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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Low hygroscopicity of organic material in anthropogenic aerosols under pollution episode in China" *by* Juan Hong et al.

Anonymous Referee #4

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This manuscript describes measurements of the hygroscopicity of ambient aerosol measured at a surburban site in the Pearl River Delta area of China. Using a Hygroscopicity Tandem Differential Mobility Analyzer (HTDMA), the hygroscopic growth factor (HGF) for 4 aerosol sizes was measured during a 5-week intensive observation period, while an Aerosol Chemical Speciation Monitor (ACSM) measured the chemical composition of the same aerosol population. This observation period included some clear and some polluted days, with air masses that were determined to come from congested and non-congested areas of southeastern Asia. The authors observe a bimodal distribution of HGF for all aerosol sizes, which they attribute to two distinct aerosol populations, one fresh and one aged. They examine the dependence of the HGF on factors such as aerosol mass loading and O:C ratio, and determine that the HGFs observed

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Discussion paper

here have a much weaker dependence on the O:C ratio than has been previously observed, and suggest that this may be related to the different chemical composition of the local emissions.

This is an interesting paper, and a good data set, and may be publishable in ACP, but I believe the authors need to address several major concerns prior to publication.

General issues:

This paper is attempting to compare many components of the measured aerosol: four different aerosol sizes, two different HGF modes, clean vs polluted conditions, and diurnal trends. It's a complicated set of comparisons, and different sections of the paper address different things. The reader would benefit if the authors would more clearly state what each section is comparing, and only include the most relevant comparisons. For example, the first paragraph of section 3.2 discusses the diurnal trends of the mean HGF, but quickly states that there are no significant trends – probably because in the next section we can see that the LH and MH modes have opposite trends, and the mean HGF, which is the average of the two, sees these trends cancel out. So why include the mean HGF at all? Another example is at the end of section 3.3, where the authors demonstrate that the HGF dependence is different in clean and polluted conditions. If this is true, the authors should be careful in the rest of the paper to distinguish between clean and polluted conditions in their other analyses.

Secondly, the authors should identify the primary message of the manuscript and more clearly describe this result. Is it that the hygroscopicity's lower-than-expected dependence on O:C is attributed to a higher concentration of organics with larger molecular weights? If so, the authors should discuss this further. Are there experimental measurements available to support this? If this is the main conclusion, what should the reader learn from the extensive look into the dependence on inorganics, on the diurnal averages, which is what the majority of the results section is about?

Specific issues:

Line 86 - 90: What is the relevant difference here between oxidation level and the oxygenation state? Insert a sentence here detailing why oxidation level is theoretically correlated with water uptake, since this is an important part of the results.

Line 90 - 92: It is stated that the knowledge of the dependency of hygroscopicity on oxidation level is unknown in urban China. Since this is the main focus of the paper, include a line indicating why this environment is different.

Line 152 - 173: More details about the HTDMA should be included in this section. The second DMA is operating in SMPS mode? How fast/frequent are the scans and therefore what is the time-resolution for retrieval of the HGF? How frequently does the first DMA cycle between the 4 diameter set points? How are doubly- and triply-charged particles that are transmitted by the first DMA handled? Are the particle size distributions plotted in the bottom frame of Figure 2 from SMPS scans by the first DMA or from some other technique?

Line 178: What are 'Ambient-improved' ratios? Either define this term or leave it out and direct the reader to a reference.

Line 184 - 186: Briefly state what the simplified approach is. Is all the BC assumed to be in PM1? Or a weighted fraction?

Line 188: The line "individual size bins" is confusing. I assume the authors are referring to the 4 sizes selected by the first DMA? Replace with something similar to "the ACSM measures only accumulation mode aerosol, and therefore the Aitken mode particles may have a different chemical composition".

Line 191: Briefly state what instrument was measuring the PM2.5 chemical concentrations. An AMS?

Line 209 - 212: What is the justification for assuming the aerosol is completely neutralized? What would the effect be on the results be if it were not completely neutralized?

Line 272 - 282: See comment in General Comments. The paragraph is perhaps unnecessary. What can be learned from looking at the diurnal profile of the mean HGF that isn't learned from looking at the MH and LH components separately?

Line 296 - 299: What is the justification for the assertion that the MH mode particle experience a decrease in HGF during the day because they are uptaking less photoreactive species. Do typical reaction rates or back-of-the-envelope calculations support this assertion? Which species are involved? If this is true, how do the authors reconcile the fact the O:C ratio sharply increases during the day, and this paper indicates that there is at least a somewhat positive correlation between O:C and HGF?

Line 305: The authors state here that Hong 2015 and Cai 2017 report that the boundary layer height has an effect on aerosol populations, but later on line 378, they suggest it doesn't. This disagreement should be addressed more fully.

Line 323: The authors state that they can only compare HGFs from the HTDMA and ACSM for larger particles. But they have also demonstrated that larger and smaller particles behave differently. The authors should address any hypotheses for how HTDMA and ACSM might agree for smaller particles.

Line 325: State why HGF is expected to positively correlate with the inorganics/(organics + BC) ratio.

Line 349 and 352: The authors state the percentages 64% and 21% in reference to the back trajectories without discussing where these numbers come from. Furthermore, more information about the trajectories would be helpful, such as error bars on those percentages.

Line 354: Is there an observed increase in ACSM organics on days when the trajectories indicate air masses are arriving from the inland areas? If not, why is that?

Line 390: Do the authors have a suggestion for why this trend (HGF depends on O:C more during clean days than polluted days) is observed? It seems like an important result, yet isn't discussed extensively in the conclusions. Additionally, why is the parameterization of the HGF-to-O:C relationship not done separately for clear vs polluted days?

Line 401: Is there an operational definition for suburban aerosol? Does this just mean an aerosol population that is somewhere between typical urban and rural characteristics?

Line 402 - 405: More detail about the residual fit should be added here. Is the ZSR prediction compared to all the HTDMA measured HGF? Of all sizes? Or just the polluted or clear days? Are different values derived depending on the subset of measured data to compare to?

Line 415: Why was the ACSM not measuring size-selected aerosol in this study, as was done in Yeung et al?

Line 425: More information should be included about how this parameterization was derived. What parameters were allowed to vary, and what was the parameter that was minimized? Is a R2 of 0.51 significantly better than 0.5? In the next paragraph, an improved parameterization is introduced by allowing SOA density to vary. Which parameterization is better? Why does the conclusion section only mention this first parameterization?

Line 430: What is the justification for stating that the hygroscopicity of organics isn't affected by the presence of inorganics?

Line 444: How are the authors accounting for error here? Presumably there is error in the measurement, which propagates through to the derivation of the parameterization.

Line 490: Have the authors plotted the HGF vs the concentration of certain inorganics? Say, vs ammonium sulfate or sulfuric acid to see if there is a larger trend for compounds known to be more hygroscopic?

Figure 2: Remove the dates from under each frame and just put them under the bottom frame. Color bar for the top four frames should be labeled. Additionally, it seems as

though the MH and LH modes both have diurnal cycles between <1 and > 4. If this is simply because the total number of particles has a diurnal profile, it would be easier to see this if it was normalized to the total number of particles. In the bottom plot, because there is only one point on the y-axis, it's hard to see that it's in log space. The boundaries (i.e. 10 -1000 nm) should be indicated, with ticks to show that it is logarithmic.

Figure 3: See Comment on line 272. It's possible that this figure is not needed.

Figure 4: Is this separately out for polluted or clear days? Why not?

Figure 5: What happens if these plots are made with MH or LH HGF instead of the mean?

Figure 7: The colors for these trajectories should be labeled more clearly, and described more fully in the caption and also in the manuscript. Do they represent one representative trajectory? Or a weighted average? What was the spread on those individual trajectories?

Grammatical/Minor:

Line 102: What does "purposes" mean here? Do you mean "properties"?

Line 107: PRD, not RPD

Line 155: Tan et al. 2013b doesn't appear to be in the listed references. Neither is Tan 2013a

Line 159: Why denote the dry mobility diameter as D0? Why not "Dp (0% RH)"?

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