We thank the two 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 revised the manuscript accordingly (in *italic text*).

Response to Referee #1

Specific comments:

RC1-1: Line 107: add the full form to be consistent with other parameters.

Responds: Added.

RC1-2: Line 138: abbreviation seems odd, INA may be better?

Responds: We thank the referee for this suggestion. We have replaced the abbreviation "NIA" with "INA" in the revised manuscript.

RC1-3: Line 164: This value seems a bit different from the one mentioned in the abstract, please check and rectify.

Response: Checked and corrected.

RC1-4: Line 168: But avg BC contribution is 10%, rephrase the sentence.

Response: Thank the referee for pointing this out. We have changed the statement "largely influenced" to "potential influences" in this sentence.

Line 156: "In addition, BC was observed to correlate well with PM$_1$ and PM$_{2.5}$ (r^2 = 0.59 and 0.62, respectively), suggesting the potential influences of regional biomass burning plumes in the Yangtze River Delta on aerosol particles at this site (Zhang et al., 2015)."

RC1-5: Line 174: I don't think it's much lower, values are pretty much within uncertainty limits.

Response: We agree with the referee that the average PM$_1$ concentration at our site is not much lower than the values at Mt. Yulong and Mt. Waliguan. Therefore, we have revised it to "slightly lower".

Line 159: "For example, the average PM$_1$ concentration at SH site is much lower

compared to Mt. Wuzhi (10.9 ± 7.8 µg m^{-3}) at a similar altitude, and is also slightly lower than those at higher altitude mountains such as Mt. Yulong (5.7 ± 5.4 µg m^{-3}) and Mt. Waliguan (9.1 ± 5.3 µg m^{-3})."

RC1-6: Line 180: 16.7% is not notably lower than 14%, authors need to check such instances of erroneous conclusions and correct them.

Response: Thank the referee for pointing this out. We have carefully rechecked and corrected the relevant text.

Line 166: "Notably, a relatively lower contribution of sulfate to PM$_1$ was observed (16.7 %) in this site compared to other mountain sites except Mt. Yulong (23.7 %−38.1 %), which was likely attributed to the significant reduction of SO$_2$ emission in China during the past decade (Wen et al., 2023)."

Line 171: "Additionally, at Mt. Tai, even though the contributions of sulfate were high, nitrate still accounted for considerable fractions (14.9 %−29.0 %) that were comparable to or higher than this site (14.7 %)."

RC1-7: Line 189: reference?

Response: A reference that supports our statement has been added.

Line 172: "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 (Li et al., 2018)."

RC1-8: Figure 3: How about adding one for EP1?

Response: Added as suggested.
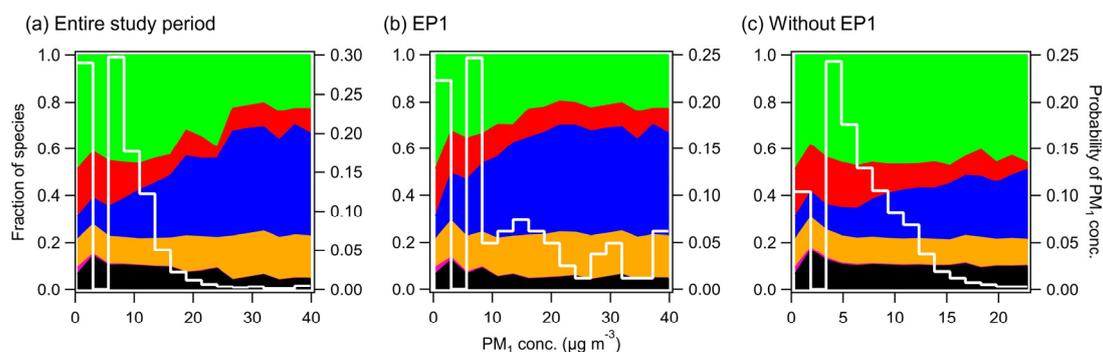
Figure S3. Variations in aerosol composition as a function of PM$_1$ mass concentration

and the probability density of PM$_1$ during (a) the entire study period, (b) EP1, and (c) the period excluding EP1.

RC1-9: Line 223: I guess BLH values are easily obtainable from different satellite/re-analysis data products now, authors should use them and quantify the values here.

Response: Thank the referee's comments. In the revised manuscript, we have obtained the hourly BLH data in November 2022 from the fifth generation ECMWF atmospheric reanalysis of the global climate (ERA5) datasets. As shown in Figure S6, the noon peak of BLH (up to ~1450 m at around 13:00) was consistent with the peaks of organic and nitrate concentrations. The following texts have been added to the revised manuscript as suggested.

Line 93: "In addition, the particle depolarization ratio (PDR) was measured by a particle monitoring lidar (Science of Light Technologies Co., Ltd) to analyze the vertical distribution of aerosol particles, and the hourly boundary layer height (BLH) was obtained from ERA5 reanalysis dataset."

Line 192: "In addition, as depicted in Fig. S6, these noon peaks coincided closely with the diurnal variation in BLH (reached up to ~1450 m at around 13:00). Consequently, the relatively elevated concentrations of organics and nitrate at noon might also be related to the development of atmospheric boundary layer, which facilitates the mixing of air masses transported from polluted areas."
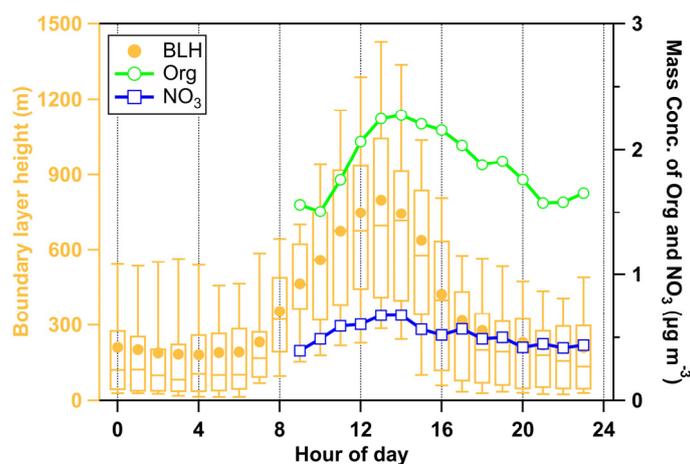
Figure S6. Diurnal variations of boundary layer height along with mass concentrations of organics and nitrate. Data of organics and nitrate before 9:00 was omitted for ease of comparison.

RC1-10: Section 3.2: This section should come before/after fig.2. Authors also needs to explain better on what basis they are defining clean/low/high PM periods. What's the PM cutoff values? and why?

Response: We thank the referee for their suggestions. We have moved Figure 3 to the supplement, so this section now follows Figure 2. High PM_1 periods were defined as times when average PM_1 concentrations exceeded the 75th percentile of the overall PM_1 concentration distribution, while low PM1 periods were defined as times when average PM_1 concentrations fell below the 25th percentile of the distribution

Line 206: "These five elevated PM_1 episodes exhibited mean PM_1 concentrations ranging from 5.2−9.8 μg m^{-3}, which were close to or exceeded the 75th percentile of the overall PM_1 concentration distribution (5.6 μg m^{-3}). Meanwhile, a clean period (C1) with a mean PM_1 concentration of 1.3 μg m^{-3}, falling below the 25th percentile of the distribution (1.6 μg m^{-3}) was also selected for comparison."

RC1-11: Figure 4: RH marking is not clearly visible.

Response: We have revised this figure.
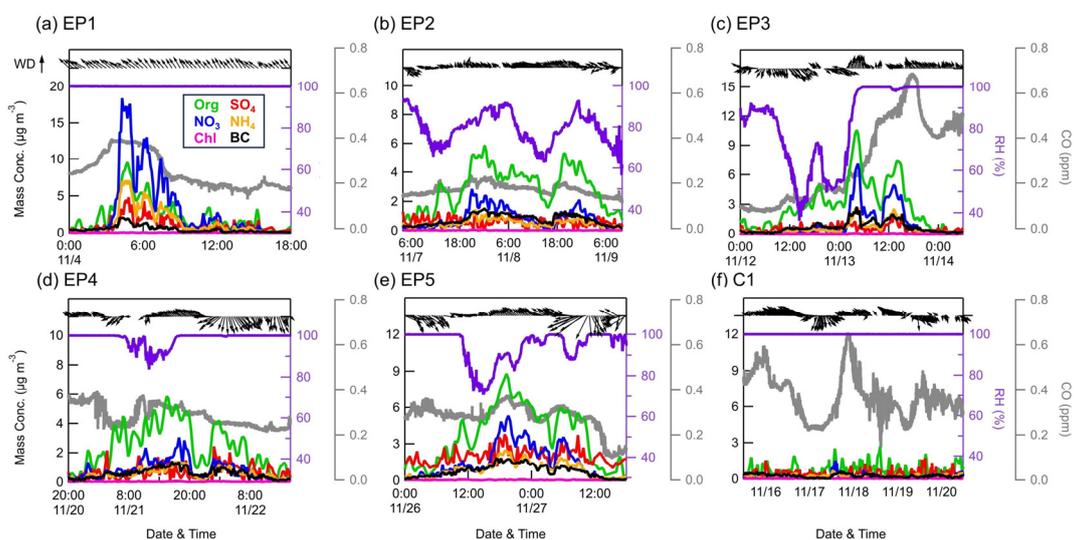
Figure 3. Temporal variations of the concentrations of PM_1 species measured by ACSM, as well as CO (grey lines), RH (purple lines), and WD/WS (black arrows) during six events.

RC1-12: Figure 4: explain the colors.

4

Response: An explanation on the colors in this figure has been added (please see RC1-11 for detailed revision).

RC1-13: Line 289: concentrations.

Response: Revised.

RC1-14: Line 293: What new insights AMS could provide over ACSM, please explain that briefly here.

Response: A brief explanation of has been added.

Line 288: "To further explore the impacts of cloud scavenging and cloud evaporation on aerosol characteristics, we selected C1 and EP5, during which NR-PM$_1$ was also measured by AMS, for additional investigation on aerosol size distributions, elemental ratios, and HRMS in this section."

RC1-15: Figure 6: are the difference statistically significant?

Response: Thank the referee for pointing this out. Following the referee's comment, we conducted t-tests to determine whether the differences are statistically significant. The p-values of OM/OC, OC, and OSc during C1 and EP5 (1.22277E-08, 2.83308E-09, and 3.76611E-08) are much lower than α=0.05, indicating that these differences are indeed statistically significant.

RC1-16: Figure 7: MO-OOA with BC? a highly oxidized SOA vs a primary emission marker? how does this comparison help?

Response: We appreciate the referee's comment. Previous AMS measurements have consistently shown that MO-OOA correlates well with sulfate due to their similar volatile nature. However, we found that the correlation between MO-OOA and sulfate at this site was not significant (r² = 0.32). Instead, we observed a significant correlation between MO-OOA and BC, suggesting that MO-OOA precursors and BC may originate from the same air masses and transport together to this site. Zhu et al. (2016) also reported a strong correlation between LV-OOA and BC, but a weak correlation with sulfate at a mountain site in southern China (Figure R1-1). Their findings indicate that

OOA precursors likely share the same sources with BC, such as biomass burning and ship emissions. Given the significant role of biomass burning in aerosol composition in southeastern China (Fang et al., 2023; Liang et al., 2020), the most plausible explanation is that organics coated or mixed with BC, emitted from biomass burning, undergo aging processes to form MO-OOA during transport. This finding provides new insights into the chemical composition and mixing state of aerosols in background regions. Furthermore, this could explain why ONs are more associated with MO-OOA than LO-OOA (please see RC1-18). Future measurements will utilize Soot-Particle AMS (SP-AMS) to further investigate this relationship.

To clarify this, we have added the above discussion to the revised manuscript.

Line 355: "However, unlike numerous previous studies, there was a limited correlation between MO-OOA and sulfate ($r^2 = 0.32$) during the sampling period. Instead, MO-OOA exhibited a moderate correlation with BC ($r^2 = 0.52$). This correlation between OOA and BC was also observed at a mountain site in southern China (Zhu et al., 2016), suggesting common air masses for MO-OOA and BC, such as biomass burning. Given that biomass burning plays an important role in aerosol composition in southeastern China (Fang et al., 2023; Liang et al., 2020), it is plausible that MO-OOA may have coated or mixed with BC particles emitted from biomass burning, undergoing extensive aging processes during long-range transport."
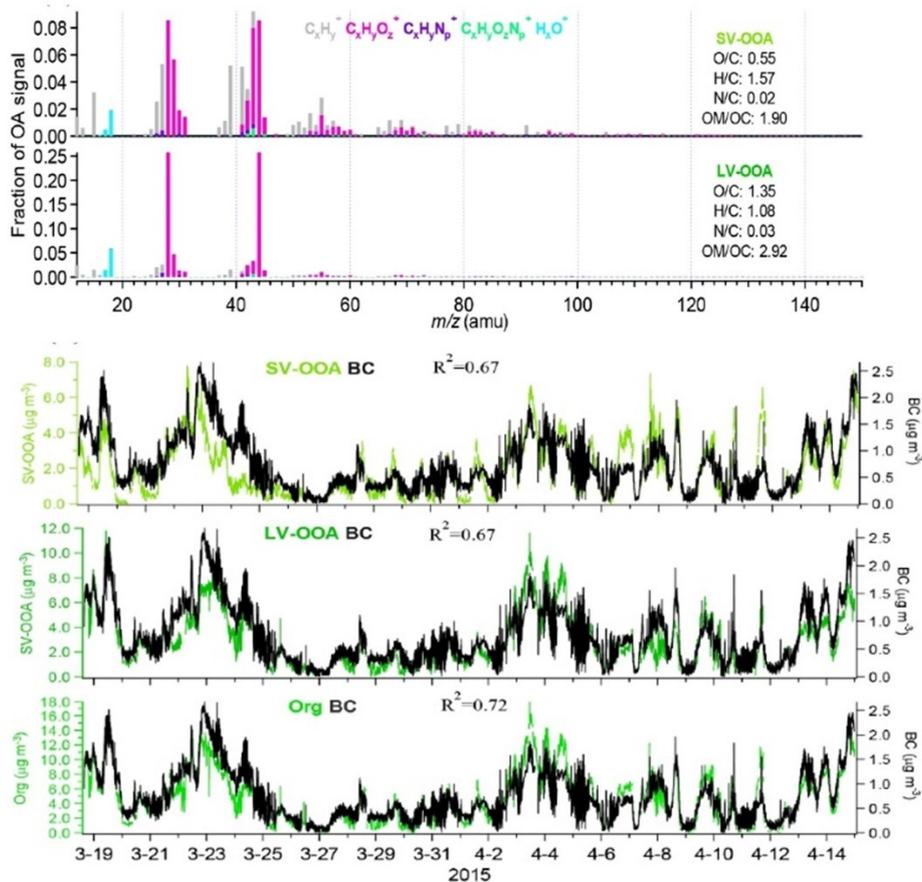
Figure R1-1. Profiles and time series of OA components and their correlations with BC at a mountain site in southern China (modified from Zhu et al., 2016).

RC1-17: Figure 7: Weren't NO3 and SO4 are part of inputs to PMF?

Response: We apologize for our oversight. It is evident that almost all nitrate and sulfate ions were assigned to INA and SO$_4$-OA, respectively. Therefore, there is no need to examine the correlation of INA and SO$_4$-OA with external factors. In the revised manuscript, we have removed the time series of nitrate and sulfate from this figure..

RC1-18: Line 383: Is BC coating happening here? what's the proof/indications?

Response: Thank the referee's comment. As we mentioned in RC1-16, the significant correlation of MO-OOA with BC may be the most likely reason here. Therefore, we consider that BC may be coated during the aging processes of transport. To clarify this, we have modified this sentence.

Line 372: "Since MO-OOA also correlated considerably with BC, one possible reason for this was that ONs formed and mixed with MO-OOA components during the aqueous

aging processes of MO-OOA-coated BC particles."

RC1-19: Fig. 10: which one is the dependent variable? I think its O/C.

Response: We agree with the referee that O/C is the dependent variable. Therefore, we have swapped the x-axis and y-axis of this figure.
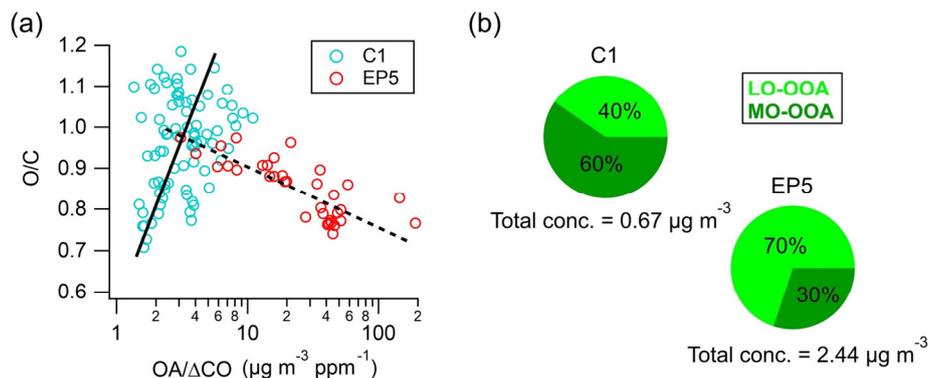
Figure 9. (a) Scatter plot of O/C as a function of OA/ΔCO values and (b) mass concentrations and proportions of MO-OOA and LO-OOA during C1 and EP5.

RC1-20: Line 423: Please substantiate the claim about cloud evaporation period.

Response: Thank the referee for pointing this out. In section 3.2, we have already provided evidence supporting the occurrence of cloud evaporation during EP5, as demonstrated through case studies and particle lidar data analysis. We have clarified it in this sentence.

Line 404: "Moreover, considering the cloud evaporation process in this period (as discussed in Sect. 3.2), the negative correlation between OA/ΔCO and O/C in EP5 also implies that the less oxidized OA previously formed or incorporated into cloud droplets can be released during cloud evaporation."

RC1-21: Line 428: The statement is unclear to me.

Response: We apologize for the confusion brought to the referee. Kroll et al. (2009) and Lambe et al. (2012) found a transition of functionalization to fragmentation oxidation process during aerosol aging. Briefly, functionalization reactions at lower SOA ages produce low volatility VOC that are able to condense to form SOA. However, further oxidation leads to fragmentation reactions, breaking carbon-carbon bonds in

particle-phase organic compounds. This process generates small molecules and high volatility VOCs that evaporate from particles, resulting in SOA mass loss. Lee et al. (2012) further found that this oxidation pathway could also happen in cloud water through aqueous oxidation, especially for OA with O/C > 0.6. Therefore, we hypothesize that MO-OOA in cloud droplets has undergone fragmentation reactions that significantly reduced its mass before cloud evaporation. This may elucidate why cloud evaporation tends to release less oxidized OA rather than more oxidized OA observed in our study. In response to the referee's comment, we have rephrased this sentence in the revised manuscript.

Line 409: "Conversely, more oxidized OA in cloud droplets may have further undergone fragmentation reactions that decrease the carbon content of particles, resulting in their mass loss (Kroll et al., 2009; Lee et al., 2012). This made MO-OOA much less likely to be reintroduced into the atmosphere through cloud evaporation."

RC1-22: In ambient, this could be due to different relative contributions of various types of OAs. I don't think such strong mechanistic statement could be made only based on slope under ambient conditions.

Response: We thank the referee's comment. It is true that ambient OA is a complex mixture of OA from different sources and processing, and interpreting the oxidation pathway based only on slopes of H:C vs. O:C may not be rigorous. The different types of OAs transported to this site may also alter the H:C and O:C values. However, this slope may still reflect that these OA have experienced the corresponding reaction pathway during atmospheric dilution and evolution before arriving at this site. For example, Ng et al. (2011) and Chen et al. (2015) have examined these slopes for numerous ambient and laboratory data. They both found that ambient data generally follows a trajectory in the Van Krevelen diagram toward higher oxidation states, and the fits of ambient data can provide simplifications and insights for OA evolution. In addition, the significant correlation coefficient of this fitted line ($r^2 = 0.62$) in this study also supports our statement. In response to the referee, we have clarified this in the revised manuscript.

Line 413: "The slope of H/C to O/C in the present study was -0.66 (r^2 = 0.64). Despite potential variations in the relative contributions of different types of OAs transported to this site, this slope still suggests the addition of carboxyl functional groups during OA evolution to some extent (Heald et al., 2010)."

Response to Referee #2

General comments:

RC2: The authors present aerosol composition measurements at a background mountain site in southeastern China. They report clear evidence of aerosol cloud interactions – namely that the dissipation of clouds around/above the station led to an increase in aerosol mass concentration and changes the particle composition.

My main concern with the original manuscript was that first other factors impacting the aerosol concentration and composition at the site had to be excluded before interpreting the observed changes with respect to aerosol cloud interactions. In the revised manuscript, the authors divided their data set into more individual events and provide a much more detailed analysis of the air mass origin for each of these events. With this and other supporting measurements, they can now show that for some of the identified events the changes in the aerosol are indeed most likely affected by cloud processes. This is a very valuable finding and may provide the basis for more detailed studies at this very interesting station.

While most of my questions and suggestions have been addressed, there are still a few minor points in the revised manuscript that need to be clarified. After these minor revisions, I recommend this manuscript for publication.

Response: We gratefully appreciate the referee for reading our manuscript carefully and giving the positive comments and valuable suggestions. Our point-by-point responses are listed below.

Minor comments:

RC2-1: The new information of the PDR shows that for EP5 the station was not in-cloud but rather that there was a low-level cloud above the station (1000 m above sample site) and RH ~100% at the sample inlet. This is different for the C1 case where the station was inside a persistent fog/cloud layer according to the PDR data. This means that during EP5, the instruments were sampling an aerosol population below a cloud and not in a cloud. This should be clearly stated when the PDR information is mentioned.

However, this observation may make things even more interesting. How low does a cloud need to be to have the scavenging/evaporation effect? Or is it the RH=100% below that cloud that creates the relevant conditions? Is the evaporation/release of aerosol material the same for an inside cloud/fog situation and the low-level cloud above case? I do understand that these questions are beyond the scope of this work and will need further long-term measurements at this (and other similar) locations. These are just things that occurred to me after thinking about the PDR data.

Response: We thank the referee's comments. It is true that we did not consider the possible height dependence of cloud evaporation. Tao et al. (2018) found that cloud evaporation could happen at 500−1000 m above the ground and transport the released particles to the ground through turbulent mixing. This is consistent with our study that the cloud evaporated at 1000 m above this site. In contrast, during C1, this site was inside the cloud layer with no evaporation occurring. Figure S12 presented the boundary layer heights (BLH) during the sampling period. The persistent cloud coverage in C1 blocked the solar radiation to the ground, thereby hindering the development of boundary layer. However, during EP5, as the cloud evaporated, the BLH also increased to ~1500 m, the vertical mixing processes within the boundary layer would thus transport the released particles upward and downward. Therefore, we hypothesize that the determining factor of cloud evaporation is solar radiation rather than cloud height. If once the solar radiation and other environmental conditions are sufficient to evaporate the cloud at this site, the boundary layer would also develop correspondingly, enhancing the vertical mixing of released aerosols during daytime.

In response to the referee, we have added a detailed discussion to the revised manuscript. *Line 255: "However, there were several observations and questions raised that cannot be conclusively confirmed with the current data we have. First, it should be noted that there was a difference in cloud height between C1 (~0 km) and EP5 (~1 km, before evaporation), indicating that aerosol particles were sampled within and below the cloud during C1 and EP5, respectively. With no cloud evaporation occurring during C1 with low-level clouds, it raises the question of how evaporation processes affected by cloud height. Tao et al. (2018) found that cloud evaporation could occur at*

500−1000 m above the ground, and the released aerosols can be transported to low levels through strong turbulent mixing. This finding agrees with our observation at EP5, where the development of the boundary layer likely promoted the vertical transport of released aerosols to this site (Fig. S12). In contrast, during C1, the severe cloud layer hindered solar radiation, leading to generally lower boundary layer heights. These clouds might be identified as orographic clouds formed by the moist air masses lifted and saturated under the influence of complex mountain terrain (Houze, 2012). If once the solar radiation and other environmental conditions are sufficient to evaporate the cloud at this site, the boundary layer would also develop correspondingly, enhancing the vertical mixing of released aerosols during daytime. Therefore, we hypothesized that clouds with heights lower than the developed boundary layer heights (up to ~2 km, Fig. S12) may all have the probability of evaporating and increasing the observed aerosol mass at this site."
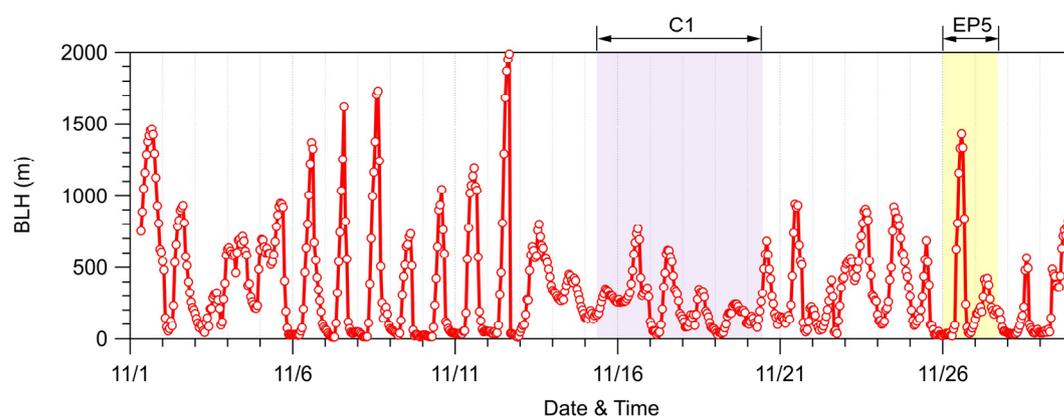
Figure S12. Time series of boundary layer height during the sampling period.

RC2-2: The arguments about the release of aerosol material from evaporating cloud droplets seems consistent. But I wonder how this all adds up. The authors point to a study which estimated that less than 20% of the total aerosol mass resides in fog droplets >2.5 um (Kuang et al. 2024), which sounds reasonable. When the cloud is there at the start of EP5, only the interstitial particles are detected – up to 20% of the aerosol mass is "hiding" in cloud droplets. When the cloud evaporates, the increase in aerosol mass for EP5 is much more than 20%. If the organic content in fog droplets found by

Kuang et al. is indeed representative for the present study as well, how can the evaporation of cloud/fog release so much more aerosol material?

Response: Thank the referee for pointing this out. After careful consideration, there might be several reasons that cloud evaporation released more aerosols during EP5:

1) The polluted air masses were transported to higher altitudes than this site and entered the cloud. In contrast, an RH of 100% near the sampling inlet, despite the lack of clouds, may result from the continuous uplift of clean and moist air from lower elevations. This condition is not conducive to particle accumulation. Therefore, the aerosol concentrations in the cloud above the site might be higher than those around the sampling inlet below.

2) New particle formation and activation of CCN may be facilitated at high altitudes, potentially increasing the aerosol concentration in the cloud (Sellegri et al., 2019).

3) The evaporation processes can also trigger some aqueous SOA formation processes such as oligomerization, thereby generating extra SOA in the cloud droplet (Lim et al., 2010; Brégonzio-Rozier et al., 2016).

Taken together, the cloud above this site during EP5 may contain more aerosols than the sampling inlet below, which could release much more material after evaporation. However, we cannot precisely identify the sources for these additional aerosol particles based on our current data, we thus seek for the referee and editor's tolerance. The above discussions have been added to the revised manuscript, and we thank the referee again for bringing these points to our attention.

Line 268: "Second, as discussed in Sect. 2.1, the sampling uncertainties caused by cloud droplets accounted for up to 20% in our study, yet significantly more aerosol particles were observed to release after the evaporation of the cloud above this site during EP5. This was likely attributed to the polluted air mass transported and entered the cloud at higher altitudes. In contrast, at the altitude near the sampling inlet, the continuous uplift of clean moist air masses from lower elevations might be unfavorable for aerosol accumulation. Moreover, the evaporation itself could also trigger some aqueous SOA formation processes such as oligomerization (Lim et al., 2010; Brégonzio-Rozier et al., 2016), thereby contributing additional aerosol mass within the

cloud droplets."

RC2-3: Section 3.2: The description and classification of the different episodes is good. I interpret the information in this section in the way that only for EP2, EP4, and EP5 the effect of air mass or source changes can be excluded for the interpretation of the impact of RH/cloudiness on the aerosol composition and concentration. (EP1 is the nitrate case and EP3 has an air mass change coinciding with and RH change). Are both EP1 and EP3 excluded from the further analysis? E.g., is EP1 and EP3 data included in Fig S7.

Response: Thank the referee for pointing this out. We have excluded EP1 and EP3 from Fig. S7 (now is Fig. 9), and the results are consistent with our previous results.
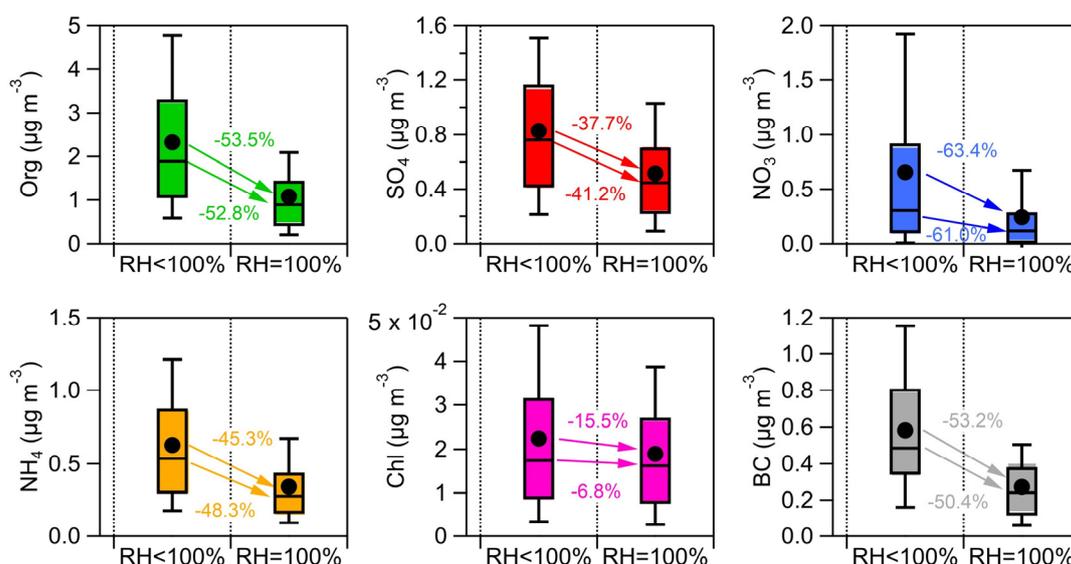
Figure S9. Comparison of mean and median mass concentrations of PM_1 species under conditions of RH below and at 100 % over the campaign without EP1 and EP3.

RC2-4: The authors show that for airmasses with (anthropogenic) emissions from cities (EP2, 4, 5) the cloud/fog evaporation mechanism is important. Unfortunately, the only clean period C1 is at the same time a true in cloud/fog period. Were there no times that could be classified as "clean" but had RH<100%? I understand that the station is new, and the data set is limited. But this would be an important aspect to check in the future: is there also a "release" of aerosol material if a clean airmass cloud/fog evaporates?

Response: Thank the referee's comments. Due to the lack of PDR data, we cannot find a clean period with cloud evaporation. However, we noticed a period (from 10 Nov to 12 Nov) that seems to be a clean and RH < 100 % period with relatively low CO and particle concentrations. In addition, this period also contains ~12 hours with RH = 100 %, indicating the possible existence of cloud evaporation processes. The back trajectory analysis showed that the air masses originated from the sea, which might be less influenced by anthropogenic emissions (Fig. S13). Correspondingly, the aerosol concentrations at this site only showed slight elevation. Therefore, we hypothesize that less aerosol will be released if a clean air mass cloud evaporates. In response to the referee, we have pointed out this limitation in the revised manuscript.

Line 274: "Third, these selected periods were all under the influence of anthropogenic-emission-related air masses, while whether the evaporation of a clean air mass cloud can impact the aerosol concentrations at this site remains unknown. However, we noticed a period (from 10 to 12 November) that seems to contain a cloud evaporation process as indicated by the suddenly reduced RH (from 100 % to 70 %) during the daytime on 11 November, but still exhibited relatively low PM_1 levels (2.6 µg m^{-3} on average). The back trajectory analysis further showed that the air mass during this period originated from the sea, which was less influenced by anthropogenic emissions. As a result, it is likely that a small amount of aerosol can also be released even if a clean air mass enters the cloud and evaporates. Despite these issues that need to be addressed through future long-term measurements and model studies, the cloud evaporation phenomenon we observed might still be a plausible mechanism to explain certain aerosol episodes in this and other similar regions."
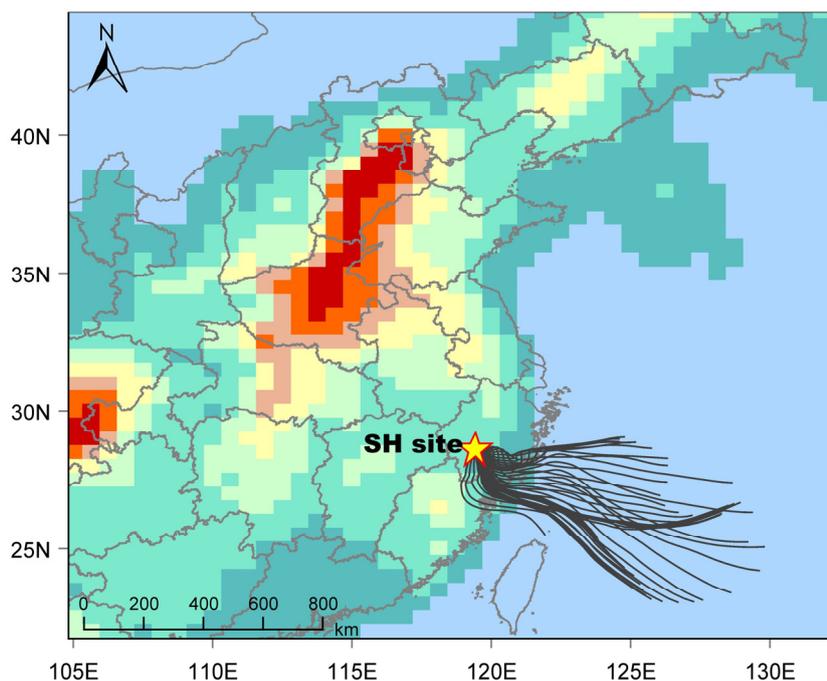
Figure S13. 72 h air mass back trajectories calculated at 1 h intervals at the SH site during 10−12 November.

RC2-5: Original comment RC3-8: Detection limits (DL) cannot be taken from literature but must be determined for the individual instrument. The literature values can provide a general guideline for the instrument type. But a specific instrument may differ cause by a number of instrument specific settings (e.g., tuning or duty cycle settings). For the AMS, the necessary data should be available as the same particle filter on inlet time period that is needed for the gas-phase $CO2$ correction can be used. Simply calculate the standard deviation of the noise during those periods. Usually 2 or 3 times that standard deviation value is used as DL (DeCarlo et al., 2006). If no such filter period is available, a period with low particle concentration can be used. Highly smooth that time period and calculate the difference between the smoothed and measured values. The standard deviation of those differences is a very good approximation of the instrument noise.

Response: Following the referee's suggestions, we select C1 to perform the DL estimation for ACSM and AMS. The results are shown in Table S2, and a brief discussion has been added to the manuscript.

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Line 105: "Additionally, the detection limits of ASCM and AMS for different NR-PM$_1$ species are determined as 3 times the standard deviation of mass concentrations during a period (16−20 November) with low particle concentration (Table S2)."

Table S2. The detection limits (in ng m^{-3}) of ACSM (20 min average time) and AMS (1 min average time) for different NR-PM$_1$ chemical species that are determined as 3 times the standard deviation of mass concentrations during a period (16−20 November) with low particle concentration.

Species	ACSM	AMS
Org	1500	97
SO$_4$	497	18
NO$_3$	166	28
NH$_4$	201	37
Chl	130	7.6

RC2-6: Section 2.2.1: It took me a while to get what "ACSM data during the sampling period was corrected by using the regression coefficients between ACSM and AMS." was about. The reply to reviewers comments document contains more details but the manuscript text does not explain why this correction was applied. The ACSM was systematically higher than the AMS and thus the authors decided to scale the ACSM to the AMS value. This should be clearly stated. Omitting the problematic Chlorine case, the factor seems to be rather similar for all species indicating a general overestimation of the instrument. (See my note on calibration below for a potential explanation.) Such a general offset should be stable throughout the measurement period and the applied correction is valid.

Response: We apologize for the inconvenience caused to the reviewer. An explanation of using AMS data to correct ACSM data has been added to the revised manuscript.

Line 108: "The concentrations of NR-PM$_1$ species measured by ACSM correlated well with those measured by AMS ($r^2 = 0.35–0.97$) except for chloride ($r^2 = 0.18$). This is likely due to the low ambient chloride masses at this background site, which are close

to or below the ACSM detection limit. Also, the non-ideal vaporization behavior of chloride in ACSM may be another reason (Crenn et al., 2015). Notably, similar slopes (0.62–0.74) were found for organics, sulfate, nitrate, and ammonium measured by AMS and ACSM, indicating a general overestimation of ACSM. The possible systematic bias in IE calibration of ACSM using ammonium sulfate could be a potential cause. Therefore, the ACSM data during the sampling period was scaled to the AMS data using these linear regression slopes."

RC2-7: Text S1: I do not disagree with selecting the 4-factor solution in this case. But in their argument about why the selected 4 and not 3 or 5 factors, the absolute value of Q/Qexp is not as relevant as the fact that a meaningful factor is introduced when going from 3 to 4 factors and that Q/Qexp is not significantly decreased when the number of factors is increased further. Q/Qexp only approaches unity if the values in the error matrix are indeed representative of the true measurement errors and all other assumptions of the PMF model are also met. This should be the case if the measurement error of the AMS (or ACSM) is well characterised. But if any additional sources of error are introduced (not related to counting statistics), the Q/Qexp values will plateau at values larger or smaller than 1. Both Ulbrich et al. 2009 and the original PMF papers (Paatero and Tapper, 1994) point out this and other issues with simply using Q/Qexp ~1 as a selector and suggest to rather use the shape of the Q/Qexp vs number of factor curve. I.e., using that a strong change in Q/Qexp with increase in factor number means that significantly more of the variability of the data is explained and thus the solution is better.

Just adjust the wording in text S1 to take the emphasis away from Q/Qexp ~1 as the main criterion for a good PMF solution.

Response: We appreciate the reviewer for this comment. We have revised the relevant text as suggested.

Text S1: "Typically, a strong change in Q/Qexp value with the addition of a factor indicates that more of the variability of the data is explained (Patero and Tapper, 1993; Ulbrich et al., 2009)."

"The four-factor solution, with a Q/Qexp value of 1.0065, can separate two OOA factors with different elemental ratios and temporal variations. Continuing to increase the factor number cannot significantly decrease the Q/Qexp."

Specific comments:

RC2-8: Line 54f: "…while smaller particles remained growth through the process of gas-to-aqueous partitioning." I do not understand the term remained growth in this context. Please rephrase.

Response: We have rephrased this sentence for clarity.

Line 53: "Chen et al. (2021) reported that fog scavenging was more efficient towards particles with aerodynamic diameter larger than 700 nm in Taiwan mountain regions, while the majority of smaller particles remained unscavenged and could grow into larger particles through gas-to-aqueous partitioning."

RC2-9: Line 93: The sentence about PDR seems to be broken/missing.

Response: We apologize for our negligence of this sentence. The missing sentence has been added to the revised manuscript.

Line 93: "In addition, the particle depolarization ratio (PDR) was measured by a particle monitoring lidar (Science of Light Technologies Co., Ltd) to analyze the vertical distribution of aerosol particles."

RC2-10: Line 194: The term "PM1 episode" is ambiguous in this context. I guess the authors mean something like "elevated PM1 episode". Also clarify which criteria were used for selection of the 5 "PM1 episode". Is it only based on PM1 concentrations? Or was there another criterion (e.g. high BC).

Response: Thank the referee's suggestions. We have changed it to "elevated PM1 episode". The elevated PM_1 episodes were defined as periods when average PM_1 concentrations exceed the 75th percentile of the overall PM_1 concentration distribution, while clean periods were defined as periods when average PM_1 concentrations fall below the 25th percentile of the distribution. The criteria has been added to the revised manuscript.

Line 206: "These five elevated PM₁ episodes exhibited mean PM₁ concentrations ranging from 5.2−9.8 µg m⁻³, which were close to or exceeded the 75ᵗʰ percentile of the overall PM₁ concentration distribution (5.6 µg m⁻³). Meanwhile, a clean period (C1) with a mean PM₁ concentration of 1.3 µg m⁻³, falling below the 25ᵗʰ percentile of the distribution (1.6 µg m⁻³) was also selected for comparison."

RC2-11: Line 230 "… scavenged by the increasing RH and strong wind." I still struggle with this interpretation in this sentence. Is "scavenge" really the right term here? And is the increasing RH the key factor or rather the formation of a new cloud/fog layer? Note that for EP5, the aerosol concentration continues to increase after the minimum of RH in the early afternoon of Nov 26th.

Response: Thank the referee for pointing this out. We have replaced the word "scavenge" to "remove". Also, we agree with the referee that the term "RH" is inaccurate and ambiguous here. The formation of new clouds may be the primary factor contributing to the reduction in particle concentrations.

Line 246: "These particles were then gradually removed by new cloud formation and strong wind (WS > 4 m s⁻¹)."

RC2-12: Line 249 (original comment RC 3-41) "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." I have to apologize for my clumsy phrasing of the original comment. I think we understand each other but the sentence is a bit weird. I recommend phrasing it as: "… suggests that the aerosol is likely externally mixed with small particles dominated by organics, which may further…."

Response: This sentence has been rephrased in the revised manuscript following the referee's suggestion.

RC2-13: Line 270: "… time series and box plots…" There are still no box plots in Fig 6. Please adjust the wording in the main text.

Response: We have corrected this sentence in the revised manuscript.

RC2-14: Fig 6: This Figure will be easier to read if the legend in the middle includes all used symbols. Yes, the axes are coloured accordingly, but the black open symbols are sometimes for the right and sometime for the left one.

Response: The legends which contain all used symbols have been added to this figure.
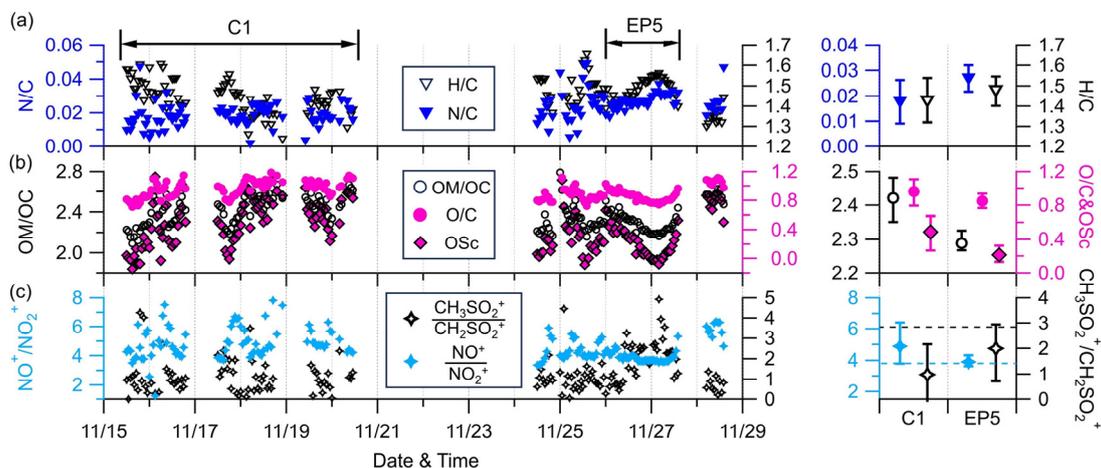
Figure 5. Time series of 1-hour averaged (a) N/C and H/C, (b) OM/OC, OSc, and O/C, and (c) NO^+/NO_2^+ and $CH_3SO_2^+/CH_2SO_2^+$ during the AMS sampling site (left) and their mean values in C1 and EP5 (right). Only ratios determined with good S/N (i.e., organics > 0.7 µg m^{-3}) are shown.

RC2-15: Note on AMS & ACSM calibration: (nothing to change in the current manuscript, just something to consider for future measurements)

Typically, the aerosol mass measured by AMS and ACSM instruments are reported relative to nitrate (nitrate equivalent mass). I.e., the IE calibration is conducted with ammonium nitrate (AN). Then the RIE of ammonium, sulphate, and other species are calculated in relation to that nitrate value. If I remember correctly, the ACSM (with standard vaporiser) has a little bit of a sluggish response to ammonium sulphate (AS). I.e., the signal decreases slower than the AN signal when switching to the particle filter. This is only visible when looking at the raw time series data before averaging over the 20 sec no filter/with filter intervals. This slightly slower response decreases the difference between the no filter/with filter values of sulphate. I.e., the calibration indicates that the instrument is a little bit less sensitive than it really is. This is normally not an issue as AS always behaves like that and RIE handles this. But if the sulphate

measurement is used to determine IE, it may lead to a systematic bias. Hence, the ACSM values would be a bit overestimated – as it seems to be the case in these measurements.

This might cause some differences between AMS and ACSM if the AS measurement is used for IE determination – especially if the AMS is referenced to an AN calibration. In an ideal case, one would perform both AN and AS calibrations on a regular basis. But if there is only time for one, I recommend doing the AN one in the future. This has the added benefit of obtaining the NO^+/NO_2^+ ratio for inorganic nitrate. That value can then be used to estimate the organonitrate contribution from ACSM data without the need for PMF analysis. Using AN also avoids the pains of accounting for the RH dependent collection efficiency of AS in a standard vaporizer.

Response: We appreciate the referee's valuable and constructive suggestions, which are very helpful for our further studies. We will perform the AN calibration for AMS and ACSM in future measurements.

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