

Interactive comment on “Volatility measurement of atmospheric submicron aerosols in an urban atmosphere in southern China” by Li-Ming Cao et al.

Anonymous Referee #2

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This manuscript describes the volatilities of the PM₁ chemical components by using the Thermo-Denuder – Aerosol Mass Spectrometer (TD-AMS) system, along with the positive matrix factorization (PMF) analysis. The results make some very important implications on the atmospheric chemistry of aerosol particles, as there appears to be the first report about such study under a polluted environment in China. Overall, the content of this study fits within the scope of ACP. I agree that this campaign was well-designed. However, the authors need to consider making more further clarifications/evidences to support a couple of ambiguous discussion and/or conclusions in this paper. Numerous corrections on the text editing are needed, including reference and abbreviation formats as well as language issues, etc. Please the authors carefully

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check that throughout the manuscript. Therefore, a major revision is needed before it would be accepted in ACP.

General comments:

When I finished reading this manuscript, I feel like that the authors did not fully analyze such comprehensive data set, then, highlight the new findings during the discussion of this study, because I found a lot of “consistent comparison” between this study and previous studies. I can understand that the authors would like to support your results/discussion/conclusions, and I am not saying that you should not do that. But the authors should try to find something new as those comparisons with previous works.

More analysis could also be done to understand such data set. For example, for both TD-path and non-TD-path data: temporal variations (PM1 species and PMF-OA factors)? chemical changes under the different environment conditions? any evidence for potential origins of changing volatilities for these species (e.g., what's difference between marine and continental air masses)? Variations of size distributions, rather than averaged ones? etc.

More details of experimental materials need to be shown wherever in the main text or supplementary. What's the duration for TD-path data? It's easier to understand for readers if the authors could show that, for instance, in the time series of Figure 2a. What's the time resolution of your measurements during the campaign? How did the authors calibrate the AMS, and what were the results, e.g., values of IE, RIENH4, RIESO4? The authors should show one figure for the relationship between measured NH4 and predicted NH4 for TD-path and non-TD-path data, respectively.

I am not convinced by the state of a finding about “. . .that HOA, rather than BBOA or COA, could be a potentially important source of LO-OOA. . .”, as shown in the abstract and the main text elsewhere, just based on current TD-AMS-PMF results. The authors should perform more analysis to support that. For example, typical cases analysis?

Since the authors have the data of seven-wavelength light absorption, it will be useful to support your PMF-POA factors by performing source apportionment of black carbon (BC) with aethalometer data (Elser et al., 2016).

Comments/suggestions in details:

Please note that abbreviations should be used in the same format throughout all the manuscript. For example, page 1, line 12: “a TD-AMS (Thermo-Denuder – Aerosol Mass Spectrometer)” and page 1, line 20: “a hydrocarbon-like OA (HOA, ...)”. For the consistency, the authors may replace “a TD-AMS (Thermo-Denuder – Aerosol Mass Spectrometer)” by “a Thermo-Denuder – Aerosol Mass Spectrometer (TD-AMS)”. Somewhere else if the same issue should also be done.

Please define abbreviations when using it for the first time. For instance, Page 1, line 13: submicron particulate matter (PM1); page 1, line 19: positive matrix factorization (PMF). Somewhere else if the same issue should also be done.

Page 2, lines 15-25, the authors should also introduce more about the major findings reported by those previous studies. Then, the authors may tell readers the missing knowledge according to the new findings of your study.

Page 2, lines 29-31, I cannot understand the relationship of this sentence with the major story of this introduction.

Page 3, line 15, Duplicate definition for “thermo-denuder (TD)”, it has been defined in the first time in Page 2 line 15. In addition, abbreviation should be followed hereafter when it has been defined at the first time. The authors should carefully check the similar issues as others, e.g., black carbon, organic aerosol, etc.

Page 5, lines 9-15, it’s hard to read these sentences Please re-edit. The authors may introduce your data treatment procedure, then/at the same time, you could give the reference(s) to support yours as well as explain why.

Page 5, line 23, the AE-31 should be described in the experimental method section.

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More details should be also given, e.g., cutoff, and which wavelength you used for the equivalent black carbon concentration. It is same for SMPS in line 29, for which more description should be given in this section too.

Page 5, lines 24-25, “. . .due to rain. . .”, to state this, the authors should provide related rain data to prove it. And what’s the link of “. . .sulfate showed a relatively stable. . .” to this “rain case” in this sentence? and I don’t understand why the relatively stable time series of sulfate can be considered as regional transportation? The authors may perform more analysis on chemical species along with your ground-measured meteorological parameters. Also, for instance, air mass trajectory analysis would be also useful to help figure it out.

Page 5, lines 23-30: It’s hard to read such long sentence. The authors should separate it for each information what you want to discuss. Such kind of long sentences is also frequently showing somewhere in this paper. The authors should keep the similar modification.

Page 6, lines 2-3: The authors should make the plot to support this discussion. And it would be also interesting to see what’s the different ratio of measured and predicted NH₄ from TD-path and non-TD-path data.

Page 6, line 4: Double meanings between “diurnal variation” and “during the day” in one sentence. Please reword it and somewhere same is also needed.

Page 6, line 5: Please the authors provide any evidence to prove the contribution of “the activity of heavy duty vehicles” to BC in the evening. If it’s a case, and what’s the difference sources that contribute BC particles between morning and evening peaks? Indeed, I feel more like that biomass burning emissions (according to the next discussion of BBOA variations) might also contribute the evening peak of BC. That’s also one of reasons that I propose the authors to perform the BC source apportionment.

Page 6, lines 6-10: The authors should be careful to sate the nitrate variations just

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according to such diurnal peaks between BC and nitrate. For example, how did the authors indicate that the peak of nitrate after the BC one should be linked to photochemistry, and that the peak at around 14:00 is due to “the enhancement of sunlight”? And, why there was no influence of gas-particle partitioning on nitrate, as discussed only for chloride?

Page 6, lines 10-11: The authors should provide/link your evidence or any published work(s) to prove such kind of discussions. In the manuscript, somewhere else with the similar issue should be modified too.

Page 6, lines 11-13: Remove “during the day”. I cannot understand that “. . .a regional product of oxidation by SO₂ that is transported . . .”, since I did not see the transported evidence of sulfate in this study. The authors may further analyze the temporal variations along with the size distribution of sulfate, and considering meteorological influence.

Page 6, lines 14-15: Please provide the neutralization plot. It seems an odd sentence for “. . .so the diurnal variation of ammonium was influenced by sulfate, nitrate and chloride”. It’s generally true that ammonium measured by the aerodyne AMS is mainly in the form of ammonium sulfate, ammonium nitrate, and/or ammonium chloride. I do more trust that diurnal variations of ammonium can be also affected by such inorganic salts formation processes, besides other factors, e.g., atmospheric physical processes. So, the authors should reword it.

Page 6, lines 15-16: I don’t think the authors need to repeat such information of organic aerosols as already provided in the introduction before (page 2 lines 13-14). introduced before. Again, somewhere else, such kind of discussion, at least, the authors should provide reference(s) to support it. I suggest the authors to reedit and combine this sentence with the next one (lines 16-18).

Page 6, lines 19-21: It’s complicate to read here with a lot of comparison in only one sentence. Were all the averaged reference values only from Chen et al. (2015)? In

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addition, why did not the authors compare those values of O/C and H/C with some results observed under other polluted environments of China? It might make sense to understand such knowledge over the regional scale for developing countries in Asia (e.g., China).

Page 6, lines 21-25: The authors did not explain those diurnal variations of O/C and H/C. And why only a small H/C peak at noon was discussed, but no explanation at the peak during the nighttime? Do the authors think the biomass burning could also influence H/C variations, in addition to traffic and cooking emissions?

Page 6, line 26: The authors should avoid highlighting “non-refractory species measured by the AMS” too many times over the manuscript, because readers will know that after you explain it at the first time (except for the special case). Please the authors carefully check that elsewhere.

Page 6, lines 26-28: Be careful making the conclusion of averaged “approximately 500 – 700 nm in the accumulation modes” linking to “all the species” being aged particles. For example, were “all the species” including primary emissions, as below discussed HOA and BBOA, as well as fresh OOA? In addition, the authors already discussed that nitrate can be formed just after the morning traffic rush hours, so is this also included? To understand so, the authors can do the time series of size distribution of each chemical species (including both inorganic aerosols and PMF-OA factors) instead of showing here.

Page 6, line 30: I don't understand why “a similar average size distribution” of these inorganic species is because of this.

Page 6, lines 30-32: As reported the comment of 21, the authors should provide the size distribution of PMF-OA factors to prove this. In addition, how to prove “products of photochemical reactions of VOCs have a significant influence on the organic pollution.”, while rather than other formation processes?

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Page 7, lines 1-2: How to understand here, the large size of sulfate being aged and from regional transports?

Page 7, line 8: Again, “measured by the AMS”, such kind of words, does not need to be iterate.

Page 7, line 10: What does mean by “measured directly by the AMS.”? Is it meaning the measured particles from non-TD-path channel?

Page 7, lines 12-13: The authors stated “. . .of the total non-refractory species and organics all. . .”. Was this “all” including all inorganic salts and PMF-OA factors? If yes, I do not suggest saying, “the fact that they consist of various compounds with a wide range of volatilities”, then I prefer to say, “the fact that they include various compounds with a wide range of volatilities”.

Page 8, lines 4-7: I suggest the authors to separate this long sentence to be clearer.

Page 8, lines 7-11: Again, please separate this too much long sentence to be clearer.

Page 8, lines 12-14: Just as an example to separate a long sentence, “;” can be changed to “.”.

Page 8, lines 16-18: I don’t think that Jimenez et al., (2009) stated such conclusion. The authors could try to see variations of both total mass spectra and organic spectra, respectively, at different TD temperatures. The mass fraction of total NR-PM1 and total PMF-OA could be shown also.

Page 9, lines 4-5: May replace “classes/species” by “species”. And replace “the total PM1 composition” by “the total PM1 mass loading”.

Page 9, lines 5-7: Duplicate definition for “positive matrix factorization (PMF)”. I don’t think this sentence is useful here, as the authors said, “as discussed in section 2.4.”. Please remove or reword it.

Page 9, lines 7-9: Same issue, this kind of information has been shown before in “2.4

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Source Apportionment Method”. Please reword.

Page 9, lines 9-11: Same again, this information has been shown in “2.4 Source Apportionment Method”. And Duplicate definition for the abbreviation. Please reword.

Page 9, lines 11-13: How are the relative contributions of them at different TD-temperature conditions? The authors could be able to show that.

Page 9, lines 14-16: Please separate this sentence mixed with different information. For example, the authors could discuss the characteristics of your HOA mass spectrum, and give supporting reference. Then to compare your H/C value with typical ones as published, to further prove your reasonable HOA factor.

Page 9, line 16: Duplicate definition for “Black carbon (BC)”.

Page 9, lines 16-17: Please provide reference(s) to support this discussion.

Page 9, lines 17-19: “as identified by previous publications (Zhang et al., 2007a; Lanz et al., 2007; Ulbrich et al., 2009)” seems not really needed here, as compared to the last sentence. It will be more useful to compare HOA with BC from traffic emissions (as I proposed above), because biomass burning emissions can also contribute BC here. Also, this will be helpful to support the Page 9, lines 17-19.

Page 9, line 23: Related reference(s) is(are) needed to be at the end of “. . .which are mainly ionized from alkanes, alkenes and, possibly, long chain fatty acids. . .”.

Page 9, lines 23-24: “COA is characterized” can be removed. As presented “which are mainly ionized from alkanes, alkenes and, possibly, long chain fatty acids”, what are such m/z 41 and m/z 55 from for the COA factor?

Page 9, lines 25-27: I did not get the meaning by mentioning this sentence about the results of Mohr et al. (2012). The authors even did not discuss your results about ratio of “m/z 55 to m/z 57”.

Page 10, line 3: What are they different between “biomass burning” and “wood burn-

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ing”, as showing here together?

Page 10, lines 7-9: Reword this sentence.

Page 10, lines 10-12: I did not understand the relationship between “an O/C of 0.32 and showed a similar diurnal trend” and “indicating the significant influence. . .”. If the authors would like to highlight the significant role of biomass burning emissions in aerosol pollution, you should provide the relative contribution to the PM loading. And please reword line 12.

Page 10, lines 13-16: Please reword this sentence. Separate it. And don't repeat to define LO-OOA and MO-OOA as shown before already.

Page 10, lines 16-18: Please separate this sentence. And provide the evidence of both sulfate and MO-OOA from regional transports.

Page 10, lines 20-21: How to get this conclusion of “denoting their secondary nature.” only according to “. . .higher concentrations during the daytime,”. Please explain it more.

Page 12, line 5: I feel like that almost findings relative to the OA volatility in this section “were consistent with” previous studies. For example, lines 9-10 (HOA), line 12 (BBOA), line 18 (COA), page 13 line 5 (OOA). I am not saying that the authors should not compare your findings with previous ones. But, the authors should find something new or that may improve our understanding. In addition, a couple of sentences are needed to be reworded/separated. E.g., some long sentences with “;” and with many times of “which attributive clause”, etc.

Page 13, lines 8-17, and page 14, lines 13-15: As many previous studies, I do trust that some POA emissions are semivolatile, which might be a missing source of secondary organic aerosols. However, the authors did not provide direct and enough evidence that may support the conclusion of “HOA, rather than BBOA or COA, could be a potentially important source of LO-OOA” via “the oxidizing process of “Evaporation – Oxidation in gas phase - Condensation”, although according to only the volatility sequence of the

PMF-OA factors.

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