

Interactive comment on “Aerosol hygroscopic growth, contributing factors and impact on haze events in a severely polluted region in northern China” by Jun Chen et al.

Anonymous Referee #1

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Summary:

Chen et al. report data retrieved in Xingtai, a city in the southern Hebei province of China, during the late spring of 2016, to determine to what extent (if any) aerosol hygroscopic growth contributes to severe haze events. They take advantage of data from a lidar, an aerosol chemical speciation monitor (ACSM), a hygroscopicity-tandem differential mobility analyzer (H-TDMA), and supporting measurements from radiosondes, to quantify aerosol hygroscopic growth. They then seek to determine both what chemical composition promotes hygroscopic growth, and how hygroscopic growth affects visibility. They choose two time periods to contrast their analysis: Case I in the

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evenings of 05/21 and Case II on 05/23. These windows were chosen as they had similar water vapor content in the (what I assume to be the boundary layer) air, yet the evening of 05/23 yielded a higher particulate matter (PM) load (~ 95 $\mu\text{g}/\text{m}^3$ for PM_{2.5} and ~ 70 $\mu\text{g}/\text{m}^3$ for PM₁) than 05/21 (~ 45 $\mu\text{g}/\text{m}^3$ for PM_{2.5} and ~ 35 $\mu\text{g}/\text{m}^3$ for PM₁). From the timeseries in Fig. 3, it appears that Case I is more representative of a normal day in Xingtai whereas Case II is a day of more severe haze. The authors conclude the haze is due to hygroscopic growth (e.g., Fig. 4 e, f, k, l., Fig. 9, and results from the Kasten model).

Overall, the tone of this manuscript is overstated. The work presented is very important and I think should be published. It brings attention to pollution in China's rapidly industrialized cities and includes important techniques that can be deployed elsewhere in China, or eastern/southeast Asia overall. The campaign design is very good! The manuscript is however unfit for publication in *Atmos. Chem. Phys.* in present form. The authors need more description to show their command over the instrumentation, restructure their sentences to convey ideas more effectively, include error values in their data to support their conclusions, and survey the literature in more depth.

I recommend publication in *Atmos. Chem. Phys.* after they address several major and minor revisions, suggestions for which are raised below for the authors' consideration.

Major Comments:

Title: Consider an Oxford comma.

Abstract: It appears that the message of the Abstract is that PM caused by anthropogenic activity is more hygroscopic than natural PM, but this does not come across clearly. The English and sentence structure of the entire Abstract require serious revision.

Introduction: The introduction should provide a comprehensive overview of why the manuscript is relevant to the community. The central portion of the introduction should

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be moved to experimental techniques, and more literature survey or relevance should be included in the body of the introduction. For example, what other events lead to haze? Is there any proportion that suggests hygroscopic growth is a minor, moderate, or major contributor to haze – either in general or specifically in the Hebei province? What consequences does this intense haze have, either in Hebei, or elsewhere in the world?

Field campaign and instruments: I would merge this section with the Methodology section. The authors don't explain why the time resolution of the lidar is 15 min, and this may be of interest to the reader and would show that the authors have tailored their use for their study. Do other lidar users (whether they use it from the ground or satellite) use 15 min time resolution? Does it depend on how clean the air is? Is there a relationship between altitude and signal-to-noise, particularly at high altitude bins? Later we discover the AE and depolarization ratio, but they haven't been mentioned in this section. This needs to be explained in greater detail to be suitable for publication. Why are the three wavelengths chosen? What does "atmospheric Mie scattering signals" mean? That phrasing is far too vague. Are the authors hinting that UV telescopes detect Mie scattering, perhaps expected from small water droplets or aqueous aerosols? Based on wavelengths used and expected aerosol size ranges, is scattering supposedly in the Mie regime? These questions should be outlined clearly for the interested reader. Finally, whilst detailed information on the ACSM can be found elsewhere, a brief overview is necessary. Ionization scheme? Quad or TOF detection? Can an ACSM measure refractory chloride? Is this an issue? Etc. Furthermore, why is there no mention to the TDMA yet? P7 Lines 15-16 aren't enough to justify leaving out basic information of the TDMA, especially if critical data has been obtained from it.

Methodology: As mentioned above, I'd merge this section with the previous one. Whilst the authors have derived equations rather clearly, and the flow of logic is very good in this section, one major comment for this section is to expand on the Fernald method, and to discuss why (7) and (8) are used. What are the advantages and disadvantages

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of one versus the other? Which one is more commonly used? Is one more accurate for specific conditions or wavelengths than the other? Also, in Section 3.3, no mention as to how NH_4^+ mass concentrations are obtained, nor any of the other supporting measurements. Why don't the authors rearrange information e.g. in Section 3.4. to here, but more importantly, why do the authors leave out so much information on the ACSM? Also, I got lost in clearly understanding what Case I and Case II are supposed to represent. The authors need to rearrange the writing so that one sentence can describe clearly the difference between the two. As I understand, Case I is a clean day and Case II is a hazy day, and that information is clear in the Abstract but not in the body of the document. The authors also need to draw clearer attention to the fact that Case I and II were chosen on days of similar water vapor content, which is hard to understand from the text.

Results and discussion: The major concern here is that no inferences are made, except for the wind directions and the possible air mass sources for the two cases. Describing the results is insufficient for this section.

-First paragraph of page 14: the authors describe their results and conclude 'good mixing atmospheric conditions' for both cases. They fail to explain why that's important for the measurements though. Please explain why good mixing is necessary, or what does it tell us. Parcel is stable? Important for data retrieval? Is there any mention to boundary layer height? How does dilution affect aerosol load / visibility? Why are heights for Case I and II different?

-First paragraph on page 15: overstated. Also, if the authors conclude the haze is likely (from lidar data e.g., Fig. 4 e, f, k, and l) due to hygroscopic growth, I would bolster my minor comment for Fig. 4 that the data visualization is weak to support this claim / conclusion.

-Figure 7: there is no decryption on how NH_4^+ has been predicated.

-P16 Line 21 – P17 Lines 1-2: I question the validity of this assertion. If I understand

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correctly, the authors conclude the aerosols in Case II are more acidic than Case I based on the regression slopes in Fig. 7. This is overstated, particularly if it comes only from one very short time window throughout the campaign. I would challenge it's 'consistent with the results presented'. What are the errors in the slope?

I think the results can support the conclusion, but data visualization needs to be clearer and include some error analysis of sorts, whether it be a confidence interval or standard deviation. Finally, equations for kappa evaluation need to be clearly stated with the proper values from ACSM, or clearly mentioned in the text.

Conclusion: No major comments that aren't addressed in the previous section.

Minor Comments:

P2 Line 1: Remove "particles" and make "aerosol" plural.

P2 Lines 3-4: Rephrase "...the hygroscopic growth effect..." to "...hygroscopic growth...".

P2 Line 4: What do the authors mean by "contributing factors"?

P2 Lines 4-5: "rich measurements" is poor phrasing in English. The sentence in general should be refined.

P2 Line 5: Include "a city" after the comma.

P2 Line 6: This may refer to multiple instances throughout the document, but I encourage the authors to double check any improper usage of words. "most serious" is a very poor word choice, please revise. A possible solution includes "...suffers from persistent haze...", but words to that effect are encouraged.

P2 Line 7: To stay in line with the tense of the Abstract, perhaps change "are" to "were". Also, was the lidar ground-based or satellite-based? If the lidar was ground-based, please rearrange the sentence or remove "ground-based instruments".

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P2 Lines 8-9: Perhaps add 'PM' as the acronym of particulate matter? Also, what type of diameter, presumably aerodynamic diameter?

P2 Line 11: I am not sure what the authors mean by "the evolution". Are they referring to aerosol growth?

P2 Lines 13-16: I'd describe Case I before Case II and remove the unnecessary colon and replace it with 'of', for example.

P2 Line 16: Maybe use the plural, 'were'?

P2 Line 17: Why is 'cases' not capitalized?

P2 Line 18: What is an aerosol acidity value?

P2 Line 22: Why keep both 'aerosol' and 'particle'?

P3 Line 2: I'd replace "Under the same water vapor conditions. . ." with "For similar ambient RH. . ."

P3 Lines 2-5: Please rephrase or merge the two sentences using simpler language.

P4 Line 2: Again, no need for "aerosol particles", please rephrase to "Atmospheric aerosols"

P4 Lines 2-4: Reconsider the citation – as well as sentence structure – since what the authors state is well-accepted. If a citation is necessary, one of the classical textbooks should do. Also, please avoid phrasing like "most important". The sentence could be rephrased, e.g.: "Atmospheric aerosols help regulate Earth's climate, mainly by scattering or absorbing incoming radiation" or words to that effect.

P4 Lines 4-6: Again, citation may not be necessary, and both arguments can be collapsed in one sentence and citing a classical aerosol / optics textbook.

P4 Lines 8-11: Please review sentence structure, because I'm assuming what the authors wrote is not what the authors mean. Hygroscopic growth is self-explanatory, the

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result however is that the scattering properties change (I would challenge the authors that it strictly increases scattering efficiency or scattering overall).

P4 Line 13: Remove “crucial” . . . as for that matter, the second half of the sentence is a repetition.

Lines 15-16: Please revisit the sentence structure, and rephrase e.g., “. . .water uptake. This growth can be detected by . . .”

P4 Line 15 – P5 Line 18: Please see my major comments. This should belong to the experimental section.

P5 Line 20: Please rephrase “. . .gain deeper insights. . .”.

P5 Line 22: Please rephrase “A specific goal. . .”.

P6 Lines 3-4: Rephrase “together with other suites of instruments measuring a variety of aerosol properties.” with “. . .coupled with supporting measurements.”

P6 Lines 12-14: Restructure the sentence by beginning with “Raman lidar was used. . .”.

P6 Line 15: This is important and may apply to more instances throughout the document. What does “aerosol optical property” mean? It is far too vague when describing data retrieval. Do the authors mean scattering efficiency? Scattering cross-section? Scattering intensity? Extinction coefficients? Please avoid these generalizations; they are not suited for publication and imply poor working knowledge by the authors.

P6, second paragraph: This needs to be revisited. Please see my major comments regarding this section.

P7 Line 1: Change “Collocated” to “Co-located”. Please check throughout.

P7 Line 4: Perhaps mention that the ascension velocity was ‘typically’ 5-6 m/s?

P7 Line 5: Again, please change “collocated” to “co-located”.

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P7 Line 17: BJT has already been defined so I'd encourage the authors to be consistent with their acronym usage and replace "Beijing local time" with this acronym.

P8 Line 10: What does "...signal is affected by radiation..." mean?

P9 Line 5: Please insert "reasonably" such that the phrase reads "...agree reasonably well..."

P9 Line 8: If the authors state a percent error for relative humidity, why do they state an absolute error for W? Also, in ref. to equations (3) – (5), can the authors be clear why they choose to display two separate figures for W and RH? Why not combine in one figure, or why report two separate ones at all? Is it for lidar retrieval validation? Unless the percent error for RH is the actual units, not a relative (percentage-weighted) error?

P9 Line 11: Delete the first sentence, it's redundant in my opinion.

P9 Line 16: Could the authors add a little more information regarding the Fernald method for the readers?

P10 Line 8: What is a "hygroscopic parameter"? Are they empirical fits? Do they have a physical meaning? The authors cite some literature, yet do not mention quite exactly what a and b are.

P10 Line 15: Again, vague wording like "...is a key parameter..." should be avoided. Furthermore, is the literature scarce on aerosol acidity and hygroscopic growth? I'd encourage the authors to find more relevant literature to cite.

P10 Line 16: "...aerosols in the atmosphere tend to be more hygroscopic than their neutralized form..." is awfully similar phrasing to the cited literature, Zhang et al., Environ. Sci. Technol., 2007. Apart from the awfully similar phrasing, isn't there a better reference? Might I suggest, but not limit to, Zhang et al., Atmos. Chem. Phys, 2015 (doi: 10.5194/acp-15-8439-2015)?

P10 Lines 17-18: Why does high hygroscopicity of aerosols enhance light scattering?

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Also, remove “particles” and make “aerosol” plural (check throughout). Finally, relate this to P4 Lines 8-11, are the authors being consistent?

P11 Line 21: Is chloride not considered because its concentration is extremely low or because the ACSM only measures non-refractory chloride? Is this even an issue for a city as inland as Xingtai?

P12 Line 15: Perhaps “e.g., Tobin et al., 2012”?

P12 Line 16: I would replace “temporal evolution” with “timeseries”.

P12 Lines 17-18: Is this a qualitative inference from the authors, or can they provide a correlation of sorts to support their claim?

P13 Line 3: Please rephrase “. . .and since. . .”.

P12 Lines 3-4: Citing one source hardly makes hygroscopicity ‘highly’ dependent on the composition of the aerosol. Please rephrase or support with data.

P12 Lines 5-6: Unclear. Are the authors implying that hygroscopic growth diluted the organic fraction (on a mass basis) detected by the ACSM?

P13 Line 17: “. . .cropped up.” is not appropriate. Please change.

P13 Line 18: Remove “a”, and I would challenge Case I and Case II help ‘fully’ understand the phenomenon.

P14 Line 4: Why are these altitude ranges chosen? If the point of the article is to assess haze as a health or visibility issue, wouldn’t it make sense to take data below the boundary layer? Or are these heights below the boundary layer?

P13 Line 21 – P14 Line 1: Please rephrase, awkward sentencing.

P14 Lines 2-3: Can the authors explain either now or in the previous section why the AE and depolarization ratio are useful? What do they tell us?

P14 Line 15: Fix “collocated” to “co-located”, as per previous comments, unless the

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doppler lidar is collocated somewhere? Also, no need to specify again the range of the 'region of interest'.

P14 Lines 13-20: Lots of repetition, unclear and inconsistent sentence structuring, and improper use of citation, and if the authors wish to keep the citation, a more appropriate description or analysis of said 'source region' is required.

P14 Line 21: Perhaps the authors can use a symbol (abbreviation) for the scattering coefficients, rather than the words?

P15 Line 12: Remove "Specifically".

P15 Line 20: Remove "distinctly".

P16 Lines 6-7: Is this an accurate representation for aerosols in Xingtai, Hebei, or Northern China overall, or it's just a generalization? I'm not sure in the current state Lines 6-9 are necessary in this manuscript.

P16 Lines 12-13: Remove sentence.

P17 Line 1: Again, please correct "aerosol particles".

P17 Lines 19-21: Again, this is overstated. How different are kappa value of 0.557 vs. 0.610? I would encourage the authors to use phrasing like 'suggests' or 'point towards', rather than definitive conclusions, which I don't think can be made from the presented data.

P17 Line 22: Remove "ion", fix "aerosol particles", and remove "significant".

P18 Line 3: Replace "Concerning the aerosol scattering enhancement factor, during the last decade, many..." with "In the last decade, many..."

P18 Line 4: Is the nephelometer an example? Or have all studied used the nephelometer?

P18 Line 6: Replace "for use in" with "for".

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P18 Line 7: Please fix “aerosol particles”.

P18 Line 12: Please fix “aerosol particles”.

P18 Lines 13-14: Is the ‘kappa model’ supposed to be capitalized?

Tables and Figures:

Figure 1: The radiosonde line does not look dashed to me, neither does it look dashed in the legend. Please amend how the authors see fit.

Figure 2: Please keep consistency with data display. Traces should appear like they do in Fig. 1. I don’t know if displaying the difference is useful, unless at those heights where the difference is marked, it implies poor lidar performance? If so, please reflect in the main text, because it is arguable how well they agree (as per main text, P9 Lines 4-5)

Figure 3: In the caption, please explain the missing data.

Figure 4: To be consistent with the text, change “Angstrom” to “Ångström”, and I don’t know if the heights for Cases I and II should be reported to 1 decimal place, unless that is instrument precision (appears so e.g., from P14 Line 4)? Finally, I don’t know how impactful this figure is visually if the x-axes are different for Case I and II. I would suggest either keeping x-axes consistent or overlapping the traces for the two cases in one plot. Differences aren’t obvious in the current display.

Figure 5: No major comments.

Figure 6: No major comments.

Figure 7: No major comments.

Figure 8: No major comments.

Figure 9: Please fix “Aerosol particle”, but more importantly, in the caption, explain this is not data, but a model based on Eq. 3 (as per the text, unless I’m mistaken).

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Tables 1-3: No comments.

Table 4: The results weakly support the conclusions of the document. I would encourage the authors to be more transparent with their data, perhaps in a Supporting Information section. Any simple errors to report, e.g. 95% confidence intervals? How was the raw data from the H-TDMA obtained? A timeseries to serve as example perhaps?

References:

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2018-753>, 2018.

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