Review report Zhang et al. 2024 – revised version

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.

Minor Comments (lines numbers refer to the revised manuscript without tracking)

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.

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 >2.5um (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?

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 airmass 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 airmass 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.

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?

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.

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.

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.

Specific Comments

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

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.

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…."

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.

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.

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+/NO2+ 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 vaporiser.