

General comments:

This manuscript has introduced the simultaneous $f(\text{RH})$ and chemical measurements with a humidified nephelometer system and CV-ToF-ACSM for both PM_{10} and $\text{PM}_{2.5}$ conducted in the wintertime of 2018 in the north China plain. The bulk hygroscopicity parameter, κ , results were calculated from both the light scattering growth measurements and chemical compositions. The two types of bulk κ values were compared and discussed in detail, with a good comparison achieved for $\text{PM}_{2.5}$ measurements especially for polluted continental aerosol particles. Further, the authors innovatively proposed a new algorithm of deriving κ_{org} from the $f(\text{RH})$ and chemical data, favoring the direct measure of the water uptake by organic fractions with commercially available instruments and conventional data set. A pronounced diurnal pattern for κ_{org} was identified and presented for the first time in the northern China. The variation of κ_{org} was closely related to both f_{44} and f_{OOA} , signifying the importance of atmospheric oxidation processes on the water uptake by organic species. The findings reported in this study could serve as a good reference for modelling investigations on the climatic effects particularly driven by atmospheric organic aerosol particles in polluted continental regions.

The techniques used in this study are valid, and the reported hygroscopicity data are comparable to those in previous studies. In general, the quality of this manuscript is good yet could be improved, providing that some of the introduction and discussion contents (see the specific comments attached) were organized and delivered in a more logical/concise way. Also, some ambiguous expressions can be avoided, and a thorough grammar check is highly recommended. I would like to suggest its final publication upon a minor revision, with the comments specified as below.

Specific comments:

1. Page 3, line 58: "... lead to **40% changes** in predicted cloud condensation nuclei (CCN) concentration." The "40% changes" here is confusing, as which is difficult to tell from the sentence whether "a 40% increase or decrease in the N_{CCN} " was resulted from the "50% increase or decrease in κ_{org} ". The similar problem exists in the following sentence, which didn't state clearly the corresponding relationship between the average difference in aerosol radiative forcing and change in κ_{org} , e.g., which scenario ($\kappa_{\text{org}}=0.05$ or $\kappa_{\text{org}}=0.15$) corresponds to a higher radiative forcing? A straightforward delivery way is necessary to avoid ambiguity.

2. Page 3, line 65: Unlike the variation of κ_{org} itself, I'm afraid I didn't find any connection between the **importance of size-dependent κ_{org}** and the above-mentioned content. Some details and corresponding references are needed to support the importance of the size influence on κ_{org} and related climatic effects. I would recommend the authors to reorganize the context of size influence on κ_{org} , which can be combined with the information provided in the third paragraph (i.e., contents related to the HTDMA and CCN measurements).
3. Page 4, line 109: How is the ‘*mobility diameter of 800 nm*’ obtained? Related information and references are preferred for the conversion here.
4. Page 6, line 151: Based on the introduction of each instrument, only the humidified nephelometer can measure both PM₁₀ and PM₁. How can chemical compositions of PM₁₀ be measured with ACSM, which is designed with a PM_{2.5} aerodynamic lens/impactor as mentioned in Line 148? Similarly, how can SMPS measure the size distribution of PM₁₀?
5. Page 8, line 206: Why is the density for size conversion regarded as 1.7 g/cm³, the same as that of black carbon used in the calculation of κ_{org} ? According to the data reported, the organic fraction is always the predominant contributor to the particle mass of PM_{2.5}. This might suggest a smaller density for the ambient particles. Then how to evaluate the uncertainty in the related calculations?
6. Page 10, line 243: For the “*iterative calculation using the Mie theory*”, how are the chemical composition and corresponding mixing state of particles considered in the $\kappa_{f(RH)}$ calculation? This would affect the closure/comparison between derived $\kappa_{f(RH)}$ and κ_{chem} , thus the interpretation of representativeness of $\kappa_{f(RH)}$.
7. Page 11, line 266: I suppose that you were assuming black carbon as hydrophobic, rather than hydrophilic, and κ_{BC} is approximately taken as 0. Supporting references would be needed for this point and also for the density assumption of BC in Line 275. A similar typo was found for the description of ‘Dust’ in Page 12, line 309, which would be hydrophobic instead of hydrophilic.
8. Page 12, line 307: What does the ‘by’ mean: “... *influences of unidentified material by the ACSM ...*”? Are you suggesting ‘not detected by’ ACSM?
9. Page 13, line 313: “*Bulk aerosol chemical compositions and aerosol hygroscopicity $\kappa_{f(RH)}$ measurements are available, one would naturally jump to the conclusion of treating $\kappa_{f(RH)}$ as κ_{chem} to derive κ_{Org} (both are from bulk aerosol measurements).*”

A connection like a conjunction is needed for the whole sentence. “Both” here sounds ambiguous, although I would assume them to be $\kappa_{f(RH)}$ and κ_{chem} . It’s better to specify which two hygroscopicity parameters you were referring to.

10. Page 16, Line 380: Why is the $\kappa_{f(RH)}$ uniform for all the particle sizes? If yes, does it mean that $\kappa_{Dp} = \kappa_{f(RH)}$, while κ_{Dp} itself is size dependent?

11. Page 17, Line 387: What does the ‘**which**’ mean?

Line 388: Is there any consideration of choosing “two extreme cases of size-resolved κ_{Dp} ”?

12. Page 18, Figure 4(c): In comparison of the PM₁ scenario displayed in Fig.4b, $\kappa_{f(RH)}$ is generally higher than κ_{chem} , and larger discrepancies exist for the PM₁₀ case. Can you provide some hints for these results?

Line 411: “*How much does κ_{chem} differ from $\kappa_{f(RH)}$ for PM₁ and PM₁₀ samples?*” In my understanding, the κ_{chem} of PM₁₀ samples is calculated from the corresponding chemical compositions that are actually measured for PM_{2.5} instead of PM₁₀ (due to the configuration of ACSM with a PM_{2.5} impactor), when sampling with a PM₁₀ inlet. In this sense, the two hygroscopicity parameters for PM₁₀ samples would correspond to the water uptake by particles of different size ranges.

13. Page 19, Line 420: It feels like “*thus smaller particles play a more significant role in κ_{chem}* ” concluded here is a bit too early, as κ_{chem} is determined not only by the volume fraction but also by the hygroscopicity of each composition. Small particles with higher κ_{Dp} normally correspond to much higher κ values for both κ_{chem} and $\kappa_{f(RH)}$. Considering the much smaller variation range of κ_{chem} or $\kappa_{f(RH)}$ caused by Ångström exponents, influence from κ_{Dp} of smaller particles would be more significant.

Line 427: in Fig.4a, the variation ranges of κ_{chem} and $\kappa_{f(RH)}$ are much smaller than those in Fig.4b of PM₁. Can we say that the influence of the particle size distribution (as denoted by the Ångström exponent) is not that important for κ_{chem} or $\kappa_{f(RH)}$?

14. Page 20, Line 440: “... *with all κ_{chem} lower than $\kappa_{f(RH)}$.*” For the discrepancies between κ_{chem} and $\kappa_{f(RH)}$, is it also because that κ_{chem} is only derived from PM_{2.5} rather than PM₁₀ measurements? How to evaluate the effect of size-cut of ACSM especially for measurements with a PM₁₀ inlet?

Line 449: I guess ‘NR-PM_{2.5}’ and ‘NR-PM₁’ is reversely sequenced, similar to the orders of ‘PM₁₀’ and ‘PM₁’ in the following sentence of the same page and in Line 570 of Page 27.

Line 456: “*During the first period*”, is there any predefinition of the first/second or any other period (e.g., the “non-fog periods” in Page 22, Line 478)?

15. Page 26, Line 555: How is BBOA identified from COA, as the diurnal patterns of the two factors seem to be quite similar?

Technical corrections:

1. Page 2 Line 35: "...correlated with *f44* (fraction of m/z 44 in OA)"
Line 51: "...*air-pollution-related* health effects"
2. Page 3 Line 54: "...submicron aerosol particles *s* mass under dry state"
Line 70: "...come from *many* different natural and anthropogenic sources"
3. Page 4, line 82: "Studies on κ_{Org} ~~has~~ *have* already..."
Line 89: "(HTDMA) or CCN counter ~~were~~ *was* applied..."
Line 98: "...that κ_{Org} *increases* with the increase in particle dry diameter"
Line 101: Revise it into "In this study, the light scattering enhancement factors of *both* PM_{10} and PM_1 ..."
4. Page 5, line 115: "... the diurnal variation of κ_{Org} ~~are~~ *is* investigated"
Line 117: "...aerosol chemical compositions measurements ~~are~~ *is* proposed"
Line 134: "...PM impactor inlet, an MFC... and a pump ~~was~~ *were* added"
5. Page 6, line 138: "...the first PM_{10} inlet"
Line 148: "...chemical compositions *s* of $PM_{2.5}$ "
Line 150: "The inlets of group2 and group3 ~~switch~~ *es* every 15 minutes"
6. Page 7, line 166: "... after four days of continuous operation (~~3th~~ *3rd*, Dec) and..."
Line 174: "measured RHs ~~and~~ / temperatures at the inlet and outlet..."
7. Page 8, line 196: "The ~~m/z's~~ *of ion fragments of m/z* 38, 49, 63 and 66 were removed"
8. Page 9, line 222: "...averaged over each *15-minute* observation episode..."
Line 223: Change it into "...and *of* 30 minutes of SMPS, ACSM ..."
9. Page 11, line 281: "...but ~~in~~ *it*(?) is on average 30% lower ..."
10. Page 12, line 299: "...loss in semi-volatile ~~of~~ aerosol components. ACSM and the dry nephelometer had *a* similar tube length and nephelometer measurements *bears* less

uncertainty than SMPS.”

Line 305: “...and volume *fractions* of the unidentified material”

Line 309: “...~~were~~*was* not discussed before”

11. Page 13, Line 321: Delete the comma after ‘Eq.2’.

12. Page 14, Line 345: “... larger κ_{DP} generally *corresponde*s to higher...”

13. Page 15, Line 352: “The average PNSD of PM_{10} was applied in the simulation of the...”

Line 371: “... Q_{sca} can *be* expressed as $Q_{sca} = k \cdot D_p$ ”

14. Page 18, Line 400: “(which contribute most to σ_{sp} and ~~is~~*are* the part of the aerosol population *that* $\kappa_{f(RH)}$ is most sensitive to)”

Line 407: “Based on results of Eqs.8 and 20...”

15. Page 18, Figure 4: It’s difficult to tell that what the square/circle stand for? It’s preferred to point out briefly in the annotation, instead of just mentioned in the main text. In the annotation, “Gray areas represent the absolute relative differences between κ_{chem} and $\kappa_{f(RH)}$ ~~are~~ less than 10%.”

16. Page 20, Line 453: “... *where* quite clean conditions...”

Line 455: “...which show*s* that on average organics contributed most to the mass concentrations of NR- PM_1 and NR- $PM_{2.5}$ ”

17. Page 22, Line 472: “with *relatively* small differences...”

Line 474: “... ~~it~~*they* can still cause a small difference...”

Table 2: “Average (range) mass concentrations...”. Keep the same format for the names of the five species in the Table, e.g., initials in capitals.

Line 482: “(see PNSD examples in Fig.S4a)”

18. Page 23, Line 491: “~~were~~*was* a known parameter... which was~~the~~ calculated by...”

Figure 6: The unit of the f_{OA} as shown by the color bar should be %.

Line 494: “...overestimated the measured one when mass fraction*s*...”

19. Page 24, Line 514: “...Period 2 might provide us ~~with~~ a good opportunity...”

20. Page 25, Line 529: “hygroscopicity of organic aerosol generally *increas*e*s* as the oxidation level...”

Line 536: “*The time series of derived κ_{org} ~~is~~are depicted*”

Line 540: “*...changed from ~~near~~ly hydrophobic to moderately hygroscopic*”

21. Page 28, Line 581: “*...a constant κ_{org} ~~were~~was used ...*”

Line 585: “*near*ly hydrophobic...”

22. A consistent expression is always recommended in one article, while such inconsistency issues are commonly found in this manuscript. For instance, a subscript format needs to be applied for e.g., (NR-)PM₁, (NR-)PM_{2.5}, and PM₁₀. Different symbols like κ_{fRH} and $\kappa_{f(RH)}$ are used randomly. OA is defined as the abbreviation for both ‘organic aerosols’ and ‘organic aerosol’. Please check through the whole content and make corrections in all the necessary places.

23. Some shorten names (such as ACSM, NR-PM) should be defined earlier, i.e., when they appear for the first time.