Answers to Referee #3

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

The authors present a study of low hygroscopicity of organic material in anthropogenic aerosols under pollution episode in China. The data set is rich, but the manuscript has two major deficiencies that should be addressed prior to considering further review. (1) The upper particle size ranges detected by different instruments are quite different: 145 nm for HTDMA, 1um for ACSM and 2.5 um for Aethalometer. Before drawing any conclusions, the authors should consider the uncertainties caused by different size ranges of particles being measured when integrating all dataset. (2) Most of previous studies showed that the hygroscopic growth factors of secondary organic aerosols are below 1.2. For example, the water-soluble organic carbon measured by Martin Gysel et al., has a hygroscopic growth factor of up to 1.17. In this study, the HGForg below 1.1 is actually comparable to those in other observations. Therefore, the low hygroscopicity of organic material cannot be a compelling result. Reference Gysel, M., et al., Hygroscopic properties of water-soluble matter and humic-like organics in atmospheric fine aerosol. Atmos. Chem. Phys., 2004. 4(1): p. 35-50.

Reply: Yes, the Reviewer is right that the upper particle size ranges detected by different instruments are quite different. For BC measurements, 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. In addition, the ACSM-derived HGF is not sensitive to the change in BC mass concentration, which was also discussed in the uncertainty analysis in the revised manuscript; see the attached Section (text in blue).

For the other chemical components measurements, ACSM only measured their mass concentration in PM1, which should not be compared directly with the one measured by HTDMA with a certain size. However, size-resolved chemical composition information of ambient aerosols for current study is not available. Cai et al. (2017) measured size-resolved chemical composition of ambient aerosols obtained from the same measurement site during the same season of 2014. Their results showed that the average organic mass fraction of PM1 were about 16% lower than that of 145 nm particles. We therefore considered the differences of mass fraction of each component in between PM1 and 145 nm particles obtained from Cai et al. (2017) and applied this assumption with certain uncertainties into the current study to make further evaluations. A newly determined parameterization of HGForg as a function of O:C ratio 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.

For the second comment: After taking into account of the size-dependent chemical composition of aerosols, HGForg value was shifted from 1.1 to 1.26, which was close to the one as 1.18 used by Yeung et al. (2014) measuring the hygroscopicity of ambient aerosols in September 2011 at the HKUST Supersite, less than 120 km away from our measurement site. I agree with the reviewer that the determined hygroscopicity of organic material is not significantly different from the ones from other studies. However, without identifying the hygroscopicity of organic material for current environmental background, one should always use the assumed parametrization for the hygroscopicity of organic material from other environments with big cautions. In addition, this study is, to our knowledge, the first time to identify the hygroscopic properties of the organic material and their O:C dependency, which may help us understand the chemical composition, sources and aging processes of atmospheric aerosols in this region. **After considering the difference of chemical composition between size-resolved aerosols and bulk ones, we feel the previous title might not fit for the current study and we changed it to 'Mixing state and particle hygroscopicity of organic-dominated aerosols over the Pearl River Delta Region in China'.**

The revised section is attached as below:

Section 3.4

3.4 Hygroscopicity-composition closure

3.4.1 Approximations of the HGF_{org}

Hygroscopic growth factors of organic compounds in the ambient aerosols, HGF_{org}, cannot be determined from direct observations. However, by conducting closure analysis using different approximation approaches, HGForg was estimated to range widely from about 1.0 to 1.3 for various ambient aerosols in other studies (Gysel et al., 2004; Carrico et al., 2005; Aklilu et al., 2006; Good et al., 2010; Hong et al., 2015; Chen et al., 2017). In this section, we performed a closure study between the measured and predicted HGF using a PM1 bulk chemical composition from the ACSM. An ensemble-mean HGF_{org} of 1.1 was determined when the sum of all residuals (RMSE, root mean square error) between the measured growth factors and corresponding ZSR predictions reached a minimum by varying HGF_{org} between 1.0 and 1.3.

By applying HGF_{org} of 1.1, Fig. 9 (Fig. R1 in this response) compares the ACSM-derived HGF with the HTDMA-measured ones for four different-size particles, with the color code indicating the O:C ratio. It is obvious that the degree of agreement increased with increasing particle size. However, the bulk aerosols mainly represent the chemical composition of aerosol particles near the mass median diameter of the mass size distribution of ambient aerosol particles (Wu et al., 2013). The question then arises as to which extent the size-resolved chemical composition of aerosols (for instance, 100 nm and 145 nm particles) is comparable with the one of the bulk aerosol. Previous studies (Cai et al., 2017; Cai et al., 2018) reported that the average organic mass fraction of PM1 were about 25% and 16% lower than those of 100 nm and 145 nm particles respectively measured by High-Resolution AMS (HR-AMS) during the same season of 2014 at the same measurement site. Correspondingly, the average inorganic mass fraction of PM1 were about 25% and 16% higher than those of 100 nm and 145 nm particles obtained in their results. Due to insufficient information of the size-resolved chemical composition of ambient aerosols, we hence made an arbitrary assumption by applying the results from Cai et al. (2017). In this section, we considered the mass fraction of organic being 25% and 16% higher and a corresponding lower inorganic mass fractions (ammonium sulfate mass fraction is decreased) at smaller sizes (100 nm and 145 nm) compared to the bulk aerosol. In addition, we assumed a 20% uncertainty in theses suggested values, thus resulting in 25%±3% and 16±3% of elevation in organic mass fractions in the 100 nm and 145 nm particles for current study. This would lead to larger values of HGForg as 1.23 (100 nm particles) and 1.26 (145 nm particles) when assuming different chemical compositions of size-resolved particles compared to the bulk aerosols, see Fig. 10 (Fig.R2 in this response). In contrast to the results from bulk chemical composition, the closure for 100 nm particles considerably improved, as the RMSE value between the HTDMA_HGF and ACSM_HGF decreased from 1.61 to 0.87 with more than 90% of the data were within 10% closure. The closure for 145 nm particles did not show any significant improvement, with no reduction in the RMSE value. However, the newly-determined HGForg is expected to be more accurate than the one reported in the previous section, as assumptions of size-dependent chemical composition was considered even though with some uncertainties. In addition, the newly-obtained HGForg was close to the one (1.18) by Yeung et al. (2014), who studied the hygroscopicity of ambient aerosols in September 2011 at the HKUST Supersite, less than 120 km away from our measurement site.

Previous studies suggest that a single ensemble HGF_{org} approximation might not be capable of evaluating the hygroscopicity of ambient aerosols from different sources with various characteristics. Hence, the HGF_{org} approximation according to the O:C ratio was tested using the chemical composition of both bulk aerosols and size-resolved particles based on previous assumptions, respectively. To facilitate our comparison, the closure analysis was only conducted for the 145 nm particles. The relation between HGForg and the O:C ratio based on the chemical composition of bulk aerosols was obtained as follows:

$$HGF_{org} = 0.31 \cdot O{:}C + 0.88. \tag{3}$$

This closure was no better than the one shown in Fig. 9 using constant a HGForg, both being based on the chemical composition of bulk aerosols, and there was little change in the RMSE value (from 0.63 to 0.62). By taking into account of the variation of the O:C ratio, HGF_{org} ranged from 0.9 to 1.2 when using Eq. 3 with around 80% of the data having values larger than 1. This finding implies that the approximation in Eq. 3 may introduce huge errors, as 20% of the values of HGF_{org} were not physically correct. The closure considering size-dependent chemical composition of aerosols from previous assumptions is shown in Fig. 11 (Fig. R3 in this response), with a new relation between HGForg and the O:C ratio as:

$$HGF_{org} = (0.32 \pm 0.01) \cdot O{:}C + (1.10 \pm 0.04). \tag{4}$$

The closure was somewhat better than in Fig. 9 according to the slightly lower RMSE value (0.58 vs. 0.63). In addition, HGForg ranged from 1.1 to 1.4 with the varying O:C ratio, and there were no HGForg values smaller than unity, indicating that the new relation in Eq.4 seems more widely applicable than the one in Eq. 3. In general, by looking at the fitted slopes being much less than unity with consideration of all the discussion above, we are concerned that other potential uncertainties may remain in the closure analysis between the measurements and predictions. This motivates us to make a comprehensive uncertainty analysis of the hygroscopic-composition closure. It is important to note that the uncertainty analysis below is taking into account the aforementioned assumption regarding the size-dependent chemical composition of aerosols.

3.4.2 Uncertainties of hygroscopicity-composition closure

Swietlicki et al., (2008) discussed the sources of error associated with HTDMA measurements and concluded that the stability and accuracy of DMA2 RH should be controlled well to maintain the nominal RH (for instance 90%). The accuracy of DMA2 RH in our system was controlled to be 90±1%. This will result in a variability in the measured HGF of ±0.04 around the reported HGF. The bias uncertainty (2.3%) associated with RH accuracy are generally smaller than the estimated uncertainty (10%) reported in HTDMA measurements (Yeung et al., 2014). For hygroscopicity-composition closure, this biased HGF will lead to a change of 2.1% in HGForg with respect to the previously-determined value of 1.26.

Other uncertainties pertain to the densities used for organic materials and black carbon. The density value is estimated to range between 1000 and 1500 kg/m^3 for organic materials (Kuwata et al., 2012) and 1000 and 2000 kg/m^3 for black carbon (Sloane et al., 1983; Ouimette and Flagan, 1982; Ma et al., 2011). The calculated uncertainty in the ACSM-derived HGF using the density value at each extreme for organic materials and black carbon is less than 3.2% and 2%, respectively, both having relatively small effect on the determination of the constant value of HGF_{org}.

Another source of uncertainty comes from the measurement of aerosol mass concentration performed by the ACSM and Aethalometer. Bahreini et al. (2009) did a comprehensive uncertainty analysis on aerosol mass concentration measurements using an Aerosol Mass Spectrometer (AMS), having similar operating principle as the ACSM, of which systematic biases are not available. Their study reported an overall uncertainty of 30% for AMS measurements and concluded that it might be better for ground-based studies. Jimenez et al. (2018) gave accuracies of 5-10% from other AMS practitioners and claimed that these estimated accuracies might be too small. Hence, we used an overall uncertainty of 20% for the mass concentration measurements in this study. The uncertainty in the BC measurements given by the manufacture of the Aethalometer is within 5% (Hansen et al., 2005; Zhang et al., 2017). The effect of the perturbation in aerosol mass concentration of each species on the ACSM-derived HGF as well as the determination of HGF_{org} are summarized in Table. 2 (Table 1 in current response). The change in the mass concentration of sulfate exerts the largest effect on the ACSM-derived HGF as well as the corresponding HGForg, which is not surprising since sulfate contributes the highest fraction in more hygroscopic component in aerosols.

In general, uncertainties were relatively low for each individual case discussed above. It is possible that the contribution from multiple factors could reduce the overall uncertainties. The greatest uncertainty aforesaid may still arise from the chemical composition of size-segregated aerosols, since the performance of the closure and the approximations of HGForg were most sensitive to changes in the mass concentration of sulfate and organic materials in aerosols. Except for the reasons discussed previously, other factors may also cause potential effects on the hygroscopicity closure. Pajunoja et al. (2015) showed that phase state of organic aerosols, which varies with ambient conditions, might have an effect on the determination of hygroscopicity of organic fraction in aerosols. Previous studies (Suda et al., 2014) suggested that the interaction between inorganic and organic materials within the particle phase might alter the hygroscopicity of organics in mixtures and speculated that ZSR mixing rule may not hold for inorganic dominated aerosols (Hong et al., 2015).

Nevertheless, the interpretation of the hygroscopicity-composition closure and different approximation of HGF_{org} above reveals that in order to estimate accurately the properties of ambient aerosols, we might need to have precise measurements of chemistry, including the size-dependent chemical composition of the aerosols, as well as a better prediction model for HGF.

Table 1. Sources of uncertainties associated within hygroscopicity-composition closure, given in terms of three standard deviation and their corresponding contribution to the overall uncertainty in hygroscopicity-composition closure.

Parameter	Uncertainty (3 standard deviation)	Uncertainty in measurements	HGForg (relative to 1.26)
RH (DMA2)	1%	2.3% in measured HGF	3.2%
Organic density	18%	2.6% in ACSM_derived HGF	3.2%
BC density	33%	1% in ACSM_derived HGF	2%
NH4, NO3 mass concentration	20%	0.6%, 0.5%	0.8, 1.6%
SO4 mass concentration	20%	1.8%	4%
Organics mass concentration	20%	1.4%	3.2%
BC mass concentration	5%	0.1%	0.8%

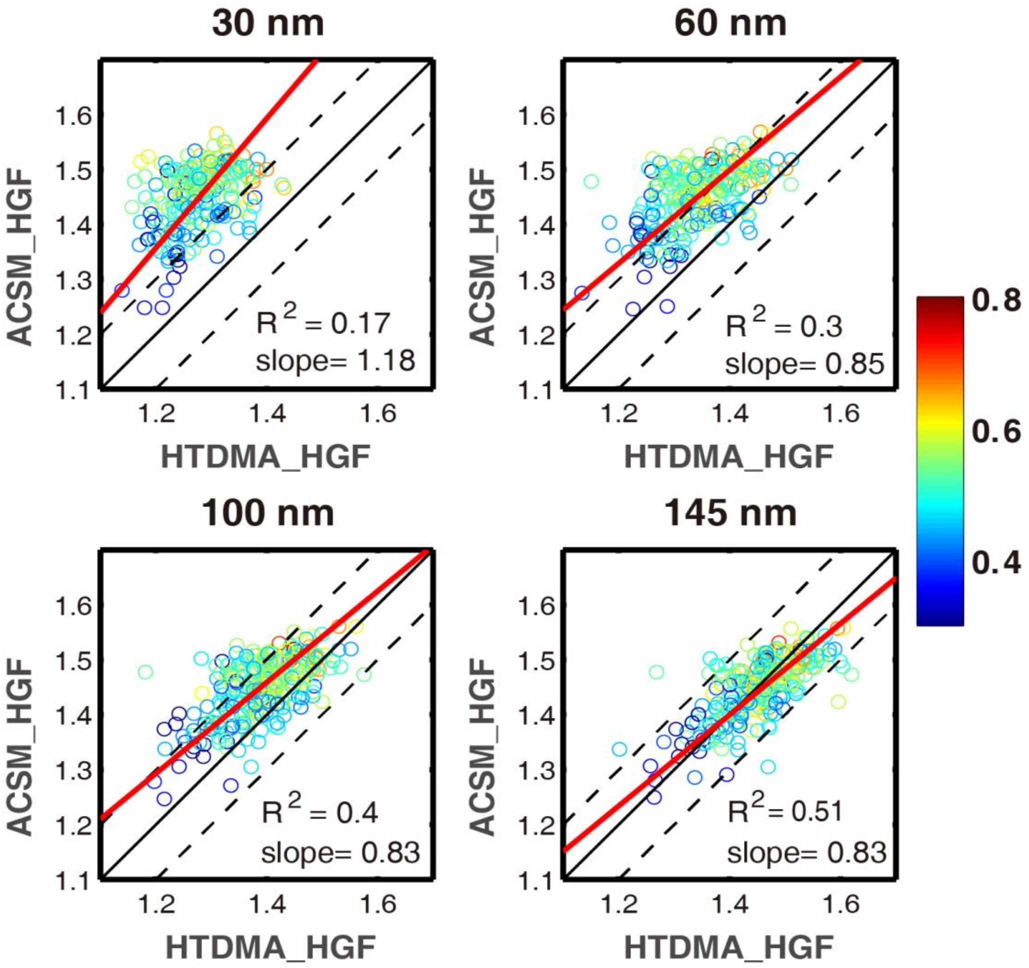

Figure R1: Closure study between the HTDMA-measured HGFs and the ACSM-derived HGFs. The black solid lines indicate the 1:1 line and the black dash lines represent ±10% deviation, while the red lines are the lines fitted to the data points. The color bar indicates the O:C ratio of the organic aerosol fraction.

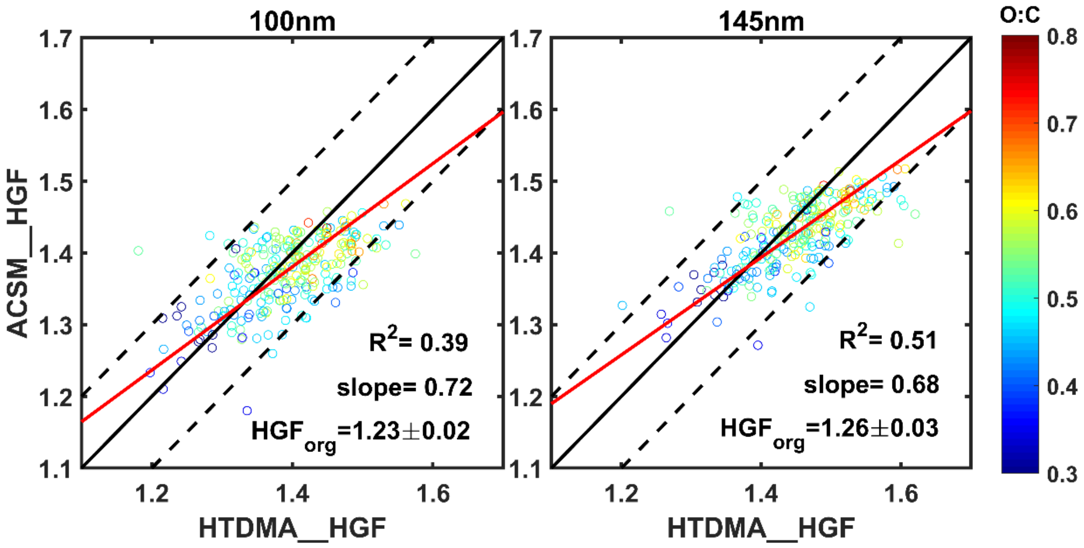

Figure R2: Closure study between the HTDMA-measured HGFs and the ACSM-derived HGFs assuming the average inorganic mass fraction of PM1 were about 25%± 3% and 16% ±3% higher and the average ammonium sulfate mass fraction of PM1 were about 25%± 3% and 16% ±3% lower than those of 100 nm and 145 nm particles. 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.

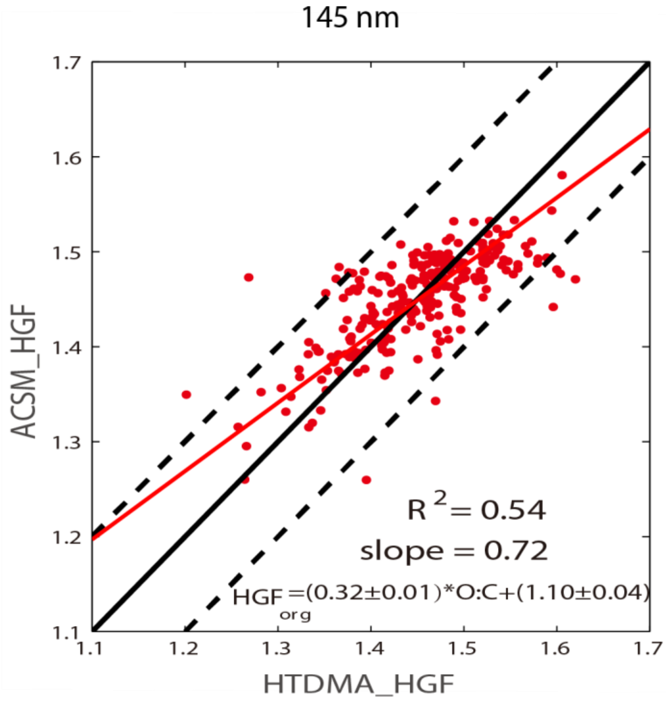

Figure R3: Closure analysis with the best fitting between the measured HGFs and the ACSM-derived ones using the O:C-dependent HGForg. The assumption of size-dependent chemical composition of aerosols was considered to determine the ACSM_derived HGF. The equation is the achieved approximation for HGForg as a function of the O:C of organic aerosol fraction.

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