

This manuscript by Cao et al. presents the field measurement results using an AMS coupled with a TD conducted in China. As the application of TD-AMS is for now still rare in the atmospheric chemistry studies in China. The manuscript contains useful and valuable information. However, this reviewer finds that in the current version, many necessary details are missing, and many statements and important conclusions appear to be over-interpreted, not well supported by the data or at least is not well explained, which limits the scientific value of the work. The current version needs major revision before its acceptance. The current version fails to make good use of the wealthy data obtained by the HR-AMS, and it should be extensively expanded. Here below I list the comments below that need to be addressed.

- (1) Please pay attentions to the in-text citation format
- (2) In the introduction part, please provide more details regarding previous TD-AMS studies, focused the important findings.
- (3) Introduction: Recently, Xu et al. (Atmos. Chem. Phys. 2014, 14, 12593-12611), and Li et al. (Atmos. Environ. 2017, 158, 270-304.) have provided a summary of the AMS studies in China, these two papers should be cited.
- (4) Section 2.2 : The authors tested the transmission efficiencies, do these numbers are included to calculate the MFR? Besides, some theoretical calculations by Chris Cappa et al. regarding TD kinetics should be included.
- (5) Section 2.2 : why not to try a higher TD temperature, for example 250C or even 300C, there can be extremely low volatility species remained at such temperatures
- (6) Section 2: Some details regarding the TD is not provided. For example, what is the temperature cycle. What is the sampling time at each TD temperature? Do the authors change the absorption materials of the TD? And how often?
- (7) Section 2: A lot of details are not provided. You should give a bit more details regarding the instrumental and operational details about the AMS, for the convenience of readers not in the AMS community. Calibrations? Relative ionization efficiencies? Size calibration? What are the chemical resolution? As your PMF includes ions with m/z up to 170, does these ions can be well separated from their neighborhood ions? Any instrument details about AE31?
- (8) Section 2.3: As this study are the very first few ones that used combined TD and Bypass data to perform the PMF analyses, more details should be provided. The determination of optimal solution, what are the profiles of other solutions? What are the diagnostic plots of the currently chosen solution? What are the correlations of the factors with external tracers? what the PMF results are if only consider bypass data, and then to show clearly what are the advantages (please provide such information in the forms of plots in details not only a brief description) by running PMF on the combined TD and Bypass data. Such details are missing, but they are very helpful and also necessary to evaluate the robustness of your PMF results.
- (9) Results: P5L20 It is not convincing to state that sulfate is due to regional transport while other species are influenced by local emissions, only based on the extents of data variability (and the variability can be quantified, from xx to xx and what are the standard deviations?)
- (10)P5L25: the BC contribution seems to be quite high in PM1 (12.2%), is this number

consistent with previous values in Shenzhen? Is this due to heavy traffic? That's also why I asked the calibration and operational details of AE31.

- (11)P5L25: Is this composition for bypass conditions or others? Please indicate it clearly.
- (12)P5L29-: Please add the meteorological parameters, such as temperatures, solar radiation data to help explain the variations of nitrate. For example, it is not convincing to state the photochemical oxidation of NO_x only based on the peak after BC peak. Similar for chloride, NH₄NO₃ is also semi-volatile, so why G/P partitioning drives chloride but not nitrate? Also, if you have PBL data, then add its diurnal pattern as well.
- (13)P6L7: Why not provide the neutralization plot? You can do that to support your statement.
- (14)P6L12: Do the elemental ratios calculated by Aiken 2008 method, or Canagaratna 2015 method? Please explain. Also, if it is from Canagaratna 2015 method, can it compare with previous values fairly?
- (15)P6L23: I don't think "which may be caused by the internal mixing and similar gas-particle distribution processes" is correct. If this is correct, then why you state previously that nitrate is due to photochemistry while chloride is due to G/P partitioning? They are not consistent.
- (16)P6L25: "products of photochemical reactions of VOCs have a significant influence on the organic pollution" I don't think your data presented till here can indicate this. And why only photochemical reactions of VOCs, and other possible processes are excluded?
- (17)P6L27: why a large size would mean a more aged sulfate? and why it would then mean it had been transported regionally?
- (18)P7L6: Is your AE31 downstream of the TD? If so, can you plot the MFR of BC measured by AE31? You didn't mention the instrument setup of AE31 at all.
- (19)P8L7: "sulphuric acid can be distinguished with temperature between 30 and 125 °C", what does this mean? I don't quite understand.
- (20)P8L10: this paragraph is not well written. You should provide the average OA MS at different TD temperatures to give more explanations. You didn't explain why a more rapid increase occurred from 150-200C, and "OA is heated and oxidized into more aged compounds", there is lacking any supporting evidence. Jimenez et al., 2009 does not say so. "the results is consistent with the OA evolution process (Jimenez et al., 2009)?" Please be more specific and not so vague, this paper doesn't have any TD-AMS results, although it does show that the O/C ratio of OA increases with the decrease of saturation of OA.
- (21)P9L15: In my viewpoint, your BC and HOA diurnal patterns don't have clear matched with traffic times. It is overall high during evening to noon, and low during afternoon, more relevant to PBL dynamics, likely.
- (22)P9L23: Your writing logistic is strange. Sometimes you mentioned the results of previous studies, normally, the readers expect to see what your data show, but instead you didn't mention them. Here, you mention the findings of Mohr et al., 2012 regarding m_z. 55 and 57, then how does them behave in your MS? If you are not going to describe them for your data, then why you mention Mohr's results? Their results don't support your explanation.
- (23)P9L28: That's what I am talking about, you mention PBL influence on COA, why not on HOA, etc.
- (24)P10L5: Do you have data of external BB tracer to use?

- (25)P10L15: What are the correlation coefficients of LOOOA with sulfate, as well as MOOOA with nitrate? Typically, the important criteria is to compare the correlations coefficients, and then postulate the assignment of LOOOA and MOOOA (although it is not always the case.) It seems like LOOOA might have a better correlation with sulfate, while MOOOA is better correlated with nitrate from Figure 4b, how to explain? What are the cross correlation coefficients between diurnal patterns of these species? A more comprehensive discussion regarding these SOA should be combined with meteorological data and also gaseous species, if available.
- (26)P10L20: When you talk about the mass contributions of different OA factors, do you mean the average contributions over all data including bypass and TD? Or it is only for the case of bypass conditions? You didn't indicate this clearly at all throughout the manuscript.
- (27)P12Section 3.4: Can you show the mass fractions of different OA factors at different TD temperatures?
- (28)P12Section3,4: As I suggested before, this section doesn't discuss any compositional changes of OA, but only mention the variations of MFR. You have an HR-AMS which can provide chemical information in very details, which is in fact the unique advantage of HR-AMS, why not do so?
- (29)P13L10: It is not that convincing to state that HOA rather than BBOA or COA was a more important source of LO-OOA, based on only the volatility sequence. The sequence may change at different temperatures, then the conclusion will change according to the logistic used here. To prove this, molecular level analyses are required. For example, a HOA tracer compound get oxidized, and the products appear in LO-OOA. Also, heterogeneous or condensed-phase reactions may also produce LO-OOA related species, it is not definitive that the reactions have to proceed via evaporation-gas-phase oxidation-condensation. Such statement is high speculative without any solid evidence in my opinion.
- (30)P13: Basically, I think it is dangerous to state the evolution process of different OA factors based on only volatility data. Even you are lacking of other data, you can perform more analyses, such as the temporal variations, compositional changes, correlations with other species, and discuss these in more details. This can be done for a few cases