject.

Response: Revised.

RC3-75: Studies can be conducted. Stations cannot be conducted.

Response: Revised.

RC3-76: Line 55f: “while the studies in southeastern China is very limited. “ First, it would be “studies are limited” second this is bad language. With that correction this would mean that there are studies, but their scope, quality whatever is limited. I guess they want to express that there is a limited number of studies available?

Response: Revised to “remain scarce”.

RC3-77: Line 63: “backward trajectory analysis” – I found back trajectory analysis to be the more common term. The documentation on HYSPLIT uses back trajectory

Response: Revised.

RC3-78: Line 63: “particle organic nitrates” – particulate organic nitrate or particle-phase organic nitrates

Response: Revised.

RC3-79: Line 75 “AMS operated under the “V-mode””: operated in V-mode. Also, v-mode is an established technical term and does not need quotation marks.

Response: Deleted.

RC3-80: Line 91: It not clear that HRMS is only referring to data from the AMS. Yes, many people know that ACSM does not provide HR data but the authors cannot assume that.

Response: Revised.

RC3-81: Line 117 “Trajectory time” : trajectory arrival time

Response: Revised.

RC3-82: Line 118 “height”: arrival height at the site

Response: Revised.

RC3-83: Line 126: “organics held the largest contribution” – bad language rephrase

Response: Revised to “organics accounted for the largest contribution”

RC3-84: Line 128: this comparison is only with other Chinese mountain sites. Not mountain site in general

Response: Revised.

RC3-85: Line 266: “studing period” – I guess they mean studying period. But even that is incorrect. In this context, it should be either “study period” or “studied period”.

Response: Revised.

RC3-86: Figure 1: The percentage values are a bit difficult to read for the Nitrate (black on dark blue) consider writing it outside of the pie chart for Mt Tai and Damaojin. Also, the Nitrate percentage value is missing for Mt Daban.

Response: Revised.

RC3-87: Line 163f: it is not clear if “this site” phrase refers to the sit in this study or the site in Chen et al ?

Response: Since we have rewritten this paragraph, this comment is no longer relevant.

RC3-88: Line 165: “AMS lens” -> Fig S4 shows ACSM data hence this must be ACSM lens in this sentence?

Response: Since we have rewritten this paragraph, this comment is no longer relevant.

RC3-89: Line 233: Mt. Bachelor ????? I guess one of the mountain names got accidentally translated from Chinese here?

Response: We re-checked the name of this mountain, and it is indeed named "Mt. Bachelor" (Zhou et al., 2019). It is located in central Oregon, USA.

References:

Zhou, S., Collier, S., Jaffe, D. A., and Zhang, Q.: Free tropospheric aerosols at the Mt. Bachelor Observatory: more oxidized and higher sulfate content compared to boundary layer aerosols, Atmos. Chem. Phys., 19, 1571-1585, https://doi.org/10.5194/acp-19-1571-2019, 2019.

RC3-90: Line 240: "Not surprise, no primary OA factor" (e.g., hydrocarbon-like OA, biomass burning OA, etc.)

Response: We have revised it to "As expected".

RC3-91: Line 243: "A nitrate aerosol factor was also separated from these OA factors, " I assume that is the NIA factor? SO inorganic nitrate factor?

Response: Revised.

RC3-92: Line 244: "Despite NIA, NOx+ ions were more assigned in MO-OOA": The presence of the NIA factor is not causal for more NOx in MO than in LO-OOA.

Response: We have changed the word "Despite" to "Apart from", and a discussion about more NO_x in MO-OOA than in LO-OOA has been added to the revised manuscript.

Text modification: Lines 322-326: "*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 possible reason is 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.*"

RC3-93: Line 255: "significant discrepancies" – differences not discrepancies

Response: Revised.

RC3-94: Line 270: "trended to increase " The English word "trend" does not work that way – rephrase

Response: Revised to "tended".