Review report Zhang et al. 2024

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.

Major Comments

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 utilise 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.
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?
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?

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 PM1 inlets leading to the detection of dried PM1 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.

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.

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 NO3 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 during P1. 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 NO3 source), one could start to interpret the reduced inorganic NO3 in the context of the clouds. One possible interpretation could be that the very water soluble HNO3 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 PM1 cutoff at ambient conditions, i.e., if only interstitial particles are sampled, see comment #4)

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

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 to low to be interpreted or used to derive parameters like ON or O/C ratios.

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.

Specific comments

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 interstital+cloud droplets were sampled.

1) 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?

2) Line 23: I do not understand this sentence. I cannot follow the argument of how the O/C change with OA/ Δ CO values implies things about the cloud processing. Also, Δ CO is not introduced.

3) 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?.

4) 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.

5) 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.

6) 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.

7) 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.

8) Line 93: Why were the default values used for RIE? Later they state that calibrations were conducted.

9) Line 94: Which compounds were used for the calibration? Ammonium nitrate? Ammonium sulfate? Both? Individually? As mixtures?

10) 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?

11) 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.

12) 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.

13) 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.

14) 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.

15) 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 realised that this section does not contain the necessary information to track what data was used.

16) 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.

17) 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)

18) 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?

19) 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.

20) 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?

21) 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.

22) 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 1 below) 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.

23) 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.
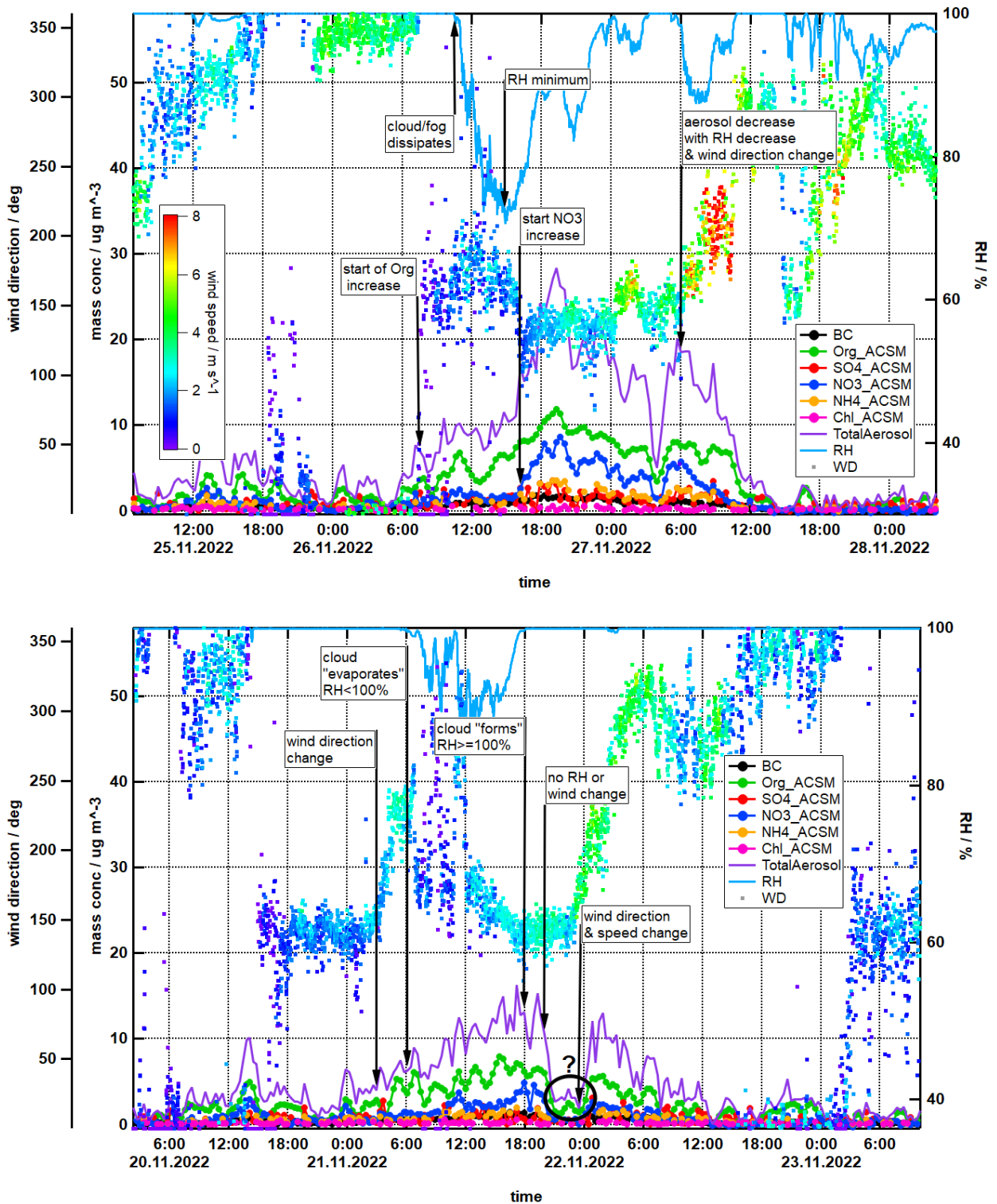
Figure 1: Detailed look at time series of BC, ACSM species, total aerosol, RH, wind direction and wind speed for the time indicated as P2 in the manuscript (Top) and Nov 20th – 23rd . Wind direction markers are colour coded by windspeed on a scale from 0 – 8 m/s.
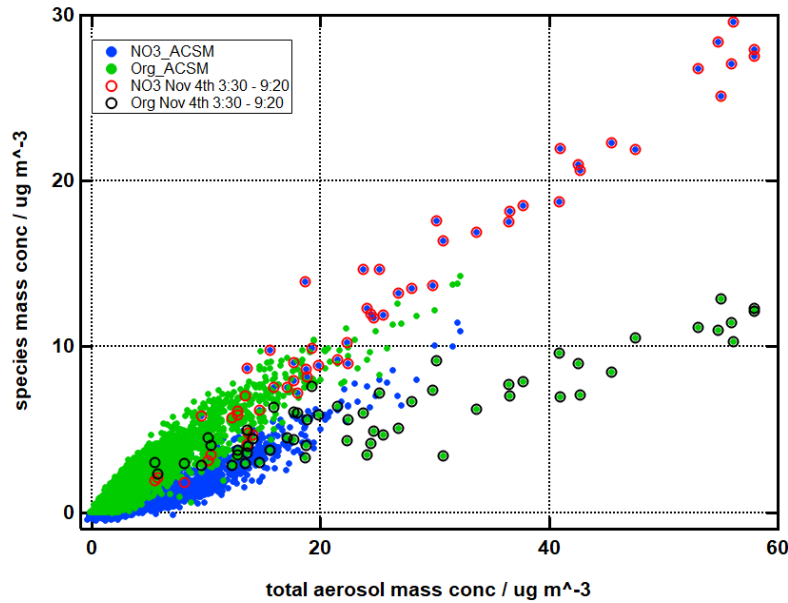
Figure 2: mass concentration of organic and NO3 as a function of total aerosol mass (NR-PM1 + BC). Red and black circles indicate data from NO3 plume on Nov 4th 3:30 – 9:20.
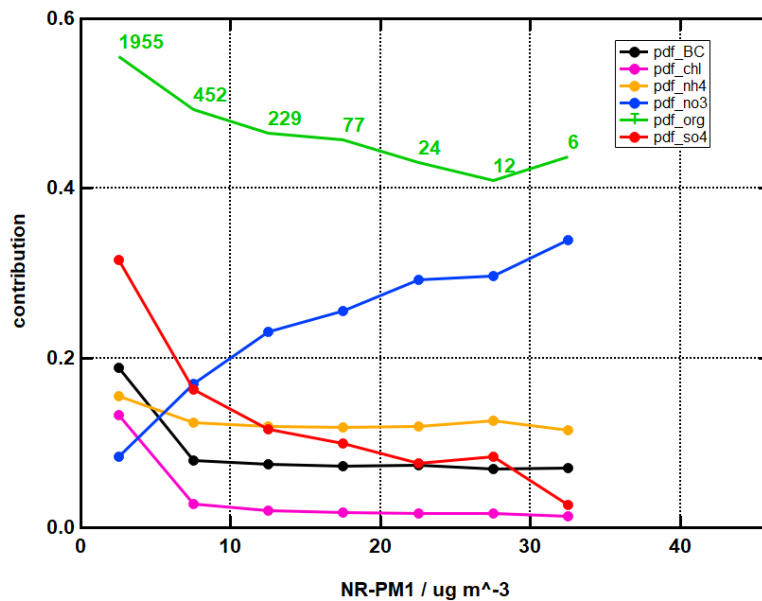
Figure 3: Relative contribution of aerosol species as a function of total aerosol mass. Data was grouped into 5ug/m^3 wide bins and data point indicate the average values for each bin. The numbers over the organic line (green) indicate the number of data points in each bin.

24) The time series of PM2.5 seems to correlate with BC? Is that the case? Does that mean something?
25) 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.
26) 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.

27) Fig S4: why are there two separate y axis? They made comparison difficult.

28) 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".

29) 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.

30) 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.

31) 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 – 11th seems 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.

32) 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).

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

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

35) 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?

36) 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.

37) 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.)

38) 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.

39) 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).

40) 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.

41) Line 264: the right reference for the Δ CO method should be (Decarlo et al., 2010).

42) 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).

43) 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.

44) 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.

45) 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.

46) Line 292: what is meant by "overlap with the aged OAs observed at other sites"?

47) 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.

48) 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.

49) 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.

50) 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?

51) Line 318f: "Remarkably, nitrate (20.9 %) and ammonium (17.0 %) exhibited unexpectedly high contributions to PM1 in this forested mountain area" Compared to the other mountain sites I found the SO4 signal remarkably LOW! Fig 1 has 50 – 66% of Inorg for all but Mt Yulong.

52) Line 322: PMF does not identify two types of SOA. It identifies 2 OOA factors and some OA associated with SO4.

53) 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?

Language:

Line 23: ratio of OA/delta CO. OA/∆ CO 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/∆ CO ratio"

Line 33: "transported to the upper layer of atmospheric boundary layer or even lower free

Troposphere" – layer of the atmospheric boundary layer

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

Line 42: characteristics over regional scales

Line 41 What impacts are referred to here?

Line 46 aerosol formation is there twice

Line 51ff "Although…" this sentence is grammatically broken - rephrase

Line 51 "several studies over mountain sites": probably at mountain sites. Or do they mean measurements literally above those mountain stations?

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

Line 53 "stations have been settled " - what do they mean? Stations were established?

Line 54f: "most of them are "

Line 55: "mainly conducted on Qinghai-Tibetan Plateau" – the verb refers back to "stations as a subject. Studies can be conducted. Stations cannot be conducted.

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?

Line 63: "backward trajectory analysis" – I found back trajectory analysis to be the more common term. The documentation on HYSPLIT uses back trajectory

Line 63: "particle organic nitrates" – particulate organic nitrate or particle-phase organic nitrates

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.

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.

Line 117 "Trajectory time" : trajectory arrival time

Line 118 "height": arrival height at the site

Line 126: "organics held the largest contribution" – bad language rephrase

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

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".

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.

Line 163f: it is not clear if "this site" phrase refers to the sit in this study or the site in Chen et al ?

Line 165: "AMS lens" -> Fig S4 shows ACSM data hence this must be ACSM lens in this sentence?

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

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

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

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.

Line 255: "significant discrepancies" – differences not discrepancies

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

References

Decarlo, P. F., Ulbrich, I. M., Crounse, J., De Foy, B., Dunlea, E. J., Aiken, A. C., Knapp, D., Weinheimer, A. J., Campos, T., Wennberg, P. O., and Jimenez, J. L.: Investigation of the sources and processing of organic aerosol over the Central Mexican Plateau from aircraft measurements during MILAGRO, Atmos. Chem. Phys, 10, 5257–5280, https://doi.org/10.5194/acp-10-5257-2010, 2010.

Kiendler-Scharr, A., Mensah, A. A., Friese, E., Topping, D., Nemitz, E., Prevot, A. S. H., Äijälä, M., Allan, J., Canonaco, F., Canagaratna, M., Carbone, S., Crippa, M., Dall Osto, M., Day, D. A., De Carlo, P., Di Marco, C. F., Elbern, H., Eriksson, A., Freney, E., Hao, L., Herrmann, H., Hildebrandt, L., Hillamo, R., Jimenez, J. L., Laaksonen, A., McFiggans, G., Mohr, C., O'Dowd, C., Otjes, R., Ovadnevaite, J., Pandis, S. N., Poulain, L., Schlag, P., Sellegri, K., Swietlicki, E., Tiitta, P., Vermeulen, A., Wahner, A., Worsnop, D., and Wu, H. C.: Ubiquity of organic nitrates from nighttime chemistry in the European submicron aerosol, Geophys. Res. Lett., 43, 7735–7744, https://doi.org/10.1002/2016GL069239, 2016.

Zhang, Y. M., Zhang, X. Y., Sun, J. Y., Hu, G. Y., Shen, X. J., Wang, Y. Q., Wang, T. T., Wang, D. Z., and Zhao, Y.: Chemical composition and mass size distribution of PM 1 at an elevated site in central east China, Atmos. Chem. Phys, 14, 12237–12249, https://doi.org/10.5194/acp-14-12237-2014, 2014.