Answers to Referee #5

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 manuscript presents results of simultaneous measurements of aerosol hygroscopicity and chemical composition in suburban site in Southern China. The measurement period covers almost 1 month and both polluted and relatively clean conditions were observed. I admit that such measurement can be very costly and labor intensive, and therefore the comprehensive set of data presented in the manuscript may carry certain value for the scientific community. However, with the current state of the manuscript, authors' main conclusion is very difficult to sink in for the readers. Authors seem to persist on determining the HGForg and large part of the manuscript is dedicated for that. However, in my opinion, it is obvious from the results that the oxidation level of organics does not affect the hygroscopicity of the suburban aerosols very much, and that might pretty much be the end of the story for HGFrog. Instead, I would like to see much more in-depth discussion on diurnal variations of LH mode in smaller particles (30 and 60 nm) and how the new particle formation and subsequent growth affects the aerosol hygroscopicity. I therefore recommend that the manuscript may be acceptable for publication in ACP after major restructuring.

Reply: We thank the referee's comments regarding the results of our manuscript. The referee is correct that the new particle formation and subsequent growth affects the aerosol hygroscopicity. At the referee's request, we did a case study of particle hygroscopicity during an NPF event.

Figure R1 shows the time series of particle number concentration, GF-PDFs of 30 and 145 nm particles and the bulk aerosol chemical composition of PM1 during 04. October, 2016, during which an NPF event was observed. The NPF event started at around 10:00 am, after which we observed a substantial increase in the number fraction of MH mode particles from 0.5 to around 1, for 30 and 145 nm particles. This indicates a clear conversion of particles from externally to internally mixing. Number fraction of the MH mode for 30 and 145 nm particles decreased again to 0.5 around 17:00 pm, which might be explained by the traffic emissions during the rush hours of the day. Meanwhile, HGF of MH mode of both 30 and 145 nm particles showed a slight decrease after the NPF event started. This suggests that the candidate for the material in the MH mode particles during NPF event may be not only sulfuric acid or ammonium sulfate but also secondary organic species. The contribution from secondary organic species, which are less hygroscopic than ammonium sulfate, may dominate in the newly formed particles to be able to reduce the HGF of pre-existing particles. Similar observations was found in other studies (Levin et al., 2012; Wu et al., 2015).

Hence, we believe the effect of NPF on hygroscopicity might be worth a study of its own. However, without size-resolved chemical composition of particles, we cannot make further conclusion based on current results. The findings above seem to be a little bit too weak to have a single section in the manuscript. To include these into this paper would make the manuscript unnecessarily long, which would even weaken the key points of this paper. At the other four referees' requests, chemical composition of PM1 should not be compared with the one from size-segregated aerosols and uncertainty analysis should be implemented into the manuscript. Hence, we adopted some assumptions and performed a comprehensive uncertainty analysis regarding the hygroscopicity-composition closure. In addition, similar analysis was done for the polluted and clean days. Finally, we revised our conclusion substantially. The revised sections are attached after the answers of all comments with color in blue.

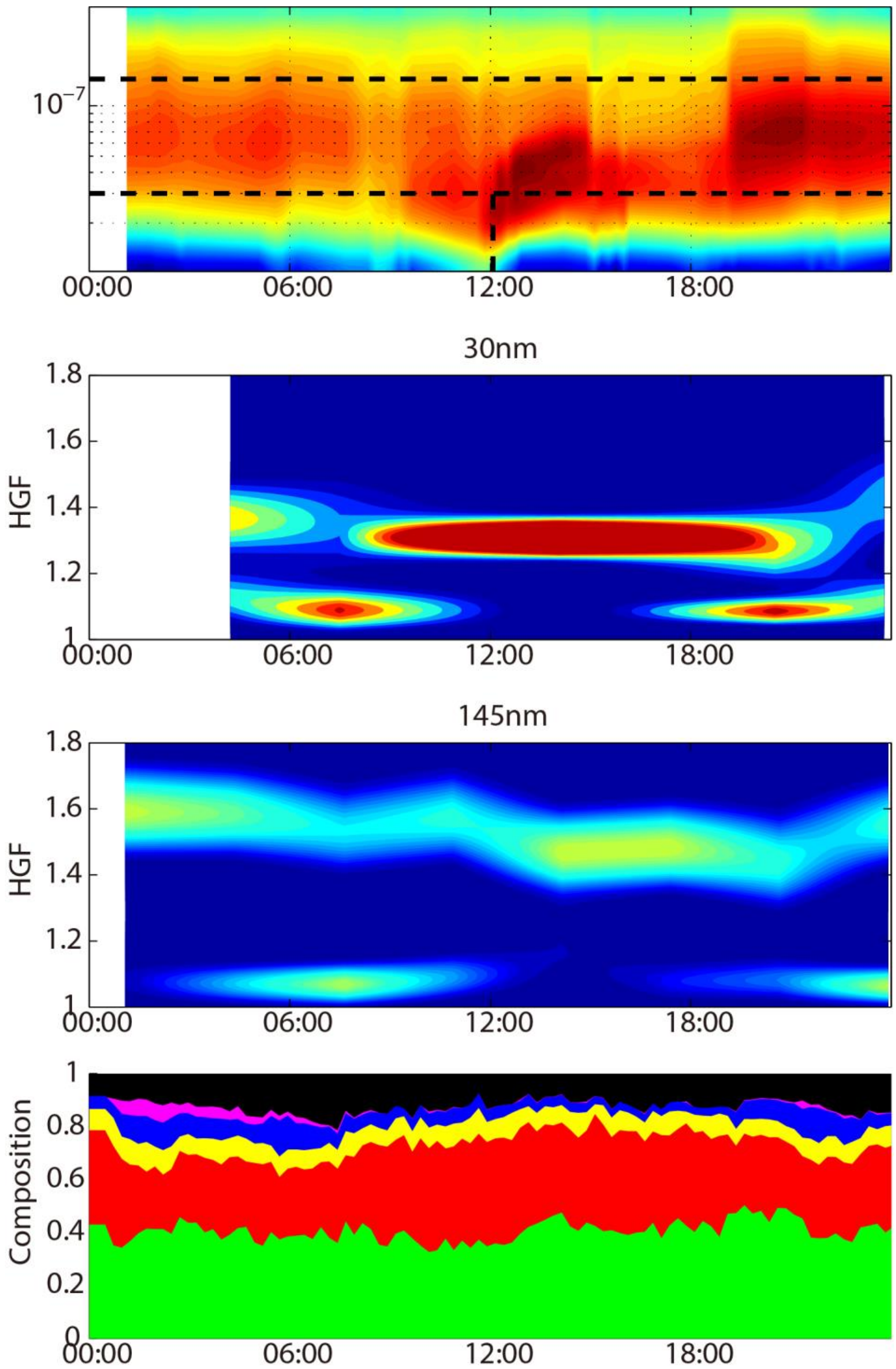

Figure R1: Time series of particle number size distribution, GF-PDFs of 30 and 145 nm particles and chemical composition of PM1 during 04, October 2016.

Specific comments
152: It is not clear from the manuscript how the HTDMA was operated to obtain the particle number size distribution (10-1000 nm) simultaneously while the instrument was measuring HGF in 4 size classes.

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. We made a mistake before in the manuscript; actually particle number size distribution within 10-400 nm was measured, but not 10-1000nm particles. We corrected it in the revised manuscript.

175-178: It is critical to indicate the calibration procedure of ACSM and what calibration parameters were used (e.g. relative ionization efficiency of SO4). Such calibration parameters can critically affect the inorganic and organic mass fractions (and therefore the ensemble HGForg of 1.1).

Reply: Ammonium nitrate (AN) particles, generated by an atomizer, was selected by a DMA with a certain size at 300 nm. The size-selected particles were then introduced into a CPC and the ACSM. After obtaining the number concentration of AN particles, we calculated their mass concentration by multiplying the density of AN. The mass concentration obtained from the CPC was then compared with the one from ACSM. Then the corresponding RIE value for NH4 was calculated. Similar procedure was performed for ammonium sulfate particles. The RIE values for NH4 and SO4 were obtained as 5.63 and 0.78, respectively. The detailed calibration procedure was described in Ng et al. (2011).

300-303: The logical basis to support the following conclusion is not clear. “In case of smaller particles (30 nm, 60 nm), HGFs of MH group particles appeared to decrease during the afternoon until about 8:00 pm, suggesting that these particles were not long-range transported, but rather secondary formed either locally or from nearby emissions.”

Reply: We changed the sentence to: ‘In case of smaller particles (30 nm, 60 nm), HGFs of MH group particles appeared to decrease during the afternoon until about 8:00 pm. This is probably attributed to the intensive traffic emissions at the time of rush hour.’

462-519: Extra caution must be taken when comparing k based on supersaturation conditions and HGF based on sub-saturated conditions. The k derived from sub- and supersaturated conditions can be quite different in some cases. In such case, the discussion on potential bias on CCN concentration may not be relevant.

Reply: The last figure in the manuscript is only an illustration for the relation between the hygroscopicity of organic material and its O:C ratio. Kappa values are not directly compared with the ones of HGF. We agree with the referee that kappa derived from sub- and supersaturated conditions can be quite different in some cases. Hence, we deleted the discussion on the calculation of CCN concentration. New discussions were included in the revised manuscript.

Technical corrections

120: "self-assembly" should appear "self-assembled"

Reply: We rephrased it to ‘self-assembled’.

188: what does it mean by “individual size bins”?

Reply: We changed it to: ‘It is necessary to note that the chemical composition of PM1 can be different from those of size-selected aerosol particles’.

330: rephrase “uncertainties of in growth factor”

Reply: We changed it to ‘uncertainties in growth factor’.

423: "as followed" should appear "as follows"

Reply: We changed it to ‘as follows’.

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. R2 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.R3 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. R4 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.

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. R5 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. R6 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 (Fig. R7 in this response). 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: $HGF_{org} = (0.32 \pm 0.01) \times O:C + (1.10 \pm 0.04)$. 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.

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%

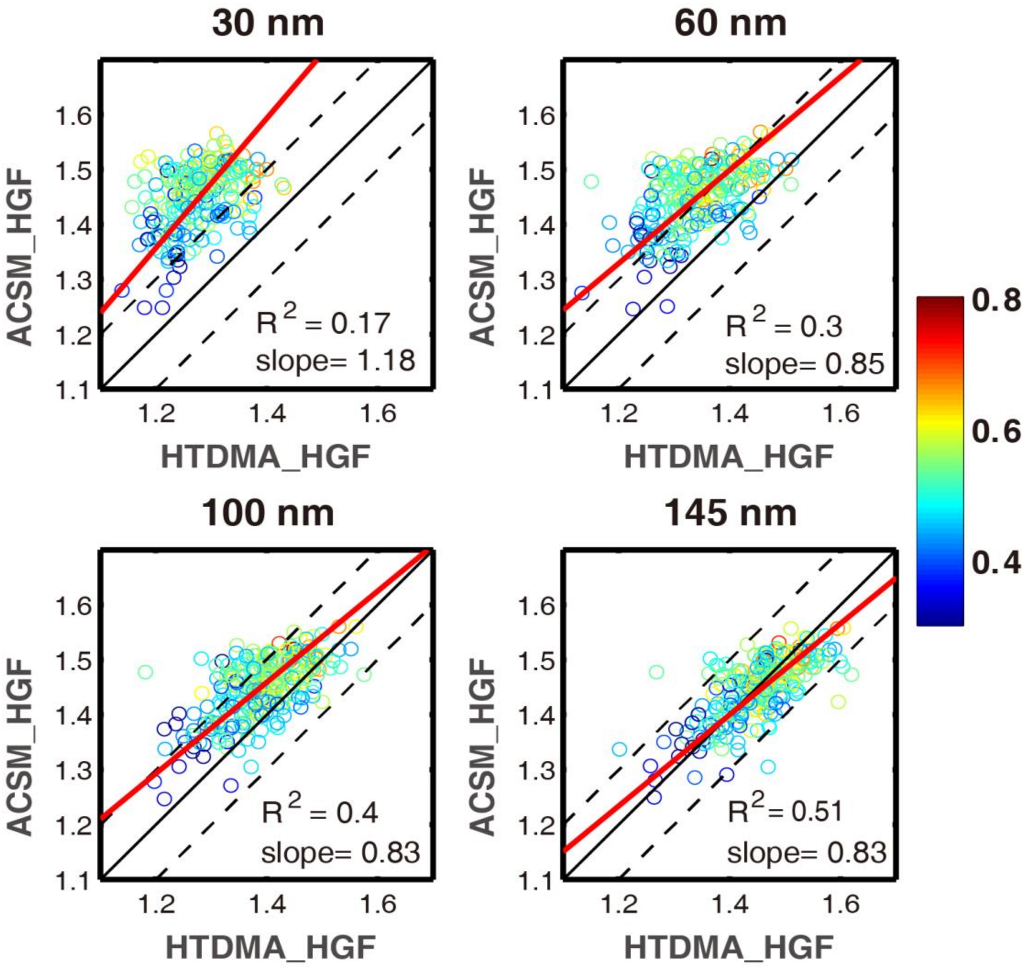

Figure R2: Closure study between the HTDMA-measured HGFs and the ACSM-derived HGFs. The black solid lines indicate the 1:1 line and the black dash lines represent ±10% deviation, while the red lines are the lines fitted to the data points. The color bar indicates the O:C ratio of the organic aerosol fraction.

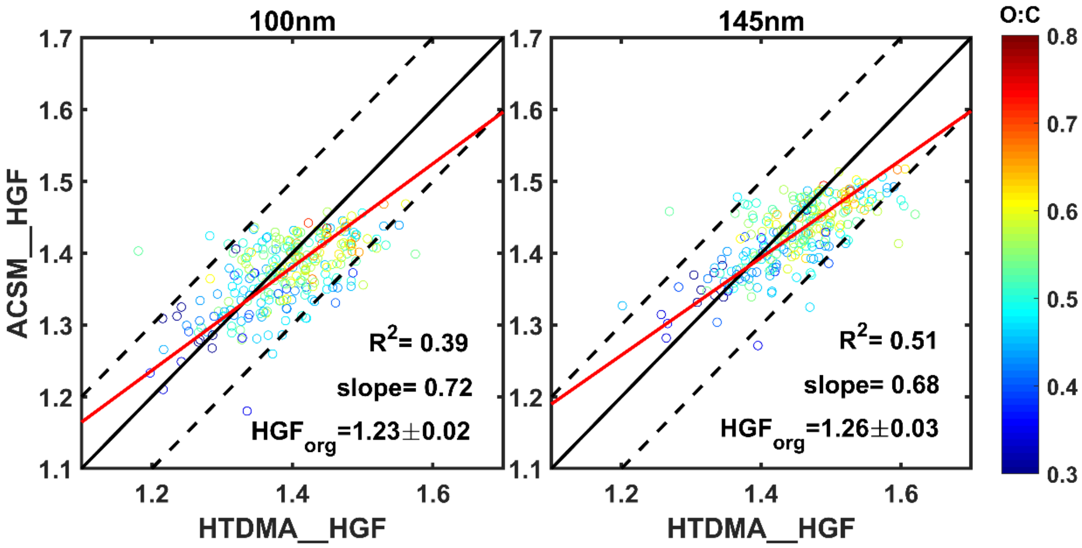

Figure R3: Closure study between the HTDMA-measured HGFs and the ACSM-derived HGFs assuming the average inorganic mass fraction of PM1 were about 25%± 3% and 16% ±3% higher and the average ammonium sulfate mass fraction of PM1 were about 25%± 3% and 16% ±3% lower than those of 100 nm and 145 nm particles. 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.

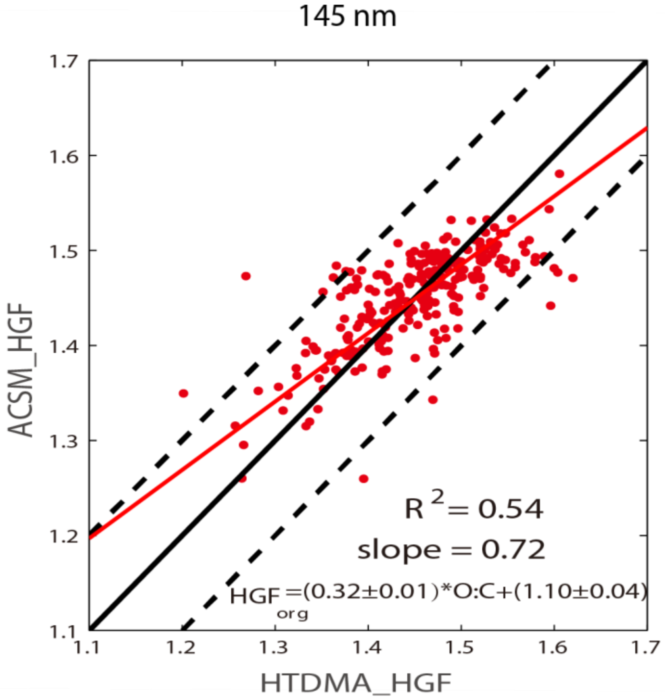

Figure R4: Closure analysis with the best fitting between the measured HGFs and the ACSM-derived ones using the O:C-dependent HGForg. The assumption of size-dependent chemical composition of aerosols was considered to determine the ACSM_derived HGF. The equation is the achieved approximation for HGForg as a function of the O:C of organic aerosol fraction.

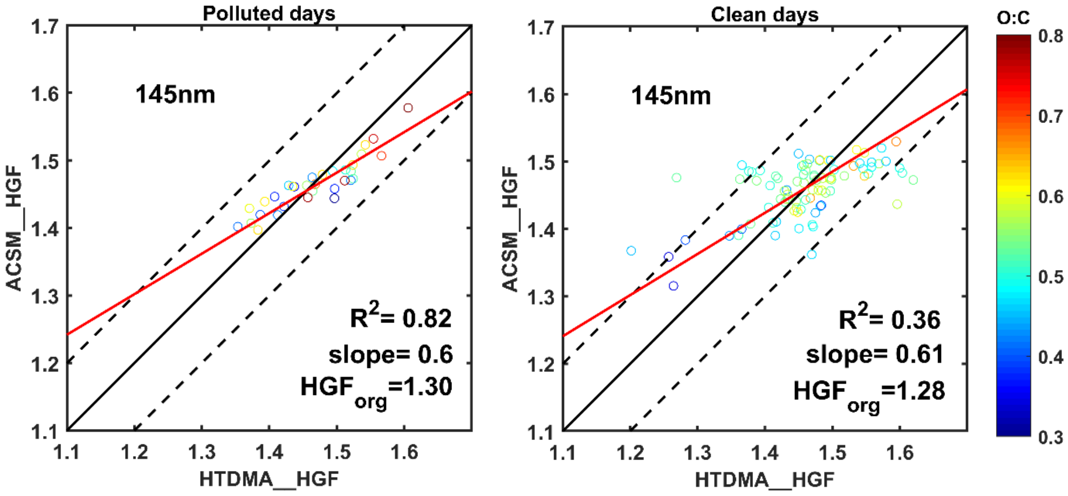

Figure R5: Closure study between the HTDMA-measured HGFs and the ACSM-derived HGFs by taking into account of size-dependent chemical composition of aerosols during polluted and clean days. The black solid lines indicate the 1:1 line and the black dash lines represent ±10% deviation, while the red lines are the lines fitted to the data points. The color bar indicates the O:C ratio of the organic aerosol fraction.

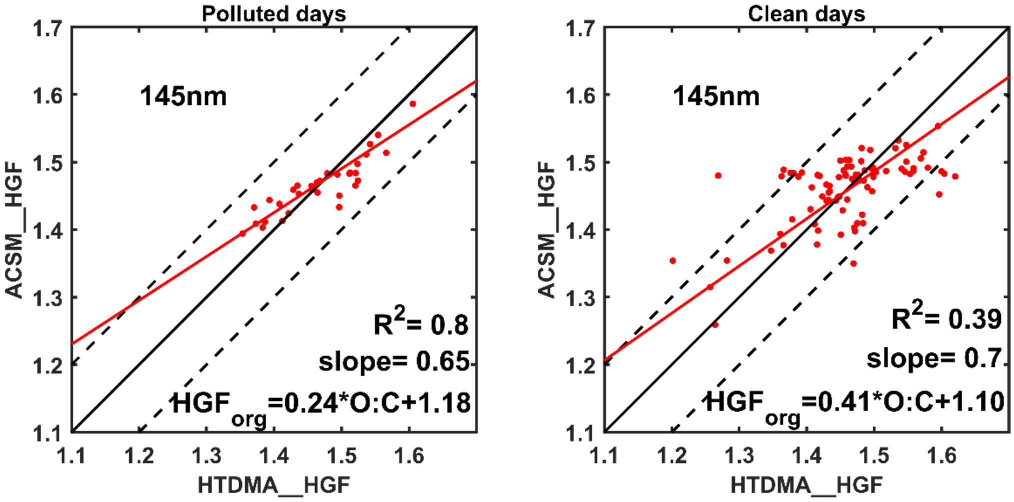

Figure R6: Closure analysis with the best fitting between the measured HGFs and the ACSM-derived ones using the O:C-dependent HGForg during polluted and clean days. The assumption of size-dependent chemical composition of aerosols was considered to determine the ACSM_derived HGF. The equation is the achieved approximation for HGForg as a function of the O:C of organic aerosol fraction.

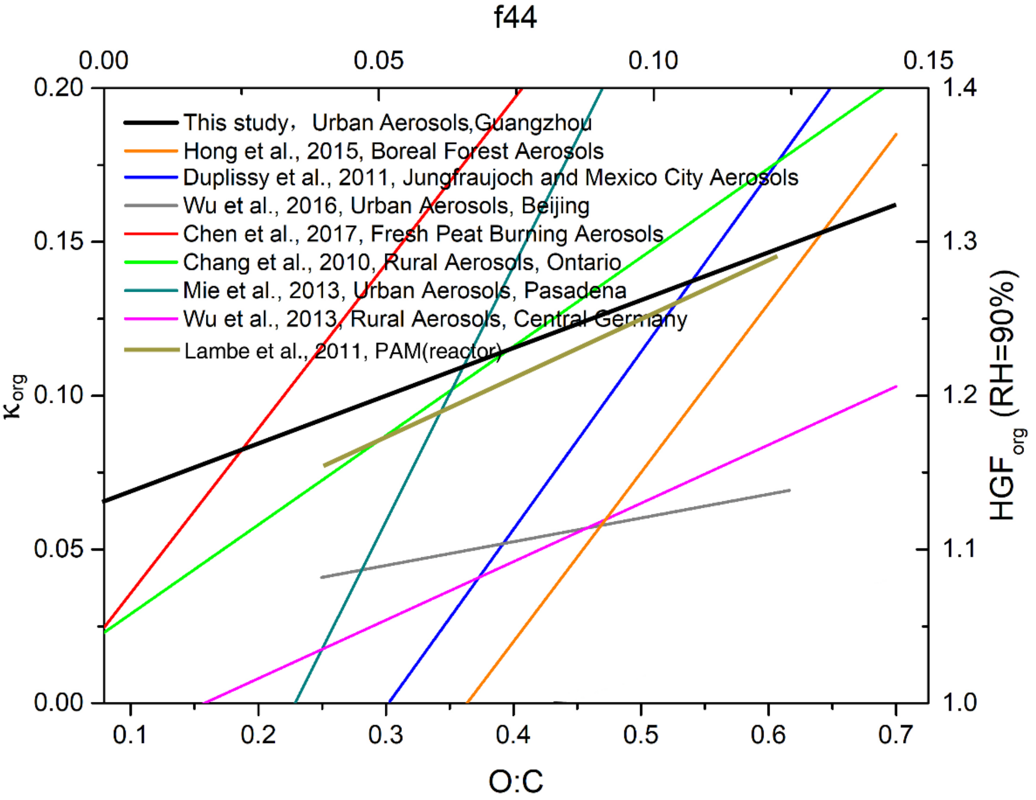

Figure R7: Comparison with earlier studies on the hygroscopicity of organic material with atomic O:C ratio (or *f44* from chemical composition data) obtained from different environmental background areas. Other studies were using derived κorg, while this study is using HGForg for the hygroscopicity of organic material.

References:

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