Answers to Referee #4

The authors appreciate the time the reviewer has spent on our manuscript, assisting us to produce a higher quality, understandable publication. The requested comments and suggestions are addressed and introduced to the revised version of the manuscript.

General Comments:

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.

Reply: We shortened the paragraph discussing the diurnal variation of the mean HGF and restructured the whole section substantially as requested. We also made closure analysis for polluted and clean days similar as Sect. 3.4 to improve the Results and Discussion section with more detailed arguments. The new section was attached below in blue.

3.4.3 Closure analysis for polluted and clean days

A similar analysis for the hygroscopicity-composition closure similar to that in Sect. 3.4.1 was performed separately for the polluted and clean days. We kept adopting the previous assumption in Sect. 3.4.1 considering the size-dependent chemical composition of aerosols in the current section. The ensemble-mean HGForg value was quite close to each other between the polluted and clean days (HGForg=1.30 and 1.28, respectively), and each closure is shown in Fig.S3 (Fig. R1 in current response). These values are similar to the one previously determined (HGForg of 1.26) for the entire experimental period. A good closure was achieved during the polluted days with a substantially high R^2 value (0.82), whereas during the clean days, the ACSM-derived HGF did not correlate well with the one measured by HTDMA, indicating that other factors, such as the O:C ratio of organic material, might have affected the achievement of the closure.

We adopted an O:C dependent hygroscopicity of organic material in the closure analysis separately for the polluted and clean days. The resulting closure is illustrated in Fig. 12 (Fig. R2 in this response). Compared with the clean days, the hygroscopicity of organic material was found to be less dependent on the O:C ratio during the polluted days. This finding is consistent with the previous discussion on Fig.8, stating that the oxidation level had a relatively stronger influence on the HGFs during the clean days compared with the polluted days. This indicates that the organic compound, even with similar hygroscopicity may contain varying chemical species resulting from different sources or atmospheric processes during these two distinct periods. As previously stated in the manuscript, the aerosol particles appeared to have been from long-range transported during the polluted days, having a longer aging history. The organic material in these aerosol particles were fully oxygenated with a similar hygroscopicity, even for different O:C ratios. However, during the clean days, the aerosol particles were mainly from local emissions, or formed locally without complex aging histories. The changes in HGForg revealed the oxidation state of this locally-formed organic material.

3.5 Synthetic Comparisons

A number of field studies have examined the relationship between the hygroscopicity of organic compounds and their oxidation level for ambient aerosols from various representative organic aerosol sources (Chang et al., 2010; Chen et al., 2017; Duplissy et al., 2011; Hong et al., 2015a; Mei et al., 2013; Wu et al., 2013, 2016). The empirical relationship obtained from our results and these earlier studies are compared and described below in Fig. 12. It is important to note two aspects before our discussion. First, Eq. 4 considering a size-dependent chemical composition of aerosols is used here for comparison, as it has a wider application than Eq. 3. Secondly, the results from other studies shown in Fig. 12 were obtained using the hygroscopicity parameter (κ_{org}) (the left y-axis), while HGF_{org} (the right y-axis) is given in this study, both representing a quantitative measure of the hygroscopicity of organic material. A thorough description to derive hygroscopicity parameter κ from HTDMA-measured HGF is given by Petters and Kreidenweis (2007).

All listed studies show that the hygroscopicity of organic matters generally increases with an increasing organic oxidation level, with significant variance in the fitting slopes among all of the empirical relationships. For aerosols from near remote (Duplissy et al., 2011; Hong et al., 2015) or rural background (Chang et al., 2010) areas, covering little or no influence from anthropogenic activities, the value of O:C exhibits a stronger impact on the water uptake ability of organic materials. This indicates that the oxidation potential from photo-oxidation in the atmosphere of these backgrounds is a critical factor in determining the characteristics of organic materials. Similar to aerosols formed from biogenic precursors, the apparent O:C dependency on the hygroscopicity of organics is obvious for peat burning aerosols (Chen et al., 2017), mostly due to the complexity in the types of biomasses.

In the suburban or urban atmosphere of megacities in China (e.g. Beijing and Guangzhou), the hygroscopicity of organic material was almost constant as shown in this study and by Wu et al. (2016), being much less sensitive towards the variation in their oxidation level. It is not surprising to observe this similar O:C dependence on hygroscopcity of organic material in the rural background areas of Germany by Wu et al. (2013). This might be explained by the fact that their measurement site is located in central Germany where anthropogenic activities cannot be neglected. Wu et al. (2016) discussed that the addition of either an alcohol or a carboxylic function could both elevate the O:C ratio of the original organic aerosols. However, the corresponding hygroscopicity of these organic products may not be increased to the same extent compared with the increase in the values of O:C. This could be a possible reason to explain that the variation of O:C of organic aerosols is not necessarily responsible for the changes in hygroscopicity. In contrast, κ_{org} of aerosols at an urban site in Pasadena, California, in US exhibited a stronger increase with an increasing O:C ratio (Mei et al., 2013). They found that the relationship of their study is in line with that obtained from HTDMA measurements of SOA formed from 1,3,5-trimethylbenzene (TMB), a surrogate for anthropogenic precursors (Duplissy et al., 2011). They also deduced that the major components in SOA from TMB photooxidation are mainly mono-acids, which are quite water soluble. It is also interesting to observe that the results by Lambe et al. (2011) showed quite similar parametrization of HGForg and O:C dependence compared with the one for current study. They used a Potential Aerosol Mass (PAM) flow reactor to study the hygroscopicity of organic aerosols from the oxidation of alkanes and terpenoids, suggesting the precursors of our organic aerosols in this study might have similar properties or same origins as these compounds in their study. The comparisons of κ_{org} or HGF_{org} as a function of O:C within these aforementioned studies suggest that anthropogenic precursors or the photo-oxidation mechanisms, might differ significantly between the suburban/urban atmosphere in China and those in the urban background of West US. This may lead to a distinguished characteristics of the oxidation products in SOA and therefore to a different relationship between κ_{org}/HGF_{org} and O:C.

4. Summary and conclusions

The hygroscopic growth factor distribution obtained in the late summer of 2016 at Panyu CAWNET station in PRD region suggests that this suburban aerosol population with a strong anthropogenic influence was almost always externally mixed. The diurnal variation of the HGF of the LH and MH mode particles of four sizes suggests that the LH mode particles were probably from local emissions, whereas the MH mode particles had a longer aging history. During daytime, an external mixing of particles decreased due to the condensation of different gaseous species onto them, which was particularly obvious for Aitken mode particles. The contribution of different species with various water affinities to the particle composition determines the variation of the mean HGF in general. However, the oxidation level of organics appeared to influence the hygroscopicity of the suburban aerosols only slightly.

The stagnant meteorological conditions favored the accumulation of pollutants originating from coastal areas in the southeast China during the polluted days. During these days, the hygroscopicity of the organic aerosol fraction was estimated to vary little despite the variability of its oxidation level. The atmosphere was cleared by the air masses from the north during clean days.

The ACSM-derived HGF correlated better with the HTDMA-measured ones for larger particles (100, 145 nm particles) compared with smaller particles (30, 60 nm particles). From the closure analysis, considering the assumption of a size-dependent chemical composition of aerosols, a new relation between the hygroscopic growth factor of organic compounds and their oxidation level was obtained for the suburban aerosols over the PRD region during the experimental periods: $HGF_{org} = (0.32\pm0.01) \times O:C + (1.10\pm0.04)$. Clearly, a moderate hygroscopicity of organic materials, with values of HGF_{org} ranging between 1.1 and 1.3, was observed and it exhibited a weak dependence on the O:C ratio for the current study. Comparison of this relation between polluted and clean days indicate that the organic material even with similar hygroscopicity during these two distinct periods may contain varying chemical species resulting from different sources or atmospheric processes.

The PRD region as one of the densely populated areas in China represents a geographical location in Asia under the subtropical marine monsoon climate system. However, these issues obtained from our results above have been discussed very little earlier, which thereby reflects a general value of our contribution. The comparison with earlier studies regarding the relationship between HGF_{org} and O:C ratio indicates that there are substantial differences, but also some similarities, in the properties of organic compounds in aerosols among different environments, especially in urban areas. This motivates us to extend our measurement network in the future to understand better the generality of the relationship between the hygroscopicity and the oxygenation of the organic compounds.

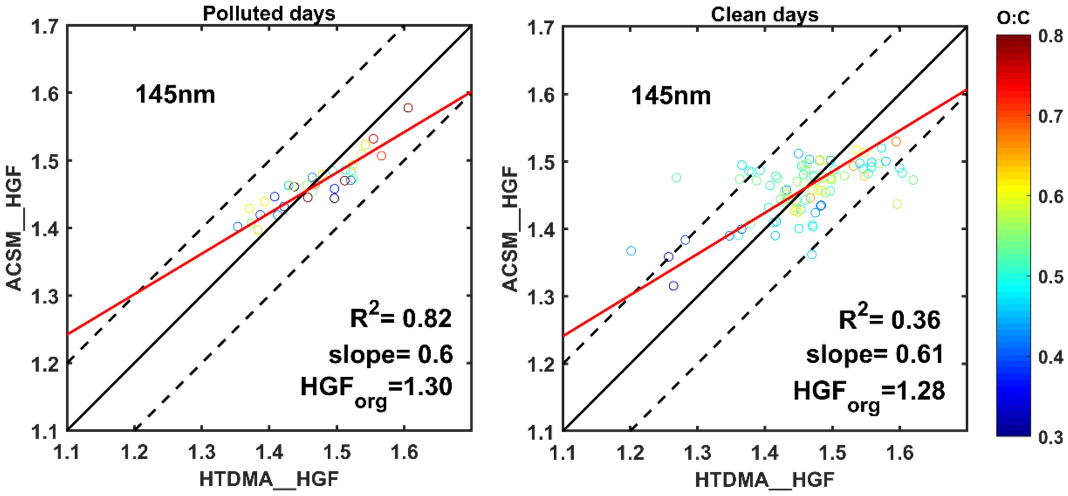

Figure R1: Closure study between the HTDMA-measured HGFs and the ACSM-derived HGFs by taking into account of size-dependent chemical composition of aerosols during polluted and clean days. The black solid lines indicate the 1:1 line and the black dash lines represent ±10% deviation, while the red lines are the lines fitted to the data points. The color bar indicates the O:C ratio of the organic aerosol fraction.

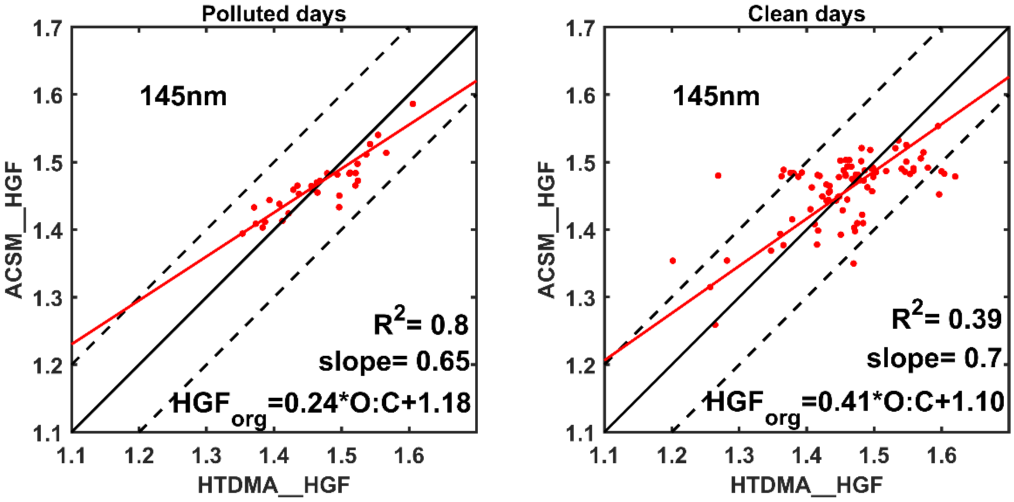

Figure R2: Closure analysis with the best fitting between the measured HGFs and the ACSM-derived ones using the O:C-dependent HGForg during polluted and clean days. The assumption of size-dependent chemical composition of aerosols was considered to determine the ACSM_derived HGF. The equation is the achieved approximation for HGForg as a function of the O:C of organic aerosol fraction.

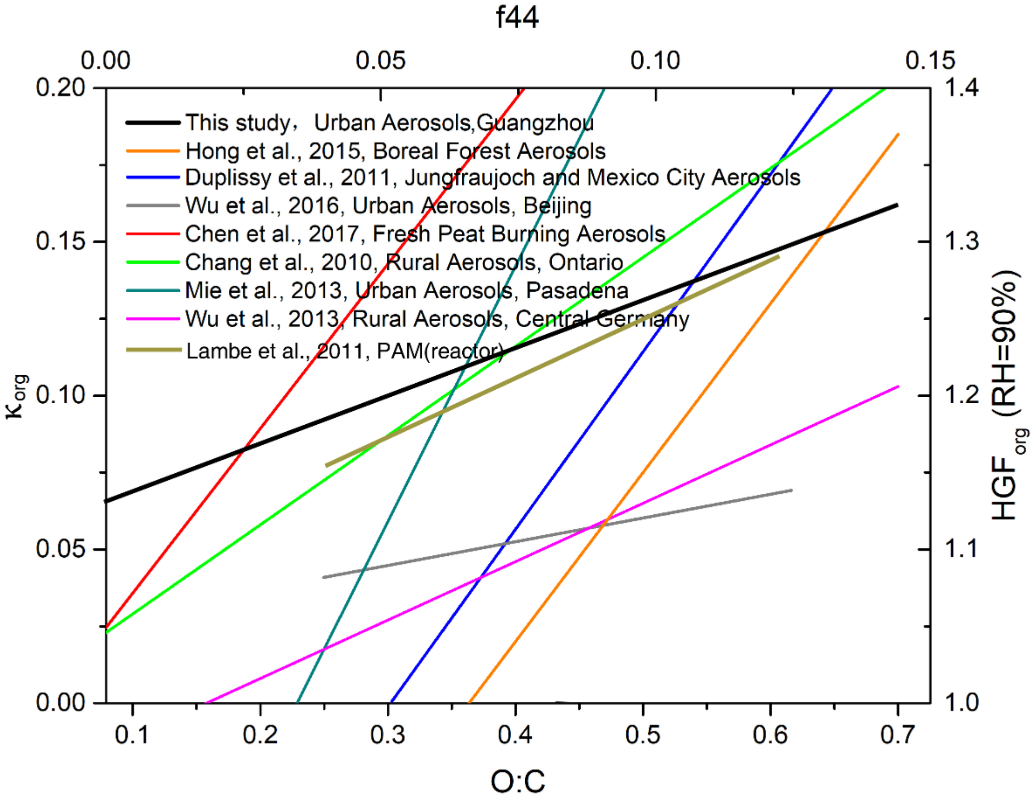

Figure R3: Comparison with earlier studies on the hygroscopicity of organic material with atomic O:C ratio (or *f44* from chemical composition data) obtained from different environmental background areas. Other studies were using derived κorg, while this study is using HGForg for the hygroscopicity of organic material.

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?

Reply: According to the other Referees' comments, ACSM-derived HGF should not be directly compared with the one measured by HTDMA, since ACSM measured the bulk aerosols while HTDMA measured the HGF of size-resolved particles. We therefore considered the differences of mass fraction of each component between PM1 and 145 nm particles obtained from Cai et al. (2017), which measured the size-resolved chemical composition of ambient aerosols obtained from the same measurement site during the same season of 2014 and applied this assumption with certain uncertainties into current study to make further evaluations. A newly determined parameterization of HGForg and its O:C dependency was given in the revised manuscript. We also performed a comprehensive uncertainty analysis for the hygroscopicity-composition closure and gave potential reasons from other sources of errors associated within this study. After taking into account of the size-dependent chemical composition of aerosols, HGForg value was shifted from 1.10 to 1.26. We hence feel our previous discussion and arguments were improper given. Hence we deleted related arguments at several places and modified the body text. However, even considering the size-resolved chemical composition of aerosols, we still observed the hygrosocpicity of organic compounds in current study has a relatively low O:C dependence. This finding is not completely unexpected, as Wu et al. (2016), which studied the particle hygroscopicity in the urban atmosphere of Beijing, also observed similar relation between HGForg and O:C ratio of ambient aerosols. Wu et al. (2016) discussed that the addition of both alcohol and carboxylic functions could elevate the O:C ratio of organic aerosols but form species with different hygroscopicities. This could be a possible reason to explain that the variation of O:C of organic aerosols is not necessarily responsible for the changes in hygroscopicity. This comparison and possible discussions were given in the revised manuscript.

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.

Reply: The oxygenation state is a more robust measure of the degree of oxidation for organic aerosols, while oxidation level, represented as O:C, sometimes may be affected by hydration and dehydration processes taking place in the atmosphere. We added a sentence into the text: They found that the oxidation level or the oxygenation state of the entire organics, which directly affects their corresponding solubility in water, is the major factor drives the water uptake ability of the organic fraction in aerosols.

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.

Reply: However, knowledge on the hygroscopicity of organic material and its dependency on the oxidation level of organics in urban background areas under high aerosol mass loading conditions, for instance, in China, where air pollution has become one of the top environmental concerns (Chan et al., 2008), is limited.

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 triplycharged 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?

Reply: Before operating in HTDMA mode, the second DMA was bypassed. The aerosol particles, after being introduced into the first DMA, was directed into the CPC to measure the number size distribution of the ambient aerosols. This is how we obtained the SMPS scans. Hence, the bottom frame of Fig. 2 in the manuscript is directly from our HTDMA system when SMPS mode is on. The time resolution for a whole scan including the SMPS scans and HTDMA scans of particles of 4 different sizes are around one hour. Multiply charge correction was performed during data inversion procedure according to the method introduced by Gysel et al. (2009).

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

Reply: We changed the sentence to: 'The oxygen to carbon (O:C) were then estimated by their relationship to the mass fractions of m/z44 (f44) to the total organic mass according to Canagaratna et al. (2015).'

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

Reply: Wu et al. (2009) compared the BC concentration in PM1 and PM2.5, respectively, and found that BC aerosols mainly exist in the fine particles with roughly 80% of the BC mass in PM1. Due to the limited literature data on BC size distributions in the PRD region, we used this simplified assumption by Wu et al. (2009) to estimate the BC concentration in PM1 for this study.

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".

Reply: We changed the sentence to: 'It is necessary to note that the ACSM measures the chemical composition of bulk aerosols, which may be significantly different from those of Aitken mode particles.'

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

Reply: The PM2.5 mass concentration was measured by an Environmental Dust Monitor (EDM, Grimm model 180).

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?

Reply: We are sorry that this assumption is based on previous studies (Gysel et al., 2007), which used similar ion paring scheme, and we do not fully understand the effect if the aerosols are not completely neutralized. However, we hypothesize that if our aerosols are more acid, the HGF would be higher than the predicted one as inorganic acid is more hygroscopic than its neutral form.

Line 272 - 282: See comment in General Comments. The paragraph is perhaps un necessary. 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?

Reply: We deleted this paragraph as suggested.

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?

Reply: I did not say they uptake less photoreactive species but rather organics from stronger photoreaction during daytime. These newly formed organics are expected to be less hygroscopic than those aged organics, which were already present in the aerosols. Here, these arguments are only related to MH mode particles, however, taking into account the mean HGF of the whole aerosol population as Fig.3 in the manuscipt, HGF values were actually higher during daytime, which is consistent with the increase in O:C during daytime.

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.

Reply: We deleted the sentence used in Line 305, but kept the one in line 378, since the logic in line 378 is more reasonable.

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.

Reply: Size-dependent chemical composition should be considered into the derivation of HGF by ACSM data, thus the HTDMA_measured HGF then could be compared with the one from ACSM.

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

Reply: Inorganics are commonly quite hygroscopic, while relative to inorganics, organic and black carbon are less hygroscopic or non-hygroscopic. Hence, the more inorganics in the aerosol phase, the larger HGF and the more organics or BC in the aerosols, HGF is relatively smaller.

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.

Reply: The method we used here was called cluster analysis. The details of this method could be found in http://ready.arl.noaa.gov/HYSPLIT.php. Thus only a brief introduction would be given here. The trajectories that are near each other are merged to a mean trajectory to represent those groups. When the clustering is complete, the change in the Total Spatial Variance (TSV) as the trajectories is merged into one cluster. The TSV is computed from the position vectors for the individual trajectory and its cluster mean trajectory. According to the change of TSV, we decide the number of cluster as an appropriate solution (In this work, the number is 5). The percentage of the cluster means that how many back trajectories can be represent by this cluster. According to the principle of cluster analysis and the limit of this model, the standard deviation of those percentages is not available from the model calculation directly.

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?

Reply: This is not clearly seen in current study, as the meteorological conditions also influence the concentration of organics in aerosols, for instance, the stagnant weather conditions during Sep 22 to 27 with low wind speed favor the accumulation of atmospheric pollutants including the mass concentration of different species. However, the air masses were mainly from coastal areas.

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?

Reply: As previously stated in the manuscript, during polluted days, the aerosols appeared to be from long-range transported, having longer aging history. The organic material in these aerosols were fully oxygenated, even with various O:C ratio. However, during clean days, the aerosols was mainly from local emissions or formed locally without complex histories. The changes in HGForg revealed the oxidation state of theses locally formed organic material. The closure analysis to determine the parameterization of the HGF-to-O:C relationship was done separately for clean and polluted days as above.

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?

Reply: Yes, compared to urban area, it has a lower density area that separate residential and commercial areas from one another. It could be part of the urban area or exist as a separate residential community within commuting distance of a city.

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?

Reply: The ZSR prediction is compared with all the HTDMA-measured HGF. Moreover, this is also done for the polluted and clean days. This is included in the revised manuscript.

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

Reply: ACSM is an instrument that cannot measure the size-resovled chemical compositon of aerosols, while Yeung et al. (2014) used a High Resolution Time-of-Flight Aerosol Mass Spectrometer, which is capable to obtain the size-segregated chemical composition information. Unfortunately, during our experimental period, we did not have HR-ToF-AMS.

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?

Reply: We gave a revised section illustrating the closure analysis, which should be much easier to understand and more reasonable. In addition, we did not include other parametrization in the conclusion, but did a comprehensive uncertainty analysis to discuss the potential errors associated within the closure analysis.

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

Reply: As the results shown, HGForg was actually quite constant during the whole study, while the concentration of inorganics varied. This is an indicator that the hygroscopicity of organics in our study is not observed to be 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.

Reply: We performed a comprehensive uncertainty analysis for the hygroscopicity-composition closure and gave potential reasons from other sources of errors associated within this study, which is shown in the revised manuscript.

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?

Reply: This is actually illustrated in Fig. 5 and Fig. 8. However, to plot the HGF vs. the concentration of ammonium sulfate may not indicate the complexity of ambient aerosols, as there was also substantial amount of organic species, which is less hygroscopic than ammonium sulfate. Even with increasing amount of ammonium sulfate concentration in aerosols, the increase of organics will cancel out the effect on HGF. This is the reason to plot HGF vs. Inorganics/(Organics+BC) is more reasonable.

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.

Reply: I modified the figure as suggested.

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

Reply: I deleted this figure and the corresponding text in the revised manuscript.

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

Reply: We put this figure in the supplement.

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

Reply: We understood the reason why the referee asked us to do this. However, the particle phase chemical composition data as well as the O:C ratio is the mean value of the bulk aerosols. Information of the chemical composition of each mode is not available. Poor correlation was observed when plotting the MH and LH HGF with the bulk aerosol composition.

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?

Reply: These trajectories are the clusters, which represent a group of back trajectories. The meaning of cluster has been answered in the former comments.

Grammatical/Minor:

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

Reply: I changed to 'properties'.

Line 107: PRD, not RPD

Reply: Yes, thanks for pointing it out and we revised it as suggested.

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

Reply: I added them into the reference list in the revised manuscript.

Line 159: Why denote the dry mobility diameter as D0? Why not "Dp (0% RH)"?
Reply: It is a conventional way to denote the dry mobility diameter as D0, hence, I keep it as it is.

References:

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