

Answers to Referee #1

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.

Major comments:

1. The authors should describe the exact calculation procedure possibly with mathematical equations for lines 171–174.

Reply: After obtaining GF-PDF, which was represented as $c(\text{HGF})$, ensemble mean hygroscopic growth factor (HGF), number fraction of particles at each mode and spread of each mode was calculated according to the following equations (Gysel et al., 2009; Tan et al., 2013):

$$\text{HGF}_{\text{mean}} = \int_0^{\infty} \text{HGF} \cdot c(\text{HGF})d(\text{HGF}). \quad (1)$$

$$\sigma = \left(\int_0^{\infty} (\text{HGF} - \text{HGF}_{\text{mean}})^2 c(\text{HGF})d(\text{HGF}) \right)^{1/2}. \quad (2)$$

$$\text{NF}^{a,b} = \int_a^b c(\text{HGF})d(\text{HGF}). \quad (3)$$

Here HGF_{mean} is the number weighted mean hygroscopic growth factor, σ is the standard deviation of GF-PDF, which is used as a measure for the degree of mixing of aerosol population. $\text{NF}^{a,b}$ represents the number fraction of particles in which $a < \text{HGF} < b$.

2. Sect. 2.1 and later. The arrival height of 150 m above the ground is unusually low; the model performs better at higher levels. This raises the question about the representativity of these air mass trajectories. The authors should discuss and explain their selection, and/or present similar trajectories for at least 2 more but larger arrival heights (e.g. one just below and one above the free troposphere). In addition to that, the authors should describe how the major trajectory clusters and their frequency in Fig. 7 were exactly derived from the individual trajectories.

Reply: We agree with the Referee that the HYSPLIT model might not work properly when the arrival height was set as low as 150 m in this study. Hence, we performed the back trajectory analysis using the HYSPLIT model for arrival heights at 300 m, 700 m, and 1000 m above ground level. For each arrival height, cluster analysis was then used to group these entire 156 back trajectories into mean trajectories, e.g. clusters for different experimental periods of interest. The basic principle is to merge trajectories that are spatially near each other and form groups or clusters that represent these trajectories. This is computationally achieved by minimizing the differences between trajectories within a cluster and maximizing the differences between clusters. The detailed equations and calculation procedure are available in the HYSPLIT Tutorial document (https://www.ready.noaa.gov/documents/Tutorial/html/traj_clus.html). Results of the back trajectory analysis for different arrival heights are illustrated in Fig. R1, Fig. R2 and Fig. R3 in this response respectively. There are no significant differences between the results of the three different arrival heights. However, we agree with the referee that the arrival height of 150 m is fairly low for this model. Therefore, we switched to the arrival height of 700 m, which is the

mean height of the boundary layer in Guangzhou during the experimental period according to the data obtained from European Centre for Medium-Range Weather Forecasts (ERA Interim).

3. Aerosol particles are usually grouped as: nearly hydrophobic ($\kappa < 0.10$), less hygroscopic ($\kappa \approx 0.10-0.20$) and more hygroscopic ($\kappa > 0.20$; Liu et al., Atmos. Chem. Phys., 11, 3479–3494, 2011). The authors may want to follow this attitude, or explain and argue for their own classification.

Reply: It is a conventional way to group aerosol particles into three characterized modes regarding their hygroscopicity. However, this might not always be the case; for instance, the other studies categorized aerosol particles into two modes, namely non or less-hygroscopic mode and more-hygroscopic mode (Aklilu et al., 2006; Gysel et al., 2007; Tan et al., 2013; Yeung et al., 2014; Wu et al., 2016) according to their own data. We tried to fit the measured data into three modes; however, the fitting procedure failed for more than half of the data sets. Two-mode fitting procedure was then carried out for all of the data.

4. The authors are requested to extend the MS with explicit discussions and estimations of the uncertainty of their major conclusions to prove their significance. In addition to that, smaller changes throughout the MS, e.g. showing standard deviations of slopes for correlation lines in Fig. 10, and similar amendments could also be adopted.

Reply: Yes, we have made a comprehensive uncertainty analysis regarding our measurements and calculations. Hence, the whole section (Sect. 3.4) was revised as shown below. **In addition, 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’.**

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. R4 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 $25\% \pm 3\%$ and $16\% \pm 3\%$ 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.R5 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. R6 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 $90 \pm 1\%$. 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/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 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.

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%
NH ₄ , NO ₃ mass concentration	20%	0.6%, 0.5%	0.8, 1.6%
SO ₄ mass concentration	20%	1.8%	4%
Organics mass concentration	20%	1.4%	3.2%
BC mass concentration	5%	0.1%	0.8%

Minor comments:

1. It is disturbing that the terms aerosol – aerosols – aerosol particles are not used in a consistent manner in the MS (e.g. lines 64, 123 and 196). The authors should decide which option to use, and should adopt it in a coherent nomenclature.

Reply: I decided to use ‘aerosol particles’ in the revised manuscript to keep a coherent nomenclature.

2. Abbreviation PM stands for particulate matter (as correctly stated in line 191), but it is sometimes used for particle matter mass (e.g. line 30). The explanation of the abbreviation should be given at its first occurrence, and it is redundant to repeated it e.g. in lines 232– 233. Furthermore, simply write for instance: “The PM_{2.5} mass concentration varied” instead of “The range of particle mass concentration (PM_{2.5}) varied”. In addition, it is not the range that varies.

Reply: I revised the sentence to: ‘The PM_{2.5} mass concentration varied from 20 to 180 ug/m³, with relatively low values (roughly below 50 ug/m³) during most of the time.’

3. Hygroscopicity usually refers only to sub-saturated conditions. Clarify line 65, or give appropriate references to back your statement.

Reply: References were added to back up my statement: ‘Aerosol hygroscopicity describes the interaction between aerosol particles and ambient water molecules at both sub- and super-saturated conditions in the atmosphere (Topping et al., 2005; McFiggans et al., 2006; Swietlicki et al., 2008).’

4. Micron (e.g. lines 98, 177, Fig. 2) is not an SI unit. Micrometer should be used instead.

Reply: ‘Submicron’ was used as a definition for particles with sizes smaller than 1 micrometer in diameter. This term is widely used in aerosol research articles.

5. Remove the repetitions in lines 102–103 considering lines 67–69.

Reply: I modified the sentence to: ‘Hygroscopicity, as an important physico-chemical property of atmospheric particles (Cheng et al., 2008; Gunthe et al., 2011; Cheng et al., 2016), has also been implemented into extensive campaigns in densely populated areas, such as North China Plain (Massling et al., 2009; Liu et al., 2011) and Yangtze River Delta (Ye et al., 2013).’

6. Revisit “self-assembly” (line 120) and “self-assembled” (line 152), or use perhaps laboratory made instead.

Reply: Yes, I used ‘self-assembled’ in the revised manuscript.

7. Clarify lines 185–189.

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.

8. Abscissa of Figs. 1 and 2 shows rather Date than Time, and its format of e.g. “(dd-MM)” could also be indicated. Furthermore, they could have (this) consistent format in both figures. In Fig. 2, how can be the probability in colour coding larger than 1? Explain or modify this. Put word space between measured values and their units everywhere in the figures. Extend the interpretation of your finding and discussions related to Fig. 2 within the frame of the conjunct conclusions of Cheung et al., *Atmos. Chem. Phys.*, 16, 8431– 8446, 2016.

Reply: Figure 1 in the manuscript was modified and is shown below as Fig.R7. The color bar in Fig. 2 in the manuscript indicates the probability density rather than probability. To integrate the probability density along the x-axis, we will obtain the probability value. This is why its value is larger than one. All figures were modified as suggested.

9. Figures 3, 4 and 6 show the mean diurnal variations; the label of abscissa should be consistent in three figures; remove the tick label at 25; the time unit as “(HH)” should also be indicated; “during this study” is redundant; avoid abbreviations in the figure captions everywhere. Extend the interpretation of your finding and discussions related to the diurnal plots (Figs. 3 and 4 or Sects. 3.2 and 3.3) within the frame of the similar recent data of Enroth et al., *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2017-926>, 2017.

Reply: I moved Fig. 3 in the manuscript into supplement material part and modified all figures as suggested. Brief comparison was made between our results and Enroth et al. (2018).

10. All correlation scatter plots should have squared layout to facilitate better their interpretations.

Reply: Figures with x-axis and y-axis indicating the same variables was modified to have squared layout, see Fig. 4, Fig. 5 and Fig. 6.

11. Rounding off strategy should be revised throughout the MS; e.g. of R2 in the figures or HGForg in lines 399, 405 (“1 and 1.3”), 515 or 546.

Reply: Yes, I modified all values I used in the whole manuscript to follow consistent strategy.

12. Lines 288–289: Remove “black carbon or” from “with black carbon or soot”.

Reply: Yes, I removed ‘black carbon or’ from the text.

13. Line 319: Consider writing “compounds with different water uptake ability” instead of “compounds of different water uptake ability”.

Reply: Yes, I changed it to ‘compounds with different water uptake ability’ in Line 319 in the revised manuscript.

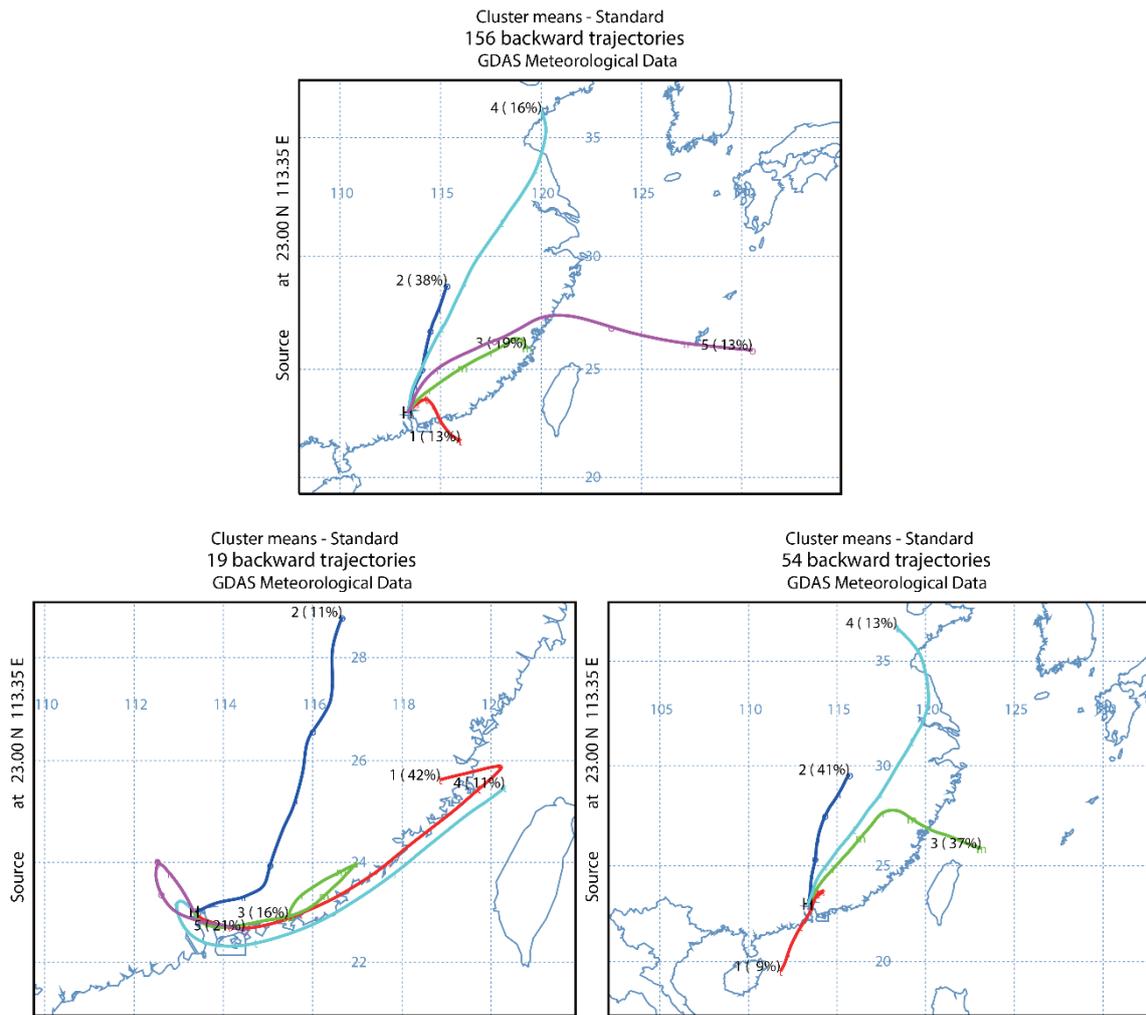


Figure R1: The major clusters for the 72-hour backward trajectory simulation for air masses arriving at the CAWNET Panyu site at a height of 300 m. The upper panel shows the results throughout the whole observational period, while the lower panel on the left side shows the one during polluted days and the one on the right-hand side is for clean days.

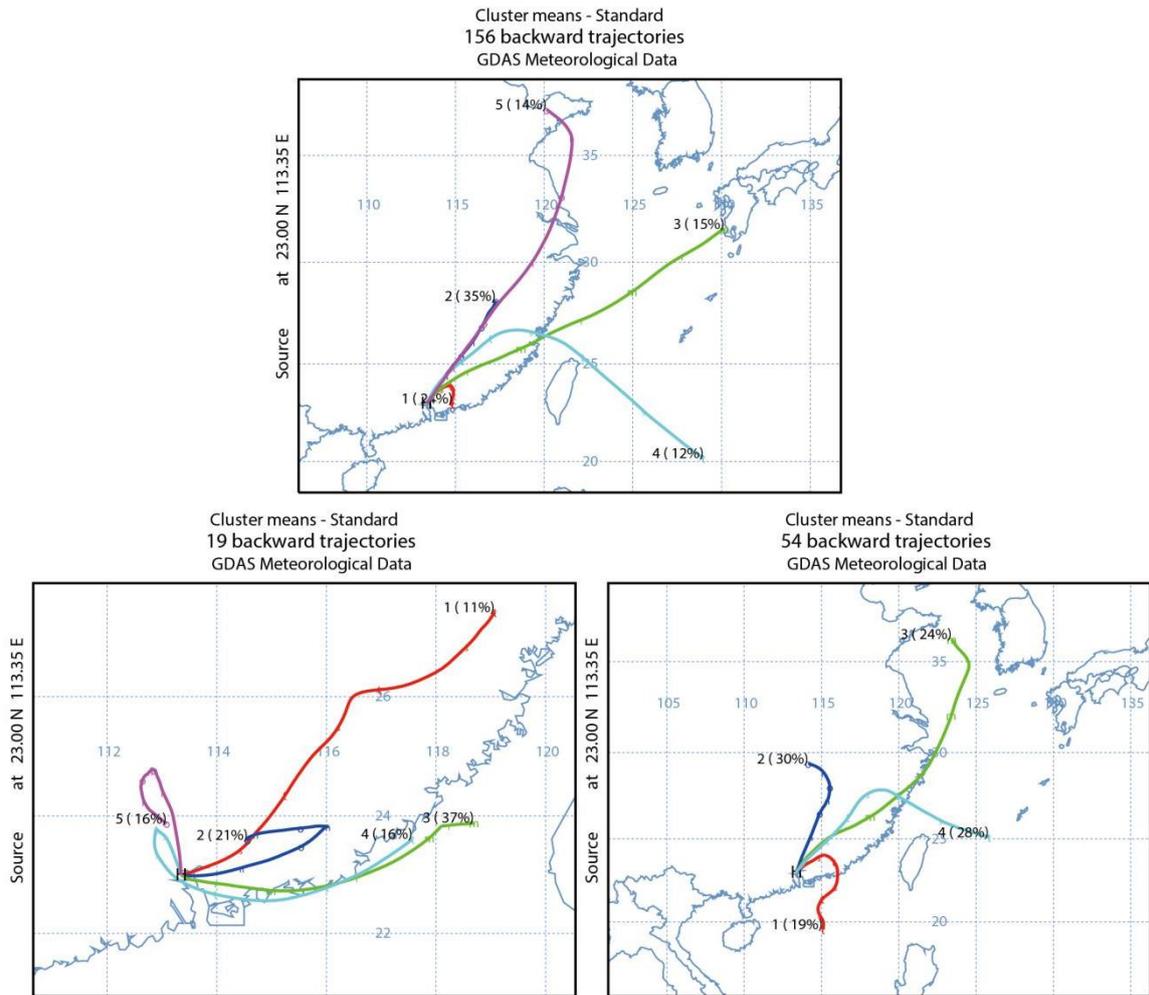


Figure R2: The major clusters for the 72-hour backward trajectory simulation for air masses arriving at the CAWNET Panyu site at a height of 700 m. The upper panel shows the results throughout the whole observational period, while the lower panel on the left side shows the one during polluted days and the one on the right-hand side is for clean days.

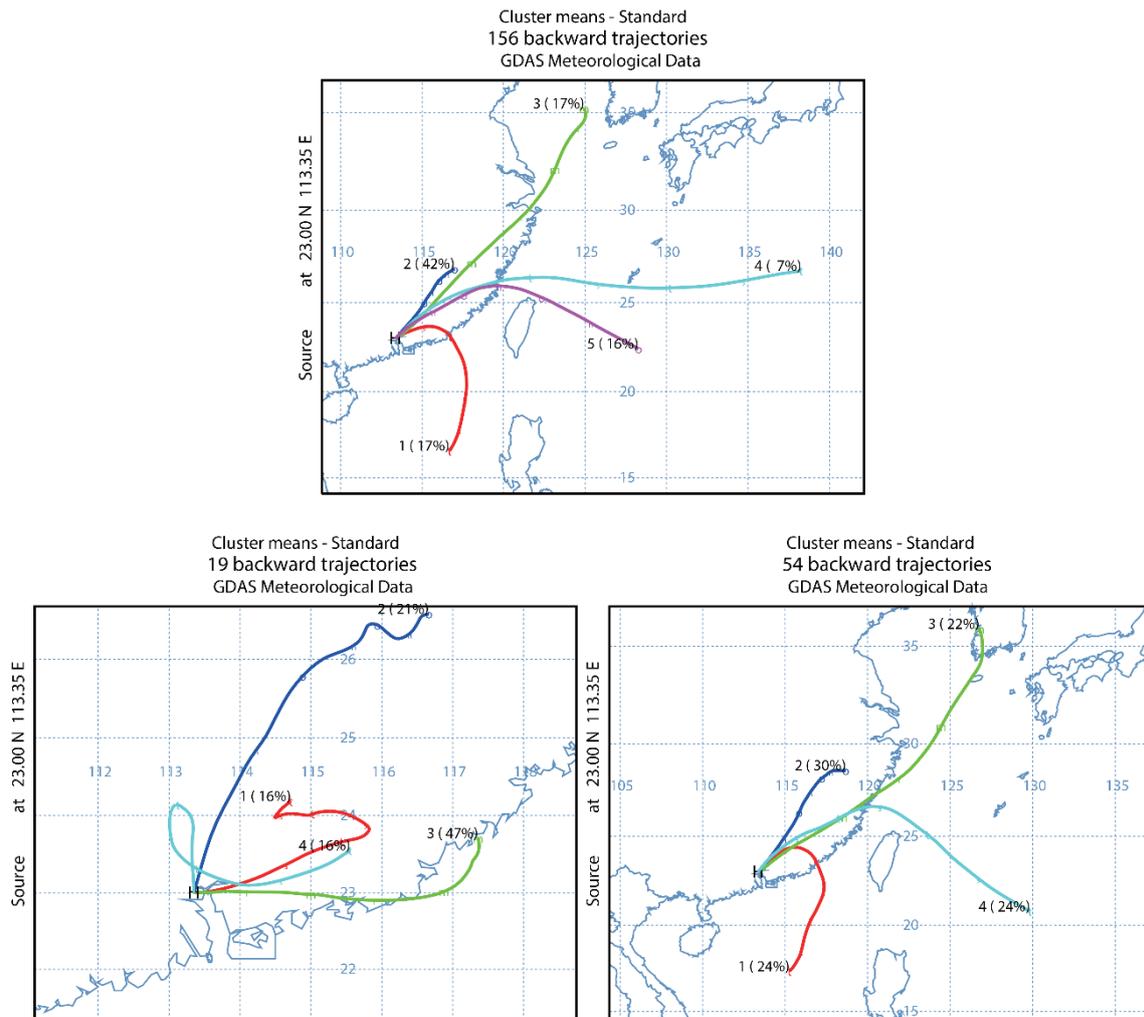


Figure R3: The major clusters for the 72-hour backward trajectory simulation for air masses arriving at the CAWNET Panyu site at a height of 1000 m. The upper panel shows the results throughout the whole observational period, while the lower panel on the left side shows the one during polluted days and the one on the right-hand side is for clean days.

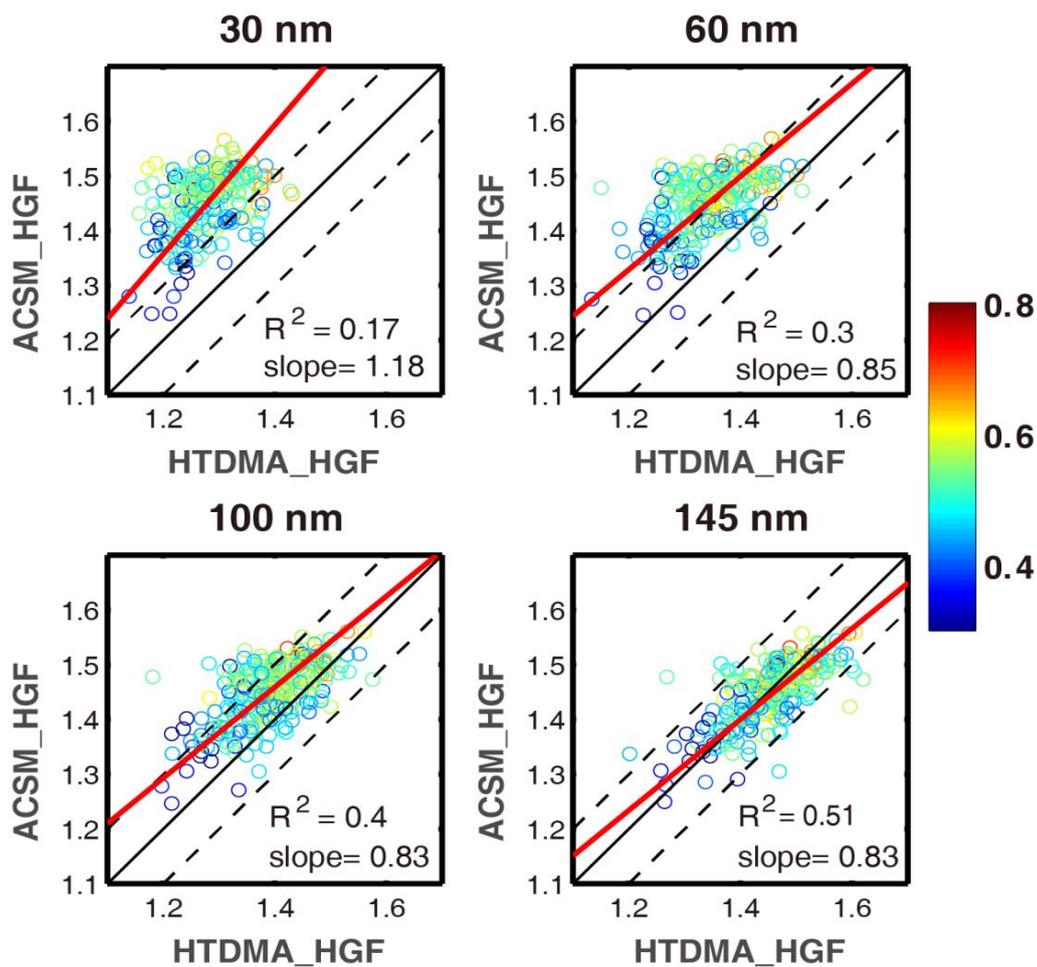


Figure R4: Closure study between the HTDMA-measured HGFs and the ACSM-derived HGFs. The black solid lines indicate the 1:1 line and the black dash lines represent $\pm 10\%$ deviation, while the red lines are the lines fitted to the data points. The color bar indicates the O:C ratio of the organic aerosol fraction.

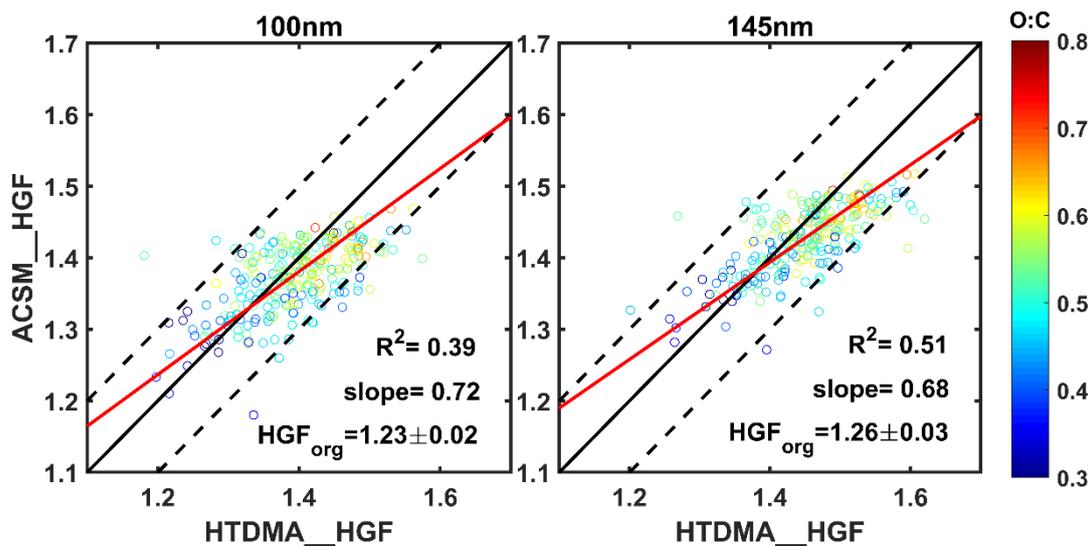


Figure R5: Closure study between the HTDMA-measured HGFs and the ACSM-derived HGFs assuming the average inorganic mass fraction of PM1 were about $25\% \pm 3\%$ and $16\% \pm 3\%$ higher and the average ammonium sulfate mass fraction of PM1 were about $25\% \pm 3\%$ and $16\% \pm 3\%$ lower than those 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.

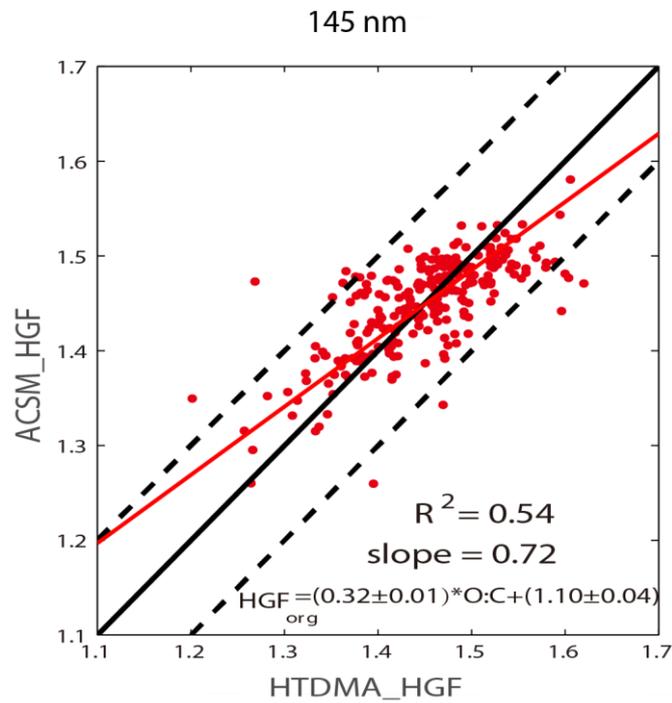


Figure R6: Closure analysis with the best fitting between the measured HGFs and the ACSM-derived ones using the O:C-dependent HGF_{org} . The assumption of size-dependent chemical composition of aerosols was considered to determine the ACSM-derived HGF. The equation is the achieved approximation for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

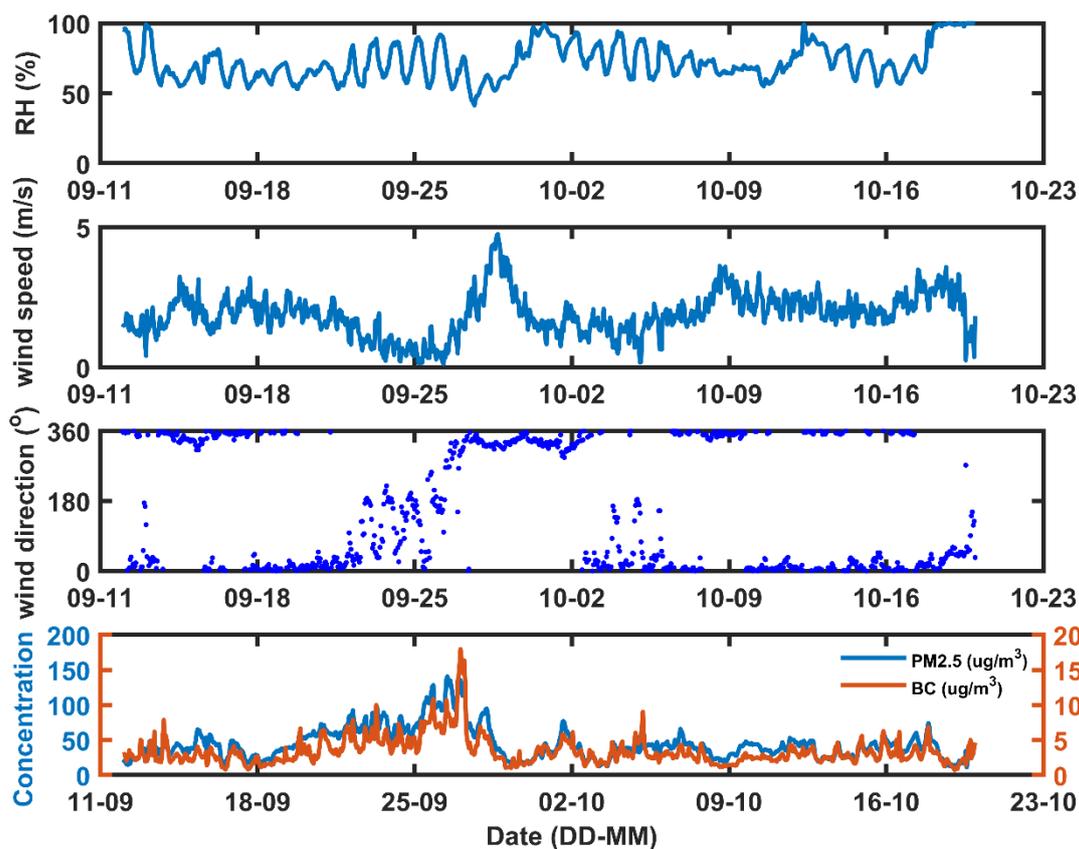


Figure R7: Time series for relative humidity (RH), wind speeds, wind directions and concentrations of PM2.5 as well as BC concentration (bottom panel).

References:

Aklilu, Y., Mozurkewich, M., Prenni, A. J., Kreidenweis, S. M., Alfarra, M. R., Allan, J. D., Anlauf, K., Brook, J., Leaitch, W. R., Sharma, S., Boudries, H. and Worsnop, D. R.: Hygroscopicity of particles at two rural, urban influenced sites during Pacific 2001: Comparison with estimates of water uptake from particle composition, *Atmos. Environ.*, 40(15), 2650–2661, doi:10.1016/j.atmosenv.2005.11.063, 2006.

Bahreini, R., Ervens, B., Middlebrook, A. M., Warneke, C., de Gouw, J. A., DeCarlo, P. F., Jimenez, J. L., Brock, C. A., Neuman, J. A., Ryerson, T. B., Stark, H., Atlas, E., Brioude, J., Fried, A., Holloway, J. S., Peischl, J., Richter, D., Walega, J., Weibring, P., Wollny, A. G. and Fehsenfeld, F. C.: Organic aerosol formation in urban and industrial plumes near Houston and Dallas, Texas, *J. Geophys. Res.*, 114(16), D00F16, doi:10.1029/2008JD011493, 2009.

Cai, M., Tan, H., Chan, C. K., Mochida, M., Hatakeyama, S., Kondo, Y., Schurman, M. I., Xu, H., Li, F., Shimada, K., Li, L., Deng, Y., Yai, H., Matsuki, A., Qin, Y. and Zhao, J.: Comparison of Aerosol Hygroscopicity, Volatility, and Chemical Composition between a Suburban Site in the Pearl River Delta Region and a Marine Site in Okinawa, *Aerosol Air Qual. Res.*, 17(12), 3194–3208, doi:10.4209/aaqr.2017.01.0020, 2017.

Cai, M., Tan, H., Chan, C. K., Qin, Y., Xu, H., Li, F., Schurman, M. I., Li, L. and Zhao, J.: The size resolved cloud condensation nuclei (CCN) activity and its prediction based on aerosol hygroscopicity and composition in the Pearl Delta River (PRD) Region during wintertime 2014, *Atmos. Chem. Phys. Discuss.*, (June), 1–55, doi:10.5194/acp-2018-339, 2018.

Carrico, C. M., Kreidenweis, S. M., Malm, W. C., Day, D. E., Lee, T., Carrillo, J., McMeeking, G. R. and Collett Jr., J. L.: Hygroscopic growth behavior of a carbon-dominated aerosol in Yosemite National Park, *Atmos. Environ.*, 39(8), 1393–1404, doi:10.1016/j.atmosenv.2004.11.029, 2005.

Cheng, Y. F., Wiedensohler, A., Eichler, H., Heintzenberg, J., Tesche, M., Ansmann, A., Wendisch, M., Su, H., Althausen, D. and Herrmann, H.: Relative humidity dependence of aerosol optical properties and direct radiative forcing in the surface boundary layer at Xinken in Pearl River Delta of China: An observation based numerical study, *Atmos. Environ.*, 42(25), 6373–6397, doi:10.1016/j.atmosenv.2008.04.009, 2008.

Chen, J., Budisulistiorini, S. H., Itoh, M., Lee, W.-C., Miyakawa, T., Komazaki, Y., Yang, L. D. Q. and Kuwata, M.: Water uptake by fresh Indonesian peat burning particles is limited by water-soluble organic matter, *Atmos. Chem. Phys.*, 17(18), 11591–11604, doi:10.5194/acp-17-11591-2017, 2017.

Cheng, Y., Zheng, G., Wei, C., Mu, Q., Zheng, B., Wang, Z., Gao, M., Zhang, Q., He, K., Carmichael, G., Poschl, U., and Su, H.: Reactive nitrogen chemistry in aerosol water as a source of sulfate during haze events in China, *Science Advances*, 2, 10.1126/sciadv.1601530, 2016.

Enroth, J., Mikkilä, J., Németh, Z., Kulmala, M. and Salma, I.: Wintertime hygroscopicity and volatility of ambient urban aerosol particles, *Atmos. Chem. Phys.*, 18(7), 4533–4548, doi:10.5194/acp-18-4533-2018, 2018.

Good, N., Topping, D. O., Allan, J. D., Flynn, M., Fuentes, E., Irwin, M., Williams, P. I., Coe, H. and McFiggans, G.: Consistency between parameterisations of aerosol hygroscopicity and CCN activity during the RHaMBLe discovery cruise, *Atmos. Chem. Phys.*, 10(7), 3189–3203, doi:10.5194/acp-10-3189-2010, 2010.

Gunthe, S. S., Rose, D., Su, H., Garland, R. M., Achtert, P., Nowak, A., Wiedensohler, A., Kuwata, M., Takegawa, N., Kondo, Y., Hu, M., Shao, M., Zhu, T., Andreae, M. O., and Pöschl, U.: Cloud condensation nuclei (CCN) from fresh and aged air pollution in the megacity region of Beijing, *Atmos. Chem. Phys.*, 11, 11023-11039, <https://doi.org/10.5194/acp-11-11023-2011>, 2011.

Gysel, M., McFiggans, G. B. and Coe, H.: Inversion of tandem differential mobility analyser (TDMA) measurements, *J. Aerosol Sci.*, 40(2), 134–151, doi:10.1016/j.jaerosci.2008.07.013, 2009.

Gysel, M., Weingartner, E., Nyeki, S., Paulsen, D., Baltensperger, U., Galambos, I. and Kiss, G.: Hygroscopic properties of water-soluble matter and humic-like organics in atmospheric fine aerosol, *Atmos. Chem. Phys.*, 4, 35–50, 2004.

Hansen, A. D. A., Rosen, H. and Novakov, T.: Real-time measurement of the absorption coefficient of aerosol particles, *Appl. Opt.*, 21(17), 3060, doi:10.1364/AO.21.003060, 1982.

Hong, J., Kim, J., Nieminen, T., Duplissy, J., Ehn, M., Äijälä, M., Hao, L. Q., Nie, W., Sarnela, N., Prisle, N. L., Kulmala, M., Virtanen, A., Petäjä, T. and Kerminen, V.-M.: Relating the hygroscopic properties of submicron aerosol to both gas- and particle-phase chemical composition in a boreal forest environment, *Atmos. Chem. Phys.*, 15(20), 11999–12009, doi:10.5194/acp-15-11999-2015, 2015.

Jimenez, J. L., Campuzano-Jost, P., Day, D.A., Nault, B.A., Schroder, J.C., Cubison, M.J.: Frequently Asked Questions for AMS Data Users, http://cires.colorado.edu/jimenez-group/wiki/index.php?title=FAQ_for_AMS_Data_Users, accessed month-year", 2018.

Kuwata, M., Zorn, S. R. and Martin, S. T.: Using Elemental Ratios to Predict the Density of Organic Material Composed of Carbon, Hydrogen, and Oxygen, *Environ. Sci. Technol.*, 46(2), 787–794, doi:10.1021/es202525q, 2012.

Liu, P. F., Zhao, C. S., Göbel, T., Hallbauer, E., Nowak, A., Ran, L., Xu, W. Y., Deng, Z. Z., Ma, N., Mildenberger, K., Henning, S., Stratmann, F. and Wiedensohler, A.: Hygroscopic properties of aerosol

particles at high relative humidity and their diurnal variations in the North China Plain, *Atmos. Chem. Phys.*, 11(7), 3479–3494, doi:10.5194/acp-11-3479-2011, 2011.

Ma, N., Zhao, C. S., Nowak, A., Müller, T., Pfeifer, S., Cheng, Y. F., Deng, Z. Z., Liu, P. F., Xu, W. Y., Ran, L., Yan, P., Göbel, T., Hallbauer, E., Mildenerger, K., Henning, S., Yu, J., Chen, L. L., Zhou, X. J., Stratmann, F. and Wiedensohler, A.: Aerosol optical properties in the North China Plain during HaChi campaign: an in-situ optical closure study, *Atmos. Chem. Phys.*, 11(12), 5959–5973, doi:10.5194/acp-11-5959-2011, 2011.

Massling, A., Stock, M., Wehner, B., Wu, Z. J., Hu, M., Brüggemann, E., Gnauk, T., Herrmann, H. and Wiedensohler, A.: Size segregated water uptake of the urban submicrometer aerosol in Beijing, *Atmos. Environ.*, 43(8), 1578–1589, doi:10.1016/j.atmosenv.2008.06.003, 2009.

McFiggans, G., Artaxo, P., Baltensperger, U., Coe, H., Facchini, M. C., Feingold, G., Fuzzi, S., Gysel, M., Laaksonen, A., Lohmann, U., Mentel, T. F., Murphy, D. M., O'Dowd, C. D., Snider, J. R. and Weingartner, E.: The effect of physical and chemical aerosol properties on warm cloud droplet activation, *Atmos. Chem. Phys.*, 6(9), 2593–2649, doi:10.5194/acp-6-2593-2006, 2006.

Pajunoja, A., Lambe, A. T., Hakala, J., Rastak, N., Cummings, M. J., Brogan, J. F., Hao, L., Paramonov, M., Hong, J., Prisle, N. L., Malila, J., Romakkaniemi, S., Lehtinen, K. E. J., Laaksonen, A., Kulmala, M., Massoli, P., Onasch, T. B., Donahue, N. M., Riipinen, I., Davidovits, P., Worsnop, D. R., Petäjä, T. and Virtanen, A.: Adsorptive uptake of water by semisolid secondary organic aerosols, *Geophys. Res. Lett.*, 42(8), 3063–3068, doi:10.1002/2015GL063142, 2015.

Ouimette, J. R. and Flagan, R. C.: The extinction coefficient of multicomponent aerosols, *Atmos. Environ.*, 16, 2405–2419, 1982.

Sloane, C. S.: Optical properties of aerosols – Comparison of measurements with model calculations, *Atmos. Environ.*, 17, 409–416, 1983.

Suda, S. R., Petters, M. D., Yeh, G. K., Strollo, C., Matsunaga, A., Faulhaber, A., Ziemann, P. J., Prenni, A. J., Carrico, C. M., Sullivan, R. C. and Kreidenweis, S. M.: Influence of Functional Groups on Organic Aerosol Cloud Condensation Nucleus Activity, *Environ. Sci. Technol.*, 48(17), 10182–10190, doi:10.1021/es502147y, 2014.

Swietlicki, E., Hansson, H. C., Hämeri, K., Svenningsson, B., Massling, A., McFiggans, G., McMurry, P. H., Petäjä, T., Tunved, P., Gysel, M., Topping, D., Weingartner, E., Baltensperger, U., Rissler, J., Wiedensohler, A. and Kulmala, M.: Hygroscopic properties of submicrometer atmospheric aerosol particles measured with H-TDMA instruments in various environments—a review, *Tellus B Chem. Phys. Meteorol.*, 60(3), 432–469, doi:10.1111/j.1600-0889.2008.00350.x, 2008.

Tan, H., Yin, Y., Gu, X., Li, F., Chan, P. W., Xu, H., Deng, X. and Wan, Q.: An observational study of the hygroscopic properties of aerosols over the Pearl River Delta region, *Atmos. Environ.*, 77, 817–826, doi:10.1016/j.atmosenv.2013.05.049, 2013.

Topping, D. O., McFiggans, G. B. and Coe, H.: A curved mult-component aerosol hygroscopicity model framework: Part 1 - Inorganic compounds, *Atmos. Chem. Phys.*, 5, 1205–1222, 2005.

Wu, D., Mao, J., Deng, X., Tie, X., Zhang, Y., Zeng, L., Li, F., Tan, H., Bi, X., Huang, X., Chen, J. and Deng, T.: Black carbon aerosols and their radiative properties in the Pearl River Delta region, *Sci. China Ser. D Earth Sci.*, 52(8), 1152–1163, doi:10.1007/s11430-009-0115-y, 2009.

Wu, Z. J., Poulain, L., Henning, S., Dieckmann, K., Birmili, W., Merkel, M., van Pinxteren, D., Spindler, G., Müller, K., Stratmann, F., Herrmann, H. and Wiedensohler, A.: Relating particle hygroscopicity and CCN activity to chemical composition during the HCCT-2010 field campaign, *Atmos. Chem. Phys.*, 13(16), 7983–7996, doi:10.5194/acp-13-7983-2013, 2013.

Wu, Z. J., Zheng, J., Shang, D. J., Du, Z. F., Wu, Y. S., Zeng, L. M., Wiedensohler, A., Hu, M. and Wu, Z.: Particle hygroscopicity and its link to chemical composition in the urban atmosphere of Beijing, China, during summertime, *Atmos. Chem. Phys.*, 16, 1123–1138, doi:10.5194/acp-16-1123-2016, 2016.

Ye, X., Tang, C., Yin, Z., Chen, J., Ma, Z., Kong, L., Yang, X., Gao, W. and Geng, F.: Hygroscopic growth of urban aerosol particles during the 2009 Mirage-Shanghai Campaign, *Atmos. Environ.*, 64, 263–269, doi:10.1016/j.atmosenv.2012.09.064, 2013.

Yeung, M. C., Lee, B. P., Li, Y. J. and Chan, C. K.: Simultaneous HTDMA and HR-ToF-AMS measurements at the HKUST supersite in Hong Kong in 2011, *J. Geophys. Res.*, 119(16), 9864–9883, doi:10.1002/2013JD021146, 2014.

Zhang, X., Ming, J., Li, Z., Wang, F. and Zhang, G.: The online measured black carbon aerosol and source orientations in the Nam Co region, Tibet, *Environ. Sci. Pollut. Res.*, 24(32), 25021–25033, doi:10.1007/s11356-017-0165-1, 2017.