

Answers to Referee #2

The authors appreciate the time the reviewer has spent on our manuscript. We thank a lot for the concern raised by the reviewer here, which we agree we did not notice and consider earlier. Hence, we tried our best to include more assumptions and uncertainty analysis into the revised version of the manuscript to address the comments and suggestions given by the reviewer. We hope the new version of the manuscript could meet the requirements for publication.

Major comments:

1. The closure analysis compares ACSM-derived HGF for PM1 with HTDMA-measured HGF for 100 nm or 150 nm particles. What was the mode diameter (in terms of mass diameter distribution) of the particle population of PM1? If the mode diameter of PM1 is significantly larger than 150 nm, the estimated HGFor_g can be significantly underestimated, because the hygroscopicity generally increases with the diameter in the accumulation mode (at least for 50 nm - 250 nm particles as shown in Liu et al. ACP, 2011). Is it possible that the estimated low HGFor_g is affected by such systematic biases?
2. In line 328-331, the authors mentioned that the measured HGF values for the mixed particles are less sensitive to the relationship of HGFor_g vs. O:C. I agree with this statement. However, this implies that the derived parameterization of $HGFor_g = 0.3 \cdot O:C + 0.87$ can be highly uncertain, as it is not well constrained by the measurements. I noticed that including the O:C-dependent HGFor_g only improves R^2 from 0.5 to 0.51 (Fig.10 vs. Fig. 9 d). In this case, what is the error associated with the parameterization?
3. Related to the previous comment, the proposed relationship would predict a HGFor_g value smaller than 1 for $O:C < 0.43$. This can be misleading. What does a HGFor_g value smaller than 1 mean? Is it just because of measurement errors?
4. The authors reported a best-fit HGFor_g value of 1.1. The error bar associated with this value should also be reported. Errors of both HTDMA and ACSM, along with the uncertainties associated bulk vs size-resolved closure analysis should be considered.
5. Even if the HGFor_g = 1.1 value is accurate, I don't think the authors can assert that "the CCN concentrations would probably be over-predicted as the organic material in these locations appeared to be close to hydrophobic". Several studies have shown that the CCN activity can be higher than that predicted based on the HGFor_g at 90% RH. For example, alpha-pinene-derived SOA has a kappa value of 0.04 at 90% RH, but the CCN-derived kappa value is around 0.1 (see Pajunoja et al., 2015), which is close to the value used in climate models. The low hygroscopicity derived at subsaturation regime does not necessarily indicate a low CCN activity.

Reply: As all of the above questions are related to each other, so we combine the answers to address the comments together.

Number size distributions of particles within the diameter range 10 - 400 nm was measured by the SMPS in this study. Hence, the mode diameter in terms of mass diameter distribution of the particle population of PM1 could not be determined directly. Cai et al. (2017) showed that the mode diameter of the mass size distribution of the chemical composition of PM1 was around 300 - 400nm for aerosols sampled at the same measurement site during the same season of 2014.

We agree that the estimated HGF_{org} could be underestimated. However, size-resolved chemical composition information of ambient aerosols for current study is not available. We therefore considered the differences in the mass fraction of each component between PM₁ and 145 nm particles obtained from Cai et al. (2017) and applied this assumption with certain uncertainties (20%) into the current study to make further evaluations. A newly determined parameterization of HGF_{org} 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.

With the new derived parameterization between HGF_{org} and O:C, the values of HGF_{org} values were all above one, which also indicates the size-resolved chemical composition taken from the above-mentioned assumption is physically reasonable. **The revised sections are attached below.**

We agree with the reviewer that the sentence ‘the CCN concentrations would probably be over-predicted as the organic material in these locations appeared to be close to hydrophobic’ is not properly used here. Hence, we deleted it.

In conclusion, 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’.

Technical issues:

1. The authors should describe how the HTDMA was calibrated and how the data inversion was performed. Did the measurements reproduce literature values for pure compounds (e.g., ammonium sulfate)? Was the RH sensor calibrated? Did the two DMAs have a shift in diameter? Was the DMA transfer function considered in data inversion?

Reply: The calibration of the HTDMA was mainly performed in two steps. First, the electronic mobility diameter of particles was calibrated using polystyrene latex (PSL) spheres with known sizes. If the mean particle sizes measured by both DMAs are within the nominal sizes of PSL spheres, then both DMAs are capable to select a certain sized particles with required accuracy. Otherwise, the voltage supplying to either DMA should be corrected by using a voltage reference (here, as a voltage meter) until the nominal sizes of PSL is reached. Secondly, calibration of the HTDMA was done by measuring the HGF of ammonium sulfate at a certain RH, for instance, 90%. The measured HGF values for ammonium sulfate at a certain RH should match rather well with theoretical predictions (for instance, HGF=1.71 at RH=90%) with about 2% deviation. The measured growth factor distributions were fitted into bimodal lognormal distributions using a corrected data inversion approach by TDMAfit algorithm assuming all particles follow a Gaussian distribution (Stolzenburg, 1988; Stolzenburg and McMurry, 2008). The detailed description about calibration of the instrument and the data inversion approach for this study is given in Tan et al. (2013).

2. The O:C ratio was estimated from the f₄₄ measured by a quadrupole ACSM, presumably less accurate than that measured by the HR-ToF-AMS. What was the uncertainty of the ACSM-derived O:C?

Reply: The uncertainties of the HR-ToF-AMS to quantify O:C have been discussed previously and are approximately to be ± 0.1 as determined by Aiken et al. (2007) and Aiken et al. (2008). The uncertainties of determining O:C for ambient aerosols using other types of AMS and

ACSM is not well established. However, Ng et al. (2011) demonstrated that due to the similarities with AMS, all the methods that already have been developed for the usage of AMS are capable to be used in ACSM data analysis. The largest differences between ACSM and AMS are that ACSM is not designed to measure mass size distribution and it has a lower detection limit (e.g. 0.2 ug/m³). Hence, we believe that the O:C ratio determined by ACSM should not deviate significantly than the one from AMS.

3. Figure 2. Unit of the color bar is missing.

Reply: We added the unit for the color bar in Fig. 2 in the manuscript.

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, HGF_{org} 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 these 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 HGF_{org} 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 HGF_{org} 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 HGF_{org} 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 HGF_{org} and the O:C ratio based on the chemical composition of bulk aerosols was obtained as follows:

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

This closure was no better than the one shown in Fig. 9 using constant a HGF_{org}, 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 HGF_{org} and the O:C ratio as:

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

The closure was somewhat better than in Fig. 9 according to the slightly lower RMSE value (0.58 vs. 0.63). In addition, HGF_{org} ranged from 1.1 to 1.4 with the varying O:C ratio, and there were no HGF_{org} 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 HGF_{org} 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/m³ for organic materials (Kuwata et al., 2012) and 1000 and 2000 kg/m³ 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 HGF_{org}, 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 HGF_{org} 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 HGF_{org} value was quite close to each other between the polluted and clean days (HGF_{org}=1.30 and 1.28, respectively), and each closure is shown in Fig.S3 (Fig. R4 in current response). These values are similar to the one previously determined (HGF_{org} of 1.26) for the entire experimental period. A good closure was achieved during the polluted days with a substantially high R² 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. R5 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 HGF_{org} 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. R6 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 hygroscopicity 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 HGF_{org} 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 $\kappa_{\text{org}}/\text{HGF}_{\text{org}}$ 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: $\text{HGF}_{\text{org}} = (0.32 \pm 0.01) \times \text{O:C}$

+ (1.10±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%

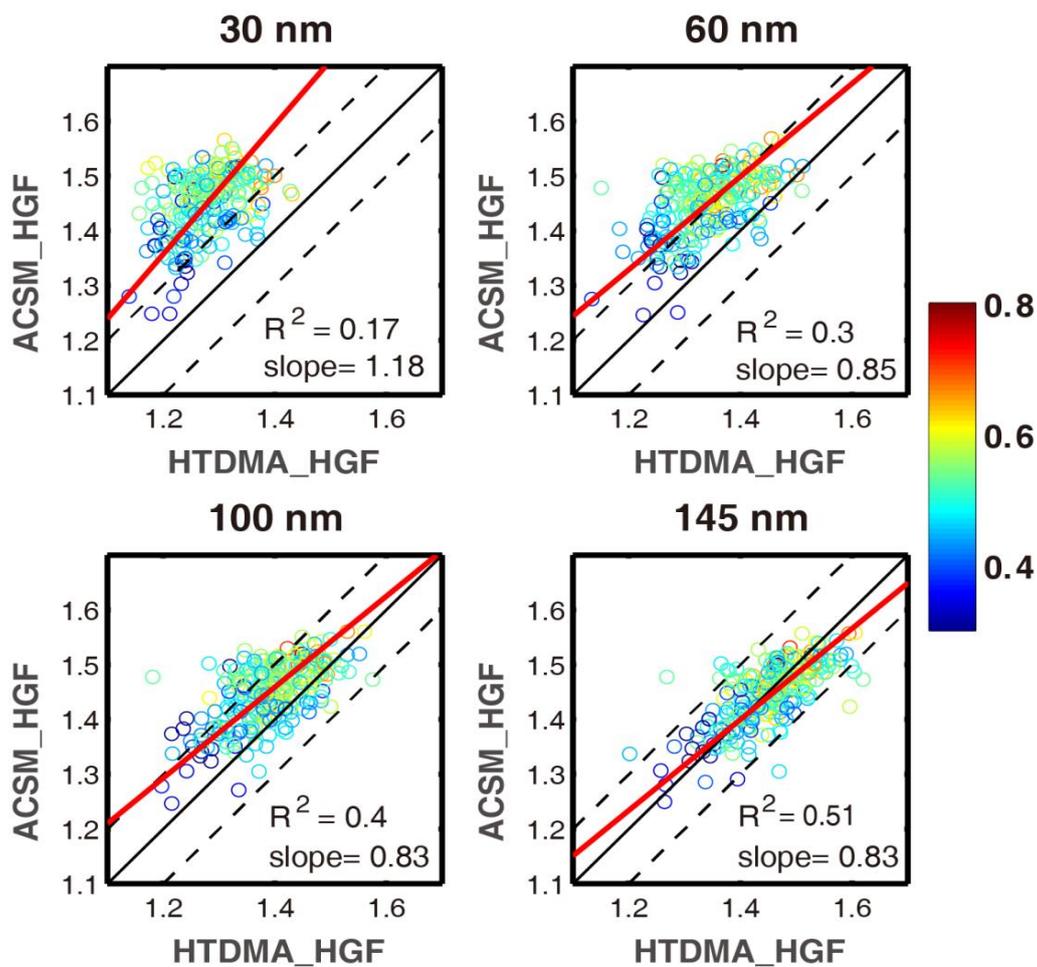


Figure R1: Closure study between the HTDMA-measured HGFs and the ACSM-derived HGFs. The black solid lines indicate the 1:1 line and the black dash lines represent $\pm 10\%$ deviation, while the red lines are the lines fitted to the data points. The color bar indicates the O:C ratio of the organic aerosol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

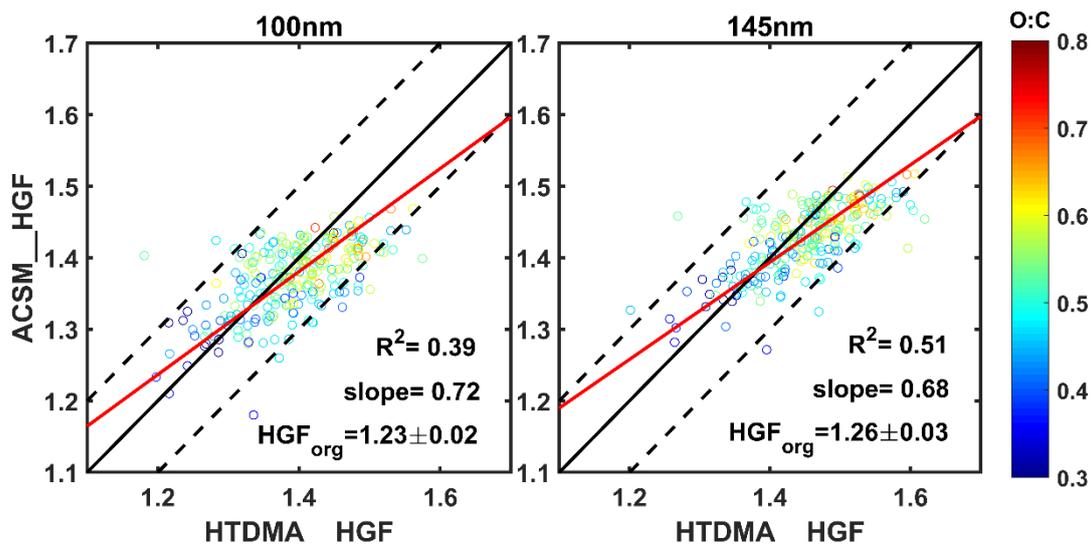


Figure R2: Closure study between the HTDMA-measured HGFs and the ACSM-derived HGFs assuming the average inorganic mass fraction of PM1 were about $25\% \pm 3\%$ and $16\% \pm 3\%$ higher and the average ammonium sulfate mass fraction of PM1 were about $25\% \pm 3\%$ and $16\% \pm 3\%$ lower than those of 100 nm and 145 nm particles. The black solid lines indicate the 1:1 line and the black dash lines represent $\pm 10\%$ 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.

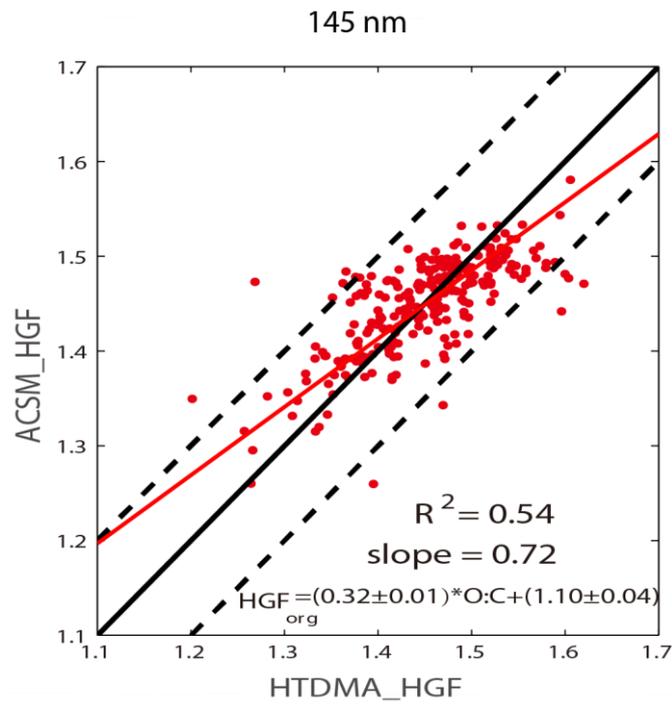


Figure R3: Closure analysis with the best fitting between the measured HGFs and the ACSM-derived ones using the O:C-dependent HGF_{org}. The assumption of size-dependent chemical composition of aerosols was considered to determine the ACSM-derived HGF. The equation is the achieved approximation for HGF_{org}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

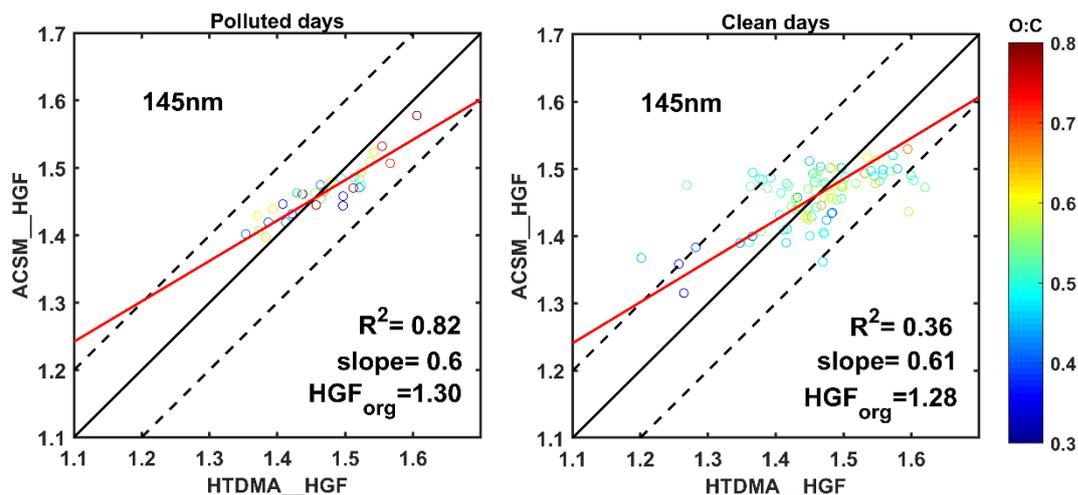


Figure R4: Closure study between the HTDMA-measured HGFs and the ACSM-derived HGFs by taking into account of size-dependent chemical composition of aerosols during polluted and clean days. The black solid lines indicate the 1:1 line and the black dash lines represent $\pm 10\%$ deviation, while the red lines are the lines fitted to the data points. The color bar indicates the O:C ratio of the organic aerosol fraction.

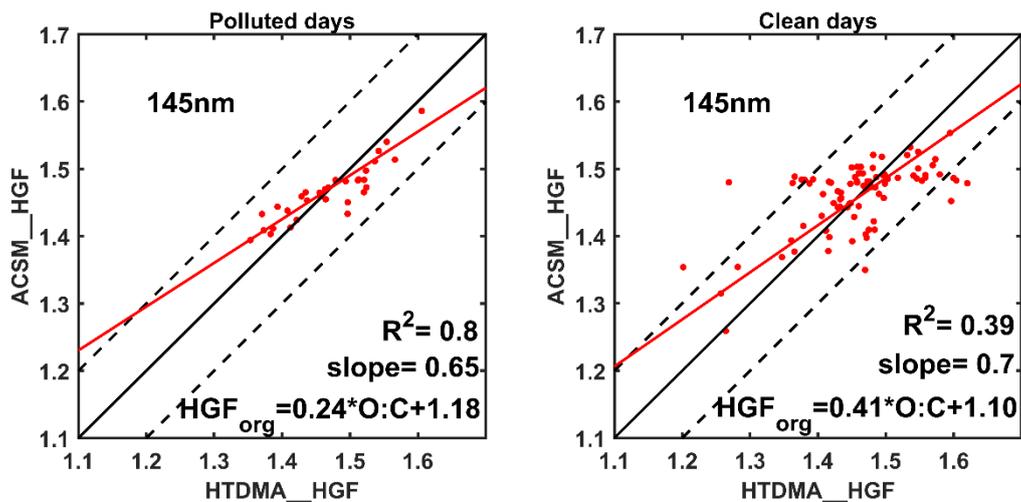


Figure R5: Closure analysis with the best fitting between the measured HGFs and the ACSM-derived ones using the O:C-dependent HGF_{org} during polluted and clean days. The assumption of size-dependent chemical composition of aerosols was considered to determine the ACSM_{derived} HGF. The equation is the achieved approximation for HGF_{org} as a function of the O:C of organic aerosol fraction.

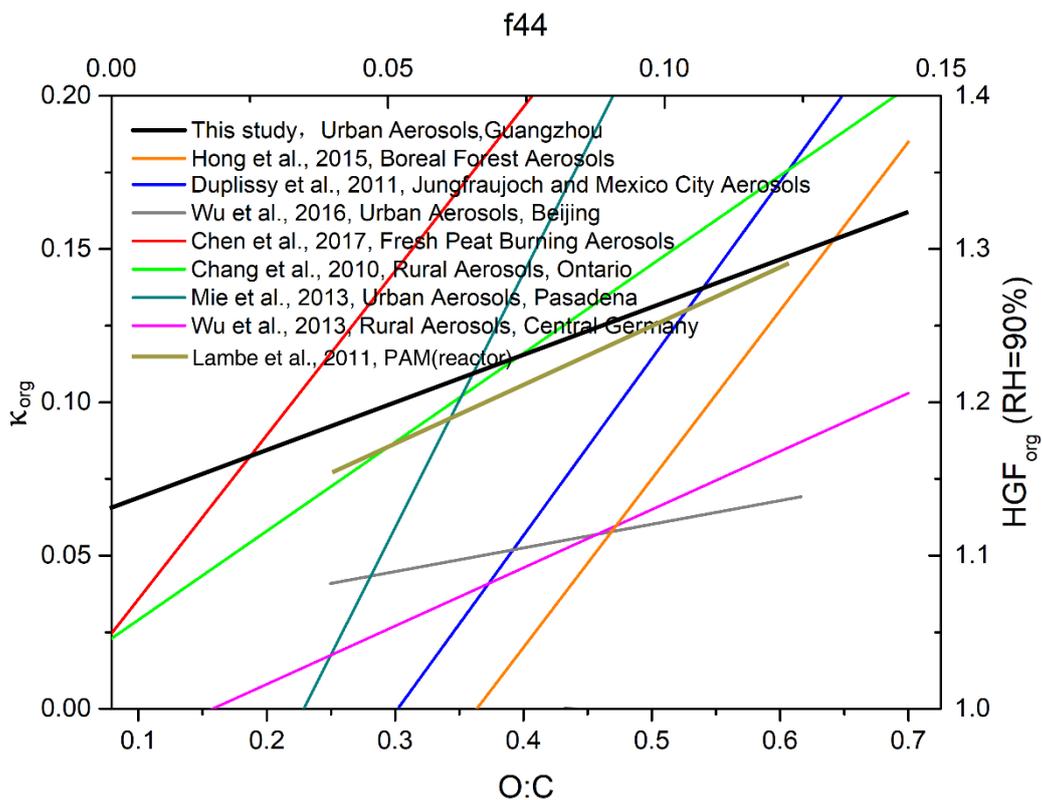


Figure R6: Comparison with earlier studies on the hygroscopicity of organic material with atomic O:C ratio (or *f44* from chemical composition data) obtained from different environmental background areas. Other studies were using derived k_{org} , while this study is using HGF_{org} for the hygroscopicity of organic material.

References:

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